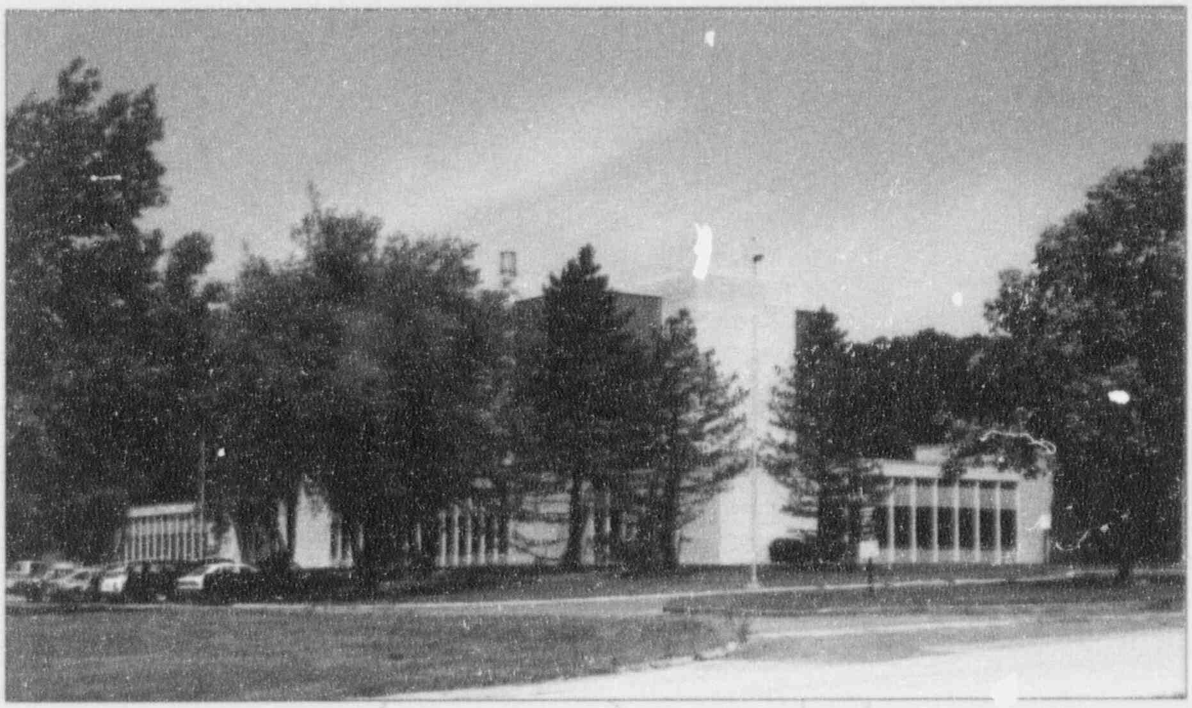
Enclosure To Comment No. 122

MU RESEARCH REACTOR

UNIVERSITY OF MISSOURI - COLUMBIA



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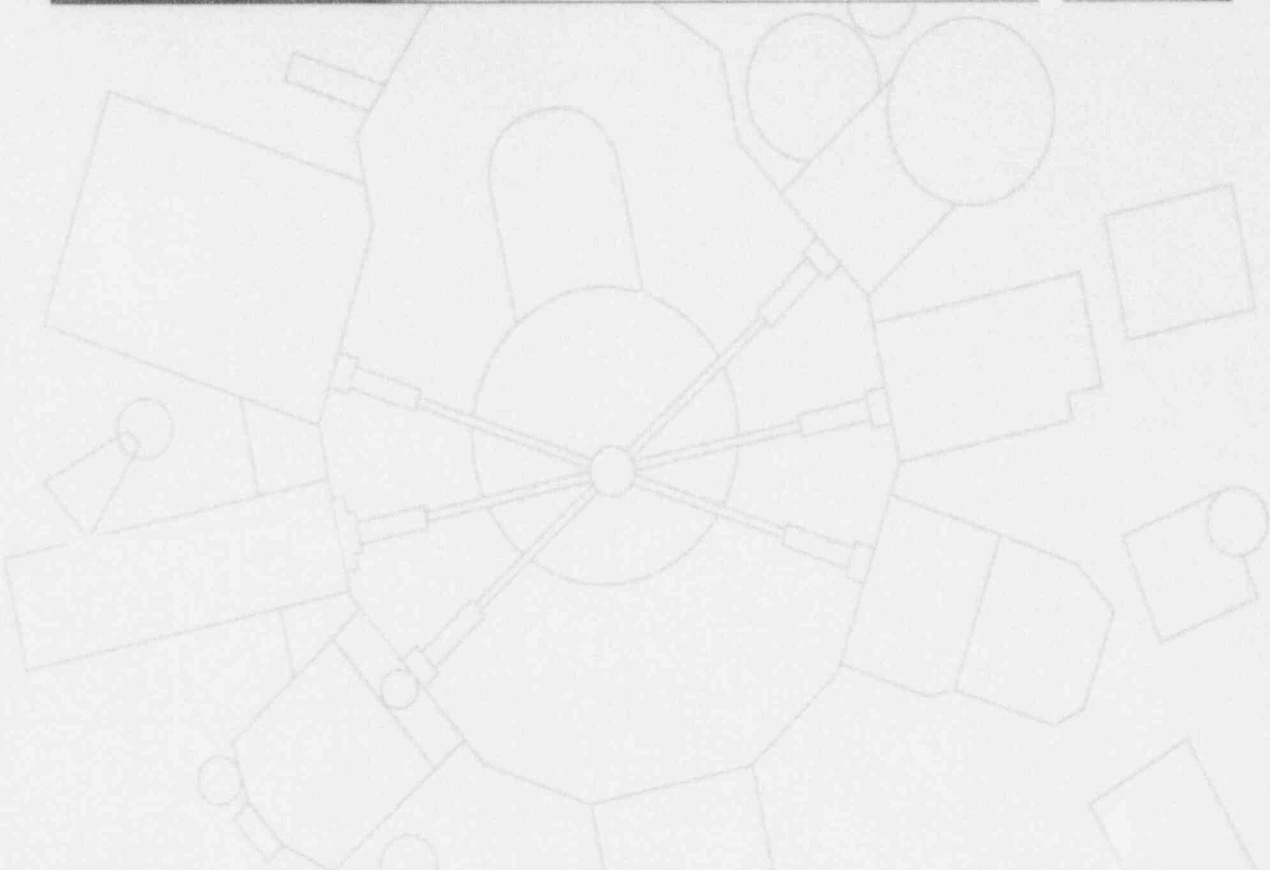

1992 ANNUAL REPORT

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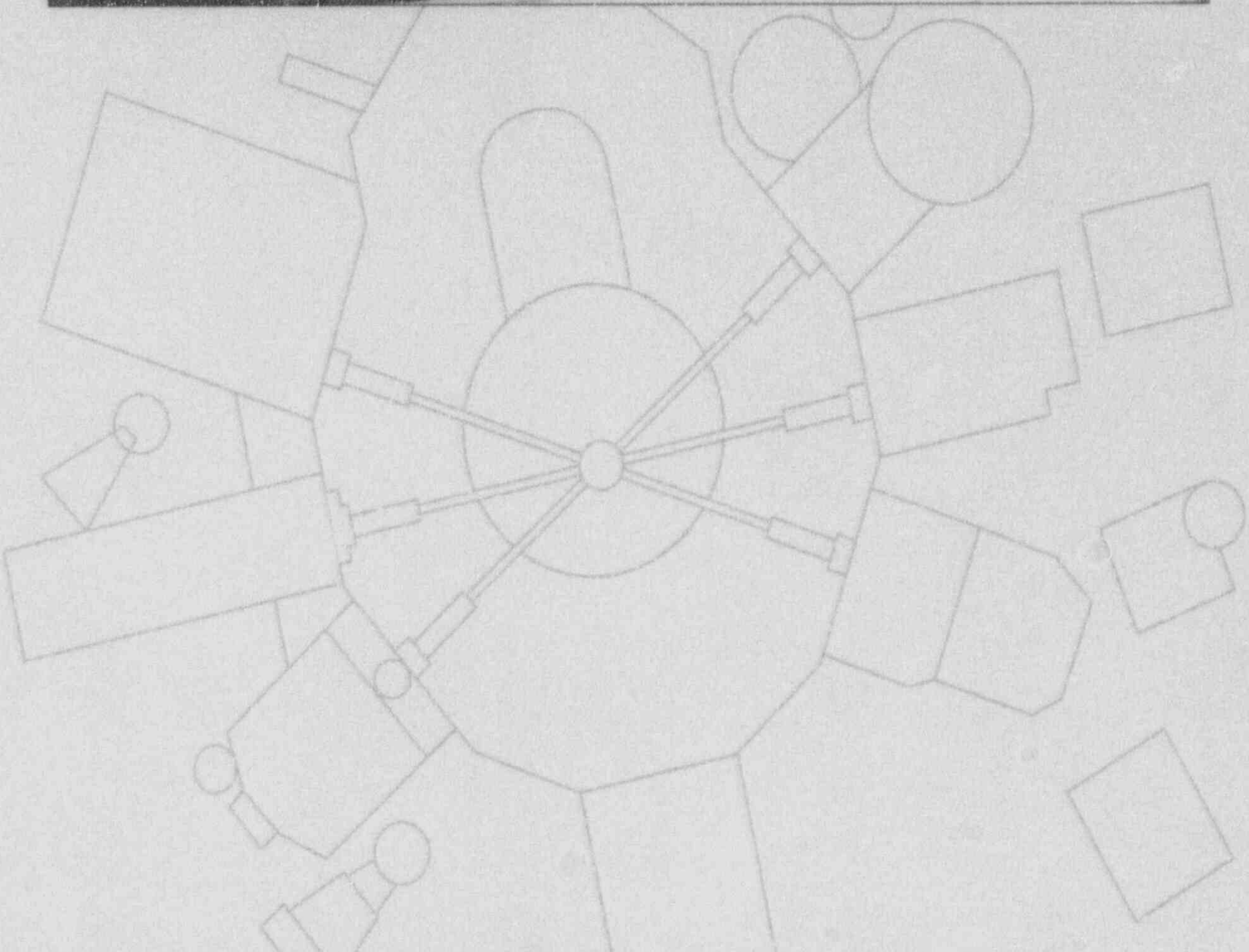

1992 ANNUAL REPORT

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PREFACE

October 13, 1991 marked the twenty-fifth anniversary of operation of the University of Missouri Research Reactor Center (MURR), the highest power and most versatile research reactor located on a university campus in the United States.

MURR was born in 1956 out of the farsighted vision of a Reactor Study Committee commissioned by University President Elmer Ellis and chaired by then MU Engineering Dean, Huber O. Croft. The members of this committee–Dean Croft, Dean Elmer Kiehl, Dean Vernon Wilson, and Physics Professor Newell Gingrich–recommended constructing a research reactor with a power in excess of one megawatt. In 1960 the committee selected Ardath Emmons as the first director. Dr. Emmons also led the design team that formalized the concept of building a light water moderated research reactor of innovative design including a central flux trap irradiation facility. Other irradiation positions for producing isotopes and other uses were provided for in the graphite reflector surrounding the center beryllium neutron reflector element. The reactor was planned initially to be five megawatts and to accommodate a later power upgrade to 10 megawatts. Six beamtubes were provided for neutron scattering and other types of research utilizing neutron beams. The design plan also called for an extensive laboratory building of 51,000 square feet associated with the reactor to contain radiological research laboratories and offices. Several of the laboratories were equipped with pneumatic tube remote irradiation facilities.

Construction of the research reactor and associated laboratory building began in 1963 with funding from both the Federal Government and the State of Missouri. It was completed and commenced operation in 1966. Power levels and operating schedules were gradually increased, and in 1969 the reactor began operating a 100 hour/week schedule at 5 megawatts. In 1974 the reactor power was increased to 10 megawatts, and in 1977 operation of the reactor for 155 hours per week was initiated. This schedule has been maintained since and represents an enviable on-line record of more than 90 per cent of available time over the past 15 years.

Over the intervening twenty-five plus years of operation of MURR, the number, quality, and breadth of reactor-based research programs have grown enormously. Much of this expansion occurred under the 14 years of leadership by Dr. Robert Brugger, MURR Director from 1974-1988. The number and scope of research programs which utilize the MURR Center or isotopes produced by the Reactor are unequaled by any other facility.

This annual report for fiscal year (FY) 1992 presents research and operational accomplishments of the reactor and its associated users and collaborators. As discussed later in this report, the year 1992 brought forth a number of new achievements as well a new organizational structure for the research groups. A MURR newsletter entitled *From Out of the Blue* was commissioned in May, edited by Ms. Ronita Dinger.

The format of this report consists of summary descriptions of the major programs, projects, and accomplishments in FY92. Publications, patents, talks, degrees awarded, and other information follow in tabular form. The intensive efforts of Ms. Christine Errante in editing and organizing this report and of Ms. Brenda Johnson for compiling the monthly reports and generating the appendices are gratefully acknowledged.

Note: Much of the above historical information about MURR was obtained from notes graciously provided by Professor Newell S. Gingrich, a member of the original Reactor Study Committee.

James J. Rhyne
Director, MU Research Reactor Center

FOREWORD

The University of Missouri Research Reactor Center (MURR) is a multi-disciplinary research and education center operated by the University of Missouri-Columbia (MU) and is located in Columbia, MO, USA. The Center provides MU with opportunities for research and graduate education in the neutron-related sciences and engineering that are unmatched at any other US university. The central focus of this research Center is a 10 MW light water moderated reactor that is the highest power university research reactor in the United States. The reactor has been in operation for 25 years (commencing October 13, 1966), and since 1976 has had a 156 hour per week operating schedule maintained more than 90% of the available time. MURR has a full-time staff of 112 including 21 PhD scientists, and additionally supports 40-50 students many of whom are engaged in graduate thesis research using the reactor or its isotope products. The reactor is a separate research Center, not part of any MU department, and reports to the Vice Provost for Research, Dr. J.D. Sheridan.

MURR has a three-fold mission of promoting basic and applied research in neutron-related science and engineering, providing an educational opportunity for students in these fields, and providing radiation and isotope production services for public and private consumers. The breadth and quality of the research programs, available facilities and equipment are comparable to those found in the US national laboratories.

The scientific programs at MURR span a broad spectrum of research disciplines and techniques and include archeometry, neutron and gamma-ray scattering, neutron interferometry, neutron activation analysis, human and animal nutrition, epidemiology and immunology, radiation effects, radioisotope studies, health physics, and nuclear engineering. The reactor research groups are made up of both MURR staff and researchers at the University of Missouri System's (UM) four campuses. Most of the programs also have strong ties to other universities, and national and industrial laboratories. MURR takes its role very seriously as one of the premier reactor facilities providing educational opportunities for students interested in programs related to neutron sciences and engineering.

The operation of the reactor is under the direction of the Reactor Operations Manager and operations group who also handle in-pool and flux-trap sample irradiation procedures. This group has close association with the Facilities Operations Manager and group responsible for improvements and maintenance of the reactor and associated instrumentation facilities. Within Facilities Operations are engineering design and mechanical and electronics shops. The Instrument Development group provides engineering of new electronic instrumentation. Computer software and hardware support, including the facility-wide computer network and central computer, are under the direction of the Computer Development group. Health physics surveillance and support for the diverse irradiation and laboratory facilities at MURR is provided by a Health Physics Manager and group.

The Center has a Service Applications group, headed by the Assistant Reactor Director for Technical Applications (a new title this year), that provides radiation services and produces a wide range of radioisotopes for educational institutions, state and federal agencies, and industrial and commercial concerns. Income derived from this service work provides over 50% of the support for the education and research programs within the MURR Center.

The organization of the research groups at MURR was restructured during this past year. What had been five groups were reorganized into four research program areas encompassing 10 research groups. As shown in the organization chart on page 5, the programs are Neutron Materials Science, Biomedical, Nuclear Analysis, and Reactor Nuclear Engineering. The group names reflect the scientific focus of the researchers rather than the techniques or specific instruments used by the group members. In addition to delineating more clearly the scientific foci of the groups, the reorganization provided added opportunities for outstanding scientists to assume leadership roles, which for the first time includes as group leaders MU faculty who have active reactor-based research, but are not directly employed by MURR. A list of individuals in the groups is given at the beginning of each group's section of this report. MURR scientific staff hold titles of Group Leader, Research Scientist, and Senior Research Scientist. Active participants in MURR research programs from other MU departments or other universities have titles (new this past year) of Research Investigator and Senior Research Investigator.

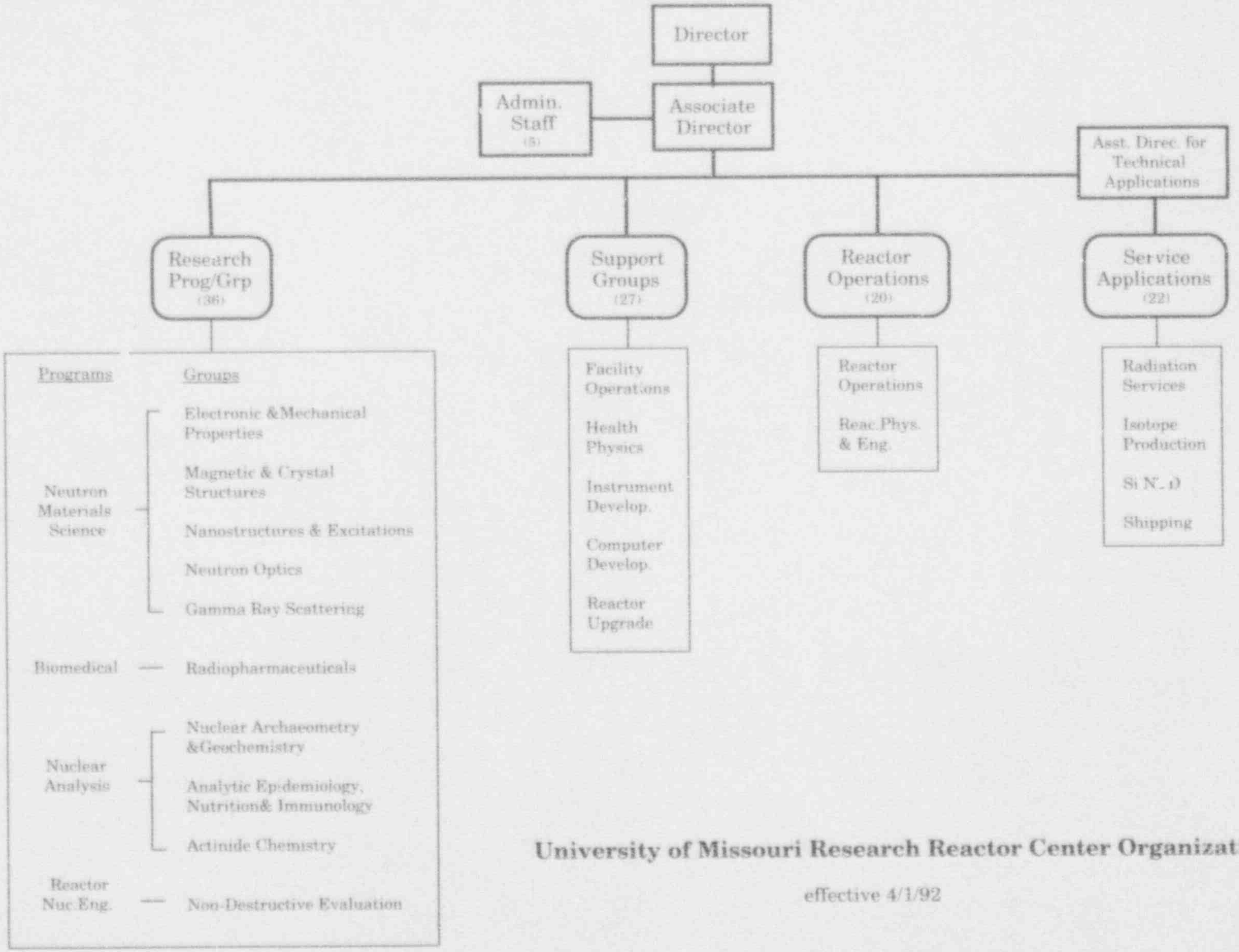
The operation, oversight functions, and enhancements to MURR facilities and programs are reviewed and approved by a Reactor Advisory Committee consisting of members from MU departments, MURR, other UM campuses, and outside members. This committee, advisory to the MURR director, has subcommittees for Reactor Safety, Isotope Use, Reactor Service, Gemstone License, Procedure Review, and Reactor Research.

The MURR Center is in the planning and development stages for a major three-part upgrade of its facilities including a laboratory and experimental building addition with a 20 x 30 m guide hall, a new fuel design, an increase in its power level, and a cold neutron source. A schematic design plan for the expanded research building was completed during this year. When finished, this building will alleviate a critical space limitation that is constraining further major program expansion in MURR's educational and research functions. As part of a continuing interim solution to the space shortage at MURR, a third temporary office building was installed to create 11 new offices and a conference room.

The MU Research Reactor Center represents a major research and education resource for the University, the State of Missouri, and the nation. It is the goal of MURR is to provide MU faculty, researchers, and outside collaborators access to and opportunities for new types of research activities and facilities not readily available at other institutions. It is a facility to enhance the international reputation of the University, and to provide the catalyst for MU to be the leader in the education of future generations of neutron scientists and engineers.

DIRECTOR'S OFFICE

James J. Rhyne, Director
J. Charles McKibben, Associate Director
William F. Reilly, Assistant Director
Christine M. Errante, Administrative Associate II
Ronita A. Dinger, Information Specialist
Rita E. Bonney, Administrative Assistant
Brenda S. Johnson, Senior Secretary
Dana A. Ward, Senior Receptionist



University of Missouri Research Reactor Center Organization

effective 4/1/92

STAFF RECOGNITION

A reception was held in the MURR lobby on April 24 to honor the nine MURR staff members who reached or will reach University employment milestones during the calendar year 1992. These MURR Center employees were also formally recognized at a Campus-wide Staff Recognition ceremony held April 7 in Jesse Auditorium.

FIVE YEARS	
G. Lawrence Jesse	Service Applications
Brenda Johnson	Directors Office
Leonard Manson III	Service Applications
Paul Miller	Service Applications
Leslie Powell	Health Physics
William Reilly	Directors Office
TEN YEARS	
Anthony Schoone	Reactor Operations
FIFTEEN YEARS	
Charles Anderson	Reactor Operations
Ronald Berliner	Instrument Development/ Neutron Materials Sciences

At the reception, Director Jim Rhyne expressed appreciation to all MURR staff for their valuable contributions to the MURR Center.

RESEARCH GROUPS AND ACTIVITIES

July 1991 - June 1992

- 219 faculty and 110 graduate students from 50 departments within the University of Missouri System, and 98 departments from 73 other universities, in addition to 24 governmental and 24 industrial laboratories performed research involving the MURR Center

- 104 journal publications from University of Missouri and other universities resulted from MURR research

- 23 invited and 47 contributed papers were presented at professional meetings based on research at MURR

- 58 grant/contract proposals were awarded or are ongoing during the year for MURR research

BIOMEDICAL PROGRAM

RADIOPHARMACEUTICALS GROUP

RADIOPHARMACEUTICALS GROUP

PERSONNEL

Alan R. Ketring, Group Leader and Senior Research Scientist (Research Assistant Professor of Radiological Sciences)
Gary J. Ehrhardt, Senior Research Scientist (Research Assistant Professor of Radiological Sciences)
Wynn Volkert, Senior Research Investigator (MURR) and Professor of MU Radiological Sciences
Karl Kopicka, Visiting Professor (Nuclear Research Institute, Rez, Czechoslovakia)
Lynn M. Stringham, Senior Research Specialist
Renee J. Hammar, Senior Research Laboratory Technician
Kavita K. Katti, Senior Research Laboratory Technician
Melissa Evans-Blumer, Research Laboratory Technician
James P. Lanigan, Research Laboratory Technician (transferred to Service Applications August 1991)
Anita R. Maves, Senior Secretary

FACULTY ASSOCIATES

Robert M. Brugger, Professor of MU Nuclear Engineering
Rodney Curtis, Resident of MU Surgery
Delbert E. Day, Professor of UMR Ceramic Engineering
Richard A. Holmes, Professor of MU Radiological Sciences and Chief of Nuclear Medicine, Harry S Truman Veterans Administration Hospital
Silvia S. Jurisson, Assistant Professor of MU Chemistry
Kattesh V. Katti, Research Assistant Professor of MU Radiological Sciences
Jimmy C. Lattimer, Associate Professor of MU Veterinary Medicine
Stanley E. Manahan, Professor of MU Chemistry
Jerome W. Olaughlin, Associate Professor of MU Chemistry
Gilbert Ross Jr, Professor of MU Surgery
Elmer O. Schlemper, Professor and Chair of MU Chemistry

For the past decade the MURR Radiopharmaceuticals group (formerly Radioisotope Applications) has been working with scientists from the University of Missouri, other universities and private industry to develop the use of reactor-produced radioisotopes for medical therapy. This research has led to developments in the areas of labeled microspheres, bone tumor-seeking agents, and high-specific-activity isotopes for therapy using labeled monoclonal antibodies. Nearly one thousand experimental patient treatments have now been completed using radioisotopes produced at MURR by the Radiopharmaceuticals (RP) group. Our collaboration with the departments of Radiology and Chemistry has resulted in the establishment of the Center for Radiological Research in facilities near the Ellis Fischel Cancer Center and a Radiopharmaceutical Chemistry Minor for MA and PhD Chemistry students.

MICROSPHERES FOR THE TREATMENT OF CANCER AND ARTHRITIS MURR and UM scientists have been involved in the development of glass, activatable microspheres for the treatment of liver cancer since the early 1980s. These microspheres are tiny glass beads slightly larger than the diameter of the smallest blood vessels or capillaries. When injected by catheter into the artery which carries blood to the liver tumor, the microspheres are trapped in the capillary bed, where they deliver a high localized radiation dose to the cancerous tissue.

Early work in collaboration with Theragenics Corporation produced Y-90 TheraSphere®, now approved for general use in Canada. More recent research focuses on developing Sm-153-glass microspheres for the presurgical treatment of kidney cancer and Re-188/Re-186 labelled protein microspheres for the treatment of rheumatoid arthritis (see **PRACTICAL RADIOTHERAPY FOR RENAL CELL CANCER**). In the latter treatment, called radiation synovectomy, radiolabelled protein microspheres appropriately sized for uptake by the inflamed synovial membrane are injected directly into the joint.

In all of these treatments, it is important to minimize or eliminate the leakage of the radioisotopes from the target tissue to other normal tissues until the radioisotope decays. This can be accomplished by the use of non-biodegradable glass microspheres (as in the case of the liver and kidney cancer treatments) or by using short lived radioisotopes such as Re-188 ($t_{1/2}$ = 17 hours) that will decay before any significant leakage occurs (as in the case of biodegradable protein microspheres for the treatment of arthritis). We are also studying glass compositions with other isotopes and biodegradable glass formulas for the treatment of cancer, arthritis and other diseases.

BONE CANCER TREATMENTS Metastatic bone cancer affects approximately 200,000 patients in the US each year, primarily by spreading from other common cancer sites such as lung, breast and prostate. Since the early 1980s MURR scientists in the RP group have collaborated with University of Missouri scientists and others outside the University to develop agents for treating the intense pain associated with metastatic bone cancer. We have supplied and continue to produce Re-186 for the pain palliation agent Re-186-HEDP (hydroxyethylene-diphosphonate), first developed at the University of Cincinnati and currently under evaluation as a potential drug product by Mallinckrodt Medical (see **Re-186 AND Re-188 HEDP CLINICAL TRIALS OF A NEW BONE CANCER AGENT**). Trials are also underway using MURR-supplied Re-188 as Re-188-HEDP for bone cancer patients at the University of Cincinnati.

The RP group was also instrumental in developing Sm-153 EDTMP (ethylenediamine-tetramethylenephosphonate) at MU, in collaboration with scientists in Chemistry, Radiology, Veterinary Medicine and School of Medicine with support from the Dow Chemical Company. Both Re-186-HEDP and Sm-153 EDTMP have demonstrated effectiveness in relieving bone cancer pain and greatly reducing or removing the need for narcotic analgesics in about 80% of the patients treated in clinical trials. These agents are currently in the final phases of clinical testing at numerous sites in the US, which are sponsored by Mallinckrodt and Dow.

During the course of this work, remote processing facilities were designed and built at MURR for the safe handling of Curie quantities of beta-emitting radioisotopes. This facility and our extensive experience with Sm-153 have enabled us to develop other rare-earth radioisotopes, such as Ho-166 and Lu-177, for potential use in new therapeutic radiopharmaceuticals.

HIGH-SPECIFIC ACTIVITY RADIOISOTOPES FOR RADIOIMMUNOTHERAPY The development of monoclonal antibodies has opened new horizons for guiding radioisotopes to tumors with remarkable specificity. The need for high specific-activity radioisotopes to label these antibodies has posed new challenges. MURR and the NeoRx Corporation have combined their efforts to find new methods for producing Re-186 and Re-188 for this application. Initially centered on 17-hour Re-188 from a W-188/Re-188 gel generator invented at MURR (see **NEW PEROXIDE PROCESS FOR PRODUCTION OF W-188/Re-188 AND Mo-99/Tc-99m GEL GENERATORS**), this work has evolved to the use of 3-4 Ci/mg Re-186 produced at MURR by newly-developed targetry and methods. This Re-186 is used at four sites in the US for attachment to monoclonal antibodies and subsequent administration to patients with colon, small-cell lung, and ovarian cancers. Over 100 patient treatments have been performed to date, with the best results observed in women with ovarian cancer. High-specific-activity isotopes with appropriate half-lives and emissions are vital for radioimmunotherapy and the reactors which can produce them are few; the potential in this area provides one of the major justifications for the planned power upgrade of the MURR.

CENTER FOR RADIOLOGIC RESEARCH (CRR)

The potential significance for radioimmunotherapy using labeled monoclonal antibodies could be enhanced by improved stability of the label and by more rapid clearance from nontarget tissues. These challenges have led us to study new labeling techniques for reactor-produced radioisotopes. Limited space at MURR has forced us to obtain laboratory and office space for this program in the Allton Building near the Ellis Fischel Cancer Center. The space (five laboratories and five offices) and the project are shared by the RP group at MURR and MU Radiology. Several new ligand systems are currently under investigation (see **POTENTIAL OF NEW RADIOPHARMACEUTICALS FROM NOVEL PHOSPHORUS-NITROGEN LIGANDS**) that may enable significant advances in the labelling of monoclonal antibodies for radioimmunotherapy of malignancies.

Due to the small size of some of the chelates, these labeling techniques are also being examined for their potential to label receptor agents and small polypeptides which localize in cancer and other disease tissues. These agents may be useful in the diagnosis and treatment of disease. We are also developing magnetic resonance imaging (MRI) contrast agents by using this technology to prepare paramagnetic chemical compounds.

RADIOPHARMACEUTICAL CHEMISTRY MINOR

Adoption of a dedicated minor in Radiopharmaceutical Chemistry by the MU chemistry department has allowed seven chemistry graduate students to base their work at the MURR Center with the RP group. RP scientists and other researchers at the MURR are now involved in team teaching chemistry classes in the areas of radiochemistry and radiopharmaceutical chemistry. An RP scientist has also served as a co-advisor and co-project director for a medical physics student in the MU Nuclear Engineering Department doing research in improved brachytherapy seeds. RP scientists are active on graduate student committees and University committees, such as the Central Radiation Safety Committee, the Isotope Use Subcommittee, the Reactor Advisory Committee, and the Reactor Research Advisory Subcommittee.

The trend toward greater integration of the MURR Center into the education and research efforts of the MU campus has been clearly evident in the RP group over the past two years, setting the precedent for future interdisciplinary achievements at the University of Missouri.

PRACTICAL RADIOTHERAPY FOR RENAL CELL (KIDNEY) CANCER

Rodney Curtis, Gilbert Ross (MU Surgery), Jimmy Lattimer (MU Veterinary Medicine), Delbert Day (UMR Ceramic Engineering), Gary Ehrhardt, Jim Lanigan, Eric Volkert and Melissa Evans-Blumer (MURR)

Renal cell cancer of the kidney occurs in about 3% of new cases of adult cancer and frequently results in distant and often fatal metastases. Surgeons feel that surgical removal of the diseased kidney and accompanying tumor, while always necessary, often results in "shedding" of malignant cells into the general circulation due to disturbance of the tumor bed by the surgery. Thus it would be desirable to sterilize the tumor with radiation prior to surgery to minimize viability of shed cells and also possibly reduce the tumor in size and thus aid its surgical removal. Unfortunately, external beam teletherapy to the kidney is limited to 2,000-3,000 rads due to the need to avoid over-dosing other tissues in the abdomen, and renal cell cancer is rarely responsive to such low doses.

Intra-arterial, beta-emitting, chemically-durable microspheres offer a way to deliver substantially higher radiation doses to these tumors while sparing normal tissue. In a cooperative project between MURR, the MU School of Medicine (Division of Urology), UMR Ceramic Engineering, and the MU School of Veterinary Medicine, experiments have been conducted in which samarium-doped glass microspheres 10-20 microns in diameter have been irradiated to produce Sm-153 and then injected via super-selective catheter into the kidney in a set of 18 New Zealand white rabbits. The objective was to evaluate safety in ascending dose trials up to ~35,000 rads to the target kidney, with surgical removal of the kidney and evaluation of the animal for collateral damage to adjacent normal tissues. Animals were sacrificed and necropsied at intervals of from 4 to 14 months, with striking results. No clinical illness or clinical radiation effect was seen at any dose, while gross pathology changes were visible only in injected kidneys, primarily visible in reduced size and evidence of fibrosis. Histology showed abnormalities only in injected kidneys, being most prominent in the renal cortex and disappearing at the renal capsule. Severity of the changes was dose-related, with no changes observed in surrounding tissues such as bowel, liver, spleen, and contralateral kidney. These well-limited effects are due largely to the short-ranged beta and minimal gamma emission of Sm-153 (which is still sufficient to permit evaluation of the "patient" by conventional gamma camera imaging).

Evaluation of the next experimental step has led to the production of holmium-doped glass microspheres, as the short half life (26.8 hr) and energetic beta emissions of Ho-166 appear ideal for the larger kidneys in human renal cell cancer patients who will subsequently undergo surgery. The near one day half life of Ho-166 results in complete decay of the isotope in about a week, permitting prompt surgery without radiation risk to the surgeon. The longer maximum range (about 1 cm) of the beta particle from Ho-166 should scale appropriately to the larger size of human kidneys.

This therapy offers the hope of both aiding surgical treatment of renal cell cancer and potentially increasing the survival rate of these patients by lowering the occurence of distant metastases

Progress in this project was reported at the 1992 High Country Nuclear Medicine Conference (Society of Nuclear Medicine).

NEW PEROXIDE PROCESS FOR PRODUCTION OF W-188/Re-188 AND Mo-99/Tc-99m GEL GENERATORS USING (n,gamma) W-188 AND Mo-99

Gary J. Ehrhardt, Qixue Liang, Alan R. Ketring (MURR), Robert G. Wolfangel, David Pipes (Mallinckrodt Medical), Carl V. Hamby (New York Medical College)

Several years ago MURR scientists developed a new W-188/Re-188 radioisotope generator which could use low specific activity, 69.4-day W-188. [As W-188 must be made by double neutron capture on W-186, it is only available in relatively low specific activity.] The 17 hour Re-188 product was successfully evaluated as a radioimmunotherapy agent in mice by NeoRx Corporation, studied by researchers at Harvard Medical School and at MURR as a radiation synovectomy (rheumatoid arthritis) agent, and is being considered as a component of a bone cancer agent (see **Re-186 AND Re-188 HEDP CLINICAL TRIALS OF A NEW BONE CANCER AGENT**). Trials of this generator as a radioimmunotherapy agent have also been conducted at New York Medical College and by researchers in Italy.

A new process has been developed for production of the zirconium tungstate gels which minimizes tedious handling and should produce a more consistent stoichiometry. This process involves forming the peroxo complexes of W and Zr, which enables mixing of the two components at neutral pH without immediate precipitation. Formation of the gel is achieved by heating to decompose the peroxide complexes. This is continued to dryness, resulting in a free-flowing material which can be used to directly pack a generator column. This process also permits irradiation of W metal, avoiding problems with formation of insoluble "blue oxides," etc., encountered in irradiation of tungsten trioxide and sodium tungstate targets. The resulting gel behaves the same as previous W-188/Re-188 gel generators. Greatly improved apparatus for this process have been designed and found to cut the processing time from days to hours, while also minimizing handling time and hand dosage. A patent has been applied for on this new technology.

One of the exciting aspects of this peroxide process is that it has also been shown to be applicable to Mo-99/Tc-99m generators produced using (n,gamma) Mo-99. With the current severely restricted choice of suppliers for fission product Mo-99, great demand for this product, and ever-increasing costs associated with fission-product waste disposal, the stage may be set for a resurgence of (n,gamma) Mo-99 if it can be made equivalent to current generators. Gel generator technology provides this equivalence by permitting the loading of gram quanities of Mo onto small columns, using the fact that the column packing does not have to **adsorb** the target, but rather is made **from** the target.

A new method of calibrating generator columns has also been devised and tested successfully. This method involves counting of high energy gamma rays from Re-188 from outside the generator shielding before and after elution. In this method an efficiency calibration for the column is not required, as only a relative measurement of column activity is needed. Combined with a dose calibrator assay of the product Re-188, these data permit accurate assay of the W-188 content of the column. It has also been found that more highly enriched W-186 targets (99.7%) register accurately in dose calibrators, due to their much lower W-185 content (as observed with ~98% enriched targets). This should also increase the already high specific activity of Re-188 product, as much less stable product Re-185 should be present from the decay of W-185.

Re-186 AND Re-188 HEDP CLINICAL TRIALS OF A NEW BONE CANCER AGENT

Edward Deutsch (formerly at University of Cincinnati Chemistry, now at Mallinckrodt Medical, Inc.), Harry Maxon (University of Cincinnati) and Gary Ehrhardt (MURR)

For many years the MURR Center's Radiopharmaceutical group has supplied Re-186 radioisotope for collaborative chemical, animal, and then clinical studies of the bone cancer pain palliation agent Re-186 hydroxyethylenediphosphonate (HEDP). Re-186, which has a gamma energy similar to that of Tc-99m and is therefore highly imageable, has a therapeutic beta maximum energy of about 1 MeV and a half life of about 90 hours. As the active component of Re-186 HEDP, Re-186 has proven about 80% effective in Phase I and II trials. With the advent of practical W-188/Re-188 generators (from ORNL and MURR), interest has arisen as to whether Re-188 might prove a better isotope of rhenium for this agent.

Re-188 has a half life of 17 hours, a Tc-99m-like gamma emission, a beta energy over twice that of Re-186 (about 2.1 MeV maximum), and convenient availability from a long-lived generator. As rhenium HEDP is known to localize on the bone mineral spicules situated between microscopic sites of cancer cells, there is speculation that the greater effective cross-fire range of the betas from Re-188 may exert a stronger therapeutic effect. Clincial trials to assess toxicity and efficacy in bone cancer patients are being continued at the University of Cincinnati using Re-188 obtained by irradiation of Re-187 target materials at MURR. Significant improvements in rhenium targetry and glove box processing procedures made at MURR have made the routine supply of hundreds of mCi of Re-188 for this purpose possible without large investments in remote handling equipment. Initial results have been promising, exhibiting pain palliation in at least one patient at a much lower dose than expected.

A paper describing this work entitled *Rhenium-186 (Sn)HEDP for Treatment of Painful Osseous Metastases* (*JNM* **32**(10):1877-1881) and co-authored by Gary Ehrhardt was awarded the 1991 Journal of Nuclear Medicine Award of Merit at the 1992 Society of Nuclear Medicine Meeting. It is of note that another MURR scientist (and MU Chemistry alumnus), Alan R. Ketring, co-invented Re-186 (Sn)HEDP during postdoctoral studies at the University of Cincinnati. MURR Service Applications currently produces Re-186 for Phase III studies of this agent sponsored by Mallinckrodt Medical, Inc.

POTENTIAL OF NEW RADIOPHARMACEUTICALS FROM NOVEL PHOSPHORUS-NITROGEN LIGANDS

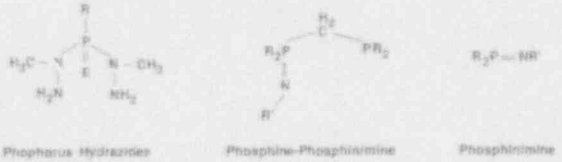
Kattesh V. Katti, Hermogenes Jimenez, Alan R. Ketring, Kavita K. Katti (MURR), Prahlad R. Singh and Wynn A. Volkert (MU Radiology)

The research conducted at the Center for Radiological Research (CRR) is focused on three major aspects:

1. Development of new ligand systems derived from novel phosphorus-nitrogen frameworks[1-9]
2. Understanding the fundamental coordination chemistry with the early and late transition metals in different oxidation states[3-4]
3. Application of this chemistry to metallic radioisotopes to develop a new genre of radiopharmaceuticals for therapeutic and diagnostic uses in nuclear medicine.[5-8]

MULTIFUNCTIONAL PHOSPHORUS HYDRAZIDES

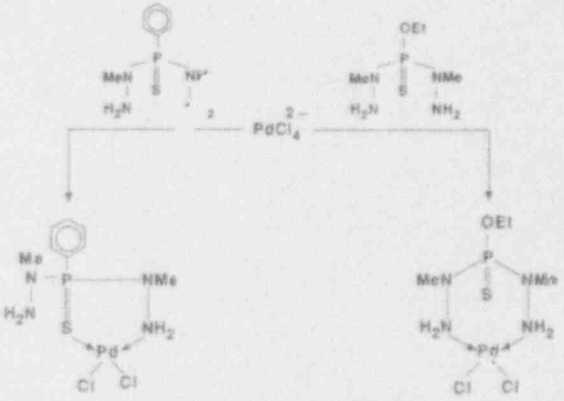
Three new classes of ligand systems developed at CRR are shown in Scheme 1. The synthetic strategies were aimed at the synthesis of all these new ligands in good yields and also to vary substituents at the phosphorus and nitrogen centers to afford extensive chemical flexibility. The phosphorus hydrazides, heterodifunctional phosphine-phosphinimines and the small molecule phosphinimines as outlined in Scheme 1 combine the ligating characteristics of hard nitrogen centers, phosphorus chalcogenides and soft π-acid phosphines to be used as versatile coordinating systems for both the electron poor (e.g., high-oxidation-state Re and Tc) and electron rich (e.g., Pd(II), Pt(II), Rh(III)) metals and their radioactive isotopes.



Phophorus Hydrazides Phosphine-Phosphinimine Phosphinimine

Scheme 1

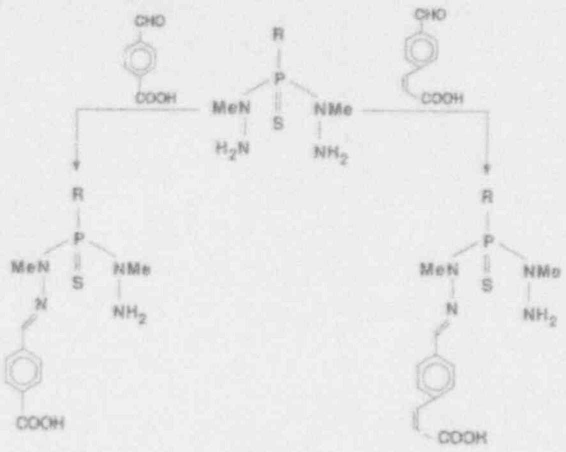
The phosphorus hydrazides showed trimodal features of coordination with rhenium as shown below. However, a distinct regioselectivity was observed in the reactions of these ligands with Pd(II) centers. For example, alkoxy substituents on phosphorus favored the bidentate coordination of the terminal hydrazides with the metal center (Scheme 2), whereas phenyl substituents showed chelation of the metal center through the P=S and one terminal hydrazide unit. Therefore, a systematic tuning of substituents on the phosphorus should allow the generation of specific regioselective features in this class of compound.



Scheme 2

As part of our ongoing research, we have explored the reactions of $^{109}PdCl_4^{2-}$ ($t_{1/2}$ = 13.5h; β_{max} = 1030 keV) with the phosphorus hydrazides. The phenyl and the ethyl phosphorus hydrazides as outlined in Scheme 2 produced ^{109}Pd chelates in high yields (85-90%) and these complexes have been found to be stable in aqueous media at pH 7 for 48h. We foresee further developments in the use of ^{109}Pd based radiopharmaceuticals in nuclear medicine if high specific activity ^{109}Pd can be produced.

The controlled Schiff base coupling reactions with the phosphorus hydrazides produce new bifunctional chelating agents (BFCA) in which one of the terminal hydrazides is functionalized with a benzoic acid unit (Scheme 3). The coordination chemistry of these BFCA's have clearly given important insight into the metal-ligand bonding features. The metal is bound to the P=S unit and one of the hydrazine centers (Figure 1) leaving the -COOH group free for attachment to proteins, peptides or other biomolecules.



Scheme 3

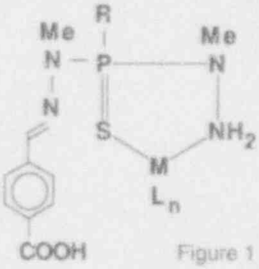


Figure 1

The BFCA was shown to readily complex ^{99m}Tc and this "preformed" bifunctional chelate (BFC) was conjugated to IgG (70% yield). The stability of both the ^{99m}Tc-BFC and the labeled protein was shown to be excellent. These studies provide strong evidence for the potential utility of multifunctional phosphorus hydrazide (BHP) based ligands to form BFCAs or BFCs for labeling proteins (or other biomolecules) with ^{99m}Tc or ^{186}Re/^{188}Re. Studies with various BHP derivatives will be initiated which may provide an important and unique method for producing ^{99m}Tc or ^{186}Re/^{188}Re complexes with desirable biolocalization properties.

HETERO-DIFUNCTIONAL PN-LIGANDS

Recent studies in our laboratory with ligands containing one phosphinimine group (or its isoelectronic analogue, phosphine oxide) as well as one phosphine group demonstrate their versatility in forming BFCs or BFCAs with ^{99m}Tc, ^{186}Re/^{188}Re, ^{109}Pd and other transition metal radionuclides. The presence of both the P=N and phosphine functionalities on the same molecule provide a vehicle to complex metals (both early and late transition metals) with a wider variety of oxidation states than ligands containing only one of these functionalities. The versatility of using this general ligand design has been demonstrated by the fact that several variations in the backbone structure are tolerated without significant compromise in complex stability. For example, we have recently shown that substitution of a bridging N-atom for the bridging C-atom in PCPN results in stable ^{99m}Tc and ^{186}Re complexes.

An important characteristic of these ligands is that they form complexes in a 1:1 ligand:metal ratio with Re and Pd despite the fact that they are only bidentate. In addition, the terminal N-atom can be derivatized with a side chain and still participate as a donor atom to form stable metal complexes. A particularly relevant example of this was demonstrated by results with an aryl isothiocyanate-PCPN (Figure 2) ligand system.

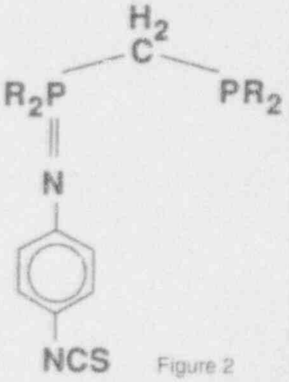
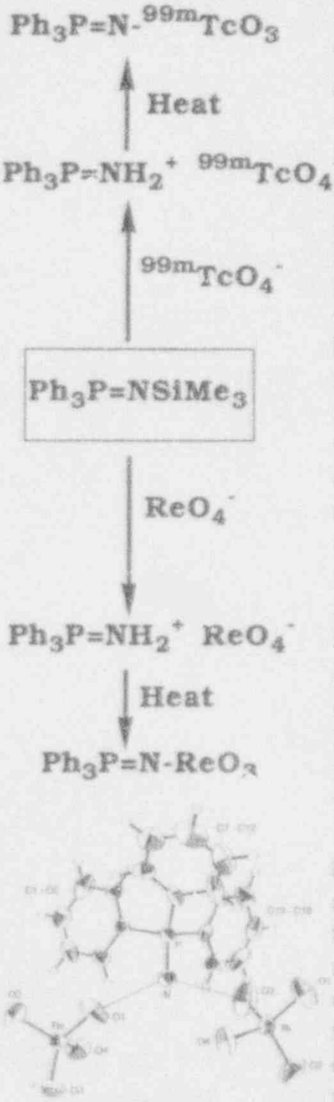


Figure 2

Studies with this BFCA show that it forms a very stable ^{99m}Tc-chelate in 95±2% yield. The ^{99m}Tc-PCPN BFC was conjugated to IgG [9]. No release of ^{99m}Tc was measurable after incubating this ^{99m}Tc-protein-conjugate at or near physiological pH for 24 hr. Clearly, these types of ligands hold immense potential for use in preparing stable radiolabeled bioconjugates. Structure-activity relationships (SARs) with appropriate derivatives will be essential to identify the specific structures that will produce radiolabeled agents with optimal properties for medical applications.

PHOSPHINIMINES

Simple phosphinimine ligands with the general structure shown in Figure 3, also exhibit the ability to form ^{99m}Tc and ^{188}Re complexes.



Scheme 4

These ligands are electronically very versatile as they can act as 1, 2, or 4 electron donors to transition metals resulting in M-N multiple covalent bonds where M is the metal. Using $Ph_3P{=}N - SiMe_3$, we have recently demonstrated that a compound with Re(VII) in a 1:1 metal:ligand ratio can be formed, i.e., $Ph_3P{=}N\text{-}ReO_3$, as determined by NMR, IR and X-ray crystallography (Scheme 4).

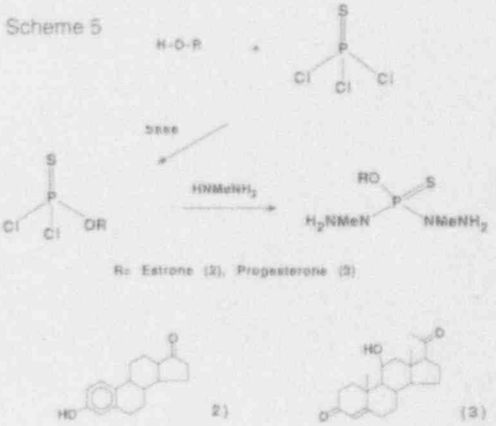
Studies with ^{99m}Tc and ^{188}Re indicate that small, neutral-lipophilic compounds with the same chromatographic properties as the Re complex are formed which infers structural similarity of the radiolabeled compound at tracer levels with the Re complex obtained at macroscopic levels. Further studies with this unique and exciting ligand system will be performed, however, the requirement that these ligands must be labeled in non-polar solvents may limit, to some extent their use with macromolecules. On the other hand, they may be particularly effective for use in the design of new ^{99m}Tc or ^{188}Re/^{186}Re receptor agents that are comparatively small compounds.

NEW CHEMICAL APPROACHES FOR LABELING RECEPTORS

The radiolabeling of receptors is an area of considerable current interest with significant clinical implications. Even though progress with radiolabeled receptors has been made, the utility of these agents in humans has been limited for a variety of reasons. For example, the chemical reactions employed to radiolabel a receptor frequently involve the incorporation of a ligating unit on the receptor backbone. Such chemical linking processes may abnormally modify the binding affinity, *in vivo* uptake and specificity of the receptors. Therefore, for widespread applicability of radiolabeled receptors, the complexities of ligating a receptor and its subsequent radiolabeling techniques must be carefully addressed.

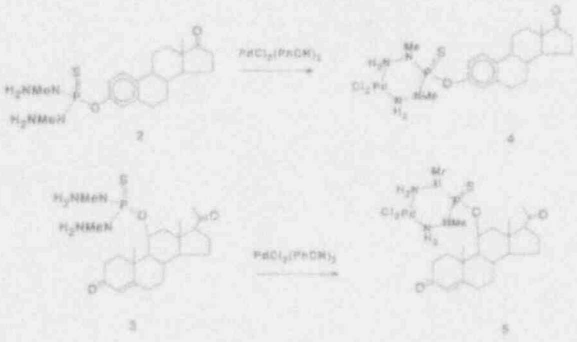
In the last six months, staff at the Center for Radiological Research have developed an elegant synthetic strategy to incorporate a new genre of multifunctional ligating units on model receptor backbones as shown in Scheme 5. The simple, straightforward and

Scheme 5



one pot synthetic discovery we have made (Scheme 5) is in sharp contrast to the multiple and often difficult reaction steps that are being used by other workers in the US and across the world to incorporate ligating units onto the receptors.

We have investigated the fundamental coordination chemistry of the progesterone and estrone functionalized phosphorous hydrazides in order to understand the ligating properties with the transition metals (Scheme 6). The complete ^{1}H, ^{31}P and ^{13}C NMR spectroscopic analysis and single crystal X-ray structures of model compounds have clearly revealed that Pd(II) is bound to the receptor functionalized phosphorus hydrazides in such a way that the receptor is not in itself involved in direct bonding interactions with the metal.



Scheme 6

Recent studies in our laboratories have demonstrated that the sequence of reactions outlined in Scheme 2 can be extended to produce the ^{109}Pd (t 1/2 = 13.5h; Eß= 1030 keV) analogues of the receptor functionalized phosphorus hydrazides. The ^{109}Pd complexes of **2** and **3** are formed in high yields (>90%) and are stable in aqueous media (pH 7) for 24h. The ^{109}Pd complexes of **2** and **3** are assumed to have structures similar to **4** and **5**.

The aforementioned studies have clearly established the efficacy of functionalizing model receptors on multifunctional phosphorus hydrazides and have also proven the formation of stable complexes with the metallic radioisotopes of diagnostic and therapeutic importance. At present, efforts are underway to label Octreotide (a somatostatin analogue) or other peptide receptor agents with Tc-99m, with the ultimate objective of developing new receptor agents that can selectively bind several types of cancer cells (e.g., small cell lung Ca, carcinoid, etc) that express somtostatin receptors.

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NUCLEAR ANALYSIS PROGRAM

NUCLEAR ARCHAEOLOGY AND GEOCHEMISTRY GROUP

ANALYTIC EPIDEMIOLOGY, NUTRITION AND IMMUNOLOGY GROUP

ACTINIDE CHEMISTRY GROUP

NUCLEAR ANALYSIS PROGRAM

J. Steven Morris, Coordinator
Charles A. Fairfax, Senior Research Specialist
Ginger L. Koziatek, Research/Laboratory Technician
Lynn R. Smith, Senior Secretary

Groups in the Nuclear Analysis Program
Nuclear Archaeometry and Geochemistry
Analytic Epidemiology, Nutrition and Immunology
Actinide Chemistry

During its 25 years of operation, the MURR Center has evolved with high quality, diverse, nuclear analysis programs which reflect the strengths of the MU campus and support the operation, revenue generation and academic mission of the MURR Center. This has been accomplished by a MURR-based scientific staff in collaboration with MU faculty and a very extensive set of scientists from other universities, government agencies and industry. All three components are critical to the success of the Nuclear Analysis Program (NAP).

MURR's Nuclear Analysis Program is a comprehensive one which includes integrated components of research, teaching and service. The central focus is the utilization of neutron activation combined with gamma-ray spectroscopy to provide users the full range of neutron activation analysis (NAA) techniques (i.e., prompt gamma-ray NAA, epithermal NAA, instrumental NAA and radiochemical NAA) as well as fate of tracer studies utilizing tissue culture and animal models. MURR's NAP has a well qualified staff, limited–but high-quality laboratories–and state-of-the art instrumentation. These Program components are summarized in Table 1.

The range of NAP research efforts reflects the strengths of the MU Campus including its Eminence Programs such as Food for the 21st Century (FF21C) and Molecular Biology. In addition, in this and previous fiscal years, NAP research projects have been the largest component of the DOE funded reactor sharing program. Reactor sharing allows other US universities access to neutron applications in their research programs.

The NAP research programs heavily emphasize the unique capabilities of the MURR Center. For example, the NAP biochemistry program maintains several isotope production networks featuring radionuclides that are not routinely available elsewhere, i.e., Se-75 (~1000 Ci/g), Cu-64 (~250,000 Ci/g) and Ca-47. These networks provide isotopes for NAP research, a mechanism for collaboration with other high-quality research programs, a service to the MU campus, and funding opportunities. In today's competitive research environment there is an ever increasing need to develop interdisciplinary approaches to attract and sustain adequate funding. Recognition of this need is evident in the NAP research programs. Each individual research area defined here has evolved beyond just the exclusive use of nuclear methodologies. For example, the Actinide Chemistry group has developed an excellent inert atmosphere facility and routinely utilizes sophisticated high temperature, non-aqueous electrochemistry methodologies to study thermodynamic properties of the actinides. The archaeology research uses microscopy techniques and applies cutting-edge principal components analyses to statistically test its extensive element composition database which contains both source materials and artifacts. The epidemiology area develops and applies nuclear and atomic spectroscopy techniques to study chronic trace-element and mineral imbalance. The biochemistry, nutrition and immunology collaborations utilize state-of-the-art tissue culture and small animal laboratories and instrumentation at MURR to advance the use of nuclear methodologies in these disciplines.

The primary teaching responsibility of the NAP is training students in 20 or more academic departments using the Program's substantial resources. In addition, NAP staff members routinely assist in teaching a wide variety of courses.

Service is the third leg of the mission triad. Here the Nuclear Analysis Program is also very active in supporting reactor operations, in providing the quality assurance programs required by exempt licenses, and in making NAA services available to other universities, government agencies and industry.

Funding for the Nuclear Analysis Program is derived from grants (55%), MURR grant incentive fund (12%), service contracts (11%) and MURR general operating support (22%), totaling $1,014,321 in FY 92.

NAP RESEARCH In the *Life Sciences*, the NAP has programs in analytic epidemiology, biochemistry, immunology and nutrition. These programs actively collaborate with MU departments such as Agronomy, Animal Science, Biochemistry, Biology, Chemistry, Child Health, Food Science and Human Nutrition, Immunology, Microbiology, Obstetrics & Gynecology and Veterinary Medicine, as well as many outside research universities and agencies such as Harvard, Johns Hopkins, Washington University, USDA and NIH-NCI.

Nutritional epidemiology research uses NAA and flame and furnace atomic absorption in collaboration with the major US centers for epidemiological research to study the relationships between chronic deficiency of trace-element nutrients and elevated intakes of toxic trace elements and human disease outcomes such as cancer, cardiovascular disease and adult onset diabetes. The MURR component focuses on the linkage between dietary intake of minerals and trace elements and the resulting concentration of these nutrients in tissue monitors.

The biochemistry and nutrition areas utilize unique isotope production capabilities at the MURR Center, with nuclear and atomic spectroscopy, in combination with biochemistry methodologies. These techniques examine macro- and trace-element requirements, interactions and binding by proteins using both tissue culture and whole animal models. This research emphasizes the combination of the utilization of high specific activity radiotracers (some of which are uniquely produced at MURR), cell culture and small animal models, with innovative application of gamma-ray spectroscopy to quantify and image radiotracers of the trace elements under consideration.

The nutrition program supports MU's Graduate Nutrition Area Program and the FF21C Nutrition Emphasis Area. Faculty, students and staff from MU departments of Agronomy, Animal Science, Biochemistry, Child Health, Food Science and Human Nutrition utilize MURR's neutron activation analysis, isotope production, gamma-ray spectroscopy and small animal facilities to study a wide range of nutritional interests such as trace-element interactions, calcium metabolism and endophyte infestation of Missouri fescues.

The radio-immunology research studies the production of monoclonal antibodies, their labelling with radionuclides while retaining immunological activity, and their use as radioimmunodiagnostic and radioimmunotherapy agents in the diagnosis and treatment of human diseases. The current focus is the development of a human monoclonal antibody capable of the sensitive diagnosis of ovarian cancer *in vitro* through the reaction with ovarian cancer antigens present on the cell membrane. There is close collaboration with the MU Departments of Immunology, Microbiology, Obstetrics and Gynecology, Biochemistry and Food Science and Human Nutrition.

In the *Anthropological* and *Geological Sciences*, the NAP has programs in nuclear archaeology and geochemistry, which, in addition to the MU departments of Anthropology, Art and Archaeology and Geology, also interact with major US universities and agencies with emphases in archaeometry through an NSF funded center for nuclear archaeology. The Archaeometry Laboratory at MURR combines NAA and microscopy techniques in archaeological research requiring determination of the sources of raw materials utilized by prehistoric peoples. Collaborations include projects with Harvard, Tulane, Dartmouth, Arizona, Arizona State and the Smithsonian Institution.

The objective of the nuclear archaeology program is to determine the geographical origins and ages of artifacts from various cultures. Heavy emphasis is placed on the sourcing capability of multi-element neutron activation analysis. The extensive "Missouri Collection" of Mesoamerican obsidian is one example of the comprehensive collections of source materials that are under investigation at MURR.

The objective of the geochemistry program is to develop petrogenetic models to study the chemistry and thermodynamics associated with the formation of the earth's oldest geological formations. Neutron activation analysis is utilized as a means of studying rare earth anomalies.

Actinide chemistry research was initiated at MURR in January 1990. In addition to MU departments of Chemical Engineering, Nuclear Engineering and Chemistry, the Actinide Chemistry group also collaborates with Rockwell International, DOE, the Central Research Institute of the Electric Power Industry (CRIEPI) of Japan and Kawasaki Heavy Industries (KHI) of Japan. The Actinide Chemistry group uses electrochemical measurements to determine the thermochemical and thermodynamic properties of the actinide metals (specifically U, Np, Pu and Am) and the metal chlorides in fused eutectic salt mixtures and cadmium that exist in nuclear fuel wastes and spent nuclear fuels. A specific aim is to determine the theoretical and practical feasibility of using an electrochemical process that would be carried out in molten salt solutions to separate the actinide elements from the complex radiological and chemical mixtures in which they exist.

NAP SERVICE In addition to its research activities, the NAP provides services to other MURR groups, MU faculty, students and staff, and to outside universities, agencies and industries–especially Missouri industry. NAP service programs can be characterized by three distinct groups/techniques/facilities: the Co-60 Irradiation Facility, neutron activation analysis (NAA) services, and the Quality Assurance Program.

The Co-60 Irradiation Facility provides gamma-ray irradiation services in support of academic and industrial research in the following ways:

- impairing the immune system to provide a baseline for immunology research
- TLD dating of archaeological artifacts
- biochemical inactivation of biological tissues and receptors
- sterilizing a wide variety of biological samples
- development of vaccines
- eradicating endophyte from infected fescue seed
- inducing chromosomal damage in maize
- inducing ESR in calcite; to study irradiation sterilized foods
- generating fruit fly mutations; to generate free radicals in Sm-153
- testing and calibrating avalanche detectors
- creating defects in semiconductor materials
- testing MOSS devices in a high gamma radiation field
- inducing polymerization of acrylics.

NAA services to industry are primarily in response to the domestic need for analysis of high-purity materials. Materials routinely analyzed include silicon (starting, refined and finished materials), graphite, quartz and boron.

The NAP Quality Assurance Program provides the quality assurance expertise necessary to obtain and sustain licensed activities such as the MURR exempt materials license which is critical to the MURR's revenue generation. The QA program includes gamma-ray spectroscopy and other nuclear analysis support for the exempt release of byproduct materials to the unlicensed general public, such as neutron color-enhanced gemstones and neutron transmutation doped silicon.

Table 1 The Nuclear Analysis Program at the MURR Center

COMPONENT	DESCRIPTION
Staff	14 full-time staff members including 5 research scientists, 4 research specialists, 4 research laboratory technicians and a secretary approximately 50 faculty, students and outside scientists collaborators
Laboratories	2 counting labs, 4 radiation labs with pneumatic-tube irradiation facilities, 3 sample preparation/project labs, an actinide chemistry lab, a small-animal research area, a tissue culture lab and a Co-60 irradiation facility
Major Equipment	Canberra/Nuclear Data-based gamma-ray spectroscopy system operating through acquisition-interface modules (AIM) attached via Ethernet to a multi-node Microvax computer network including six low-background spectrometers, four shielded spectrometers with sample changers, two shielded spectrometers in pneumatic-tube labs, one x,y-energy spectrometer and one PGNAA spectrometer Other major equipment: • atomic spectroscopy equipment including flame, furnace and ICP systems • sample preparation equipment including analytical balances, a micro-balance, static and centrifugal freeze-driers, microwave wet-acid digestion system, muffle furnaces and binocular and petrographic microscopes • biochemistry equipment including a UV-visible spectrophotometer, electrophoresis systems, column chromatography equipment with fraction collectors and a refrigerated centrifuge • tissue culture equipment including a biological safety cabinet, incubators, microscopes (inverted and fluorescence), autoclave and an ELISA reader • small animal surgery equipment, housing and metabolic units • actinide chemistry glove box with an argon inert atmosphere and equipped with Ni-train and Dri-train argon purification systems, computer controlled electrochemistry instrumentation and a high-temperature thermowell

NUCLEAR ARCHAEOLOGY AND GEOCHEMISTRY GROUP

PERSONNEL

Michael Glascock, Group Leader (Adjunct Assistant Professor of Nuclear Engineering)
Hector Neff, Research Scientist (Adjunct Assistant Professor of Anthropology)
Robert Benfer, Senior Research Investigator (MURR) and Professor of Anthropology
Michael O'Brien, Senior Research Investigator (MURR), Professor of Anthropology and Associate Dean of Arts & Sciences
Kathleen Slane, Senior Research Investigator (MURR) and Professor and Chair of Art History & Archaeology
Marcus Rautman, Senior Research Investigator (MURR) and Associate Professor of Art History & Archaeology
Peter Nabelek, Research Investigator (MURR) and Associate Professor of Geological Sciences
Jeffrey Denison, Senior Research Laboratory Technician
Sindy Hays, Research Laboratory Technician

The Nuclear Archaeology and Geochemistry Group, or "the Archaeometry Group" as we are frequently called, uses neutron activation analysis (NAA) to characterize thirty or more trace elements in artifacts and geological specimens. These data are used by archaeologists to determine the origins of artifacts and by geochemists to investigate the natural processes that created different types of igneous, sedimentary, and metamorphic rocks. The Archaeometry Group has an extensive network of student and faculty collaborators from MU and other universities.

Major accomplishments during the past year include:

- obtaining a three-year renewal of our NSF Archaeometry grant, enabling continued support for five or six students
- analyzing over 3,000 new samples from 25 research projects, further enhancing the utility of our archaeological databank
- developing an abbreviated-NAA procedure that reduces the cost and effort of sourcing obsidian artifacts from Mesoamerica
- beginning the incorporation of the hydration dating technique to supplement our NAA sourcing method on obsidian
- obtaining a unique collection of obsidian artifacts from the Valley of Oaxaca in southern Mexico
- sponsoring a visiting doctoral candidate student from the University of Michigan who used NAA to study ceramic materials from Peru
- beginning a long-term study of pottery and ceramic raw materials from Mississippian occupations in SE Missouri.

In addition, Dr. Neff published an edited volume entitled *Chemical Characterization of Ceramic Pastes in Archaeology* that reports data from MURR and other universities.

MESOAMERICAN STUDIES

Hector Neff, Michael D. Glascock, J. Michael Elam (MURR), Frederick J. Bove (University of California-Santa Barbara), Mary Thieme (Vanderbilt) and Ernesto Gonzalez-Licon (Instituto Nacional de Antropologia e Historia-Mexico)

One long-term research focus of MURR's Archaeometry Group under the direction of Michael D. Glascock is the archaeology of the region known as Mesoamerica, a designation which refers to northern Central America and the southern half of Mexico (Figure 1). The Archaeometry Group's first large-scale project, initiated in the early 1980s, was an effort to characterize by neutron activation analysis (NAA) all of the major Mesoamerican obsidian sources. The resulting source database permits identification of the geographic origin of obsidian cutting tools found on archaeological sites throughout Mesoamerica.

Current Archaeometry Group research activities in Mesoamerica center on the Valley of Oaxaca, Mexico and Pacific coastal Guatemala. NAA combined with obsidian hydration rim dating are providing fresh perspectives on the prehistory of both regions.

In the Valley of Oaxaca, J. Michael Elam, PhD candidate in Anthropology at MU, is exploring how acquisition of obsidian (which was unavailable locally) changed at several sites during the period 500 BC - 1520 AD. Interestingly, Elam has found that the histories of source utilization differ between sites in different parts of the Valley. The

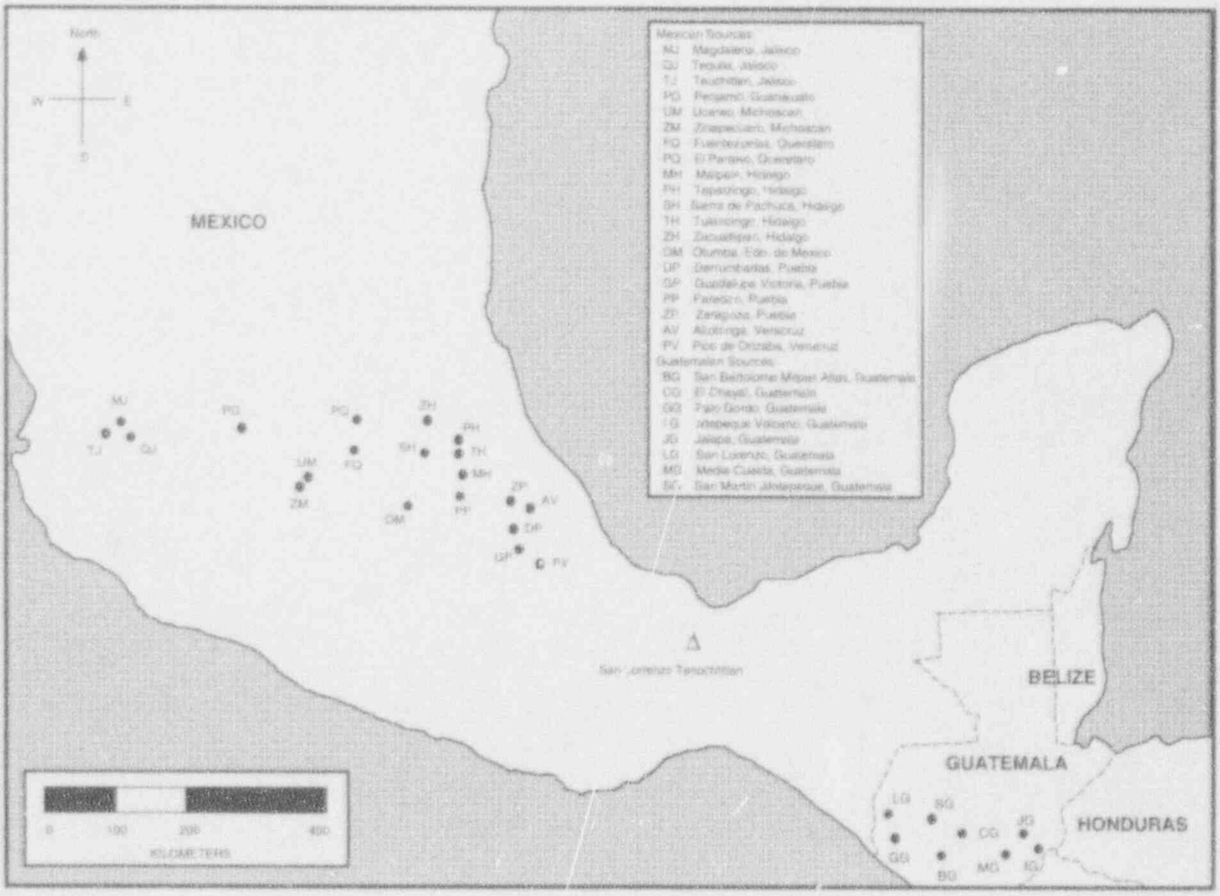

Fig. 1 Map showing obsidian sources in Mesoamerica.

500 or so obsidian dates to be generated by the project will also provide evidence pertaining to a controversial hypothesis that the entire region was nearly abandoned between about 700 and 1100 AD.

Another project, now in the planning stages, will combine NAA of modern and prehistoric pottery from the Valley of Oaxaca in an investigation of ceramic resource use and pottery consumption from 500 BC to the present. The modern pottery comes from four villages whose ceramic traditions can be traced to their pre-Columbian roots. Prehistoric pottery will come from the great Zapotec capital of Monte Alban, whose inhabitants consumed vast quantities of pottery from all over the Valley of Oaxaca over a period of about 2,000 years. The prehistoric pottery will be sourced to different Valley subregions by comparison with the analyzed modern pottery. A major goal of the project is to trace the waxing and waning of Monte Alban's ties with various parts of its hinterland.

In Pacific coastal Guatemala, where there are no modern potters, sourcing prehistoric pottery requires systematic survey and sampling of clay and temper sources. Temper is any non-plastic material mixed into the clay. Hector Neff, a research scientist at MURR, has been working with Guatemalan and US collaborators since 1987 to amass both the required source sample database and a database of analyzed pottery. Here, as in the Oaxaca study, there are two general research goals. The first is to learn how humans exploited certain natural resources such as ceramic raw materials. The second is to determine how humans interacted with each other over the past 3,500 years or so.

Of course, the goals just mentioned require precise control over the chronology, and this is where obsidian comes in. Next to pottery, obsidian is the most abundant artifact category in Pacific coastal Guatemala. Thus, using obsidian hydration rim dating, it is possible to obtain many calendrical dates from archaeological excavations in this region. Taking the number of dates per unit of time as a rough index of intensity of occupation, extremely detailed information on the population history of particular sites can be generated. For example, the stair-step pattern of dates at the important coastal site of Balberta (Figure 2) suggests several periods of intense occupation, the last one around 300 AD. The importance of the obsidian dating evidence is highlighted by the fact that the few existing radiocarbon and archaeomagnetic dates from Balberta only document the period from 0 - 500 AD.

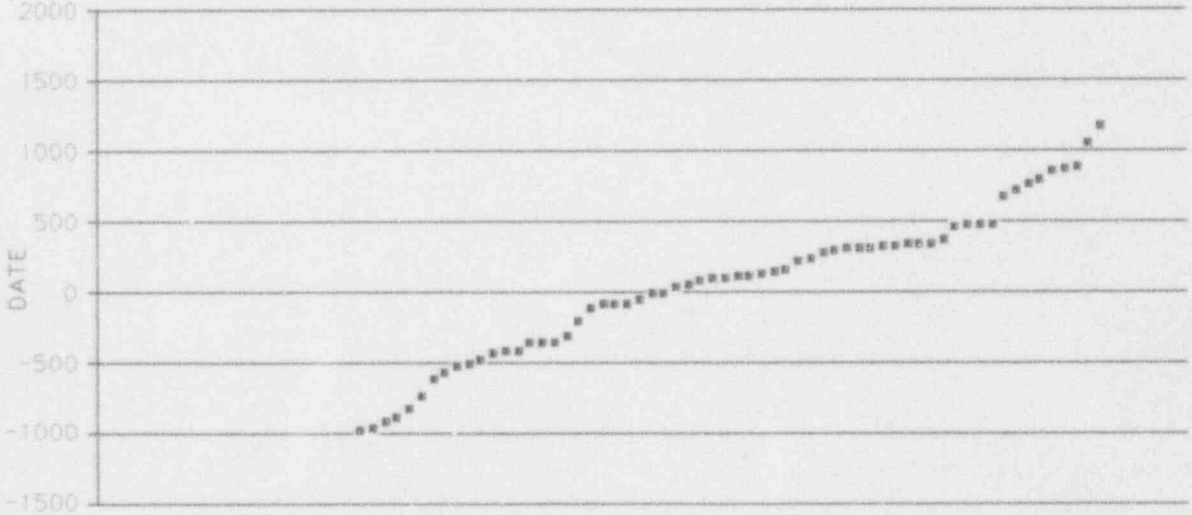

Fig. 2 Dates for obsidian artifacts from Balberta as determined by obsidian hydration analysis.

EXAMINING GEOGRAPHIC VARIATIONS IN OBSIDIAN SOURCE COMPOSITION AT SAN MARTIN JILOTEPEQUE, GUATEMALA

Michael D. Glascock, Hector Neff (MURR) and Geoffrey E. Braswell (Tulane)

Although the vast majority of obsidian sources are highly homogeneous in chemical composition, a few sources are known that exhibit compositional variation according to location around the source. Provided enough information concerning the geographic variation in composition is obtained, archaeologists have hypothesized that it might be possible to trace artifacts to the specific subregions of the sources from which they were quarried. This knowledge might assist archaeologists in obtaining information concerning chronological changes in obsidian acquisition for specific sources.

This collaboration between MURR's Archaeometry Laboratory and Tulane University has sought to examine obsidian from the large volcanic source near the city of San Martin Jilotepeque located in the highlands of Guatemala. Braswell collected 75 source specimens from geographically recorded locations that were distributed around the city of San Martin Jilotepeque. A map showing many of the collection sites is shown in Figure 1. The source specimens were analyzed by MURR to determine a 28-element chemical fingerprint. To facilitate examination of the data, principal components analysis was applied to the matrix of trace-element concentrations. A scatterplot of the first and second principal components is shown in Figure 2. The source data are shown as individual samples and are compared to a 95 percent confidence ellipse generated from artifacts previously sourced to San Martin. The interpretation of this scatterplot is that only certain areas around the source at San Martin were being exploited by prehistoric miners. The shaded area shown on Figure 1 identifies the quarrying area that coincides with these artifacts. The result of this study is that we have confirmed that sub-source areas within a complex and variable source, like the one at San Martin, can be identified and examined.

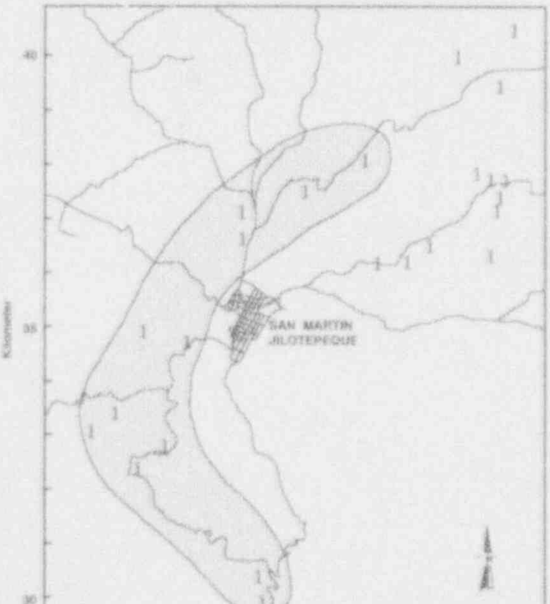

Fig. 1 Sample locations for obsidian source materials surrounding the village of San Martin Jilotepeque, Guatemala. The shaded area indicates the region where raw materials were apparently mined by the prehistoric inhabitants to produce the artifacts being found by modern-day archaeologists.

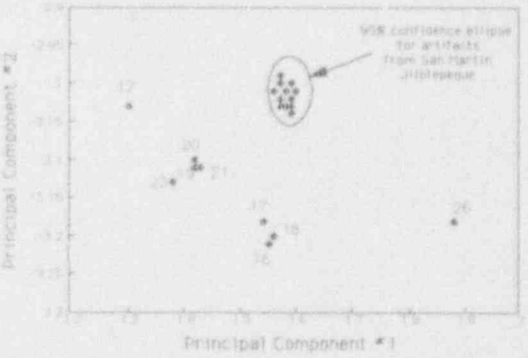

Fig. 2 Scatterplot of principal component #1 versus principal component #2 showing the correspondence between artifacts from San Martin Jilotepeque and analyzed source specimens.

CHEMICAL CHARACTERIZATION OF MISSISSIPPIAN POTTERY AND CLAYS FROM THE SOUTHEAST MISSOURI LOWLANDS AND ADJACENT OZARK UPLANDS

Hector Neff, Michael D. Glascock, James W. Cogswell (MURR), James E. Price, Michael J. O'Brien (MU Division of American Archaeology) and Mark J. Lynott (National Park Service)

An ongoing project funded by the National Park Service is assessing the feasibility of using chemical characterization of pottery and raw clays to study prehistoric ceramic production and exchange in southeast Missouri. The first 100 specimens analyzed by neutron activation analysis at MURR came from locations indicated on the accompanying map (Figure 1).

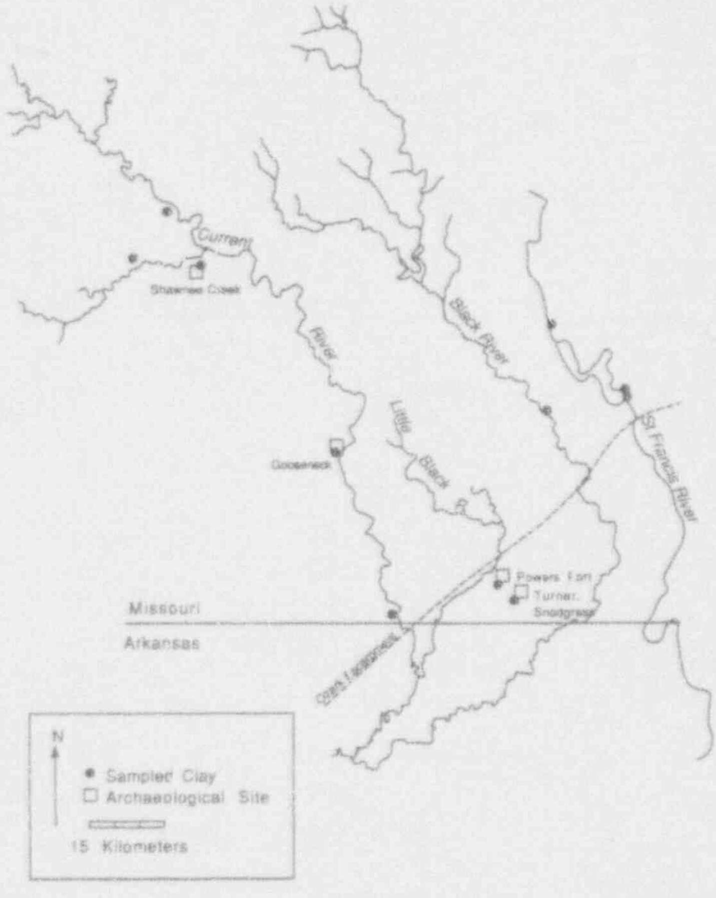

Fig. 1 The study region in southeast Missouri showing archaeological sites and clay sampling locations. The Ozark escarpment indicates the separation between the Ozark uplands and the Mississippi Valley lowlands.

Several tantalizing results have emerged from the pilot study. First, both clays and raw materials manifest two distinct compositional profiles, one associated with the Ozark uplands and one associated with the Mississippi Valley lowlands. Therefore, ceramic resources do manifest sufficient geographic patterning to permit sourcing of prehistoric pottery, at least on the broad level of uplands vs. lowlands. Related to the upland vs. lowland dichotomy, pottery from the lowland sites of Powers Fort and Turner/Snodgrass pertained exclusively to the lowlands, while pottery from the upland site of Gooseneck pertained to both uplands and lowlands. This evidence suggests a difference in geographic scope of economic activities between the upland and lowland inhabitants.

On a finer scale of geographical resolution, compositional distinctions are also recognizable within the lowlands. For instance, Turner/Snodgrass pottery analyzed in the pilot study was found to be compositionally distinct from the pottery of Powers Fort, which is located only 6 km from Turner/Snodgrass. Thus, lowland ceramic production and consumption was even more restricted in geographic scope than suggested by the upland vs. lowland contrast mentioned above.

SOURCING OBSIDIAN ARTIFACTS BY AN ABBREVIATED-NAA PROCEDURE

Michael D. Glascock and Hector Neff (MURR), in collaboration with more two dozen archaeologists working in Mesoamerica

Obsidian is a naturally occurring volcanic glass similar in composition to granite but lacking in crystalline structure. When struck by a skilled craftsman, the glass yields conchoidal-shaped fractures and produces some of the sharpest edges of all lithic materials. As a result, obsidian was a highly-prized natural resource for prehistoric peoples who used it to manufacture sharp-edged tools, weapons and ornaments.

Although the glass occurs naturally in only a few regions, there is much evidence for the existence of exchange networks during prehistoric times. In particular, the pre-Columbian inhabitants of Mesoamerica mined and traded obsidian from source areas to numerous sites that archaeologists have discovered more recently.

Since the early 1980s, the Archaeometry Laboratory at MURR has been involved in an effort to characterize by NAA all known Mesoamerican obsidian sources. The NAA database consists of a 28-element compositional fingerprint for every source specimen analyzed. Currently the source database includes fingerprints for more than 800 source specimens from approximately 40 sources located throughout Mesoamerica.

Applying the same procedures to artifacts as to source specimens permits near 100 percent success in determining the geographic origins of obsidian artifacts found at archaeological sites. Unfortunately, the cost of generating a full 28-element fingerprint is far higher than most archaeologists are able to afford. In response, the Archaeometry Laboratory recently developed an abbreviated NAA procedure, one relying on the measurement of short-lived elements, that succeeds in sourcing about 90 percent of the artifacts from this region. A scatterplot of Mn vs. Na is shown in Figure 1 where more than 500 artifacts from Guatemala, Belize, El Salvador and Honduras are compared to the 95 percent probability ellipses for the three main obsidian sources in Guatemala. The ten percent of artifacts not sourced by the abbreviated NAA procedure can still be submitted to the full 28-element analytical procedure such that we retain the ability to source nearly 100 percent of the artifacts from Mesoamerica. Thus, we have developed a method that increases sample throughput, provides the same rate of success in determining source, and greatly reduces the cost of NAA for archaeologists.

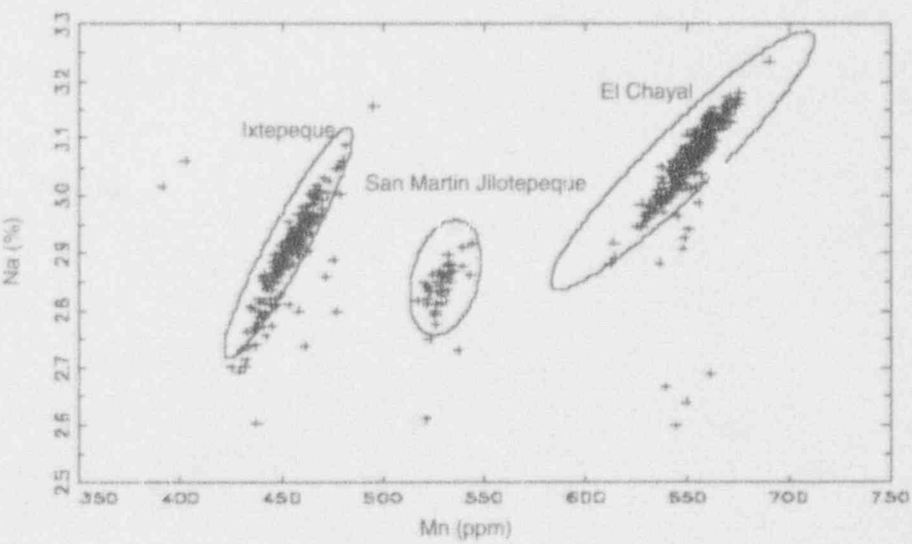

Fig. 1 Scatterplot of Mn versus Na for obsidian artifacts from archaeological sites in Guatemala, Belize, El Salvador, and Honduras.

ANALYTIC EPIDEMIOLOGY, NUTRITION AND IMMUNOLOGY GROUP

PERSONNEL

J. Steven Morris, Group Leader and Adjunct Professor of Chemistry
Kurt Zinn, Research Scientist
Tandra Chaudhuri, Research Scientist
Harold Anderson, Senior Research Investigator (MURR) and Professor of Science, Stephens College
Dennis T. Gordon, Senior Research Investigator (MURR) and Professor of MU Food Science and Nutrition
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Roger A. Sunde, Senior Research Investigator (MURR) and Professor of MU Food Science and Nutrition
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Vickie Spate, Senior Research Specialist
Dan Trokey, Senior Research Specialist
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Maddie Mason, Research Specialist
Ginger Koziatek, Senior Laboratory Technician
Cheryl Reams, Research Laboratory Technician

FACULTY ASSOCIATES

Gretchen M. Hill, Associate Professor of MU Food Science and Nutrition
S.R. Koirtyohann, Professor of MU Chemistry
Alfred S. Llorens, Professor Emeritus of MU Gynecology Oncology
Gregory A. McDonald, Assistant Professor of MU Microbiology
Boyd O'Dell, Professor Emeritus of MU Biochemistry

The Analtyic Epidemiology, Nutrition and Immunology group is a part of the Nuclear Analysis Program and is involved in research, teaching and service. The research function of the group has been described elsewhere in this report (see **NUCLEAR ANALYSIS PROGRAM** also) and is summarized in Table 1.

The Analytical group is also responsible for operation and utilization of the MURR Cobalt-60 Irradiation Facility and supports the regulatory requirements of the exempt license under which irradiated gemstones are released.

Table 1 **Research Summary of the Analytic Epidemiology, Nutrition and Immunology Group**

METHODS	MODELS	EXAMPLES
Radioisotope Production	Small Animal	Selenium–an essential trace element
Gamma-Ray Spectroscopy	Cell Culture	
Neutron Activation Analysis	Human hospital-based	Radioimmunodiagnosis of ovarian cancer
Atomic Spectroscopy	Human population-based	Fluoride and osteoporosis
Biochemical Analyses		

NUTRITIONAL STUDIES INVOLVING TRACE ELEMENTS

Kurt Zinn, Tandra Chaudhuri, Daniel Trokey (MURR) and Dennis Gordon (MU Food Science and Nutrition)

For the past two years MU and MURR researchers have used MURR's unique *in vivo* whole body counter and scanner. This gamma-ray spectroscopy instrument includes a germanium detector and is therefore capable of high resolution and simultaneous measurement of several gamma-ray emitting radionuclides. MURR scientists and MU collaborators have relied on this instrument to investigate nutritional questions, particularly those related to trace element metabolism.

Typically the rat model has been used, with varying test diets to determine their effect on the retention of the radionuclides. Several radionuclides are administered to the animal, and the gamma rays are detected with the instrument. Each gamma ray is associated with a specific radionuclide and has a measured count rate. The count rates are normalized to the rate following dosing and are reported as percent retention. Because all measurements are performed simultaneously, the scanner has reduced the numbers of animals needed to conduct meaningful studies.

The following two collaborative research projects were conducted in FY92 using these techniques.

Fe-Cu-Zn Interaction Previous collaborations between Professor Gordon and MURR researchers investigated dietary copper (Cu), iron (Fe) and zinc (Zn) interactions. That work suggested a negative interaction of excess Fe on Cu utilization, although it was not clear whether the interaction might change with rat age, or with time that rats were fed Fe-supplemented diets.

To investigate this interaction further, 24 male weanling (21 d) Sprague-Dawley rates were divided into three groups and fed test diets identical in Cu and Zn levels but with different Fe concentrations. The test diets were the same basal diet recommended by the American Institute of Nutrition supplemented with either 25 µg, 125 µg or 250 µg of Fe. The radionuclides ^{67}Cu, ^{65}Zn and ^{59}Fe were administered by gavage at three dosing intervals (6 hours, 10 days, 20 days) after the rats were started on their respective test diets. The ^{67}Cu and ^{65}Zn were available at MURR since a regular production schedule is maintained for research programs. Following dosing, the *in vivo* retentions of the radionuclides were measured at six time intervals.

There was a significant effect of high dietary Fe on percentage of ^{67}Cu retention ($p<0.05$) over time for all three dosings (Figure 1). In contrast, the ^{65}Zn was not significantly affected by dietary Fe level (Figure 2). As expected, high dietary Fe decreased the percentage of ^{59}Fe retention (Figure 3).

Consistent with previous studies, our investigations this year reproduced a negative

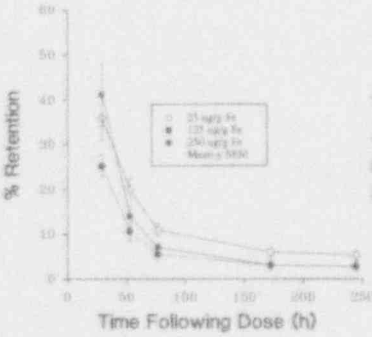

Fig. 1 Percent retention of ^{67}Cu over time.

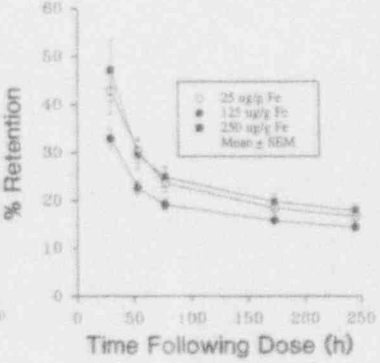

Fig. 2 Percent retention of ^{65}Zn over time.

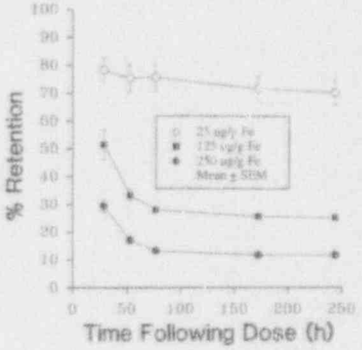

Fig. 3 Percent retention of ^{59}Fe over time.

interaction of Fe on ^{67}Cu retention in the rat. The interaction did not change with rat age, and was not minimized as rats were fed the diets for longer periods of time. The results indicate that ^{65}Zn retention is not adversely affected by high Fe level, but that high Fe significantly decreases ^{59}Fe retention. Future work will focus on the mechanism of the interaction, particularly at the level of the intestine.

Dietary Fiber and Trace Element Metabolism

Dietary fiber (DF) is a topic of keen interest in nutrition. While NIH guidelines suggest that Americans should increase their daily consumption of dietary fiber, there have been reports that DF has a negative effect on mineral absorption. MU/MURR collaborative research has set out to measure the effect of fiber on mineral bioavailability by studying the effect of DF on the retentions of trace element radionuclides. Seven groups of male weanling rats, six per group, were fed diets containing calcium (Ca), phosphorus (P), magnesium (Mg), iron, zinc, manganese (Mn) and copper at threshold levels adequate to sustain growth. An additional control group was fed a diet with no DF but with mineral concentrations recommended by the American Institute of Nutrition as a control diet. Diets with threshold mineral concentrations were prepared to contain 0, 5, 10 and 15% total dietary fiber (TDF) from soy hull cellulose and 5 or 10% TDF from wheat bran.

Rats were fed their respective test diets *ad libitum* for 11 days and were dosed with radionuclides by stomach gavage. The mixture contained ^{47}Ca, ^{54}Mn, ^{59}Fe, ^{65}Zn and ^{75}Se. The retentions of all radionuclides was measured over the next 10 days. All radionuclides could be measured simultaneously because of the high-energy resolution of the detector system. Figure 4 demonstrates the gamma-ray spectra plotted with log of counts versus gamma-ray energy. The right inset clearly shows the resolution of two gamma-ray peaks that differ in energy by only 5 keV (^{59}Fe and ^{47}Ca). The gamma rays for determining retention are shown in Table 1.

Table 1 Gamma Rays For Determining Dietary Fiber Retention

^{67}Cu	1297	keV
^{65}Zn	1115	keV
^{59}Fe	1099	keV
^{54}Mn	835	keV
^{75}Se	264	keV

Percent retention of the radionuclides after 10 days for rats fed the diets with threshold mineral concentrations supplemented with either cellulose or wheat bran averaged: ^{47}Ca, 78%; ^{54}Mn, 5%; ^{59}Fe, 70%; ^{65}Zn, 60% and ^{75}Se, 40%. These values were not significantly different from retention values in rats fed a DF-free diet. The results of this investigation strongly indicate that neither cellulose nor wheat bran impaired mineral absorption or retention. Furthermore, the utility of multiple radionuclides for evaluating dietary fiber has been demonstrated.

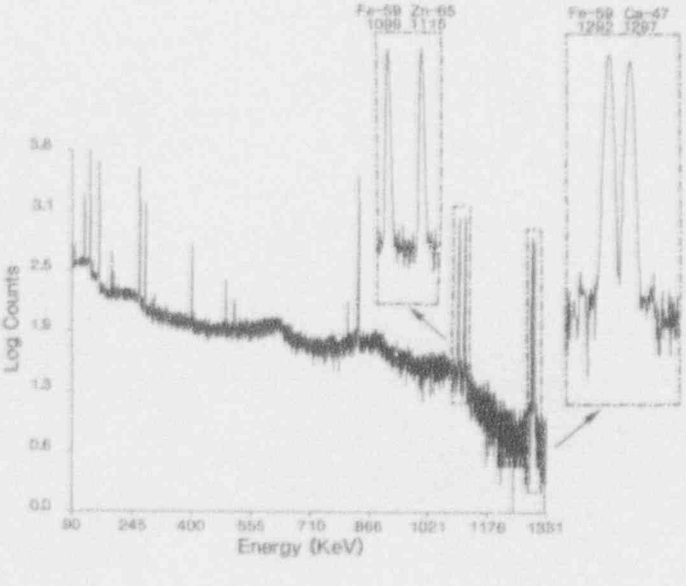

Fig. 4 Gamma-ray spectra from a rat that was dosed with the five radionuclides. Each peak is associated with the decay of one of the radionuclides.

RADIOIMMUNODETECTION AND THERAPY OF OVARIAN CANCER

Tandra Chaudhuri, Kurt Zinn, Steven Morris (MURR), Gregory McDonald (MU Microbiology), Alfred Llorens (MU Obstetrics and Gynecology) and T.K. Chaudhuri (University of Texas, Nuclear Medicine)

Ovarian cancer, also known as silent killer, is one of the leading causes of death among American women. Owing to the lack of quality control, purity and specific binding nature of existing antibodies, false diagnoses have been all too common, with inaccurate therapy a further result. Without a reliable method for early diagnosis of ovarian cancer, dedicated research is crucial for improving methods for both diagnosis and therapy. One of our major areas of interest is the radioimmunodetection (RAID) and radioimmunotherapy (RAIT) of cancer.

Radiolabeled monoclonal antibodies have been used by numerous investigators in nuclear medicine as well as in other areas of life science research. Murine monoclonal antibodies are increasingly used for immunoscintigraphy and RAIT. However, these agents are foreign proteins to humans and produce undesirable side effects, such as Human Anti-Murine Antibodies (HAMA). In order to overcome the effects of HAMA, we have developed human monoclonal antibodies to purified ovarian cancer cell surface antigens.

One human monoclonal antibody, TC5, was found to be specific for ovarian cancer as indicated by Western blot, immunodiffusion and fluorescent antibody assays. TC5 did not react with any normal cells of the ovary, uterus, cervix or fallopian tube, nor did it react with normal lung, heart or breast tissue. This monoclonal antibody was directly labeled with Au-198 and purified by column chromatography. The purified Au-198 labeled antibody was analyzed by using native polyacrylamide gel electrophoresis (PAGE) followed by autoradiography. A distinct band was developed at 150 kDa. The radiolabeled antibody was incubated with rabbit blood at 370 for 2 hours to test the stability. Only one sharp radioactive peak was obtained in a second chromatography purification, and this peak was subjected to native PAGE. Autoradiography showed one distinct band at 150 kDa, with other silver-stained proteins from rabbit blood not radiolabeled. However, the immunoreactivity of the labeled antibody was not preserved.

We developed a method to radiolabel monoclonal antibodies, developed against human ovarian cancer cell surface antigens, with Au-198 and to retain immunoreactivity. This was accomplished by protecting antigen binding sites with affinity chromatography techniques using Sepharose 4B column. Sixty percent of the total radioactivity bound to the antibodies was recovered by this method.

Immunoreactivity of the radiolabeled antibodies was first tested in a slot blot experiment, where various types of antigens were bound to the nitrocellulose blot. Au-198 labeled antibodies were reacted to these antigens followed by autoradiography.

Au-198 labeled antibodies reacted only to the extracts of ovarian cancer and a breast cancer but not to any normal cell extracts. Binding studies using Au-198 labeled antibody were further conducted with human biopsy specimens (Figure 1). In a blind experiment, Au-198-labeled TC5 reacted strongly with six out of six specimens pathologically diagnosed as metastatic ovarian cancer (serous and endometrioid). One "normal" ovary tissue reacted strongly by our method; four months later metastatic ovarian cancer was diagnosed in this patient. Au-198-labeled

TC5 did not bind with different types of normal tissue (ovary, uterus, fallopian tube, endometrium and normal breast), but did bind to a breast cancer specimen. Our method offers higher sensitivity to detect ovarian cancer at its early stage compared to existing methods. We hope to use this highly specific human monoclonal antibody and Au-198 labeling techniques, for early diagnosis (and possibly treatment) of ovarian cancer.

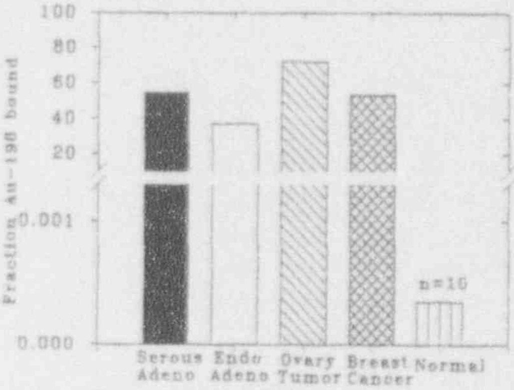

Fig 1 Au-198-labeled antibody bound to biopsy specimens

PRODUCTION OF NO-CARRIER-ADDED Cu-64

Kurt Zinn, Tandra Chaudhuri, Steven Morris, Ting-Pang Cheng, Daniel Trokey and Walter Meyer (MURR)

During the past year MURR staff developed a new technique for the production and purification of very high specific activity of ^{64}Cu. The development was sparked by collaborations with Dr. Michael Welch (Washington University), who wished to use this radionuclide for imaging with positron emission tomography (PET). In this application of PET, a monoclonal antibody directed against colorectal cancer was labeled with copper using a bifunctional chelate. A relatively high percentage of the copper atoms attached to the antibody needed to be radioactive to allow imaging in animal models. The purpose of these imaging studies was to demonstrate that the radiolabeled antibody could detect tumors in the animal model and to establish the basic research required for human testing. Hence, a very high specific activity ^{64}Cu was required. This was accomplished at MURR with the nuclear reaction ^{64}Zn(n,p)^{64}Cu. Following the production of ^{64}Cu, the zinc target had to be removed in such a manner that all metal impurities were minimized.

The fast neutron reaction producing the ^{64}Cu is only one type of reaction that can occur with the high purity zinc metal target. Thermal neutrons produce unwanted zinc radioimpurities, mainly ^{69m}Zn and ^{65}Zn. These reactions were minimized in our production method by conducting the fluxtrap irradiations under boron shielding. MURR's machine shop staff produced a special boron-nitride-lined can for these irradiations. Under the guidance of the MURR Reactor Manager, the can was tested under a variety of conditions, and this year the can was approved for running in the flux trap. Results indicate a 15-20-fold reduction in unwanted zinc radioimpurities. The benefits of this special boron-nitride-lined can go beyond producing a high specific activity ^{64}Cu: reduction of radioimpurities reduces the radiation exposure to the individual processing the ^{64}Cu; and the reduced radioimpurities result in less radioactive waste that must be packaged and shipped.

The key to producing very high specific activity ^{64}Cu is the chemical separation method. Our method employs a three-step procedure to remove the zinc target from the ^{64}Cu. First the ^{64}Cu and zinc are dissolved in concentrated HCl and taken to dryness. The chloride salts are dissolved in 1 M acetic acid and passed through a chelex column (4 ml); the ^{64}Cu remains selectively bound to the column, while the zinc impurities pass through. Following a rinse, the ^{64}Cu is eluted with 1 M HCl, and passed through an ion exchange column for final purification. As a measure of the separation method, a zinc purification factor has been calculated. This factor is the ratio of ^{65}Zn in the final ^{64}Cu solution divided by the total ^{65}Zn produced, and gives a measure of the fraction of zinc which makes it through the purification scheme. The zinc separation factor for the current technique averaged 1.3 x 10^{-7}, and with the best separation method reported in the literature[1] at 1.1 x 10^{-6}, the MURR method offers a ten-fold improvement.

A second assessment of the ^{64}Cu radionuclide is the measurement of ^{64}Cu specific activity. This ratio is the ^{64}Cu concentration divided by the copper concentration. Copper concentrations were measured using graphite furnace atomic absorption. Early runs averaged 282 mCi/µg Cu, with 7.3% of the Cu atoms being radioactive. We anticipate future runs will improve. For comparison, Mausner et al[1] reported 5 mCi/µg Cu for ^{67}Cu production, which is 0.66% of the Cu atoms radioactive. Another comparison is high specific activity ^{186}Re (3 mCi/µg), with 1.6% of the atoms radioactive.

In summary, our method to produce ^{64}Cu has resulted in a very pure radionuclide with high measured specific activity. The boron-nitride-lined can has reduced undesired thermal neutron reactions as well as radioactive waste. The separation method for purification of the ^{64}Cu was ten times better than literature reports, and could be accomplished in just 1.5 hours. The ^{64}Cu has been used in evaluating a monoclonal antibody against colorectal cancer,[2] and is currently being used in human clinical trials. Our group has also been actively involved in using this unique radionuclide to investigate copper metabolism and evaluate copper-binding proteins in animals.

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DETERMINATION OF FLUORIDE IN HUMAN NAILS VIA CYCLIC INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

J.S. Morris, V.L. Spate, C.K. Baskett, M.M. Mason and C.L. Reams (MURR)

The role of fluorine in human health has become somewhat controversial. It is widely accepted as protective against dental caries, may be protective against osteoporosis, and has been very conservatively implicated with osteosarcoma in male rats. We have found that toenails collected in population-based epidemiology studies apparently reflect fluoride intake. To our knowledge none of the geographical osteoporosis studies or fluoride therapy studies, or any osteosarcoma study known to us, have attempted to measure the individual fluoride status. Hence, it was in this context that we began experiments in our laboratory to determine if neutron activation analysis could be used to measure fluorine in human nails, and if that measurement reflects fluoride intake. Nails are essentially protein that contain a large percentage of α-keratin. We know of no reason to believe that fluoride actively participates in nail growth; however, it may be passively captured proportional to its level of intake. To evaluate this possibility, we developed an NAA method using ^{19}F(n,γ)^{20}F and applied it to toenails obtained from subjects consuming different levels of drinking water fluoride.

DETERMINATION OF FLUORIDE VIA NAA

Fluorine has one stable isotope, F-19, which can participate in several nuclear reactions that are summarized in Table 1. In the case of prospectively collected nail specimens, such as would be the situation in population-based epidemiology studies, the total sample

Table 1 Fluorine Nuclear Data

Target (Abund %)	Nuclear Reaction	σ_{th}(RI) or σ_f mb	Product	$t_{1/2}$	E_γ(KeV)	γ-Ray Abund %
$^{19}F_9$ (100)	n,γ	9.5 (39)	$^{20}F_9$	11.03 s	1633.6	100
	n,p	1.35	$^{19}O_8$	26.91 s	197.1	95.9
	n,α	7.85	$^{16}N_7$	7.13 s	6130	69
	n,2n	0.0073	$^{18}F_9$	109.8 m	511	194

quantity is quite variable and small, ranging from 10 to 100 milligrams for most subjects. These small samples and poor F-19 thermal neutron cross-section (9.5 mb) combine to make the determination of fluoride by NAA in these specimens quite challenging. Nevertheless, we have managed to develop an adequate method by summing the spectra from repeated cycles, each consisting of a 7-second irradiation, 10-second decay and 18-second count. The nuclear reactions and experimental conditions are shown in Figures 1 and 2.

Fifty milligram samples, if available, are weighed into small (~ 0.5 ml) high-density polyethylene vials (HDPE) which are placed in HDPE rabbits for irradiation. The rabbits are transferred, with a four second one-way transit time, to the irradiation position immediately adjacent to the beryllium reflector, and reactor core, through a pneumatic tube. The thermal and epithermal neutron fluxes are shown in Figure 2. Following irradiation and return of the rabbit to the laboratory, the sample vial is quickly removed and centered on the end cap in contact with a 30% HpGe detector. The samples are real-time counted for 18 seconds following a 10 second decay which is under the control of the data acquisition system. The 1633.6 KeV gamma-ray from the decay of F-20 ($t_{1/2}$ = 11.03 seconds) is measured using a Microvax-based spectrometer with a Tennelec 244 spectroscopy amplifier, Nuclear Data 599 loss-free counting module and a Nuclear Data 581 ADC. This analysis cycle is repeated for each sample 3 to 6 times at intervals of approximately 60 minutes or more. The spectral data are summed and the area of the 1633.6 KeV peak is extracted using the Nuclear Data Interactive Peak Search program. Data reduction is accomplished by standard comparison.

NAA Methods for the Determination of Fluoride
thermal and epihermal neutron capture: n,γ

$$^{19}F_9 + {}^1n_0 \rightarrow {}^{20}F_9 + \gamma \text{ (prompt)}$$

$$^{20}F_9 \rightarrow [^{20}Ne_{10}]^* + {}^0\beta_{-1} + \upsilon$$

$$[^{20}Ne_{10}]^* \rightarrow {}^{20}Ne_{10} + \gamma \text{ (delayed)}$$

for $^{20}F_9$: $t_{1/2}$ = 11.3 seconds and E_γ = 1633

Fig. 1 Nuclear reactions for fluorine

NAA Methods for the Determination of Fluoride
thermal and epithermal neutron capture: n,γ

irrad. time = 7 sec
$\phi_{th} = 8x10^{13} n\bullet cm^{-2}\bullet sec^{-1}$
$\phi_{epi-th} = 2x10^{12} n\bullet cm^{-2}\bullet sec^{-1}$
decay time = 10 sec
count time = 18 sec
sample vial in contact with detector end cap
3 to 6 cycles, > 60 minutes between cycles

Fig. 2 Experimental conditions used for the analysis of fluorine at the MURR

FLUORIDE STANDARDS Twenty microgram standards, giving 600 counts/µg/cycle, were prepared from a 100 ppm standard (RICCA, Chemical Co., Arlington, TX) by pipetting onto a small quantity of filter paper pulp substrate (Schleicher & Schuell Inc., Keene, NH) which had been placed in the HDPE vials. The F standards were then frozen and freeze-dried.

STUDY SAMPLES All toenail specimens were collected at various times as part of the Harvard-based Nurses' Health Study. The specimens were cleaned in DI water using a sonicator and allowed to air dry. When available, sample masses of approximately 50 mg were prepared. For approximately 20% of the samples, it was possible to prepare a duplicate.

RESULTS AND DISCUSSION

EXPERIMENT ONE Nail specimens from 13 women living in Holden, MA, a low fluoride area with a drinking water fluoride level of 0.09 ppm, and specimens from 12 Boston area residents, where the drinking water is fluoridated to a level of 1 ppm, were analyzed using the above described NAA method. The results are given in Table 2. The Holden and Boston means, 4.2 and 6.4 , respectively, are significantly different ($p < 0.05$). However, the ranges overlap (Table 2). One Holden subject had a toenail F level of 9.7 which was reproducible on a second set of clippings. This value is significantly higher than the means for both areas indicating that while the toenail levels generally reflect F drinking water levels, there are individuals that are misclassified.

Table 2 **Comparison of nail fluoride levels in subjects from municipalities with fluoridated and unfluoridated water supplies**

Locale	F in Water (ppm)	F in nails (ppm) range	mean*
Holden, MA	0.09	2.4 - 9.7	4.2
Boston, MA	1.0	2.7 - 10.1	6.4

* means are significantly different, $p < 0.05$

This could account for some of the inconsistencies and inconclusions reported in the geographic-based epidemiology studies where drinking water F concentrations have been used as a surrogate for tissue levels in predicting the incidence of osteoporosis.

EXPERIMENT TWO The long-term reproducibility of F in nails was evaluated. In 1983, over 60,000 participants in the Nurses' Health Study submitted a set of toenail clippings for use in nested case-control studies relating diet and disease. A second set of clippings was obtained 8 years later from 70 of the subjects; and both sets were analyzed for F in this study. We found that fluoride levels remained highly correlated (Spearman $r = 0.60$) over the eight-year period suggesting that toenails are a useful monitor of individual fluoride status.

EXPERIMENT THREE Twenty-four of the NHS participants that provided nail specimens in 1983 reside in Lynn, MA, which at that time had an unfluoridated water supply. In the ensuing years, fluoridation was implemented and a second set of nails was collected from the 24 participants eight years after the first, and several years after fluoridation began. Based on the 24 before and after samples, we observed a significant increase in toenail F levels due to fluoridation. The mean change was 2.1 ppm ($p = 0.04$) which coincidentally is practically the same difference as observed in the toenail F levels from residents of Holden, MA (a low F area) compared to Boston, MA (a fluoridated area) (Table 2).

CONCLUSIONS

Geographical epidemiological studies relating F drinking water levels to the incidence of osteoporosis are for the most part inconclusive. In cases where a low F area (~ 0.1 ppm or less) was included, the studies tend to show a relative protective effect due to fluoride. In at least one study comparing 4 ppm to 1 ppm areas, a greater incidence of fractures occurred in the higher F area. As we have shown in this study, the level of F in nails is generally correlated with drinking water levels; however, there is an overlap that can result in misclassification and may explain some of the inconclusive findings.

Fluoride therapy is controversial. Well designed and carefully conducted studies have, in one case found a significant protective effect, and the conclusive absence of such an effect in another. In both studies, increases in certain types of bone mass were reported. In the latter, cortical bone loss was also observed; and in both, there were non-trivial side effects due to fluoride therapy. Again, no attempt was made to individualize F status.

While there is no direct evidence that fluoridation of drinking water increases the risk of osteosarcoma in humans, the Delaney clause has prompted a call for further study based on the development of a small number of osteosarcomas in male rats consuming highly fluoridated water.

We have shown that neutron activation analysis can be used successfully to measure fluoride in human nails collected in population-based studies, and that nails apparently do reflect intake. The NAA method is based on cyclic NAA and could be substantially improved by reducing the 10-second decay time through development of an automated process. The method will be applied to a population-based fracture study in the near future

DETERMINATION OF TOTAL AND BOUND SE IN SERA BY INAA

V.L. Spate, M.M. Mason C.L. Reams, C.K. Baskett, J.S. Morris, and D.S. Mills (MURR)

The role of dietary selenium as a protective agent against cancer and heart disease has been studied in our laboratory in collaboration with epidemiology groups for many years using different monitors of selenium status such as blood, red blood cells, sera, plasma, hair and nails. Since 1973, MURR Center staff members have been developing methods for the analysis of selenium in biological samples using the standard comparator method. This method is based on instrumental neutron activation analysis (INAA) using the Se-77m isotope, which has a half-life of 17.4 seconds and a gamma-ray energy of 161.9 KeV.

The analysis of blood serum using the dialysis method at MURR has proven to be an accurate and long-term reproducible method. The Se component quantified using the dialysis method will be referred to in this paper as "bound Se." The development of high-purity germanium detectors and improvements in both the hardware and software used for pulse pile-up correction and peak extraction now allow for the accurate determination of Se in freeze-dried serum that has not been otherwise altered by dialysis or ashing. We refer to the Se concentration determined by this method as "total Se." The difference between "total Se" and "bound Se" is that small fraction of selenium that is either not bound to large molecules, or is free to dissociate from them during dialysis. We refer to that component as the "free Se."

We also make use of the samples prepared for "total Se" to determine the sodium concentration. This allows for the identification, and normalization if desired, of those samples that have been concentrated as a result of freezer-burn or the effects of repeated thawing, aliquoting and re-freezing that sometimes occurs with these unique sera banks, prospectively collected, and limited in amount.

This paper will discuss the standardization and quality control approaches used to identify the relatively small "free Se" pool, describe the two sample preparation methods and report on a comparison between "bound Se," "total Se" and "free Se" concentrations in 977 case-control samples.

SAMPLE PREPARATION

The study sera were retrieved from multi-year storage and shipped frozen to MURR. Samples were stored frozen until sample preparation was done in sets with 20 subjects per set. Four one milliliter vials, each vial containing approximately 0.5 ml, were shipped for each subject. The contents of the four vials were mixed to produce a composite sample having a volume of approximately two milliliters.

Duplicate samples for both the "total Se" and "bound Se" configurations were prepared from the two ml composite sample. Typically, the duplicates for the determination of "bound Se" were prepared first followed immediately by those for the determination of "total Se" and sodium. The composite sample is thoroughly vortexed immediately prior to pipetting each aliquot. Dialysis membranes 7.5 cm in length are freshly prepared, labeled with the assigned sample number and placed in the specially designed plexiglass dialysis racks with a ten-membrane capacity. In turn, each membrane is individually placed on a holder on an analytical balance and samples of approximately 0.5 ml are carefully pipeted into the dialysis membrane and the mass is determined by difference. After the "bound Se" duplicates are prepared for each sample, the duplicates for the "total Se" and sodium determinations are prepared by pipetting approximately 0.3 ml of the composite serum into pre-numbered 1.5 ml HDPE vials positioned on the analytical balance. Again the mass is determined by difference.

QC sera (SK9-90) is reconstituted to a volume of 3 ml using DI water. Replicates for the determination of "bound Se", "total Se" and sodium are prepared as described for the study samples.

Each dialysis rack is loaded with duplicate samples from four subjects and one QC serum sample. The racks are placed in DI water at 35°C for one hour. The volume ratio of DI water to sera is approximately 500:1. After dialysis of one hour, the samples are removed and the outside of the dialysis membranes are rinsed with DI water. The samples are frozen, then freeze-dried for approximately 12 hours, removed, and folded in the shape of a "Z", then placed in a numbered HDPE vial, secured with a polyethylene plug, and capped. The replicates prepared for "total Se" and sodium are frozen, freeze-dried for 12 hours, plugged and capped. Typically, the replicates and QC specimens for approximately 40 subjects are prepared per day.

RESULTS AND DISCUSSION

One hundred and ten "bound Se" and "total Se" selenium standards were analyzed on the same day to quantify the difference (net counts per μg) due to the slight differences in geometry and gamma-ray absorption. These results are shown in Table 1. SRM-1577 bovine liver samples, prepared in the two different configurations, were also analyzed using the above mentioned standard data. The bovine liver results are given in Table 2.

The data in Tables 1 and 2 were taken from standards and SRMs which were analyzed on the same day over a continuous 6 hour period. There is a small but significant ($p < 0.001$) difference (~4%) between the "bound Se" vs "total Se" counting geometries (Table 1). However, the standardization approach used accurately compensates for this difference in actual samples (Table 2).

Results for the MURR QC serum (SK9-90) previously described are given in Tables 3 and 4. The data in Table 3 were taken in a preliminary experiment to demonstrate the low variability due to sample preparation over a 10 day period. The data in Table 4 resulted from the analysis of the MURR QC sera over the 9 months during which the case-control analyses were done. In both cases the MURR QC sera, once reconstituted to 3 ml, were aliquoted to prepare three 0.5 ml replicates for the "bound Se" determination and two 0.3 ml replicates for the "total Se" determinations. In the preliminary experiment only undiluted SK9-90 serum was used whereas a random selection process including all concentrations of SK9-90 were used with the case-control analyses. All concentrations were normalized to the undiluted sera.

The 7% difference between the "bound Se" and "total Se" concentrations which remains after the geometric correction is made is statistically significant ($p < 0.05$). We have defined this as the "free Se" component which is either a low molecular weight Se compound present in sera or a fragment that dissociates during dialysis.

Significant differences between "total Se" and "bound Se" in this study were observed in both the MURR QC and study sera sets. Consequently, we have defined the "free Se" component to be the difference between these two pools. Hence, the "free Se" pool is generally that small fraction of the serum Se that is either not bound to protein, or some other high MW species, or is free to dissociate from such compounds. Of the 977 case-control samples, 651 had "free Se" components, i.e., the "total Se" was greater than the "bound Se." Twenty-three percent were greater by one standard deviation or more. We separately tested the correlation between the "free Se" with the "total Se" and "bound Se" pools. There is no significant correlation between the "free Se" and "bound Se" ($r = 0.06$); however, the "free Se" and "total Se" pools are highly correlated ($r = 0.78$).

Table 1 **Comparison of selenium standards used in the determination of serum selenium concentrations**

Geometry	Number of samples	Mean ct/ug*	Standard error	% C.V.
"Bound Se"	110	72061	123	0.2
"Total Se"	110	75081	69	0.1

* difference between geometric configurations is significant at $p < 0.001$

Table 2 **Comparison of the Se concentration in SRM-1577 Bovine Liver analyzed using "bound Se" and "total Se" standards**

Geometry	Number of samples	Mean PPM	Standard error	% C.V.
"Bound Se"	6	1.139	0.035	3.5
"Total Se"	9	1.129	0.034	3.0

Table 3 **Se concentration in "MURR" quality control sera SK9-90 sample preparation variability trial**

Treatment	Number	Mean PPM*	Standard error	% C.V.
"Bound Se"	30	0.108	0.0007	0.6
"Total Se"	20	0.116	0.0030	2.4

*difference is significant at $p < 0.05$

Table 4 **MURR quality control sera (SK9-90) analyzed over a 9 month period**

Treatment	Number	Mean PPM*	Standard error	% C.V.
"Bound Se"	174	0.106	0.0008	5.5
"Total Se"	116	0.113	0.0023	14

*difference is significant at $p < 0.00$

Table 5 **Comparison of "Bound Se" and "Total Se" concentrations in 977 sera samples from a case control study over a 9 month period**

Treatment	Mean PPM*	Standard Deviation	Low PPM	High PPM
"Bound Se"	0.109	0.0174	0.060	0.185
"Total Se"	0.115	0.0273	0.060	0.657
Total Na	3214	182	2189	4053

* "bound Se" vs "total Se" concentration means are significantly different, $p < 0.001$

CONCLUSIONS

There is a small difference between the "bound Se" and "total Se" counting geometries which was accurately compensated by the standardization approach used. The MURR quality control sera had 6% difference between the mean "bound Se" and "total Se" concentrations. It has been shown that there is also a significant difference (5.2%) between "bound Se" and "total Se" concentrations in study sera samples. A small, but real, fraction of the selenium in human serum is either not bound to high MW (12,000 D) species or is free to dissociate from them during dialysis against DI water.

A correlation (r = 0.59) between the "bound Se" and "total Se" was observed in this study. However, a stronger correlation (r = 0.78) exists between "total Se" and "free Se" concentrations with no correlation (r = 0.06) between "bound Se" and "free Se" concentrations of selenium in sera.

We would recommend the continued analysis of both "bound Se" and "total Se" to test hypotheses relating "bound Se," "total Se," and "free Se" serum selenium concentrations to disease end points. Additionally, the determination of sodium is useful to determine if the serum has been concentrated during long-term storage.

THE DETERMINATION OF SELENIUM IN URINE

C.K. Baskett, V.L. Spate, M.M. Mason, C.L. Reams, J.S. Morris

Selenium is an essential trace element in human nutrition. It is present at the active site of glutathione peroxidase, an enzyme that catalyzes the breakdown of hydroperoxides, maintaining the appropriately low levels of these oxidants in the cell. The status of selenium in humans is assessed by estimating dietary selenium intakes, by measuring selenium in tissues or excreta, or by determining glutathione peroxidase activity in blood components. Dietary selenium intake causes urinary concentration of selenium to change rapidly. It was determined that the urine selenium concentration may be a sensitive indicator of selenium intake.

This study had two major objectives. The first was to develop a method for the NAA of urine which could be applied to the large quantity of samples required in epidemiological studies. Urine samples cannot be analyzed by INAA due to the interference from the sodium contained in the urine. The method developed focused on minimizing the required chemical steps to a manageable level, while still producing useful epidemiological data regarding dietary selenium as monitored by the urine. The second objective of this study was to apply the method to a dietary trial which was designed to evaluate and correlate various dietary monitors, such as, urine, blood, sera, and toenails. This method would then extend the selenium methodologies that are already used in our laboratory for blood, sera, plasma, and nails to include urine. The procedure for selenium analysis discussed in this paper was applied to urine samples from a selenium supplementation study conducted in collaboration with Harvard Medical School.

The selenium feeding study conducted at Harvard Medical School consisted of 12 healthy, adult male subjects who were randomly assigned into three groups. Whole wheat bread was chosen as the medium for the selenium because grain is a major source of dietary selenium. The study bread contained three different doses of selenium determined by the fraction of high-Se wheat flour that was used. Subjects were requested to eat two slices of the study bread daily for one year. The control whole wheat bread contained 32.4 ± 0.2 μg selenium per two slices, the medium dose bread contained 206

± 17 µg selenium per two slices, and the high dose bread contained 388 ± 22 µg selenium per two slices. Toenails, whole blood, sera, and urine were collected at regular intervals during the feeding year and the following year.

The urine samples must be chemically separated prior to the irradiation due primarily to the large amount of sodium contained in the urine. The separation procedure used for urine is based on a co-precipitation of the selenium with arsenic. Five ml of urine was pipetted into Erlenmeyer flasks and the sample mass was determined. All urine samples were prepared in duplicate. Approximately 1 x 10^{-2} µCi of quantified Se-75 tracer solution, diluted from high specific activity Se-75 produced at MURR, was added to each flask. The flasks were heated on a hot plate to just under boiling, for approximately five minutes. Carrier solution containing 5 mg As was added to each of the flasks. The samples were heated for an additional five to ten minutes. Three ml of hypophosphorus acid (50%) was added as a reducing agent and samples were heated another five to ten minutes. A second three ml aliquot of hypophosphorus acid was added and samples were heated for an additional 20 minutes. A dark, almost black, arsenic precipitate formed during the heating. Selenium was also reduced and co-precipitated with the arsenic. The precipitate was filtered onto a 0.8 micron Nuclepore filter membrane. The flask was rinsed with deionized water and acetone to remove any residual precipitate. The Nuclepore filter membrane was folded, using plastic forceps, and placed inside a high density polyethylene MURR vial. A polyethylene spacer was placed on top of the filter membrane to maintain the same geometry in all samples and a lid was placed on the vial.

Se-75 tracer standards were prepared to check the percent yields of the separation. National Institute of Standards and Technology Urine (NBS SRM 2670) was prepared as a quality control check of the separation procedure. Both normal level and elevated level of the NIST urine were prepared. Deionized water was used for the analytical blank. The SRM urine and the blank were prepared in the same manner as the urine samples. The NIST urine was lyophilized and had to be reconstituted before pipetting. Blanks were prepared with each group of samples which went through the separation procedure.

The Se-75 standards, the urine samples and duplicates, the SRM urine, and the blanks were counted on a NaI detector, attached to a multichannel analyzer, to determine the percent yields of the Se-75 from the separation procedure. The application of the arsenic-coprecipitation separation to undigested urine specimens produced consistent yields ranging from 65 to 87%. The yields between duplicate samples were very similar.

Fifty-eight urine samples from the feeding study, along with duplicates, SRM urine, and blanks were analyzed according to the Se-77m analysis method used in our laboratories. Se-77m is useful for the measurement of selenium because of its favorable production and decay parameters. Se-77m, the isotope of interest, has a gamma-ray energy of 161.9 keV and a half life of 17.4 seconds. Each sample was placed in a shuttle rabbit and irradiated for five seconds via the MURR pneumatic tube system. The thermal neutron flux in this irradiation position is approximately 8 x 10^{13} n•cm^{-2}•sec^{-1}. The shuttle rabbit was returned to the laboratory and the sample vial was quickly removed and transferred to the appropriate counting geometry. The decay time for each sample was 15 seconds and the real time count was 25 seconds on a high resolution gamma-ray spectrometer. The spectrometry system included a Tennelec 244 amplifier coupled to a Nuclear Data 599 loss-free counting module and a Nuclear Data 581 ADC. Data acquisition and peak extraction were done using a VAXstation 3100, model 38.

The Se-77m concentrations were then determined using the standard comparator method. National Institute of Standards and Technology Bovine Liver (NBS SRM 1577) was analyzed with each batch of samples as an additional analytical quality control check. The bovine liver SRM was not subjected to the pre-irradiation separation. It was primarily utilized to ensure that the Se comparison standards are accurate.

The certified concentrations of selenium in the normal level and elevated level NIST urine are 0.030 ± 0.008 and 0.46 ± 0.03 ppm, respectively. Our determined concentration of 0.027 ppm selenium for the normal level urine is within the acceptable range. However, the coefficient of variance was rather high. Our determined concentration of 0.56 ppm selenium for the elevated urine is above the acceptable range, but the coefficient of variance was 5.6%. The SRM's had been lyophilized and consequently required reconstitution prior to analysis. The study specimens had been stored frozen. Perhaps the freeze-drying process affects the pre-irradiation separation. The certified concentration for selenium in NIST bovine liver is 1.1 ± 0.1 ppm. Our determined concentration of selenium in NBS 1577 bovine liver was 1.16 ± 0.03 ppm selenium for all analytical runs. The results of the quality control analyses are shown in Table 1.

Table 1 Quality Control Samples and Blank

Quality Control	Mean Se PPM	Std. Dev.	% C.V.	Certified Se PPM
Normal level urine (NBS 2670)	0.027	0.010	37	0.030 ± 0.008
Elevated level urine (NBS 2670)	0.56	0.03	5.6	0.46 ± 0.03
Bovine liver* (NBS 1577)	1.16	0.03	2.3	1.1 ± 0.1
Blank	0.0029	0.00076	26	

* Bovine liver SRM used as analytical QC, did not go through separation procedure.

The urine samples were analyzed in duplicate. All duplicates had a coefficient of variance of less than 5%. The concentration of selenium in the urine samples ranged from 0.0168 to 0.7071 ppm. The mean concentration of selenium in the 58 urine samples from the feeding study was 0.0904 ppm. The urine results are shown in Table 2.

In conclusion, the pre-irradiation separation which was used appears to be a useful approach for the analysis of urine for selenium using NAA. This method is useful for the analysis of selenium in urine as a reflection of diet. Useful epidemiological data can be obtained by using urine as a dietary monitor for selenium. The level of the selenium supplement could be detected in the determined urine concentration. The separation procedure can be applied as a screening method for the large sample sets required in epidemiological studies. This method gives useful results for selenium with a limited degree of chemical processing.

Table 2 Urine specimens

n Duplicates	Low Se PPM	High Se PPM	Mean Se PPM	Mean % C.V. for Dupls.
58	0.0168	0.7071	0.0904	2.364

There was a dose-response relation observed between the selenium intake and the urine selenium concentrations in the samples from the selenium supplementation study conducted at Harvard Medical School. Urine samples were compared with sera and whole blood samples from individuals in the selenium supplementation study. The selenium concentrations in the urine were highly correlated with both the selenium concentrations in sera (r = 0.70) and whole blood (r = 0.72). These relationships are shown in Figure 1.

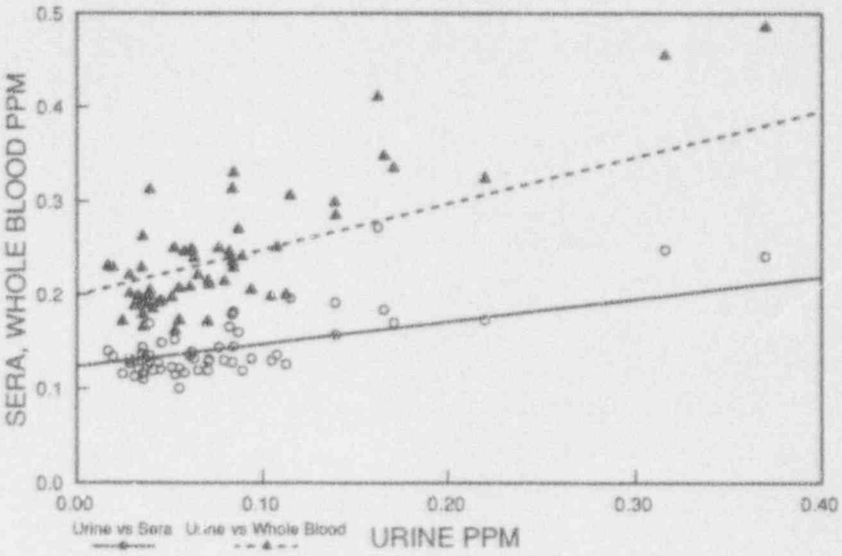

Fig. 1 Urine vs. sera and whole blood

ACTINIDE CHEMISTRY GROUP

PERSONNEL

Truman Storvick, Group Leader, Professor of Chemical Engineering
C. Leon Krueger, Research Scientist
Paul Sharp, Senior Research Investigator (MURR) and Associate Professor of Chemistry

FACULTY ASSOCIATES AND VISITING SCIENTISTS

LeRoy Grantham, Senior Chemist, Rockwell International
David Grimmett, Chemist, Rockwell International
Jeff Roy, Chemist, Rockwell International
Hajime Miyashiro, Senior Research Engineer, CRIEPI
David Retzloff, Associate Professor of MU Chemical Engineering
Yoshiharu Sakamura, Research Engineer, CRIEPI

The Actinide Chemistry Group was formed to do experimental work on uranium and the higher actinides under contract with Rockwell International, Rocketdyne Division, Conoga Park, CA. Funding comes to Rockwell International from Japan's electric power industry through contract with Kawasaki Heavy Industries (KHI) and the Central Research Institute of Electric Power Industries (CRIEPI), both of Tokyo, Japan. The Department of Energy, USA, has provided the pure uranium, neptunium, plutonium and americium metals that are used in the experimental program.

The Japanese plan to use the TRUMP-S (TRansUranic Management by Pyropartitioning-Separation) process to remove the transuranic metals from the liquid extraction residues from the PUREX process used to reprocess the spent nuclear fuel from the boiling water reactors used to generate electricity. In the PUREX process, the metal oxides in the spent fuel elements are dissolved in acid and the uranium and plutonium are recovered by a combination of liquid extraction-chemical complexing between aqueous and organic phases. The PUREX residue (about 3-4% of the total metal content of the spent fuel) contains all of the fission products, some uranium and plutonium, and the minor actinides: neptunium, americium and curium. These actinides have long half lives and present long-term radiological hazards (tens of thousands of years) which must be managed if placed in a repository. The TRUMP-S process is designed to remove the actinides from the PUREX residue. Removal and fissioning or transmuting the minor actinides produces a smaller residue stream that should require management for 200-300 years. The TRUMP-S process can also be used for "on site" reprocessing of the metal fuel that is proposed for the next generation of liquid metal cooled nuclear reactors for production of electricity.

The proposed pyropartitioning-separation process is dry. The aqueous PUREX residue stream is dried and denitrated, the metals are chlorinated and dissolved in fused salt. Processing must be done in an inert atmosphere because all of the metals are chemically active and readily form oxides, nitrides and hydrides which are insoluble in fused salt. Molten lithium chloride-potassium chloride eutectic salt provides a solvent for the metal chlorides at 400-500 C. The actinide metal chlorides can be chemically reduced to metal and alloyed into a liquid cadmium phase placed in contact with the fused salt mixture. The chemical reduction leaves the more chemically active lanthanide metals in the salt phase with a small fraction of actinide metals. The last traces of actinide will be removed from the fused salt by electrolytic reduction to an inert electrode.

The experimental program at MURR is designed to provide laboratory measurements to determine the thermodynamic and rate data required for the TRUMP-S process. Instruments have been developed to measure the standard potential of the actinide metal chloride in equilibrium with the pure metal. The electrochemical experiments use less than 10 mg of actinide chloride to conserve the available pure actinide metal and to reduce radiation levels so that the work can be done in a glove box. The experiment can be modified to measure the activity coefficient of the actinide metals dissolved in cadmium.

This year has been spent on the properties of americium chloride and americium metal in cadmium. The measurements suggest that americium chloride is present in both the +2 and +3 oxidation state in the fused salt when it is in equilibrium with pure metal. This complicates the electrochemical measurements and the interpretation of the experiments. The potential of the americium chlorides is closer to the lanthanide chlorides and this will make it the most difficult actinide to remove from the salt without removing the lanthanide metals. It is important to know this potential accurately. Interpretation of the americium measurements continues as the measurements of the equilibrium separation coefficients the pure actinides from the lanthanides begin.

NEUTRON MATERIALS SCIENCE PROGRAM

ELECTRONIC AND MECHANICAL PROPERTIES GROUP

MAGNETIC AND CRYSTAL STRUCTURES GROUP

NANOSTRUCTURES AND EXCITATIONS GROUP

NEUTRON OPTICS GROUP

GAMMA-RAY SCATTERING GROUP

NEUTRON MATERIALS SCIENCE PROGRAM

James J. Rhyne, Coordinator (acting)
G. David Peeler, Senior Research Engineering Technician
Mary B. Seewoster, Senior Secretary

Groups in the Neutron Materials Science Program:
- *Electronic and Mechanical Properties*
- *Magnetic and Crystal Structures*
- *Neutron Optics*
- *Nanostructures and Excitations*
- *Gamma Ray Scattering*

The new Neutron Materials Science Program includes groups who primarily use elastic and inelastic neutron scattering, gamma-ray scattering, and neutron radiation damage in their research work. The groups are Electronic and Mechanical Properties; Magnetic and Crystal Structures; Nanostructures and Excitations; Neutron Optics; and Gamma-Ray Scattering, and include members from MURR and from MU Physics, Chemistry, and Mechanical and Aerospace Engineering departments. Major collaborative programs are maintained with scientists from MU departments, and a number of outside universities, national laboratories, and industrial research organizations.

The scattering research makes use of the six radial beam tubes at MURR of either 4" or 6" diameter which view the core at three closely-spaced vertical positions. Because of the light water moderation, extensive use is made of cooled and room-temperature silicon and sapphire filtering in the beams before the instruments. Of the six beam tubes, five are devoted to neutron scattering facilities, and the sixth is a filtered neutron beam used by nuclear engineering.

The present beamport facilities include a high resolution powder diffractometer, a triple axis spectrometer, two single crystal diffractometers, a long wavelength diffractometer with a five detector array, a small angle scattering spectrometer in vertical geometry, and two neutron interferometers.

Significant instrumentation funding was awarded last year from the National Science Foundation (NSF), supplemented by university resources, to provide for the construction of an entirely new 15 m high-resolution horizontal geometry SANS, and the initiation of a joint program with the Ames Laboratory of Iowa State University for the upgrading and modernization of the Ames triple-axis spectrometer which was moved to MURR from the de-activated ORR. In addition to these new instruments, extensive modifications are being made in an existing diffractometer to provide a neutron reflectometer facility. Work is also underway on a new residual stress facility. It is anticipated that all of these instruments will be fully operational during the coming year, and will provide state of the art diffraction and scattering capabilities for research at MURR. A schematic drawing of the existing, new and upgraded instruments on the beamport floor is shown in Figure 1.

The MURR-MU Physics neutron interferrometry program, funded by NSF, is the only such facility in the US. The recent observation of the Aharonov-Casher effect for neutrons which has attracted wide attention was made at this facility.

In addition to neutron scattering, MURR also has a program in gamma-ray scattering which utilizes high intensity gamma sources produced by irradiation in the MURR flux trap. The facilities include a gamma-ray diffractometer MUGS, and a photon analog of a triple-axis spectrometer, QUEGS, employing the Mössbauer effect and used for quasi-elastic gamma spectroscopy. This latter instrument is currently being significantly upgraded, the result of a DOE instrumentation development grant, to include four independent gamma-ray beams and a new detector system.

An extensive effort is underway in the Electronic and Mechanical Properties group to study the effects of high energy neutron irradiation on the new ceramic superconductors and on semiconductor materials. A crystal growth effort on the YBa$_2$Cu$_3$O$_7$ has been highly successful, and the neutron irradiation produces significant enhancements in the critical current density achieved in these materials.

Summaries of the major research projects in the neutron and gamma ray scattering group and descriptions of the instrumentation upgrades are in the following pages.

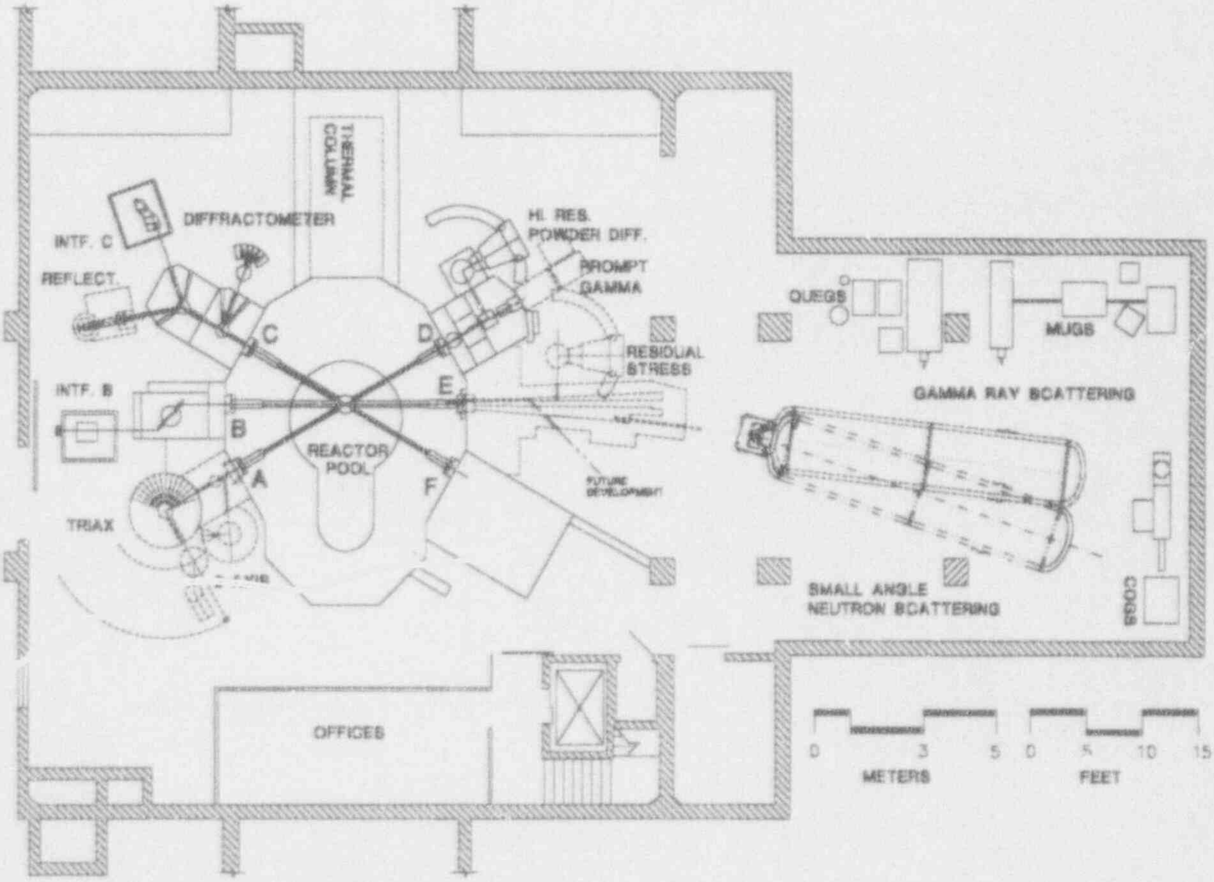

Fig. 1 Neutron scattering instruments on the beam-port floor at MURR. Instruments shown dotted are under development.

ELECTRONIC AND MECHANICAL PROPERTIES GROUP

PERSONNEL

John Farmer, Group Leader (Adjunct Associate Professor of Physics and Adjunct Associate Professor of Electrical and Computer Engineering)
Aaron Krawitz, Senior Research Scientist (MURR) and Professor of Mechanical and Aerospace Engineering
Andrew Winholtz, Research Scientist (MURR) and Assistant Professor of Mechanical and Aerospace Engineering

FACULTY ASSOCIATES

David Cowan, Professor of Physics
Jon M. Meese, Professor and Chair of Electrical and Computer Engineering

The Electronic and Mechanical Properites Group at MURR has a commonality based in materials science, but the research has a natural division between electrical and mechanical properties. The efforts in the Electronic Properties area have had a strong basis in the characterization of radiation damage in semiconductors. More recently, the focus of research has shifted to the study of defects in high temperature superconductors. The inability of these materials to carry a useful current has been a hindrance to many of the exciting applications that were originally envisioned. It is known that the introduction of irradiation defects can increase the current carrying capacity of these materials. Neutron irradiation, in particular, appears to enhance greatly the usefulness of these new superconductors. The Electronic Properties group is investigating the role of defects in the enhancement of these very exciting new superconductors.

The Mechanical Properties component of the group formally began with the addition of R.A. Winholtz in August 1991 and the change in status of Professor Krawitz after his return from a research leave in January 1992. Both are now half-time in the MAE department and MURR Center. This component is primarily concerned with neutron diffraction measurements of residual and applied stress. The current research program is focussed on the construction of a new, dedicated instrument for residual stress measurement, the establishment of a capability to measure residual stresses for industrial customers, and continued scientific research concerned with residual stress methodology and problems.

HIGH TEMPERATURE SUPERCONDUCTORS

John W. Farmer (MURR), Paul Chu (University of Houston), Marcus Kirk (Argonne National Laboratory) and Matt Kramer (Ames National Laboratory)

The Radiation Effects program at MURR has been studying the role of neutron irradiation induced defects on the ability of high temperature superconductors to carry current. The inability of the new materials to carry useful current has been a hindrance to the development of many of the exciting applications that high temperature superconductors promise. It is generally understood that the cause of the limited current capacity of the new superconductors is due to the movement of magnetic flux (fluxoids) within the sample, and recently it has been shown that neutron and other forms of irradiation greatly enhance the current capacity of these new materials. It is believed that the defects introduced by irradiation act as pinning centers which prevent or impede the motion of the fluxoids and thus lead to enhanced current capacity. MURR-based research and collaborations have focused on understanding the mechanism that produces the beneficial effects of neutron irradiation in order to optimize their beneficial effects.

Early MURR results indicate that neutrons are much more effective than most other forms of irradiation. It was postulated that the small displacement cascades (10 to 30 Å) unique to neutron irradiation are responsible for the superior effectiveness of neutron irradiation. These clusters are heavily damaged regions which act as very effective pinning centers for magnetic flux due to their near optimum size (approximately the magnetic coherence length). Materials irradiated at MURR have shown current densities among the highest in the world, currents that are adequate for most of the applications of high temperature superconductors. MURR research has also shown that neutron irradiation is far more effective than either proton or electron irradiation and may in fact be the most efficient method of all for increasing the critical current the high temperature superconductors.

More recent results indicate that the enhancement mechanism is more complicated than expected. The neutron irradiation produces a large amount of lattice expansion. More important, the expansion exhibits an non linear dependence on fluence (Figure 1). The unusual expansion suggests that oxygen vacancies play a dominant role and that it may be possible that more simple point defects can contribute the flux pinning. We have observed that the final value of the enhanced current is strongly sample dependent as well as a function of fluence. The sample dependence indicates that the irradiation induced defects are interacting with existing defects and that this interaction can play an important role in the pinning process. Further work is ongoing to understand this interaction and to find the optimum process for enhancing the critical current.

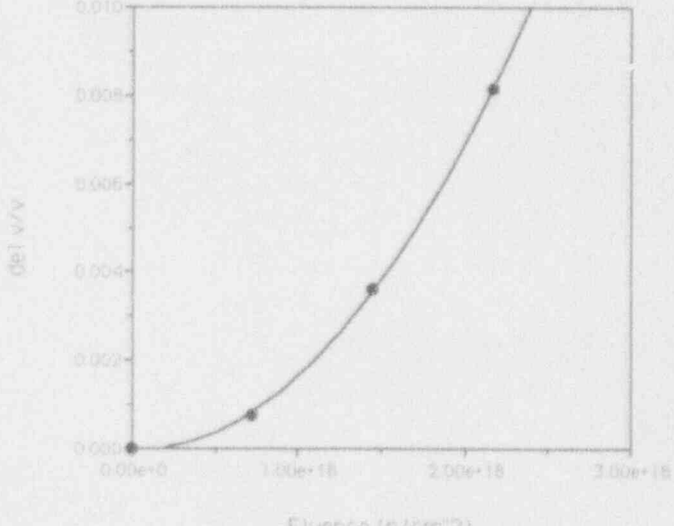

Fig. 1 The change in sample volume as a function of fast neutron fluence; the rate of change increases with fluence. This surprising result may be due to the effect of oxygen vacancies and implies that a large number of point defects are produced in neutron irradiation.

NEUTRON LINE BROADENING STUDIES

Aaron D. Krawitz and R. Andrew Winholtz
(MU Mechanical and Aerospace Engineering and MURR)

For many years studies of the broadening of x-ray diffraction peaks due to plastic deformation have been conducted using Fourier methods. The physical origins of such broadening, which correlate with a variety of physical properties, can be separated into two quantities, particle size and strain variance. In engineering alloys, particle size usually refers to the average size of dislocation cell networks (mosaic), and strain variance refers to the local variation in interplanar spacing, also due to dislocations. The object of the study is to apply the method to neutron diffraction patterns. This involves adapting an existing x-ray analysis program, determining minimum breadths and intensities suitable for analysis, and comparing results with simpler integral peak breadth methods previously used in our work. Such an application has only become possible with the construction of high resolution neutron powder diffractometers, such as the instrument on beamport D.

A series of rolled copper plate samples (22, 40, 59 and 72% reductions in cross-sectional area) have been measured, with matching standards recrystallized to remove cold work. Results indicate that the percent differential broadening (%DB) measured for the lower angle peaks, ~45°2q, is not sufficient without extended data collection time, due to the instrument resolution function. The rule of thumb for x-rays is that a minimum of 20%DB is needed for good results. Neutrons may require a higher minimum because the run times needed for statistically sufficient intensities become impractical at the lower percentages. Experiments will be also be performed on cemented carbide composites.

RESIDUAL STRESSES IN ADVANCED SOLID ROCKET MOTOR CASING

Aaron D. Krawitz and R. Andrew Winholtz
(MU Mechanical and Aerospace Engineering and MURR)

The Advanced Solid Rocket Motor (ASRM) is the next generation solid rocket booster for the NASA Space Shuttle program. A project has been funded to characterize the residual stress state associated with the welding procedure in the vicinity of the weld at representative locations, particularly in the steady state weld region. The actual sample to be investigated is a sub-scale model of an ASRM cylindrical section. The ASRM is assembled from a series of circumferentially welded sections of a high strength (HP9430) steel.

Residual stresses associated with the steady state weld will be determined in the weld and unaffected base metal regions of a subscale cylinder of approximately 36 in. diameter by 0.59 in. wall thickness. Regions of specific interest include the weld center line, first pass heat affected zone, just outside the cap pass heat affected zone, and the base metal. About five positions through the wall will be investigated. Data collection will provide information concerning:

1. The sign and magnitude of the residual stress state.
2. The directions of the principal stress axes.
3. The effect on the stresses of a post weld heat treatment on the as-welded stress state.
4. The effect of a hydroproof test on the post weld heat treated cylinder.

This work is supported by a research contract with Babcock and Wilcox.

RESIDUAL STRESS IN METAL MATRIX COMPOSITES

Aaron D. Krawitz and R. Andrew Winholtz (MU Mechanical and Aerospace Engineering and MURR) and R.J. Arsenault (University of Maryland)

Modern composite materials are typically combinations of two mechanically combined phases for the purpose of obtaining enhanced physical properties, most commonly strength. A major class of composites consists of a metal matrix reinforced with a high strength ceramic component. They are mixed, bonded at elevated temperature, then cooled. Because of the different properties of the constituent phases, the rates of contraction upon cooling are typically very different, creating what are known as residual microstresses. Neutron diffraction is particularly useful for the measurement of such stresses because of their extreme depth of penetration relative to more conventional x-rays. This enables good volume averages to be obtained, even on actual parts, and avoids surface preparation problems.

The nature and role of such residual stresses in metal matrix composites has been the subject of two studies conducted at MURR in collaboration with Prof. R.J. Arsenault at the University of Maryland. The first study involved measurements of microstresses in silicon carbide (SiC) reinforced 1100 aluminum (Al) alloy. Measurements were made on samples containing 20 volume percent (V%) SiC in the form of whiskers (length-to-diameter ratio ≈ 2, diameter ≈ 0.5 mm), platelets (major diameter-to-thickness ratio ≈ 3, diameter ≈ 70 mm) and spheres (diameter ≈ 0.5 mm). Results indicate that the average stresses in the Al matrix for whisker and spherical SiC reinforcement are close in value and greater than for platelets (Table 1). These results differ from finite element model predictions and indicate that particle spacing, as well as shape, determine the magnitude of thermal residual stresses. It is also noted that the general stress level is quite low, indicating the sensitivity of the method.

Table 1 **Thermal residual stress in the Al phase of 20 V% SiC/Al composites**

SiC Morphology	Transverse Stress (MPa)	Longitudinal Stress (MPa)
Whiskers	58±20	110±19
Spheres	94±20	100±19
Platelets	32±16	43±16

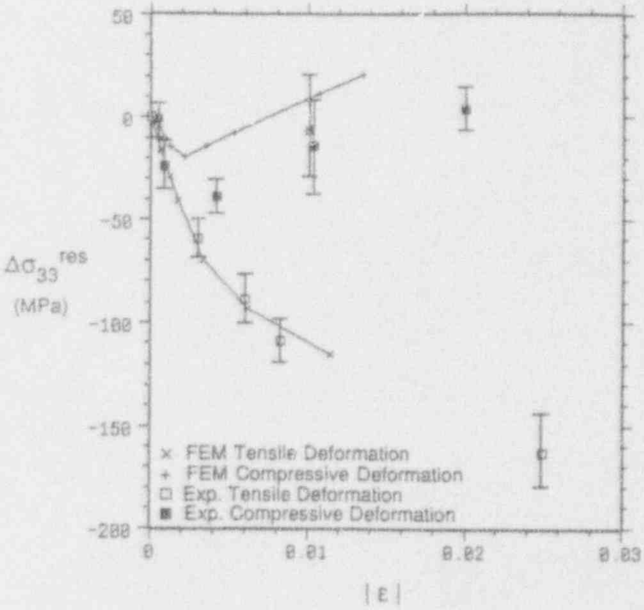

Fig. 1 Deformation-induced changes of average matrix residual stresses in the longitudinal direction in SiC/Al as a function of applied tensile and compressive strain.

The second study monitored changes in SiC whisker (20 V%) reinforced 6061 Al composites subjected to deformation. Measurements were made using the actual mechanical test samples. The changes were found to be asymmetric in response to uniaxial external tensile or compressive loads applied along the longitudinal whisker axis.

The changes in the longitudinal direction are shown in Figure 1. The initial residual stress in the undeformed samples is about 150 MPa, i.e. the changes due to tensile deformation are substantial. Finite element and analytical models were developed to predict the behavior, which was found to arise due to a gradient of plastic flow in the matrix. The relation of such an asymmetry to residual stresses has been a subject of controversy.

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RESIDUAL STRESS AND STRESS DISTRIBUTION IN TUNGSTEN CARBIDE-NICKEL COMPOSITES

Aaron D. Krawitz and R. Andrew Winholtz (MU Mechanical and Aerospace Engineering and MURR)

Tungsten carbide (WC) particle reinforced composites are the basic class of materials used for high performance cutting tool and rock drilling applications. While the most common grade is WC-cobalt (Co), in our present work, the metallurgically simpler metal nickel (Ni) is used. These materials are produced from powders sintered at high temperature (≈ 1400°C). Due to large differences in their coefficients of thermal expansion, large thermal residual micro-stresses develop upon cooling and have been characterized using neutron diffraction. In addition, diffraction peaks are observed to be significantly broadened due to a range of residual stress around an average value. The origin of this broadening is not clear but has potential implications for failure of the material in service. The purpose of this project is to investigate the nature of this broadening using neutron diffraction and finite element modeling (FEM) methods.

Preliminary FEM results indicate that the angularity of the WC particles and their distribution and interaction have a strong effect on the stress distribution and magnitude. FEM models are being developed to match previous neutron diffraction data and a new set of samples has been produced and measured. As a result of this work, the stress magnitude and distribution is understood to be related to particle shape and size, mean free metal binder spacing, local variations in metal binder spacing, and the *in situ* yield strength of the metal, which is affected by dissolved tungsten content as well as particle spacing.

Thermal residual strains in WC-Ni composites of 11, 20, 40 and 60 V% WC were measured using neutron diffraction techniques. Cell parameters and volume fractions of the phases were determined using the Reitveld profile refinement method. Gaussian fits to Ni (111, 200, 311 and 222) and WC (101, 110, 002, 102, 201 and 112) peaks were also made. The strains for the *a*- and *c*-directions in WC were compressive for each sample, as expected considering the relative thermal expansion coefficients of WC and Ni. Within experimental error, the magnitude of the compressive strain in each direction decreased as the WC volume fraction increased. The *a*-strain exceeded the *c*-strain in each sample as predicted by analytical and FEM models.

The lattice parameters for Ni increased with increasing WC. They are influenced by both the stress in the Ni phase and W in solid solution. Preliminary calculations indicate that for 8 atomic percent dissolved W, the volume-averaged strains in the Ni phase of all composites are tensile and increase with WC. These results are consistent with analytical and FEM models, and previous neutron diffraction measurements. The volume fractions of WC and Ni calculated by GSAS for each sample were in good agreement (differing by a maximum of 0.32 V%) with those determined from density measurements.

Ni peak breadth increased with increasing WC content, in agreement with FEM calculations which show that binder strain distribution increases with strain magnitude, and there is also an increase in WC peak breadth for the 11, 20, and 40 V% samples. Micrographs show an increase in particle interaction as V% increases. FEM models of isolated WC particles show the particles to only be in compression, whereas WC-Ni models involving interacting WC particles reveal substantial stress distribution in this phase, reaching into the tensile region. Such a distribution would contribute to the peak breadths. The mean free paths of the Ni of the 40 and 60 V% samples are close, suggesting that the trend of increasing strain distribution with increasing strain magnitude dominates the influence of particle interaction for composites of higher WC content.

MAGNETIC AND CRYSTAL STRUCTURES GROUP

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Ronald R. Berliner, Senior Research Scientist (Adjunct Associate Professor of Physics and Adjunct Associate Professor of Electrical Engineering)
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Mihai Popovici, Visiting Research Scientist (since September 1991)
Olivier Isnard, (Visiting) Research Specialist (since June 1992)

During the past year, this group has concentrated on research with and about the position sensitive detector (PSD) diffractometer, and with the single crystal diffractometer on beamport E. In addition, design work on the new single crystal diffractometer (sharing beamport A with TRIAX) has continued.

The most exciting results have involved the development of bent perfect crystal techniques. In place of a conventional mosaic monochromator, an elastically bent perfect Si crystal has been put into use at the PSD diffractometer. Although the effective mosaic of this crystal is about a factor of 10 smaller than for the Cu crystal perviously in use, it is used without collimation and thus receives a much higher indicent flux. This factor, plus the use of an asymmetric geometry, leads to a net gain in flux on sample. The choice of radius of curvature leads to phase space focussing effects and to better resolution than with mosaic crystals and collimation. This success has led to the consideration of these methods to other facilities including the new single crystal diffractometer, for which a double crystal monochromator in antiparallel configuration can give large intensity gains while preserving resolution. A proposal to DOE to develop these methods has been prepared.

Research with the PSD has concentrated in three areas: hard magnet materials, including nitrided phases; hydrogen (deuterium) in Nd2Fe14B and Ti based alloys and high Tc superconductors. Research in the first area produced one *Physical Review Letter* and an invited talk at the Intermag Conference (St. Louis, April 13-16, 1992), while the second area featured the exchange visit of Olivier Isnard from the CNRS Grenoble, to carry out measurements previously scheduled for the ILL and no longer possible there due to the outage of the reactor. The high Tc area has seen work on both 1-2-3 compounds and BISCO related phases.

Single crystal diffraction studies included cis $[Co(en)_2(NO_2)_2]NO_2$, $MgHPO_3\Sigma 6H_20$, $NaH_5(PO_3)_2$ and $YBa_2Cu_3)_{6.5}$. The analyses on all of these are in progress.

BENT PERFECT CRYSTAL MONOCHROMATORS ON THE PSD II HIGH RESOLUTION NEUTRON DIFFRACTOMETER

M. Popovici, W.B. Yelon and R. Berliner (MURR)

Two bent perfect crystal monochromators were succesively implemented at MURR on the high resolution powder diffractometer with position sensitive detection. Large-diameter silicon plates were bent in pneumatic devices in which vacuum or air pressure was used to produce spherical curvatures by pressing the crystals against O-ring surfaces. The curvature was controlled either by the pressure differential or by a spherical mechanical limit.

The old configuration of the instrument used a 15' in-pile collimation and a Cu (331) monochromator. The in-pile collimation was removed and the Cu monochromator was replaced by an elastically bent Si plate 3 mm thick, set in reflection from (511) planes. The plate had a [100] orientation and thus the reflection geometry was asymmetric. Data for two specimens were compared in detail: an annealed Ni sample, 1/8" diameter and 2" high, and a standard Al_2O_3 specimen 1/4" diameter and 2" high, which we use for instrument calibration. With the Cu monochromator the resolution was reasonably good, but a noticeable l/2 contamination was present. With the silicon monochromator, an improvement in resolution, despite the somewhat shorter wavelength, was apparent for both samples, with no l/2 peaks. Normalized for equivalent counting times, the Ni peaks at high angle were 50% higher with the silicon monochromator than with the copper.

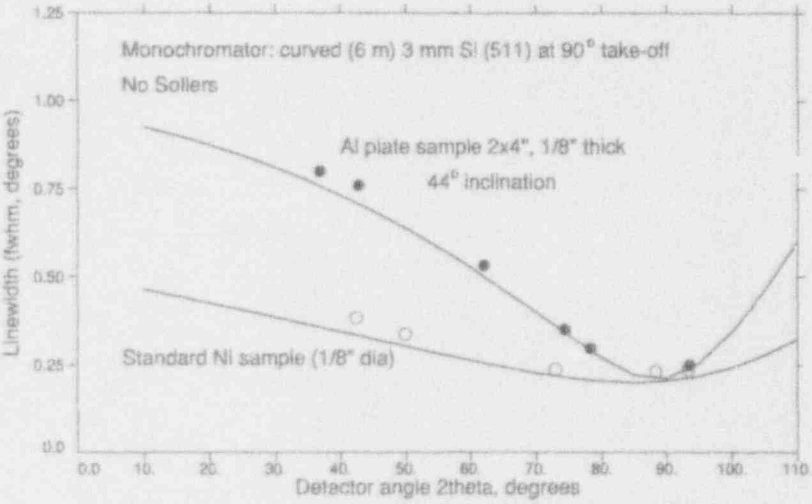

Fig. 1 Bent perfect monochromator on MURR PSD, plate vs. cylindrical sample

The computations were found to give a fairly good description of the measurements. As an example, Figure 1 shows experimental and computed linewidths for two cases, the 1/8" diameter Ni specimen and a large plate Al specimen 1/8" thick, oriented to optimize the resolution at high angle.

A second Si monochromator was subsequently installed in this instrument. The crystal plate was cut obliquely from a [100] ingot to give a more pronounced beam condensation effect for the (511) planes. The crystal was 6 mm thick and curved by a 90 psi pressure to a radius of 14 m. The resolution at high angle was preserved and there was a notable improvement in both resolution and background at low angle, as well as a further gain of 2.5 in peak intensities. This configuration is close to the best achievable with elastic bending and is expected to be our normal one for the immediate future. At the present time, however, the monochromator is used with a (331) reflection for residual stress measurements and for structure determinations of magnetic materials. The larger wavelength puts close to 90° the diffraction line (211) convenient for stress determinations in steels, and allows better separation of the crowded lines with significant magnetic information in the materials under study. This illustrates a useful option available, that of choosing from more than one plane of a given crystal.

A relatively straightforward extension of this case would use elastic bending in the horizontal plane and pseudo-bending in the vertical plane to create a polygonal approximation to a curvature for strong vertical focusing (radius of 1.25 m needed). This would give an additional intensity gain of almost two on the existing diffractometer. The mechanics of such a system are, however, more complex and development of ideas are underway. Thicker crystals could be used, with an intensity gain proportional to thickness and little resolution loss, if they could be bent to the necessary radii. Thick silicon, however, cannot elastically support such radii and the current system is close to the breaking limit.

NEUTRON POWDER DIFFRACTION STUDIES OF MAGNETICALLY HARD PHASES

W.B. Yelon, H. Xie (MURR), G.J. Long, O.A. Pringle, W.J. James (UMR Physics), K.H.J. Buschow (Philips, Eindhoven), F. Grandjean (University of Liège, Belgium), G.C. Hadjipanayis (University of Delaware), D.J. Sellmyer, S. Jaswal (University of Nebraska), O. Isnard, D. Fruchart (CNRS, Grenoble), F. Pourarian (Central Methodist University)

The largest use of the high resolution powder diffractometer has been for research in magnetism. This has been an interest area at MURR for the past 16 years and our excellent facility and expertise has led to a growing circle of interactions. There is a continuing interest in new Fe based compounds with Curie points comparable to $Nd_2Fe_{14}B$. These include nitrides of rare earth (R) compounds R_2Fe_{17} and $RFe_{12-x}T_x$ (T=transition metal), substituted R_2Fe_{17} compounds and others.

Reasearch on the nitrides has revealed the location of the nitrogen atoms in the crystal structures.[1,2] In all cases the N atoms are found only in the single largest interstitial site, and the occupancy can be varied from 0 to about 90% full. The lattice expansion depends upon the occupancy as do the magnetic properties. Band structure calculations carried out in relation to the NPD measurements suggests that the dominant effect is the change in band structure with Fe-Fe distance and not any effect of electron transfer with the N atoms. For a given rare earth (e.g., Nd), it is found that the Curie point increases (perhaps linearly) with average Fe-Fe bond length (Figure 1).

The substitutional systems $R_2Fe_{17-x}T_x$ (T=Al,Si) have been studied. Both systems show enhanced Curie points compared to the pure Fe compound and our research has focussed on the origin of this effect. It is found that the Al compounds show strong lattice expansion while the Al shows a diminished preference for the c-site, the one most strongly connected with good magnetic properties (Figure 2). These two effects overcome the effect of Fe dilution. On the other hand, the effect of Si is more mysterious; the lattice does not expand and the site preference effects do not clearly provide an explanation for the enhancement. Further studies of substitutional compounds

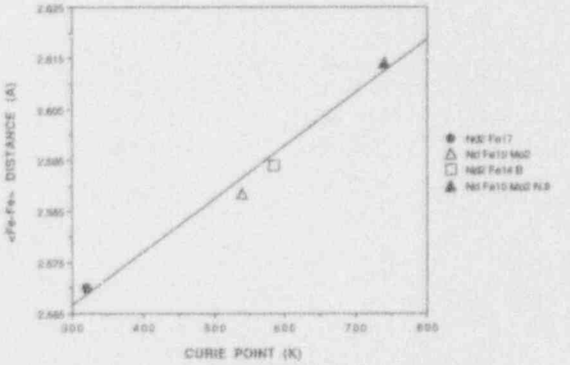

Fig. 1 Curie point versus average Fe-Fe bond length in Fe rich Nd based compounds

are underway and may provide insight into this problem and lead to the development of better materials.

The effect of D in $Nd_2Fe_{14}B$ has been studied as well. Samples prepared in France with D content varying from 0-4/f.u. are being studied between 10K and room temperature in order to determine both the D site occupancy, thermal motion and the changes in magnetic moments with concentration and temperature. These sample are unique in being well crystallized, single phase specimens and the final results of the study should be quite interesting and definitive. The analysis of these data are being performed using a Rietveld code brought from the ILL which allows magnetic refinement of multiphase samples. While less user friendly than the GSAS program, it should be a useful addition to our library.

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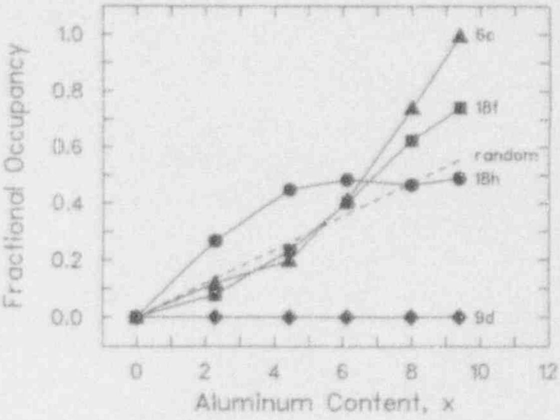

Fig. 2 Fractional occupancy of Al on the four Fe sites in $Nd_2Fe_{17-x}Al_x$

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NEUTRON MICROFOCUSING BY DOUBLE REFLECTION ON CURVED PERFECT CRYSTALS

M. Popovici and W.B. Yelon (MURR)

Double crystal monochromators with two identical flat crystals in parallel (+,-) setting are quite common, both for neutrons and synchrotron radiation (SR). The parallel setting is known to be non-dispersive: all radiation diffracted by the first crystal, regardless of wavelength, automatically has the right conditions to be diffracted by the second one. An equivalent of this setting exists too for diffraction from two bent crystals. The neutron results on the optics of diffraction by two bent crystals[1] were particularized to the point-source SR case.[2,3] Computations for the SR case have shown that for curved crystals, the distinction between parallel (+,-) and antiparallel (+,+) configurations is no longer relevant from the intensity point of view. An essential difference, however, is that the full beam intensity can be easily condensed into a narrow focus in the antiparallel setting. We returned to the neutron case in a recent paper[4] for an examination similar to that presented for SR.[3]

For a neutron double monochromator required to image the source at the sample position, the design relations were derived by accounting for the condition of efficient double diffraction in the focusing equations. The configuration with the strongest demagnification appears indeed to be that with crystals in antiparallel (+,+) setting. The instrument resolution can be adjusted to put the minimum of the diffraction linewidths at a desired position. The small radii of curvature necessary for microfocusing can be achieved with plastic bending techniques only. The peak reflectivity decrease on bending then becomes important. This restricts the choice of useful planes to strongly reflecting ones.

As an example, a microfocusing double monochromator for stress scanning has been considered. In such measurements the strain is probed by recording a diffraction line in the vicinity of 90° for a sample volume limited (by beam-defining slits) to several cubic millimeters. Computed data on a configuration meeting the requirement of narrow diffraction lines in the vicinity of 90° are shown in Figures 1 and 2. The distances from source to first crystal and between crystals were fixed to the actual values for a MURR beamport where a stress instrument is to be installed. The instrument is being designed to have a single, elastically bent, Si (511) monochromator in strongly asymmetric reflection. For the double monochromator, plastically bent Ge (400) crystals in symmetric reflection were considered. Vertically, the first crystal was taken to be flat and the second crystal optimally curved.

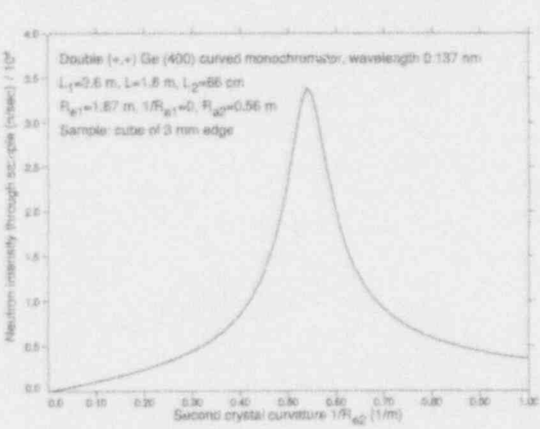

Fig. 1 Intensity through sample (3x3 mm area) vs. second crystal curvature, for a double monochromator with plastically bent Ge (400) crystals in antiparallel setting (thermal source flux 10^{14} n/cm²/sec).

The computed minimal beam widths are below 6 mm in the diffraction plane and 1 cm in the vertical plane. The neutron intensity through a 3x3 mm sample (source flux 10^{14} n/cm²/s) is shown in Figure 1 as a function of the second crystal horizontal curvature. At maximum, it is 34 times larger than that computed for the elastically bent Si (511) monochromator. The radii of curvature should be set quite accurately, with a precision of about 2%. The dependence of the powder diffraction linewidths on the detector angle is shown in Figure 2 both for the single Si (511) monochromator and the double Ge (400). In the microfocusing case the lines broaden considerably on going away from 90°. At minimum, the linewidths are still worse than in the single monochromator case. However, in a limited range around 90°, there is a large intensity gain from spatial focusing (factor of about 15) that more than compensates for the broader linewidths. The linewidths in Figure 2 were computed for no sample broadening. When the sample broadening is important, as sometimes happens, the intensity gain may approach that of the intensity through sample.

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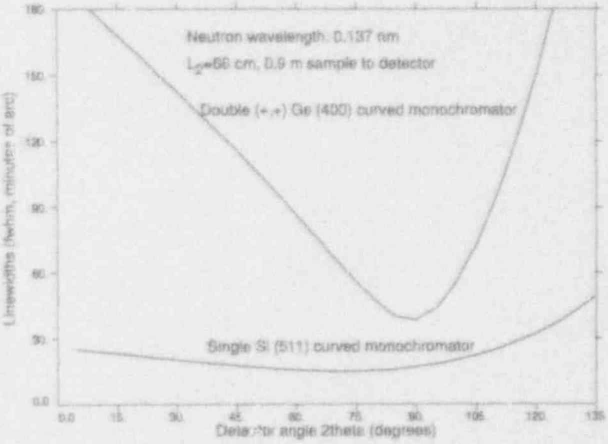

Fig. 2 Neutron powder diffraction linewidths vs. detector angle for the configuration considered in Figure 1 and for an elastically bent single Si (511) monochromator.

DETERMINATION OF REFLECTIVITY CURVES OF MULTILAYER NEUTRON MONOCHROMATORS

M. Popovici, B.J. Heuser, W.B. Yelon (MURR) and J.E. Keem (Ovonic Synthetic Materials Co.)

To anticipate the performance of a synthetic multilayer monochromator in a SANS instrument under construction at MURR, four Ovonics Ni-Ti multilayers deposited on Si [100] thin wafers were investigated. Two of the multilayers had 60 Ni-Ti bilayers with 10 nm nominal spacing; the other two had 30 bilayers with 9 nm spacing. The measurements were made on the MURR 3-axis neutron spectrometer by rocking the multilayers against the Si (111) monochromator.

The initial results were confusing; conventional rocking curves appeared to depend on whether the face (deposition side) or the back of the multilayer was measured and on the portion of its surface that was exposed to the beam. The multilayers turned out to be curved (convex), due to deposition-induced stresses. Further checks indicated that the layer spacing was also non-uniform over the surface. It became clear that the conventional technique of measuring and interpreting double crystal rocking curves should be adapted to the peculiarities of synthetic crystals: due to the shallow Bragg angles the separation of the incident and reflected beams is small; the multilayers may be curved and, unlike natural crystals, may have d-spacing non-uniformity.

The most convenient measurement configuration was found to be that with tight collimation in front of a narrow detector and with measuring the neutrons **transmitted** through the multilayer. Formulae for the interpretation of such rocking curves were derived, multilayer curvature and lateral gradient of layer spacing accounted for. The radii of curvature R and the lateral gradients g were determined by scanning the multilayer surfaces. By measuring the reflection from the face and back of the multilayers, R and g could be determined separately. Radii of curvature of several tens of meters and lateral variations in layer spacing of the order of 1% cm^{-1} were found in all cases. Consideration of their effects turned out to be essential in understanding observed rocking curves.

Figure 1 shows the results of diagnostic measurements for a 30-layer along one of the [110] directions of the support wafer. The changing slopes of the variation of the transmission rocking curve position on translating the multilayer give corresponding changes in the rocking curve peak and width values. Minimal widths and best peak values were expected to correspond to some optimal slopes (shown in Figure 1) and this did indeed happen, as seen in Figure 2.

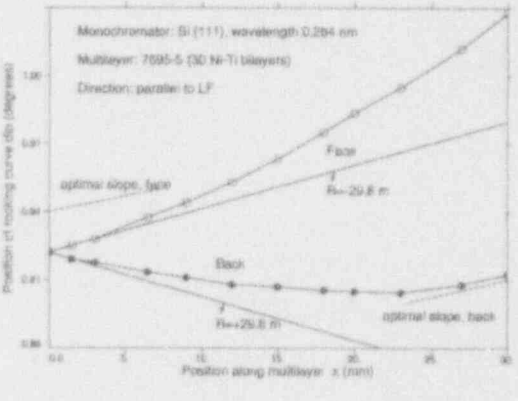

Fig. 1 Variation of the Bragg angle scale dip position of the transmission rocking curve on translating a 30-layer parallel to its large flat (LF).

The measured rocking curves showed some structure between the total reflection range and the outer dips due to Bragg reflection for the nominal d-spacing, indicating that the Fourier transform of the multilayer contrast distribution is different from zero not only in the proper Bragg range. Data obtained under different conditions showed that the correction procedures were adequate and gave consistent reflectivity parameters. The observed peak reflectivities were 80% for 30-layers and 94% for 60-layers. No real discrepancies were noticed between measured and theoretical widths (~0.8 mrad).

A detailed account of this work will be presented at the International Symposium *"Neutron Optical Devices and Applications,"* San Diego, July 1992.

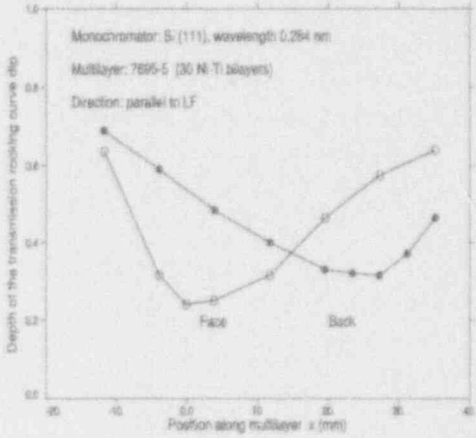

Fig. 2 Variation of the depth of the rocking curve dip on translation ǁ to LF (same multilayer).

DESIGN COMPUTATIONS FOR SYNCHROTRON RADIATION CURVED TWO-CRYSTAL MONOCHROMATORS

M. Popovici and W.B. Yelon (MURR)

The design of curved two-crystal monochromators for synchrotron radiation (SR) has been considered in two papers[1,2] starting from earlier neutron optics results.[3] Both for neutrons and SR, one can ensure that all the radiation diffracted by the first crystal automatically have the right conditions to be diffracted by the second one. For this to happen, one has to choose the right curvature for the second crystal in the SR case, and the right curvature **and** the right reticular spacing in the neutron case. The difference is due to the fact that in the diffraction plane an SR source is sharp while a neutron source is usually wide.

The first paper discussed the problem of overcoming the heat load problem in SR double monochromators by suitably curving the second crystal to accommodate the curvature of the thermally distorted first crystal. The second paper discussed also the possibilities of achieving large demagnifications. Results of computations with a neutron optics program adapted to the SR specifics were presented in both papers. The adaptation consisted merely of working with appropriate perfect crystal reflectivity parameters and with effective crystal thicknesses given by the absorbtion depths. An automatic generation of those parameters will have to be implemented to make the program suitable for general use in SR computations.

The heat load problem In flat SR double monochromators, there is a loss of intensity as the flat second crystal no longer efficiently diffracts the beam given by the thermally distorted first crystal. Our computations referred to the monochromator on the A_2 beamline at the Cornell High Energy Synchrotron Source (CHESS) for which the heat load effects are well documented. Figure 1 shows the rocking curve width and peak intensity vs. the second crystal curvature R_1 for two radii of curvature of the first crystal. It is seen that by curving the second crystal to the right radius, the rocking curve width is practically restored to its flat-crystal value. The peak intensities are slightly higher than two flat crystals, due to the reflectivity increase on curving. For a heat load ten times higher, computations showed that the lattice spacing gradient in curved crystals no longer allows the rocking curve widths to be restored to their flat-crystal values, but the residual broadening is accompanied by a marked intensity increase. This effect is related to the large absorbtion depth of silicon (about 100 microns at 20 keV) and for germanium it is virtually absent. This makes silicon double monochromators superior to germanium ones at high energies and high heat loads.

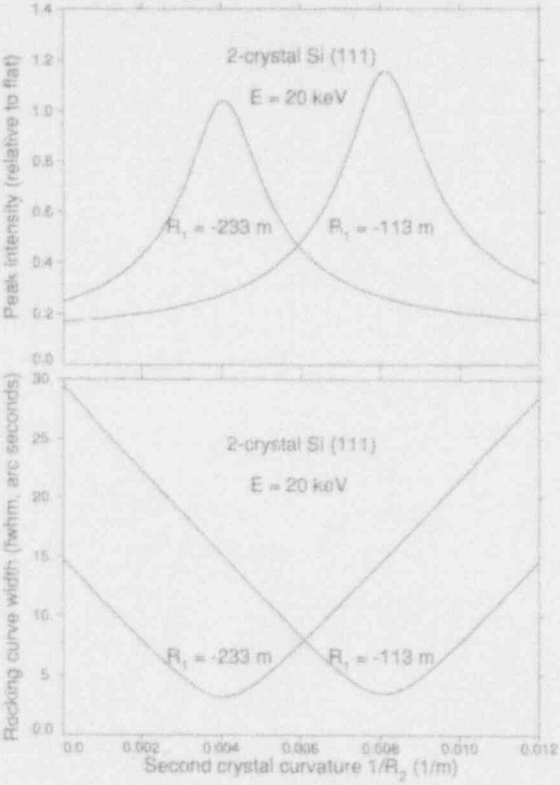

Fig. 1 Computed dependence of the rocking curve widths and peak intensities on the second crystal curvature R$_1$ for the geometry of the CHESS A$_2$ beamline. The radii of curvature of the first crystal correspond respectively to the gallium-cooled (-233 m) and water-cooled (-113 m) Si (111) double monochromators set to 20 keV at a power load of about 0.4 W/mm^2.

Microfocusing With identical crystals in antiparallel symmetric reflection, large demagnifications can be obtained just by correctly choosing the distance between the two crystals. Computational results are shown in Figure 2 referring to a possible optimal configuration for microfocusing on the same CHESS A$_2$ beamline with a mixed Si+Ge monochromator (the smaller absorption depth of germanium gives a lower aberration due to beam penetration).

It is seen that the setting of crystal curvatures should be very precise and the focus is expected to be very sharp (5 µm). In contrast to the situation for flat crystals, in the (+,+) configuration the full beam intensity is higher than for two crystals in parallel (+,-) position. The beam flux (number of quanta per unit area) at focus is expected to be larger than in the present configuration by a factor of about 1000. A further enhancement by a factor of 100 is possible through saggital focusing.

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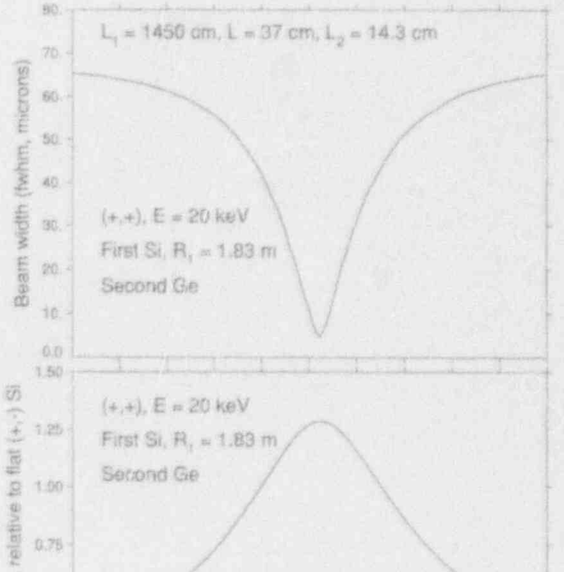

Fig. 2 Computed beam intensity and width at focus for a possible configuration with strong image demagnification on the same geometry, using a first Si and a second Ge (111) curved crystals of optimal curvatures.

NANOSTRUCTURES AND EXCITATIONS GROUP

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The research work of this group is primarily focused toward disordered (amorphous or crystalline) materials and materials with a substructure of order 10 or more lattice spacings. The group in concert with other groups in the Neutron Materials Science Program has the responsibility for the development of the new 15m Small Angle Scattering spectrometer (SANS) and also for the redesign and upgrade of the Ames Triple Axis spectrometer (TRIAX) in a collaborative effort with C. Stassis and A. Goldman from the Ames Laboratory of Iowa State University. Both the SANS and TRIAX programs are the recipient of Instrument Development Grants from the National Science Foundation for construction of these state of the art instruments. The SANS is funded under NSF grant DMR-8915199 in the amount of $395,000 (including MU match amount) and the TRIAX under NSF grant DMR-9103383 in the amount of $419,000 (including UM, MU and Ames Laboratory matching amounts).

PHYSISORBED MONOLAYERS

K.W. Herwig (MURR and MU Physics), P. Dai, S. Wu, B. Matthies (MU Physics) and H. Taub (MU Physics and MURR)

Our investigations have focused on the question of how the steric properties of a molecule influence its behavior when physisorbed onto a solid substrate. The physical size, shape, and flexibility of a molecule can play a definite role in determining such properties as the melting behavior of a monolayer film[1] and the growth mode of multilayer films. This issue poses a problem of considerable interest in surface science and impacts on such basic areas as the theory of wetting as well as technological problems related to lubrication and adhesion.

We have sought to address this problem by investigating the behavior of a series of isostructural rod-shaped molecules, the *n*-alkanes (C_nH_{2n+2}), physisorbed on graphite. Our goal has been to determine the effect of the molecule length (n) on the monolayer melting behavior and the wetting properties of multilayer films. In the following, we describe two experiments each designed to address one of these issues.

Quasielastic scattering from hexane physisorbed on graphite Elastic neutron diffraction studies have demonstrated that the melting transition in the *n*-alkanes physisorbed on graphite differs greatly among representative molecules although they have similar solid monolayer structures.[2,3] The first three members of the series with an even number of carbon atoms (ethane, butane, and hexane) have a herringbone structure in their solid submonolayer phase, with a melting temperature which increases monotonically with molecule length. However, the butane monolayer exhibits an abrupt, first-order melting, while the hexane monolayer retains a large degree of translational order even at room temperature.

Molecular dynamics simulations[1,4] have given welcome insight into the differences in the melting behavior of these two alkane monolayers while suggesting a general melting mechanism in monolayers of flexible, rod-shaped molecules. In both cases, melting requires the formation of vacancies in the film by motion of the molecule perpendicular to the substrate. In butane, this occurs by tilting of the molecule, lifting one end slightly off the surface, to create the required excess area. For the longer hexane molecule it appears that this excess area is created by a conformational change of the molecule which reduces the size of its footprint on the surface.

We have employed quasielastic neutron scattering (QNS) to probe the dynamical behavior of these two molecules adsorbed on graphite both above and below the melting temperature. These experiments have been carried out on the QENS spectrometer at Argonne National Laboratory's Intense Pulsed Neutron Source. In summer 1992 we were allocated two weeks of beam time which we used to continue earlier experiments. Our goal was to return with a complete (in the sense of covering a broad range of temperatures and momentum transfers) set of data for a near monolayer coverage of hexane on graphite.

Typical spectra in QNS contain an elastic component whose strength is related to the fraction of molecules which, on the time scale set by the instrumental resolution and length scale set by the momentum transfer, are static. The second component is typically assumed to be Lorentzian in shape, with a Doppler broadened width associated with the diffusive behavior, either rotational or translational, of the molecule.

Figure 1 shows the widths of this Lorentzian component for both hexane and butane as a function of reduced temperature, T_R, which is the sample temperature divided by its melting temperature, T_M. To within 10 K of T_M, physisorbed butane exhibits no observable quasielastic scattering on either of the two length scales defined by the momentum transfers of Figures 1(a) and (b). In contrast, hexane clearly exhibits quasielastic scattering at temperatures well below T_M.

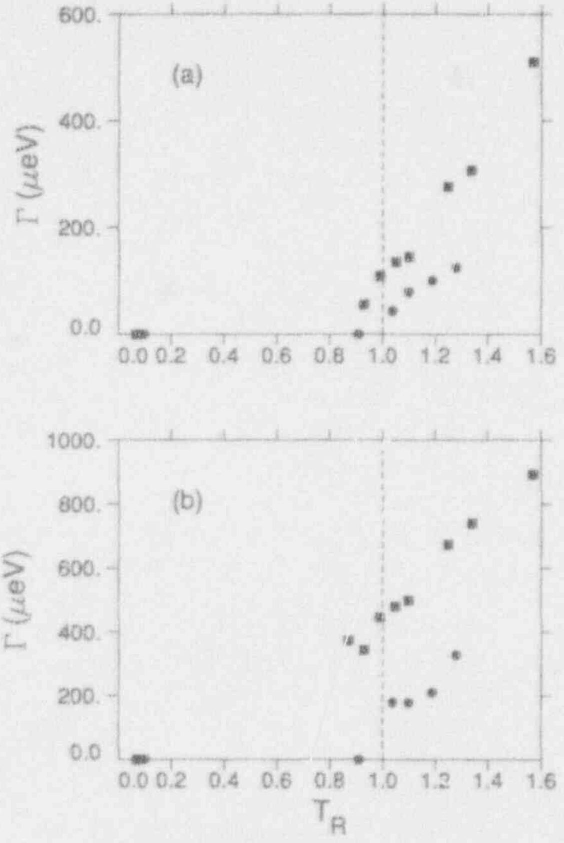

Fig. 1 The HWHM, G, of the Lorentzian component in the quasielastic spectra of butane (filled circles) and hexane (filled squares) physisorbed on graphite versus reduced temperature. The dashed line indicates the melting temperature in both plots. Plot (a) shows the data collected at a momentum transfer Q of 1.2 Å^{-1} while plot (b) shows the data for Q = 2.5 Å^{-1}.

These data are entirely consistent with the diffraction data which demonstrated that melting in butane occurs in an abrupt first-order manner while the melting behavior in hexane occurs over a much broader temperature range. The source of the quasielastic scattering observed in hexane below melting remains a question to be answered by the computer simulation. A likely candidate is the conformational change of hexane predicted by the simulation to occur prior to melting of the monolayer. A more complete interpretation awaits calculation of the intermediate scattering functions from the simulation results.

The multilayer growth of butane on graphite

In the past year, our measurements using the C-port diffractometer have focused on the layer-by-layer growth of butane on graphite. Previous diffraction studies[3] demonstrated that butane forms a herringbone structure with a unit cell similar to the (011) plane of bulk butane (the largest lattice spacing mismatch is approximately 5%). Because of this similarity to the bulk solid, we believed butane was a likely candidate for growing crystalline multilayer films on graphite.

During the course of our experiment, we collected approximately 100 data sets, covering a range of temperatures from 11 to 138 K, and a range of coverages, Θ, corresponding to the material required to form 0.9 to 6.0 monolayers (ML). The large "background" associated with the substrate and the relatively weak signal from the adsorbed molecules required long experimental runs in order to obtain good statistics in the difference data. Approximately two days of beam time were required to obtain a good diffraction pattern at the lower coverages.

Figure 2 shows the low temperature, 11 K, data for all the coverages studied. Diffraction peaks associated with the monolayer are observed as part of the diffraction pattern at all coverages studied, indicating the continued presence of only one layer of crystalline film on the graphite surface. The strong diffraction peaks which continuously increase in strength are associated with the growth of bulk crystallites. Analysis of this data indicates that bulk crystallites begin to grow when Θ exceeds 1.8 ML.

Contrary to our expectations, it appears that a herringbone monolayer forms a poor template for epitaxial growth of crystalline overlayers. The reason for this may be that in bulk butane substantial tilting (greater than 11 deg) of the molecules occurs relative to the (011) plane while butane lies flat on the graphite substrate. This difficulty of accommodating to the molecular orientation in the monolayer may be a critical factor in achieving multilayer growth.

The intensity of the diffuse scattering in regions where no Bragg peaks from either the film or the bulk crystallites are present may be associated with the amount of disordered material present in the sample cell. This intensity clearly starts to increase when Θ exceeds 1 ML. This observation indicates that some disordered material is present in the film at the onset of bulk growth (at Θ = 1.8 ML), and the amount continues to increase during bulk growth.

Analysis of the temperature dependence of the diffraction pattern at Θ = 2.2 ML demonstrates that the monolayer component persists to 128 K, 12 K above its melting temperature at Θ = 1 ML. This effect can be interpreted as evidence for molecules in a second layer, stabilizing the first layer so as to raise its melting temperature. These molecules must be disordered with respect to molecules in the first layer as they do not affect the monolayer diffraction pattern.

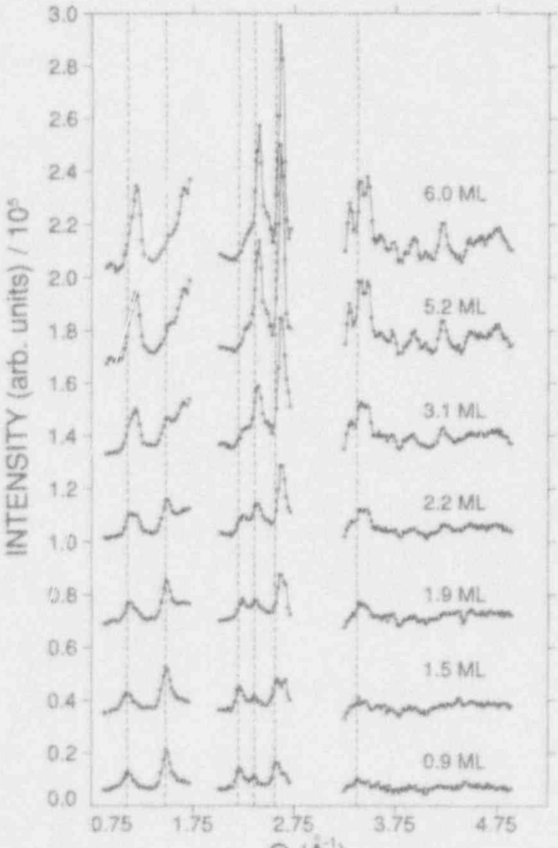

Fig. 2 Elastic neutron diffraction data from butane adsorbed on graphite. The background signal due to the bare graphite and aluminum sample cell has been subtracted, and data have been suppressed in regions of strong substrate scattering. The data points have been connected for clarity. Dashed lines indicate the location of the diffraction peaks associated with the monolayer film.

The melting temperature was determined at two coverages, Θ = 2.2 and 3.1 ML. All the diffraction peaks, including those associated with the bulk crystallites, vanish at a temperature between 128 and 133 K, less than the melting temperature of bulk butane, 135 K. The diffraction peaks are replaced by very broad features consistent with a liquid film. This behavior indicates that, at least up to Θ = 3.1 ML, butane forms relatively thick liquid films on graphite although only a single crystalline layer may be grown.

In summary, we have demonstrated that butane forms only a single crystalline layer when physisorbed on graphite. There is considerable evidence in our data that molecules are randomly distributed in a second layer before bulk crystallites begin to form. The presence of this amorphous second layer increases the melting temperature of the crystalline first layer. The amount of disordered material in the film continues to increase during bulk growth. Finally, there appears to be a wetting transition near (but below) the bulk melting temperature where butane is able to form thick liquid films on graphite.

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SMALL ANGLE NEUTRON SCATTERING STUDY OF ELECTROCHEMICALLY ETCHED SILICON*

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The discovery of a form of Si with a photoluminescence efficiency much higher than bulk Si has led to a flurry of research activity. This unusual form of Si is created during electrochemical etching of oriented single crystal Si in hydroflouric acid under a variety of conditions. The resultant material is very porous and often appears red in color. Small-angle neutron scattering (SANS) measurements of four electrochemically etched, porous silicon (PS) samples have been performed. The photoemission spectrum of each sample was obtained as well.

The SANS responses of the four PS samples are shown in Figure 1. The scattering curves overlap and have been separated for clarity. [Time constraints limited the measured range of scattering angles for the G1 and G3 samples.] The SANS data can be modeled with a non-particulate, random phase model[1] over intermediate scattering angles range. The functional form of this scattering behavior is given by

$$\frac{d\Sigma}{d\Omega}(Q, L) = 8\pi\langle\eta^2\rangle \frac{L^3}{(1 + Q^2L^2)^2} \qquad (1)$$

where L is the correlation length scale of the random, inhomogeneous structures within the scattering system and $\langle\eta^2\rangle$ is the mean square scattering length density fluctuation averaged over all points within the scattering system. The low scattering angle response of M1 and G2 follows a power law behavior $d\Sigma/d\Omega \sim Q^n$, where Q is neutron wavevector transfer. The parameters obtained from fitting the data in Figure 1 are presented in Table 1.

An important physical characteristic of the M1 deserves discussion. The top layer of this particular sample material was weakly bound to the underlying PS layer. It was evident from visual inspection with a microscope that this layer had cracked and partially peeled off the underlying material. Other researchers have attributed this effect to post-etching, evaporation-induced stresses.[2] The two curves for the M1 sample are before and after removal of this top layer. A check of the vertical uniformity of this sample is then facilitated by a comparison of the two measurements.

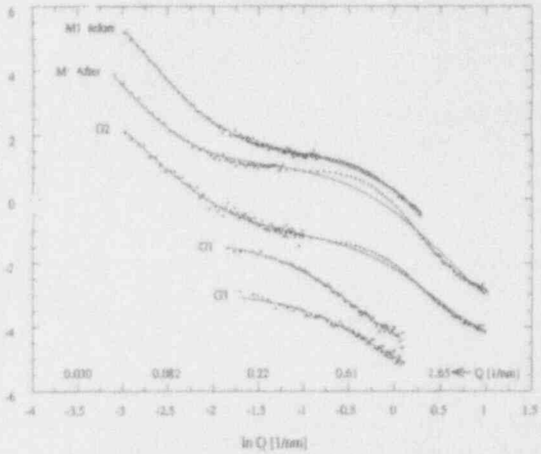

Fig. 1 SANS response of PS samples. Solid lines represent best fit of data with Eq. 1 and power law. Individual data sets have been separated for clarity.

TABLE 1 **Random phase correlation length scales and power law exponents for PS samples**

Sample ID*	L [nm]	n
M1 before	1.0	-3.5 ± 0.1
M1 after	1.1	-3.3 ± 0.2
G1	2.2	——
G2	0.9	-2.8 ± 0.1
G3	1.7	——

* Before and after measurements are before and after removal of weakly bound top layer.

In general, the M1 curves are similar; the correlation lengths and power law exponents are equal to within experimental error. Notice, however, that the plateau and shoulder are more pronounced in the M1 "after removal" curve. The observed shouldering can be interpreted as an interference effect between scattering centers that make up the PS material.[5] Given this explanation, the enhanced shouldering after top layer removal suggests that the interference effect is more clearly defined for the underlying PS material.

The possibility of microstructural anisotropy in the M1 (top layer removed) and G2 sample materials was also investigated at low to intermediate Q. Figure 2 demonstrates the effect of tilting the M1 sample 20° about a vertical axis. The G2 data showed little difference from the M1 measurement and are not shown. The horizontal and vertical sector averages in this figure are averages over pie-shaped areas on the detector deviating ±15° from the horizontal and vertical detector center-lines, respectively. The no-tilt M1 measurement, omitted for clarity, bisects the vertical and horizontal sector average curves more or less equally. Based on Figure 2, a certain degree of structural anisotropy does exist in this PS sample that extends over length scales of approximately 5 to 30 nm. It is interesting to note the microstructural anisotropy disappears beyond length scales of ~30 nm (Q values below 0.1 nm^{-1}).

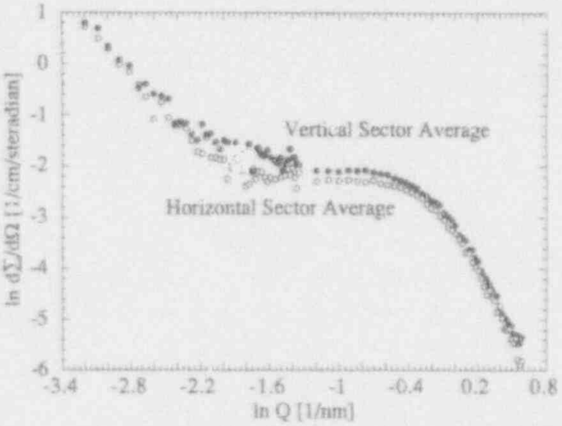

Fig. 2 Effect of tilting M1 sample (after removal of top layer) 20° about a vertical axis. Microstructural anisotropy is evident over length scales from 5 to 30 nm.

The photoluminescence spectra for all samples are shown in Figure 3. The asymmetric peak shapes from the M1 and G2 samples are due, at least in part, to the detection system efficiency going to zero at 930 nm. Notice the marked similarity between the M1 and G2 spectra, and the dissimilarity between these two spectra and those obtained from G1 and G3. Correlations between SANS determined structural characteristics and photoemission behavior can not be drawn, however, until the G1 and G3 SANS measurements are extended to lower Q.

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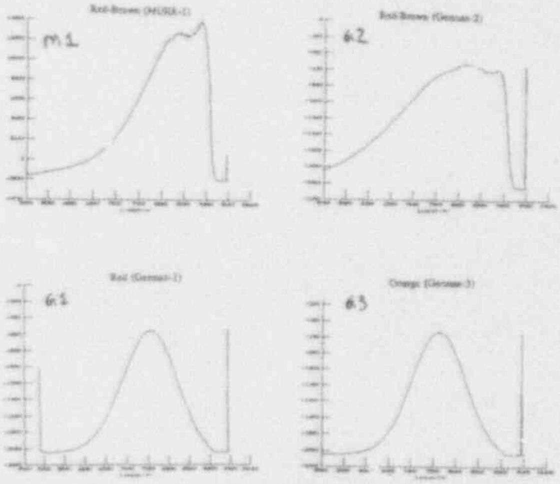

Fig. 3 Photoluminescence measurements

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* Neutron scattering experiments were performed at the Oak Ridge National Laboratory National Center for Small-Angle Scattering Research, Oak Ridge, TN and at the National Institute of Standards and Technology Cold Neutron Research Facility, Gaithersburg, MD.

SMALL-ANGLE NEUTRON SCATTERING INVESTIGATION OF DEFORMED SINGLE CRYSTAL Cu*

B.J. Heuser (MURR), J.S. King and G.C. Summerfield (University of Michigan-Ann Arbor)

The first set of measurements designed to investigate the SANS response of defects in metals were performed in May of 1992. The goal of these initial experiments was to resolve dislocation scattering in deformed single crystal Cu.

The starting material was a 5N's pure Cu single crystal ingot with a [110] cylinder axis. This ingot was supplied by Metron, Inc. of Allamuchy, NJ. The four neutron scattering samples investigated were deformed in compression along the [110] cylinder axis direction to 7%, 16%, 35%, and 54% reduction in thickness. An undeformed reference sample cut from the same ingot was measured as well. All samples were mechanically polished, with the final step using 1 micron alumina powder.

Early theoretical work[1] predicts a Q-3 scattering response from the 1/r elastic strain field of an edge dislocation. Epperson *et al*[2] have observed a Q-3 response in heavily deformed (greater than 95% reduction in thickness) single crystal Cu.

Figure 1 shows our measured, radially averaged macroscopic scattering cross section of the four deformed samples and the reference sample. Absolute cross sections were obtained by calibration against 1 mm of water. [Note that the portion of the detector shadowed by the beam stop is included in Figure 1.] The response of the reference sample is flat over the Q range, indicating the quality of the single crystal material is good. The asymptotic cross section of the reference sample is approximately 50% greater than expected from incoherent scattering from Cu. The reason for this is not clear-based on a measurement of a partially deuterated polystyrene sample in the same instrument configuration, the water calibration is correct to within 5%.

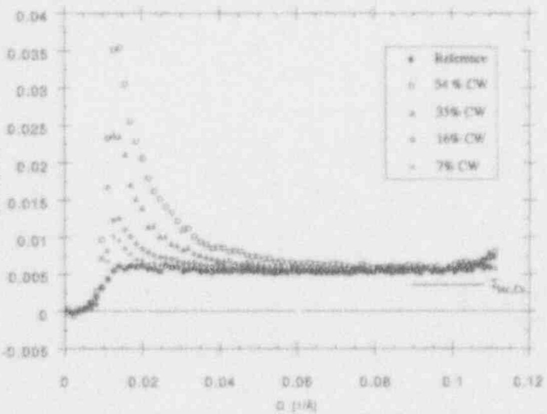

Fig. 1 dΣ/dΩ vs. Q for deformed and reference Cu

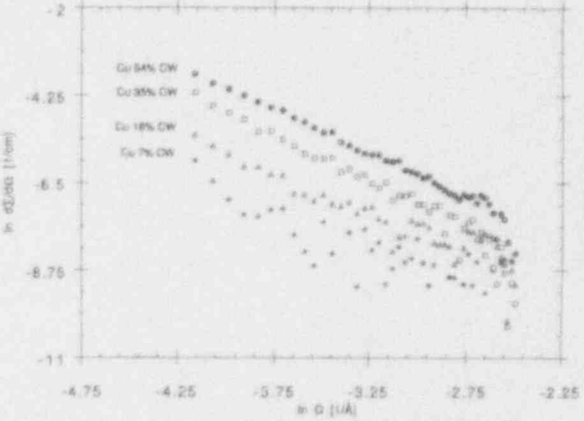

Fig. 2 ln net dΣ/dΩ vs. ln Q for deformed Cu

Figure 2 shows the radially averaged net scattering response (defined as deformed sample response-undeformed reference sample response) of the four deformed samples in ln-ln format. These data exhibit a Q$^{-2.2}$ power law over the entire measurement range, although the data for the 7% and 16% CW samples are poor. Figures 3 and 4 show the net scattering cross section of the 54% CW and 35% CW samples, respectively, with the measurement range now extending to low Q. [Low Q investigation of the 16% CW and 7% CW samples was not adequately performed in this study.] These figures demonstrate that the Q-2.2 power law is maintained to at least Q ~ 0.007 Å-1.

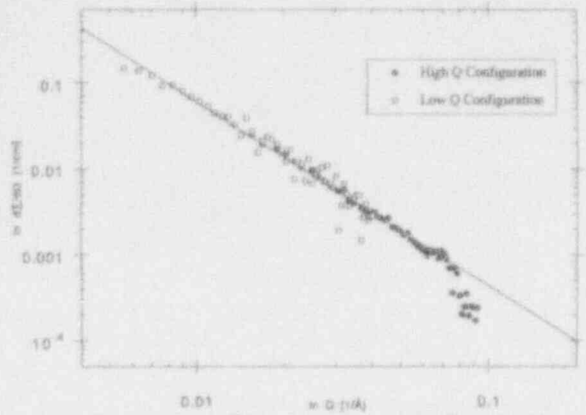

Fig. 3 ln net dΣ/dΩ vs. ln Q for 54% CW sample

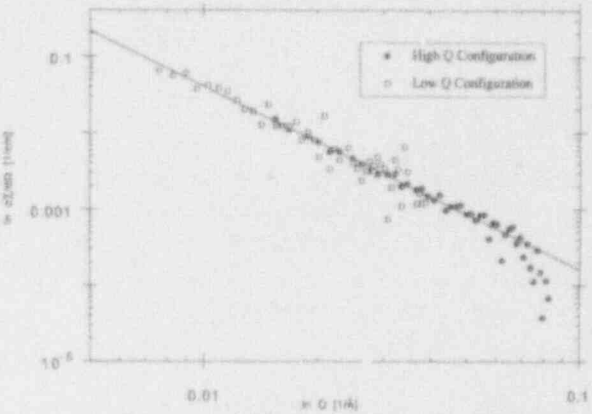

Fig. 4 ln net dΣ/dΩ vs. ln Q for 35% CW sample

The observed scattering response is not consistent with the $1/r$ dislocation strain field response discussed above. One possible explanation is that an interference between dislocation cores (which are expected to scatter like $1/Q$) and the $1/r$ strain field results in the Q^{-2} behavior. This seems unlikely, though, because this interference should depend on the degree of cold working. Furthermore, we would not expect such an interference effect to dominate over the entire measurement range.

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*Neutron scattering experiments were performed at the National Institute of Standards and Technology Cold Neutron Research Facility, Gaithersburg, MD.

SMALL-ANGLE NEUTRON SCATTERING INVESTIGATION OF DEUTERIUM TRAPPING AT DISLOCATIONS AND GRAIN BOUNDARIES IN Pd AND Nb*

J.S. King, G.C. Summerfield, J. Steinhauer (University Michigan-Ann Arbor), and B.J. Heuser (MURR)

As a precursor to measurements performed in November 1992 at Saclay, France, prototype samples were measured on the 30 m SANS instrument at Oak Ridge National Laboratory. The primary purpose of these measurements was to ensure that the sample preparation procedures employed were correct. Unfortunately, the majority of the measurements were inconclusive because of the poor counting statistics. Three measurements did provide interesting results and are presented below.

Three Nb samples were prepared from polycrystalline stock. The samples were deformed by rolling at room temperature to 62% reduction in thickness. All the samples were then heated to 700° C for 30 minutes in vacuum of 10-5 torr. Two of the samples, Nb2 and Nb3, were loaded with deuterium at 400° C to 34,400 and 13,200 atomic ppm, respectively. Sample Nb1 was not loaded with deuterium and served as a reference standard.

The net absolute differential neutron scattering cross sections (defined as loaded sample cross section minus Nb1 reference sample cross section) are shown in Figures 1 and 2. The scattering response is modeled with a combination of two power law behaviors; $d\Sigma/d\Omega \sim 1/Q + 1/Q^4$, where Q is the wavevector transfer variable. The $1/Q$ component represents deuterium trapping at dislocations forming rod-like scattering structures while the $1/Q^4$ component represents a Porod response from internal surfaces.[1] In this case we suspect that surfaces of large Nb deuteride particles are causing the Porod response. Notice that dislocation trapping is the dominant behavior of the Nb3 sample, but that a significant Porod component exists in the Nb2 measurement. The room temperature solvus of the Nb-D phase diagram is approximately 30,000 atomic ppm. The larger Porod response of Nb2 may be consistent with this deuterium solubility limit.

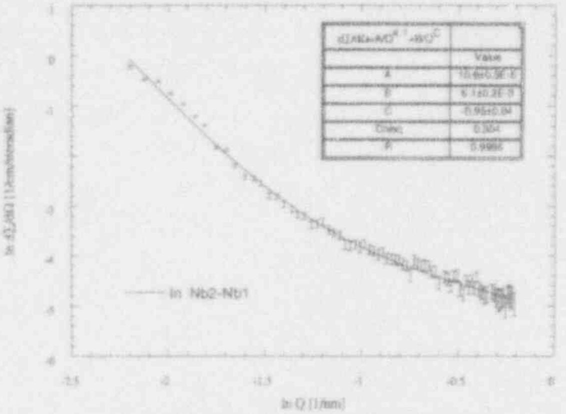

Fig. 1 SANS from Nb deuterides and from deuterium trapped at dislocations in Nb2

Analysis of the dislocation trapping component is difficult without an accurate measurement of the excess deuterium trapped at dislocations. It was not possible to measure the excess fraction during the preparation of these samples, however. The two deuterium loaded samples were deformed to the same degree and we expect the dislocation response to be of approximately equal magnitude. This is in fact what was observed.

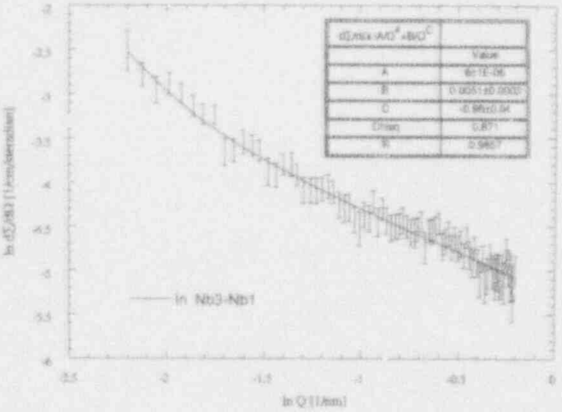

Fig. 2 Deuterium trapping at dislocations in Nb3

As discussed below, we generally expect to observe a response from deuterium trapping at grain boundaries. However, the large amount of deformation of the Nb samples will flatten out the grains. This creates a situation where most of the grain boundary area is parallel to the surface of the sample. In the standard measurement configuration, this grain boundary area will no longer be perpendicular to Q, a necessary requirement for grain boundary induced SANS.

Figure 3 shows the SANS response from a polycrystalline Pd sample, PPd1, loaded with 6400 atomic ppm deuterium. This sample was swaged by the manufacturer, and had needle-like grains that were oriented with the needle axis parallel to the incident beam direction. In this case, the majority of the grain boundary area was perpendicular to Q. The scattering response from deuterium trapped at oriented grain boundaries is $d\Sigma/d\Omega = A\delta(\mu)/Q^2$, where $\delta(\mu)$ represents an orientation average over the grain boundaries. For a Gaussian distribution $\delta(\mu) = 1/Dq$, where Dq is the FWHM of the distribution. The constant A includes the number of deuterons per unit area trapped at grain boundaries. This number is found to be approximately 0.15 deuterons/Å^2 for PPd1, compared with a value of 0.4 ± 0.2 deuterons/Å^2 for an undeformed Pd sample measured previously.[1]

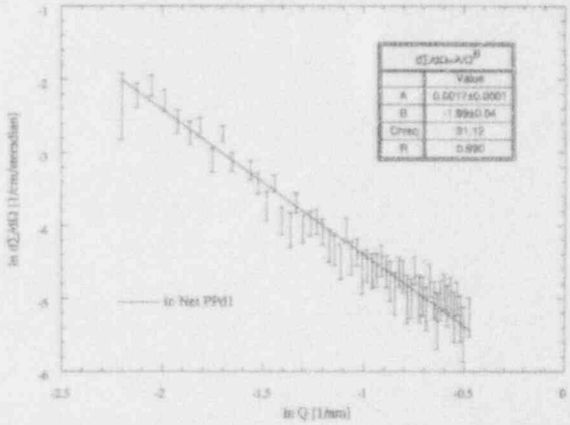

Fig. 3 Deuterium trapping at grain boundaries in PPD1

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*Neutron scattering measurements performed at the National Center for Small-Angle Scattering Research at Oak Ridge National Laboratory, Oak Ridge, TN.

STRUCTURAL ANALYSIS AND PHASE RETRIEVAL METHODS IN NEUTRON DIFFRACTION STUDIES OF BIOLOGICAL LIPIDS

D.L. Worcester (MU Biological Sciences), H. Kaiser (MURR), R. Kulasekere, K. Hamacher (MU Physics) and J. Torbet (University of Edinburgh Biochemistry)

Studies are being made of using vapor phase addition of perdeuterohexane or other short hydrocarbons to bilayer arrays of lipid mixtures as a means of obtaining structural information and phase retrieval. The method of vapor phase addition of hexane to hydrated single-component lipid bilayers was originally studied for the purposes of understanding the thermodynamics of the process, and the location of the hexane in the bilayer by perdeuteration and difference profiles.[1] The main finding of these studies was that the hexane enters the bilayers and is localized near the center of the hydrocarbon region, at a center of symmetry. In work at MURR, applications to bilayers of sphingomyelin and cholesterol and other lipid mixtures have been investigated. The interest is to determine whether the hexane is highly localized in bilayers containing cholesterol and whether the hexane in perdeutero form produces changes in diffraction intensities to 0.3 nm resolution that can be used for phase retrieval and structure analysis.

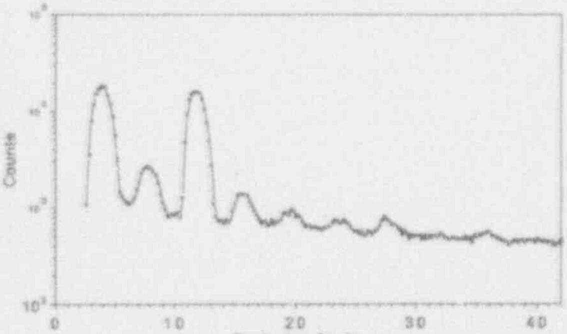

Fig. 1 Neutron diffraction pattern obtained on instrument 2XC from about 20 milligrams of a mixture of sphingomyelin and D-5 cholesterol hydrated with H_2O and with perdeutero hexane incorporated from vapor

Results for sphingomyelin and cholesterol bilayers presently show changes in diffraction through nine orders from a bilayer repeat spacing of 6.3 nm. A diffraction pattern is shown in Figure 1. Perdeuterated hexane was added in 20% volume mixture with heptadecane and the solution introduced into the lipid sample container. The solution mixture determines the vapor pressure of the hexane and hence the amount that equilibrates into the lipid bilayer sample. The density profile for bilayers of sphingomyelin, D-5 cholesterol and D-hexane is shown in Figure 2. The perdeutero-hexane is clearly seen in a narrow region at the center of the bilayer. The additional density at this location has reduced the contrast so phase assignments are mainly benefited by the "heavy atom" at a center of symmetry. This "heavy atom" must be well localized for best use.

Reference

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Fig. 2 Neutron scattering density profile for sphingomyelin and D-5 cholesterol bilayers with perdeutero-hexane introduced from the vapor phase and present as additional density at the center of the bilayer. The repeat spacing is 6.3 nm.

STUDIES OF CHLOROPHYLL CYLINDRICAL AGGREGATES AND CHLOROSOMES: ENERGY TRANSFER IN PHOTOSYNTHESIS

David L. Worcester (MU Biological Sciences), T.J. Michalski and J.J. Katz (Argonne National Laboratory, Chemistry Division)

Chlorophylls hydrated in nonpolar solvents can form hollow cylinders as aggregates. We have studied these cylinders previously using neutron scattering to determine cylinder diameters, and reported cylinders of diameters from 1.0 nm to 4.5 nm depending on chlorophyll type. Such cylinders are of interest as models of chlorophyll aggregates *in vivo* because of the cylinders reported in freeze fracture electron microscopy studies of chlorosomes in the green photosynthetic bacteria *Chlorobium* and *Chloroflexus*. Chlorosomes contain proteins and much bacteriochlorophyll-c, which serves to collect light energy and transfer it to photosynthetic reaction centers. The structural basis for these processes is of interest for understanding how efficient energy conversion is achieved.

The arrangement of protein and bacteriochlorophyll-c in chlorosomes is controversial. Some groups claim that proteins form the cylinders, whereas other groups have claimed that portein-free cylinders can be isolated. Thus, it is possible that the *in vivo* cylinders are chlorophyll aggregates such as we have studied *in vitro*, but detailed experimental comparisons of these structures are badly needed. For this purpose we are using small angle neutron scattering.

We previously reported that artificial cylinders of Bacteriochlorophyll-c were much the same size as the cylinder diameters reported for *Chlorobium* chlorosomes (≈10 nm diam.). We also found that Bchl-c cylinders were not disaggregated by drying, as were cylinders of other chlorophylls, suggesting that the ring-I hydroxyethyl group of Bchl-c, rather than water, was responsible for the cylinder aggregation.

In *Chloroflexus* the cylinders are reported to be smaller (5.2 nm diameter), a size we had not found *in vitro*. As a new study, we tested if the absence of the ring V carbomethoxy group in these chlorophylls, together with aggregation without the hydroxyl group, produces smaller cylinders. To do this we studied hydrated aggregates of Pyrochlorophyll-a. The results (Figure 1) showed smaller cylinders, which were comparable to the sizes reported from electron microscopy studies of *Chloroflexus* chlorosomes. The neutron scattering data also demonstrated some dispersion of sizes in our artificial aggregates. Neutron scattering measurements on intact chlorosomes were recently initiated as the next part of this project.

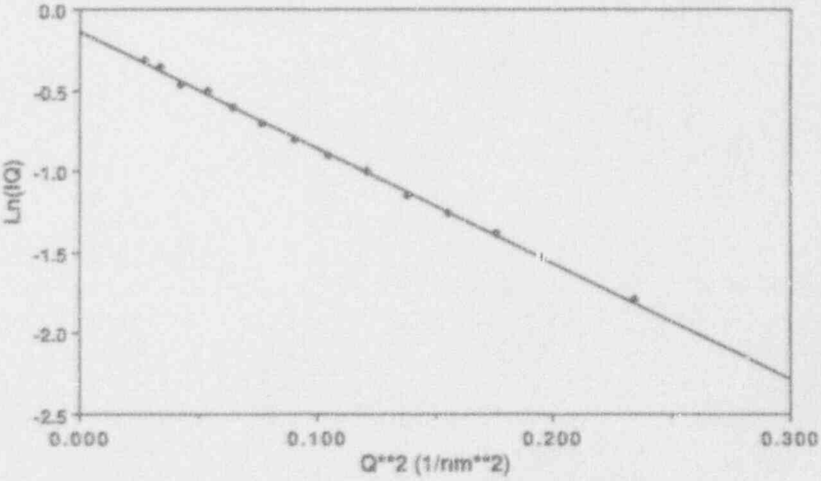

Fig. 1 Guinier-type plot of neutron scattering data obtained from hydrated pyrochlorophyll-a aggregates formed in deuterated octane. The concentration is 9 mg/ml. The cylindrical radius of gyration determined from the plot is 3.78 nm.

NEUTRON SCATTERING FROM NANODISPERSED MAGNETIC PARTICLES*

J.R. Childress and C.L. Chien (Johns Hopkins University), R.W. Erwin (National Institute of Standards and Technology) and J.J. Rhyne (MURR and NIST)

Granular magnetic materials, which consist of fine magnetic particles dispersed in an amorphous insulating matrix, display a number of unique properties as a result of their nanometer-scale microstructure.[1] However, many properties of granular magnetic composites are still poorly understood at the fundamental level, such as the exact nature of the magnetic ordering and magnetic excitations within and between the grains. A well known property of single-domain magnetic particles is the occurrence of superparamagnetic relaxation, resulting from the thermally activated fluctuation of the particle moment. Superparamagnetism can be observed on different time scales t by using different magnetic probes, such as SQUID magnetometry (t = 10 sec) and 57Fe Mössbauer spectroscopy (MS) (t = 10-8 sec). In contrast, neutron scattering has a much shorter interaction time (typically < 10-12 - 10-13 sec), and measures approximately the instantaneous spin-pair correlation function. The use of total scattering techniques (e.g., SANS) and also inelastic measurements (e.g., using a triple axis spectrometer) provide a mechanism to isolate dynamic processes from longer time structural relaxation in these nanoscale materials.

The sample examined in this study was prepared using RF magnetron sputtering, by co-deposition of Fe and Al_2O_3 from a single mixed target onto thin Al_2O_3 substrates kept at 200° C. The granular microstructure was confirmed by transmission electron microscopy, which reveals a narrow distribution of Fe particle sizes around 3 nm in diameter. The film composition of 35 vol% of Fe is well below the granular percolation concentration (= 60 vol%), and, therefore, contains well-isolated particles.[2] X-ray diffraction analysis shows that the particles retain the body-centered cubic structure of bulk -Fe. Magnetic hysteresis loops obtained a T = 5 K show a clear ferromagnetic behavior, with 60% remanent magnetization and a coercivity of about 600 Oe. For our sample, the blocking temperatures T_B above which the particles become superparamagnetic are 75 K for SQUID measurements (peak in the zero-field-cooled susceptibility and onset of irreversibilities) and 210 K for MS measurements (onset of magnetic hyperfine splitting). Above T_B, little information on the magnetic ordering can be obtained by these methods.

Previous SANS data[3] showed that the high-Q (= 1.5 nm-1) data are well described by a single Lorentzian-squared scattering function. At the smallest wavevector transfers, we find additional intensity, which was modelled by adding a $1/Q^2$ term to the scattering. The $1/Q^2$ term corresponds to a Lorentzian cross-section with a near-zero inverse correlation length, thus reflecting correlations on a scale larger than what can be measured in our experiment (roughly a few tens of nm's). It is important to note, however, that the form of this cross-section is not unique, and that further refining of the data analysis is necessary to pinpoint the functional form and correlation length associated with the low-Q scattering.

The Lorentzian-squared amplitude decreases with decreasing temperature, suggesting that this component of the scattering is dynamic in origin. The decrease is sharp below 200 K, flattening out near zero below about 120 K. The correlation length ξ shows a related temperature dependence. Starting at room temperature in the 1 nm range, ξ increases only slightly with decreasing temperature until about 200 K, below which a rapid increase is observed. However, instead of a divergence of ξ, we observe a saturation at about 4 nm, very close to the average particle size. By contrast, the amplitude of the Lorentzian component to the scattering is constant and nearly zero at high temperatures. As T is decreased below 200 K, the amplitude begins to increase and rises sharply below 75 K, a behavior typical of order parameter scattering. Because of the long correlations lengths associated with this scattering, it is suggestive of inter-particle ordering. Due to the short characteristic time scale, there is no evidence of magnetic blocking in the SANS data in contrast to that observed in the magnetization and Mössbauer results.

Additional data have been taken on triple axis spectrometers using a range of energy resolutions to study the inelasticity of the scattering. These results showed the presence of both very fast (< 10-11 sec) and much slower dynamic processes. In particular, inelastic data taken with 850 μeV energy resolution showed (1) an elastic resolution limited central peak which decreased with increasing temperature reflecting the intra-particle order parameter, and (2) a very broad quasielastic Lorentzian line which increased with increasing temperature presumably correlated with the inter-particle coupling dynamic fluctuations.

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*Neutron scattering experiments were performed at the NBS Reactor of the National Institute of Standards and Technology, Gaithersburg, MD 20899.

MAGNETIC STRUCTURE Of Dy|Sc SUPERLATTICES*

F. Tsui, C.P. Flynn, R.S. Beach, and M.B. Salamon (University of Illinois-Urbana Physics), J.A. Borchers, R.W. Erwin (National Institute of Standards and Technology) and J.J.Rhyne (MURR and NIST)

Molecular beam epitaxy techniques can be used effectively to do atomic scale engineering on layered epitaxial specimens when it is desired to control closely the number of atomic planes or two (or more) elements in adjacent layers. In addition the techniques allow the building of multilayer sequences of a prescribed number of bilayers, within each of which the number of atomic planes is closely controlled and reproducible.

Previous studies by this group[1] on c-axis growth layered structures of magnetic rare earth metals alternated with non-magnetic Y [e.g., Dy|Y] have shown that bulk-like magnetic order is developed in each of the magnetic layers and that, in the case of the periodically ordered structures [e.g., Dy], this order shows a phase coherence from one magnetic layer to the next. Such results imply a long-range spin density wave type coupling which is propagated across the non-magnetic layers. Studies of the multi-layer spin correlations show that the range of this coupling falls off inversely with the thickness of the non-magnetic layer and is consistent with the range calculated from the width of the generalized conduction electron susceptibility function (q) for Y (or Dy).[2] These functions show a sharply defined peak along the c* direction in reciprocal space at a value of reduced q corresponding to the periodicity of the magnetic ordering (for those elements having 4f spins as in Dy, or for a pseudo magnetic ordering in Y) as dictated by the caliper dimension spanning parallel sheets of the Fermi surface. Perpendicular to the c* direction the susceptibility function is centered at $q = 0$, and is quite broad corresponding to a much shorter (approximately one neighbor) coupling range in these directions. The anisotropy in (q) and its effect on the range of magnetic coupling was graphically demonstrated in superlattices of [Dy|Y] grown along a basal plane direction[2] which showed magnetic order within each magnetic layer, but a total absence of interlayer coupling in contrast to the long range interlayer coupling observed in the c-axis growth direction superlattices.

Dilute alloy studies of magnetic rare earths in Sc[3] have shown an anomalous critical concentration behavior for the onset of long range order in contrast to Y[4] alloys which show order for alloys as dilute as 1% magnetic impurities consistent with the electronic structure predictions of very long range interactions. In Sc the critical concentrations are in the range of 25% of the magnetic element necessary to induce ordering. Such large magnitudes are more closely reminiscent of nearest neighbor percolation arguments than of the long range spin-density wave coupling seen in Y systems. Such a discrepancy is not found in the calculated $\chi(q)$ functions. This anomalous difference in Y and Sc was the motivation for investigating superlattices of Dy and Sc. In spite of the large basal plane lattice parameter mismatch (> 8%) successful growth of several [Dy|Sc] superlattices was achieved.

As shown in Figure 1, the neutron experiments on [Dy|Sc] superlattices revealed only a very broad temperature-dependent magnetic component underlying the nuclear scattering which is consistent with a ferromagnetic coupling of the Dy spins below a $T_c \approx 150$ K. The magnetic state above 150 K up to the expected ordering temperature of ≈ 180 K was uncertain although no evidence of a helically ordered state was observed. The broad peaks corresponded to a correlation length for the order extending along the c-axis growth direction of less than the layer thickness. This result clearly showed that the magnetic order was not propagated through the non-magnetic Sc layers unlike the case for Y superlattices.

This puzzling lack of coherence in Sc superlattices was further investigated in field dependent studies of the scattering in which the field from a superconducting magnet was applied along a basal plane direction to artificially induce coherent order along the layers. These results at both 10K and 80K showed only a very gradual monotonic increase in net superlattice moment (layer to layer correlation) was achieved and even in fields as large as 6T, full saturation was not achieved. There was no evidence of any critical transition fields but only a smooth increase in net moment.

Future investigations into the anomalous exchange range effects in Sc superlattices are underway.

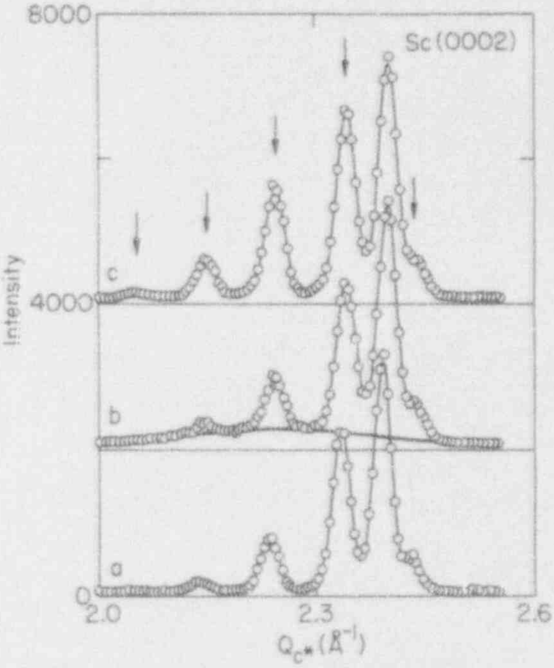

Fig. 1 Neutron diffraction scan along the [0002] direction in the superlattice $[Dy_{25\text{Å}}|Sc_{40\text{Å}}]_{86}$. Curve a is the nuclear intensity showing five structural harmonics, curve b is a zero field scan at 10K showing the development of short-range ferromagnetic order indicated by the solid line under the structural peaks, and curve c is a scan taken with a 60 kOe applied field following zero-field-cooling of the sample illustrating field-induced ferromagnetic coherence.

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*Neutron scattering experiments were performed at the NBS Reactor of the National Institute of Standards and Technology, Gaithersburg, MD 20899.

NEW MURR HORIZONTAL SANS INSTRUMENT UPDATE

Brent J. Heuser, Mihai Popovici, Ronald R. Berliner, William B. Yelon, Olawale Oladiran, David Nickolaus, Chester Edwards, James J. Rhyne (MURR), and David L. Worcester (MU Biological Sciences)

Design of the new SANS instrument is well underway. This instrument will be located at beamport E and extend into the north vestibule of the beamport floor. Work performed to date and significant aspects of the design are as follows.

A two-wire, gas filled proportional counter manufactured by ORDELA was received in April of 1992. The detector is being stored on the beamport floor until testing this winter. The detector vacuum chamber "Request For Proposals" was awarded to the Paul Mueller Company in Springfield, MO, with delivery of the stainless steel tank expected in January 1993. Detector translation within the tank will allow continuous adjustment of the sample to detector distance from approximately 1 to 8 meters.

In order to accommodate a total of three instruments, a new in-port collimator will be installed at beamport E. This collimator will have two separate channels (a SANS channel and a thermal channel) with a 3° divergence in the horizontal plane. A two channel collimator is necessary to accommodate the different incident beam sizes and filter requirements of the E port instruments. Geometric constraints, both within the beamport and on the beamport floor, dictate that the two channels are divergent. The entire beamport, including the collimator section, will be filled with He to eliminate long wavelength neutron attenuation by air and water vapor. Allowances will also be made for room temperature neutron and gamma ray filters within the beamport collimator.

The primary fast neutron and gamma-ray filtration will occur just outside of the E port biological shield. A beam stop/filter cryostat elevator will hold the neutron and gamma-ray filters in position during operation. Be and Bi filters will be used in the SANS channel at this location. A second filter position further downstream, between the monochromator device and the start of the incident collimation, also is included in the SANS instrument design.

The neutron monochromator device will consist of two Ni-Ti multilayers in an anti-parallel arrangement. The first multilayer will be a stack of approximately 4 cm long wafers in soller geometry. The second multilayer, downstream from the first, will be a single wafer approximately 2 meters long in reflection geometry. Extensive calculations indicate that this configuration maximizes the incident current while providing good instrumental resolution. In the current design, the downstream multilayer is slightly curved in the diffraction plane. This eliminates the inherent resolution asymmetry of multilayer monochromators and also provides a certain degree of phase space focusing. Testing of prototype multilayer depositions has been underway for the past six months. The results have been encouraging, with peak reflectivities of approaching 95% in the best case.

Current work on the project includes further testing of multilayer depositions, detector support and beam stop design, and beamport E shielding design.

TRIAX – MURR'S NEW TRIPLE-AXIS SPECTROMETER

K.W. Herwig, S.A. Werner, and J.J. Rhyne (MURR and MU Physics)

The MURR Center and MU Physics staff in collaboration with C. Stassis, J. Zaretsky, and A. Goldman from Iowa State University are developing a highly versatile, triple-axis spectrometer, TRIAX, on beamport A funded by the National Science Foundation, MU and the Ames Laboratory (DOE). Major components of the new instrument were delivered to MURR from Oak Ridge National Laboratory in mid-October 1991. Since that time, these components have been thoroughly refurbished, and design and construction of missing pieces initiated. When completed, TRIAX will be a world class instrument used to address a variety of problems in condensed matter physics and materials science. Here we summarize the progress that has been made on TRIAX and outline the remaining tasks.

There are several principles which govern the design and ultimately the effectiveness of the instrument. The goal is to maximize the number of neutrons incident on the sample while minimizing the neutron and gamma backgrounds in the immediate vicinity of the instrument. The first is achieved by optimizing the neutron optics of the instrument while the second is satisfied by the design of effective shielding.

While design of the neutron beam path has been completed (and construction underway), design of the surrounding shielding is only in the conceptual stage. Contained in this heavy concrete shielding will be the double-axis monochromator, the filter cryostat, and a Soller collimator elevator assembly. A beryllium single-crystal, viewed in transmission by the triple-axis instrument, seems the likely choice of double-axis monochromator. F. Ross is responsible for the double-axis instrument. The filter cryostat has been constructed and will contain three individual filters: pyrolitic graphite, beryllium and silicon. The Soller collimator assembly, designed and under construction, will contain three collimators: Cd-plated steel, an open position, and beam stop.

The largest and heaviest single component of TRIAX is the monochromator drum which rotates as the incident neutron wavelength is varied. This drum contains the monochromator assembly at its center and serves to stop stray scattered neutrons and the beam transmitted through the monochromator. The three focusing monochromators will be accessible remotely, the choice depending on the particular experiment. The focusing device--using copper, pyrolitic graphite and pressed silicon monochromating crystals--has been purchased. The system of shielding wedges that lift and lower as the drum rotates required extensive rebuilding, and this task and other refurbishing of the main drum was completed earlier this year.

Cantilevered from the monochromator drum is an arm which holds the sample and analyzer tables, the assembly of seven 1/2 inch diameter ^{3}He detectors, and associated shielding. This arm required substantial modification and rebuilding of the drives associated with the rotation axes. In addition, an entirely new shielding system for the analyzer crystal was designed and is under construction, with shielding wedges cast from a mixture of B_4C and epoxy. New detectors have been purchased and the associated shielding is being constructed. Eight stretched-film Soller collimators that will define the angular divergence of the beams scattered by the sample and analyzer crystal are nearing completion. These items are being built in the Physics Machine Shop by C. Holmes, S. Potts, R. Schlotzhauer and W. Steifermann.

B. McCracken and C. Edwards of the MURR Facility Operations group are beginning the design of the major shielding components to be located on the beamport floor. This work

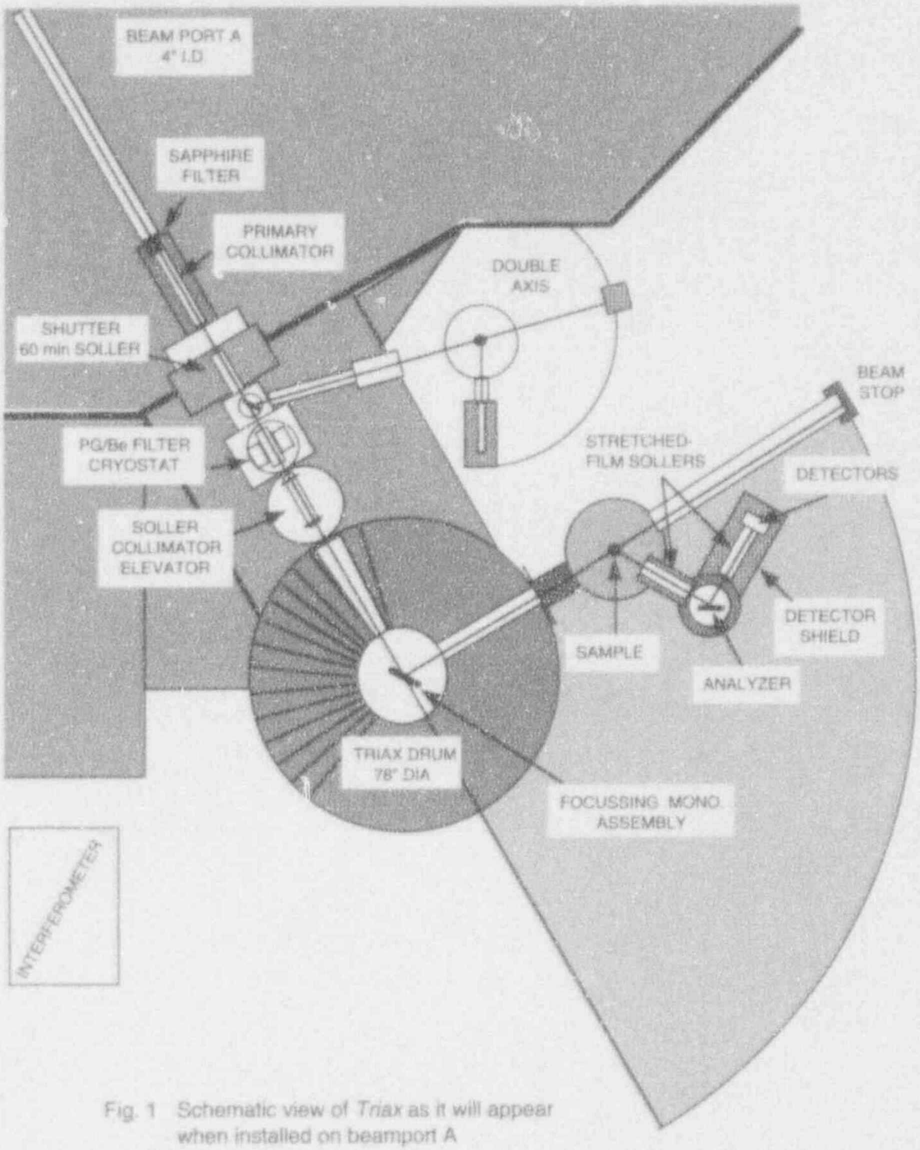

Fig. 1 Schematic view of *Triax* as it will appear when installed on beamport A

includes designing concrete shields and reworking the beamport shutter. The computer control program, based on the NeXT station, is being developed by R. Iyer and G. Moum of MURR's Computer Development group. The interface and motor drivers are being constructed by R. Berliner and his assistants in the MURR Instrument Development group. In addition, various pieces of peripheral apparatus (closed-cycle refrigerator, temperature controller, low-temperature cryostat, pyrolitic graphite filters, and monochromator and analyzer crystals) have either been purchased or will soon be ordered.

Although much work remains to be done, we believe that a realistic goal for installation and testing of the machine is March 1993, at which time all major components should be completed and ready for assembly and testing.

This work is supported by NSF-DMR-9103383 ($225,000); DOE Ames Laboratory ($50,000); MU Prime Fund ($144,000).

NEUTRON OPTICS GROUP

Samuel A. Werner, Group Leader and Professor of Physics
Helmut Kaiser, Senior Research Scientist (Adjunct Associate Professor of Physics)

This group is engaged in basic studies of the fundamental properties of the neutron utilizing two neutron interferometer instruments at the MURR Center and is also developing a neutron interferometer. The group has broad international collaborations and the interferometry program has had continuous funding from the National Science Foundation for more than 15 years. The current grant is NSF-PHY-9024608 running from June 1991 - June 1995 in the total amount of $616,000.

NEUTRON INTERFEROMETRY

S.A. Werner (MU Physics) and Helmut Kaiser (MURR)

The perfect silicon-crystal neutron interferometer has provided us with a wonderful device to observe subtle quantum mechanical phenomena and interactions with clarity and elegance. We have two neutron interferometry experimental stations at MURR.

The instrument at beamport B was constructed during 1976-1977 with NSF funding. A schematic diagram of this instrument is shown in Figure 1. A monochromatic beam is produced in a double-crystal monochromator assembly which presently utilizes two copper (220) crystals. This monochromator assembly produces a variable energy incident beam in the wavelength range of 0.9 to 2.4 Å, which is allowed to pass through a series of slits and then onto the interferometer. The interferometer, along with the ^{3}He detectors, is mounted inside an aluminum box, which provides for thermally-isolated operation of the interferometer. This box is rigidly mounted on a rotator assembly (for gravitationally-induced quantum interference experiments) inside of a large dense Benelex 70 neutron shield, which in turn rests on a vibration-isolation support consisting of 4 small Firestone pneumatic isolators. This arrangement is essential in order to provide for vibration-free and microphonic-free operation. The interferometer used in this set-up is an LLL-type device. The LLL stands for three Laue-transmission geometry reflections in the three crystal slabs which make up the interferometer.

The continued growth of the neutron interferometry program at MURR over the past 15 years has created the demand for a second neutron interferometry set-up at beamport C (Figure 2). This instrument utilizes a skew symmetric interferometer as shown in Figure 3. The beam comes from a vertically-focussing pyrolytic graphite (PG) monochromator. The wavelength is fixed at λ= 2.35 Å. The beam passes through a PG filter to eliminate order contaminations from the beam incident upon the interferometer. Recently we have carried out experiments on the Neutron Phase Echo effect, the effect of Spectral Filtering in Neutron Interferometry, and Time-of-Flight Neutron Interferometry on this instrument. The experiments formed the basis of Russell Clothier's PhD thesis research.

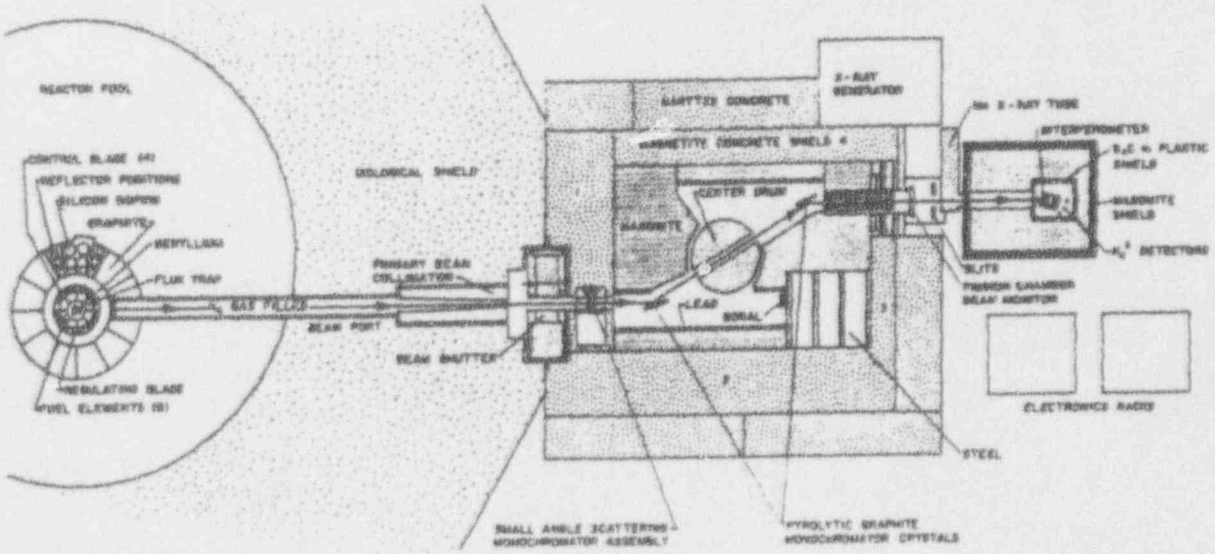

Fig. 1 Double-crystal monochromator and interferometer at beamport B.

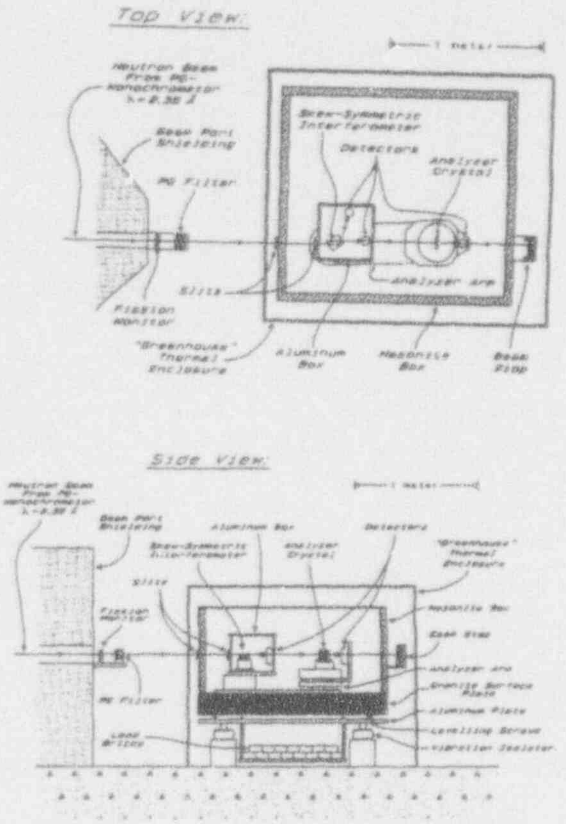

Fig. 2 Interferometer set-up at beamport C.

We have two international collaborations: with the Vienna group of Professor Helmut Rauch and his students, and with Professors A.G. Klein and G.I. Opat and their students at the University of Melbourne, Australia. The Aharonov-Casher effect experiment that attracted wordwide attention (it was written up in *Physics Today*, *Science News*, *Scientific American*, etc.) was carried out at beamport B in collaboration with the Melbourne group. The recent experiment in which we observed the scalar AB effect for the first time was carried out at beamport C in collaboration with Brendan Allman, a student from Melbourne. The neutron phase echo, spectral filtering, and time-of-flight experiments were carried out in collaboration with Harold Wölwitsch and Helmut Rauch from Atominstitut, Vienna.

We are also involved in a collaboration with Drs. Muhammad Arif and Geoffrey Greene at the National Institute of Standards and Technology in building a new, sophisticated neutron interferometry instrument at the NBSR in Gaithersburg, MD. Ken Littrell, a graduate student of ours, is doing his PhD thesis research there.

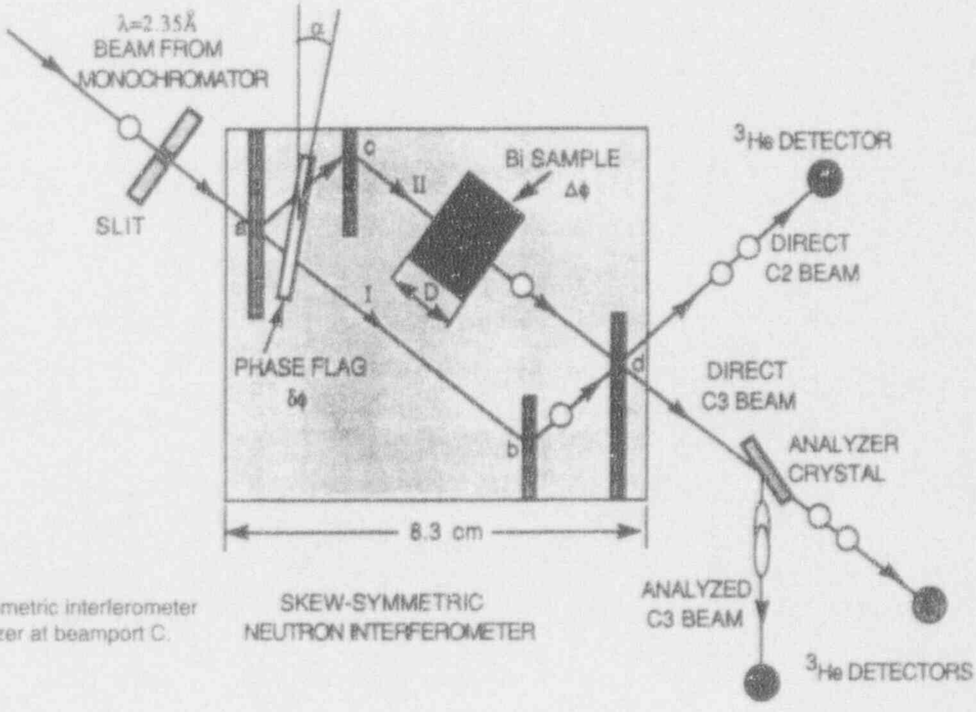

Fig. 3 Skew-symmetric interferometer and analyzer at beamport C.

A list of the experiments carried out in this program over the past 17 years is as follows:

- Observation of Gravitationally-Induced Quantum Interference (1975,1980,1985,1988)
- Observation of the Change of Sign of the Wave Function of a Fermion due to Precession of 360 Degrees in a Magnetic Field (1976)
- Observation of the Effect of the Earth's Rotation on the Quantum Mechanical Phase of the Neutron (Sagnac Effect), (1980)
- Measurement of the Energy-dependent Scattering Length of Sm-149 in the Vicinity of a Thermal Nuclear Resonance (1983)
- Measurement of the Longitudinal Coherence Length of a Neutron Beam (1983)
- Neutron Interferometric Search for Quaternions in Quantum Mechanics (1984)
- Experiment on the Null-Fizeau Effect for Thermal Neutrons in Moving Matter (1985)
- Precision Measurement of the Bound-Coherent Neutron Scattering Length of U-235 (1986)
- Observation of a Motion-Induced Phase Shift of Neutron deBroglie Waves Passing through Matter Near a Nuclear Resonance (1987)
- Observation of the Topological Aharonov-Casher Phase Shift (1989)
- Observation of the Effects of Spectral Filtering in Neutron Interferometry (1990)
- Observation of the Neutron Phase Echo Effect (1991)
- Coherence Effects in Time-of Flight Neutron Interferometry (1991)
- Observation of the Scalar Aharonov-Bohm Effect (1992)

Descriptions of several of these experiments are given in a number of widely used physics textbooks, including J.J.Sakurai's *Quantum Mechanics* and K. Krane's *Introduction to Nuclear Physics*.

During the next year we will be upgrading the instrument at beamport B. This improvement will consist of replacing the computer with a NeXT station, upgrading the interface and modifying the double crystal monochromator assembly so as to utilize a 4 inch high beam with a vertically-focussing second monochromator. This modification will yield an increase in counting rate of about a factor of 4.

Our agenda of experiments for the next three years is the following:

- Effect of the moon and the sun on quantum interference
- Aharonov-Casher effect-stage II experiment
- Multi-photon exchange between a neutron and an oscillating magnetic field
- Search for quaternions in Q.M.-stage II experiment
- Wheeler-delayed-choice experiment
- Highly precise gravitationally-induced quantum interference experiment, checking the effect of x-ray interrogation areas
- Construction of large-scale, split component interferometer
- Michelson-Morley experiment with neutrons
- Search for the Mashhoon effect (coupling of spin to rotation)

This program has been funded continuously by the Physics Division of the National Science Foundation. The current grant is NSF-PHY-9024608, June 1991-June 1995, $616,000.

ELECTROMAGNETIC ACCELERATION OF NEUTRONS

R.C.Casella (National Institutes of Standards and Technology) and S.A.Werner (MU Physics)

Discussions of topological phenomena in quantum mechanics, such as the Aharonov-Bohm (AB) and Aharonov-Casher (AC) effects, are most easily framed in the language of Dirac-Feynman path integrals. Apart from the important quantum phase, the path-integral formalism allows us to employ classical concepts such as velocity, acceleration and the orbit of a particle. This facilitates the consideration of orbits winding around topological charges, such as the magnetic flux in the AB effect and the linear charge density in the AC effect, to calculate the phase shift in an interferometer experiment. However, as recently stressed by Casella[1] in discussing variants of the neutron interferometry experiment on the AC effect carried out by Cimmino et al,[2] *it is essential to treat the neutron spin as an intrinsically quantum variable*. That emphasis is brought into focus here where we describe an attempt to search for an experiment to detect a putative acceleration of the neutron in the combined presence of electric **E** and magnetic **B** fields, as measured in the rest frame of the interferometer (Figure 1). This acceleration, recently suggested by Anandan,[3] is $\mathbf{a}=(\gamma/mc)\mathbf{E}\mathrm{x}(\mu\mathrm{x}\mathbf{B})$, where γ is the neutron gyromagnetic ratio relating its magnetic moment μ to its spin angular momentum **s**. This acceleration would produce a maximal effect on the neutron kinetic energy when μ is along its direction of motion **v**, with **E** and **B** collinear while normal to **v**. For reasonable values of $|\mathbf{E}|$ ($\approx$ 300 kV/cm) and of $|\mathbf{B}|$ ($\approx$ 0.005 T) that can be achieved within the confines of a neutron interferometer, the Anandan formula yields an acceleration $|\mathbf{a}| \approx 0.18$ cm/s². This would give rise to a measurable phase shift by current neutron interferometry techniques.

The Hamiltonian for the neutron moving in a region of space containing both an **E** field and a **B** field in the interferometer's frame is

$$H=\mathbf{p}^2/2m - \mu_o\sigma_z[\mathbf{B}-(1/mc)\mathbf{p}\mathrm{x}\mathbf{E}]\bullet\mathbf{z}.$$

Here $\sigma_z=+1$ or -1, **p** is the canonical momentum, the axis of quantization is along the unit vector **z**, parallel to the effective field $[\mathbf{B}-(1/mc)\mathbf{p}\mathrm{x}\mathbf{E}]$; and **E** and **B** are each constant. It then follows immediately, using Hamilton's equations, that the acceleration $\mathbf{a}=0$. This is contrary to the prediction of classical mechanics given by Anandan.

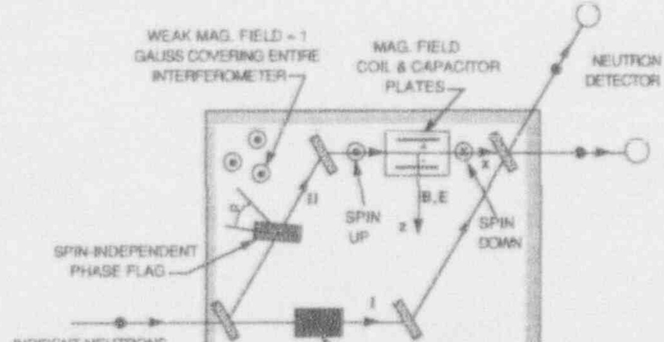

Fig. 1 Diagram illustrating a neutron interferometer arrangement to search for the putative Anandan acceleration.

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GAMMA-RAY SCATTERING GROUP

PERSONNEL

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James Mullen, Professor of Physics, Purdue University

This group at MURR is concerned with materials science research conducted with the University of Missouri Gamma-ray Spectrometer (MUGS) and the QUasiElastic Gamma-ray Spectrometer (QUEGS) instruments located on the beamport floor. Elastic scattering phenomena at photon wavelengths of 0.03 to 0.10 Å are investigated with MUGS whereas QUEGS uses Mössbauer photons with wavelengths in the 0.1 to 0.3 Å range to study non-elastic interactions in a manner that is unique in the world.

A major application of MUGS is the evaluation of crystal quality through the detailed investigation of mosaic structure by Bragg diffraction. This technique is routinely applied to various materials being considered for fabrication into detectors or monochromators. An on-going project for MUGS involves the evaluation of crystal perfection in mercuric iodide grown by EG&G-Santa Barbara for the development of the technology for room-temperature semiconductor detectors. Crystals grown in the micro-gravity environment of the space shuttle were analyzed using MUGS to determine the role of gravity in the growth of high-quality HgI_2 crystals.

Another collaborative research project using MUGS is the measurement of radiation channeling being pursued by a group from the University of Florida. This group has constructed a detector with a crystallographically oriented germanium crystal. The detector and its dewar assembly were mounted in the sample position of the MUGS instrument to search for anomalous transmission accompanying the channeling effect. In addition, the detector can be operated in coincidence with the transmission detector to analyze the energy effects of channeled radiation.

Research with QUEGS has been supported since 1983 by the Department of Energy in a collaborative program with Purdue University. These studies have ranged from fundamental line-shape analyses to anharmonic effects in sodium and tungsten. These line-shape studies have led to the most accurate values reported for interference parameters in five different Mössbauer transitions. In addition to a better understanding of the interference phenomenon, systematic trends in these measurements have implications concerning time-reversal-invariance.

Quasielastic broadening of the observed Mössbauer linewidth due to diffusive motions in pentadecane and polydimethylsiloxane have also been investigated. A substantial upgrade of QUEGS supported by the DOE and by University sources is planned for completion in Fall 1993.

MÖSSBAUER CHARACTERIZATION OF TUNGSTEN DOPED YBCO HIGH T_c SUPERCONDUCTOR

J. Day, J.G. Mullen (Purdue Physics), G. Schupp (MU Physics) and W.B. Yelon (MURR)

Many studies have been carried out on the $YBa_2Cu_3O_{7-\delta}$ (123) system since it was found to be superconducting at ~92 K in 1987. A number of these studies have dealt with modifying the 123 system by doping and then investigating the changes caused by the dopants. Besides offering insights into the physics of high transition temperature superconductors (HTSC), altering the basic structure by doping can lead to enhanced superconducting characteristics such as the transition temperature T_c or the critical current density J_c.

One such doped system is the $YBa_2Cu_{3-x}W_xO_{7-\delta}$ (123-W) superconductor. Because there was some question of where the tungsten site was located in the ^{123}W structure, we carried out a Mössbauer experiment to resolve this issue.

The Mössbauer experiment used a ^{182}Ta source and a ^{123}W, x = 0.08 absorber using the QUasiElastic Gamma-ray Scattering instrument (QUEGS) at MURR. Both the source and absorber were cooled down to liquid nitrogen temperature. The Mössbauer spectrum was collected until the baseline had approximately 520,000 counts.

Using precise line-shape analysis of the resultant spectrum, we were able to determine that there were two different tungsten sites (Figure 1). One corresponded to that of precipitated tungsten at the grain boundaries, and the other site was due to either a) tungsten substituting for copper chain sites, or b) precipitated WO_3. The precipitated tungsten can be seen in the figure as the large, center line (1), and the substituting tungsten can be seen as the hyperfine split lines (2), (3), and (4). Correcting for the large Debye temperature of the precipitated tungsten site, compared to the substituting tungsten, we calculated that approximately six times as much tungsten is located in the second site as compared with the precipitated tungsten site. Another experiment with the YBCO superconductor absorber cooled to 10 K will be performed to determine the location of the substituting site. If it turns out to be a copper chain site, the result will be exciting for superconductor research.

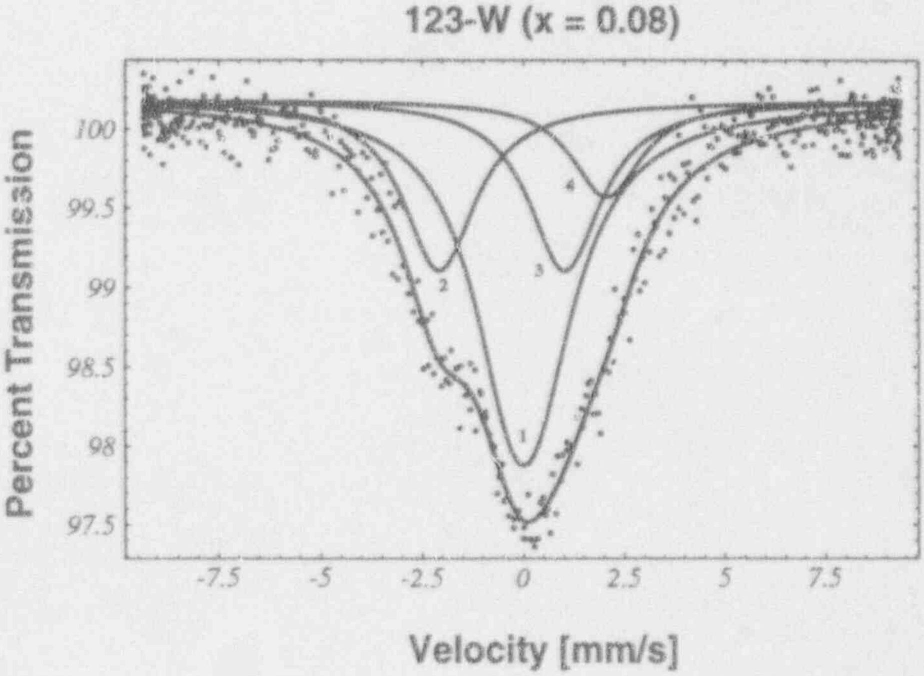


Fig. 1 ^{182}W Mössbauer spectrum for 8 atomic % W doped YBCO at 77 K

GAMMA-RAY SCATTERING STUDIES OF ALKALI HALIDES

R. Wagoner, J.G. Mullen (Purdue University Physics), and G. Schupp (MU Physics)

The change in elastic fraction of Mössbauer photons upon Bragg reflection was measured for NaCl, NaF and LiF crystals. The Mössbauer width of 2.5 meV for the 46.5-keV photons from ^{183}W allowed energy resolution not available in x-ray or neutron scattering methods. A natural tungsten foil (analyzer) was Doppler shifted first in a position between the source and crystal under study (before position) and then in a position between the crystal and the photon detector (after position). The two arrangements are shown schematically in Figure 1. Careful lineshape analysis gave the recoilless fraction, f_{sb}, for the photons incident on the absorber in the before case (which was also the same as the resonant fraction emitted from the source itself, f_s). The recoilless fraction for the after position, f_{sa}, is related to f_s by

$$f_{sa} = \frac{\varepsilon}{(\varepsilon + i)}\, f_s \ ,$$

where ε and i are defined as follows. If N is the number of Mössbauer photons incident on the crystal, εN and iN are the number scattered elastically and inelastically, respectively, into the detector. The effective fraction, F, is given by

$$F = f_{sa}/f_{sb} = \frac{\varepsilon}{\varepsilon + i} \ .$$

Our values of F for the crystals and Bragg reflections studied are tabulated in Table 1. The tungsten absorber was a natural foil 50.8 mm thick. Data were collected for the absorber and crystals at room temperature. It is noteworthy that the smallest value of F is still 0.942(5) for the (600) reflection from NaCl even though the integrated intensity of this reflection is more than an order of magnitude smaller than the (200) reflection. Large values of F are important since these crystals are widely used as monochromating filters. Further studies will focus not only on the elastic and inelastic scattering as a function of Bragg angle, but also as a function of temperature.

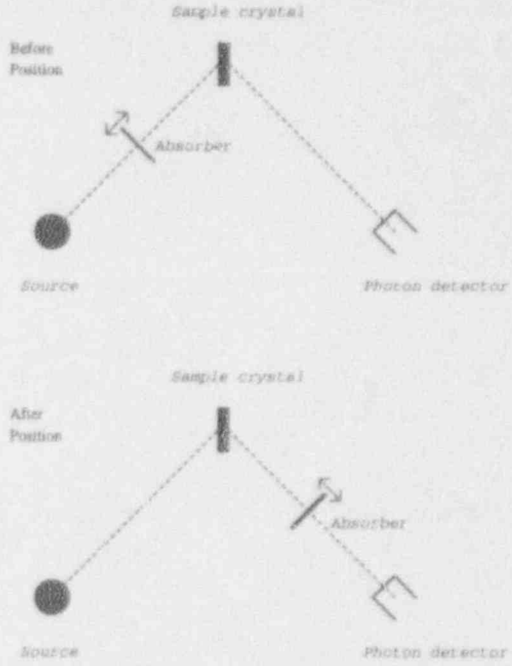


Fig. 1 Schematic geometries for the (a) before and (b) after positions used in these studies.

Table 1 **Values of F for the Crystals and Bragg Reflections Studied**

	reflection	*F* measured	*F* best fit
NaCl	(200)	0.980(4)	0.984
	(400)	0.966(4)	0.964
	(600)	0.949(6)	0.949
NaF	(200)	0.962(3)	0.973
	(400)	0.947(3)	0.945
	(600)	0.942(5)	0.939
LiF	(200)	1.000(3)	0.996
	(400)	0.991(3)	0.993

MÖSSBAUER SPECTRA OBTAINED BY MODULATING A SCATTERING CRYSTAL

G. Schupp, K. Barnes (MU Physics), W.B. Yelon (MURR) and J.G. Mullen (Purdue)

Supported by an Instrumentation Grant of \$74,065 from DOE and \$52,000 in MU matching funds, plans are underway to upgrade the Mössbauer quasielastic gamma-ray scattering (QUEGS) instrument with a new source cask to give four beam lines and a cryorefrigerator to yield $\leq$ 20 K source temperatures. In addition, the plans include a modification of our present source cask to accommodate velocity modulation of the source. This modification would allow use of a fixed Multifoil Internal Conversion Electron (MICE) detector which, in favorable cases, can give an improvement in the Mössbauer signal to continuum ratio of more than a factor of 10. Another method for obtaining the velocity modulation with a fixed MICE detector (it gets noisy when oscillated at high velocities for extended periods) is to move the monochromating filter crystal along the scattering vector (Q) while also leaving the source fixed. The difficulties associated with this approach are that the velocity component along the incident and scattered gamma-ray beams is commonly small and the scattered intensity depends on the uniformity of both the motion (lack of vibrations) and the crystal over the amplitude of its motion.

Initial measurements on oscillating the filtering crystal were recently taken which were very encouraging. Figure 1 shows a schematic drawing of the source-oscillating crystal-fixed absorber-Ge detector geometry used and Figure 2A shows the resulting multiscaling spectrum. The minima in Figure 2 occur at the turning points (positional extremes) of the oscillatory motion where the velocities are zero. Figure 2B shows an ordinary spectrum taken with the filtering crystal at rest and the absorber being oscillated along the scattered beam line. When the channels are converted to velocities via our "crank function," the width of the crystal modulated spectrum, Γ_M, is related to the usual Mössbauer width, Γ, by $\Gamma_M = \Gamma/(2\sin\theta)$, where θ is the Bragg angle for the reflection used. The data shown in Figure 2A were collected in 2.7 hours using the 400 Bragg reflection of a stressed LiF crystal; data were also collected using the 200, 600, and 800 Bragg reflections. The experimental values for Γ_M are given in Figure 2C along with the expected $\Gamma/(2\sin\theta)$ values for these measurements.

We were particulary pleased with the ease with which these data were obtained as well as with the excellent agreement seen in Figure 2C. Further experiments are planned using the MICE detector, different filtering crystals, and different scattering geometries to see if this method would be better operationally than modulating the source.

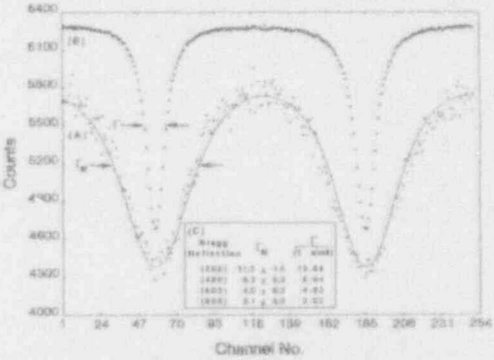
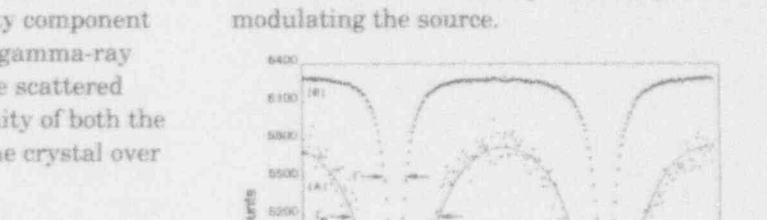


Fig. 2 (A) Multiscaling spectrum taken with the experimental arrangement shown in Figure 1. (B) Spectrum taken with the monochromating crystal fixed and the absorber oscillated along the scattered beam line. (C) Summary of the observed line widths versus the expected widths for the four Bragg reflections used.

Fig. 1 Schematic drawing for the source, oscillating monochromating filter crystal, fixed absorber, and Ge detector arrangement used in these measurements. A is the amplitude of the oscillatory motion.



REACTOR NUCLEAR ENGINEERING PROGRAM

NON-DESTRUCTIVE EVALUATION GROUP

REACTOR NUCLEAR ENGINEERING PROGRAM

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Reactor Nuclear Engineering Program Group:
Hydrogen Embrittlement and Non-Destructive Evaluation

The nuclear engineering program at MURR is intertwined with the MU Nuclear Engineering Program (MU NE) in the College of Engineering. Reactor facilities are used for laboratory classes and as the base for a significant number of research projects in collaboration with the MU NE program. A few of these research projects are described in summaries following this introduction and others are given in the *Hydrogen Embirttlement and Non-Destructive Evaluation Group* section.

MU is in the process of upgrading the research and operational capabilities of the MURR Center and associated facilities. The plans include an expanded research building that will double the laboratory space, the addition of new research programs, instrumentation and equip-ment, a cold neutron source, improved reactor systems, a new fuel design and a power increase. The work to support this effort is spread throughout various MURR groups and also involves many faculty associates.

The members of the Nuclear Engineering Program are especially involved in the new fuel and power upgrade analyses. This part of the upgrade has provided excellent graduate research projects for students in the MU NE program. Currently five PhD and one MS projects are in progress involving these areas. Since 1985 10 master's and 2 doctoral theses have been completed concerning the new fuel and power upgrades.

CONTAMINATED SHIPPING CASKS

Jay F. Kunze, Bruce Gu, George Vosnidis, Jun Lu (MU Nuclear Engineering), Michael D. Glascock and J. Steven Morris (MURR)

Spent fuel shipping casks will find major uses during the end of this decade, as spent fuel is shipped from the 110 operating power reactors to the Monitored Retrievable Storage (MRS) sites to be established by the US Department of Energy. With the shipments that have occurred over the last 15 years, there have been significant health physics control problems encountered, known as "weeping" or "sweating" of adsorbed surface contamination. A number of violations have been issued by the Nuclear Regulatory Commission (NRC) to the shipping organization when casks that were checked out as being below the surface contamination limits arrived at the receiving end well above the allowed "swipe" limits of 22,000 disintegrations per minute per 100 square centimeters.

A cooperative research effort with the Callaway Nuclear Power Plant and sponsored by Sandia National Laboratory exposed a number of 6-inch diameter pipe samples of typical shipping cask outer wall material in the Callaway Spent Fuel Storage Pool. The pool water contamination is less than 1 radioactive atom per 1012 water molecules (0.6 atoms of cobalt and 0.1 atoms of Cs-134 plus Cs-137 per trillion water molecules). The samples were then returned to the University for analysis and environmental testing to determine if weeping could be induced, and if so to try to understand the cause.

The results were most conclusive. Weeping was seen on all samples, but could not be correlated to any specific environmental condition. It appeared that time and changing environmental conditions were all that were necessary to induce weeping. Figure 1 shows a typical batch of data, from a variety of environmental tests from 0° F to 100° F, low to high humidity, and with varying temperature gradients across the pipe wall and surface stresses, simulating those that would be present in a shipping cask. As the figure shows, the weeping was as much as a factor of 10 (in a few cases as high as 100) above the initial surface contamination.

As part of the assessment, measurements on the consistency of results of swipe measurements were conducted, under a variety of conditions and using a variety of health physics "technicians." The conclusion was that variations in swipe technique were negligible compared to the weeping effects found. It was also concluded that variations over the cask surface could be significant, and, therefore, that a 100 square centimeter area for the swipes was too small an area to obtain a truly representative sample--or alternatively, that many areas of 100 sq cm needed to be swiped to obtain a representative surface contamination measurement.

Most surprising, however, was the result of the total contamination measured on the samples, something that never could be done on the actual shipping casks. In general, the measured "removable" surface contamination (as measured with a swipe) was only 1% or less of the total "fixed" contamination. Hence, the potential for just a few percent of the "fixed" contamination freeing itself to become "removable" contamination would account for the enormous potential for "weeping."

Work then continued to attempt to determine how to remove this fixed contamination. These "cleaning" methods are still being investigated, but the preliminary conclusion is that there is no commercially acceptable method of successful removal. Future work will address methods of passivating the surface so that the contamination might never occur. The magnitude of the contamination is only of the order of one radioactive ion per 100,000 surface metal atoms, and appears to reach saturation at this level. Hence, it is being assumed that the actual interactions with the surface atoms must involve dislocations or other rare aberrations on the surface.

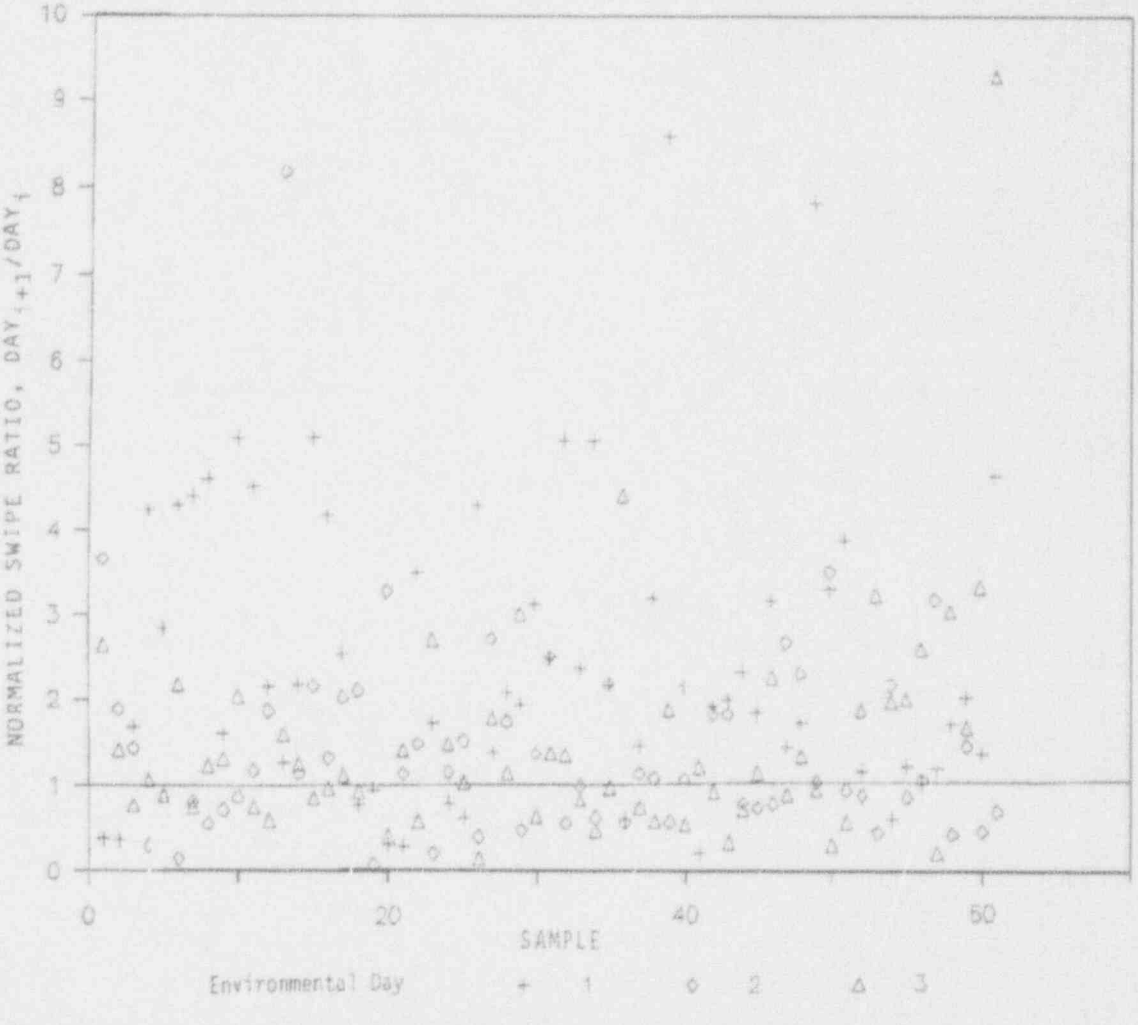


Fig. 1 Normalized swipe results. Scatter plot of "weeping" results from 61 samples (along the abscissa) following each of three days of various environmental testing. The ordinate represents the ratio of the swipe on the given day to that on the previous day, correcting for the efficiency of the swipe method and the actual amount of contamination removed by the first swipe. No weeping would be represented by a 1.0 ratio. A ratio less than 1.0 indicates either a fixing of removable contamination, a loss of some removable contamination, or merely statistical variations in the measurement. Values greater than 1.0 (by far the predominant situation) implies either weeping or statistical variations.

USING THE CODE RELAP FOR SAFETY ANALYSIS OF RESEARCH REACTORS

William Chao, Jay Kunze, Sudarshan Loyalka, Weimin Dai (MU Nuclear Engineering), Roland Hultsch, J. Charles McKibben, and Olawale Oladiran (MURR)

The loss of coolant accident (LOCA) in an operating reactor is a most unlikely event, yet one of the most severe safety concerns. This involves a major leak that results in sufficient loss of coolant that the removal of shutdown (decay) heat from the core is compromised. The credible event that would initiate such an event is a major earthquake that would result in relative movement of plant components sufficient to rupture an inlet or outlet pipe to the core vessel.

The RELAP thermalhydraulics time dependent computer code is one tool for analysis of a LOCA. The code development was sponsored by both the Department of Energy and the Nuclear Regulatory Commission, and has been certified by the NRC for analysis of LOCA for most of the commercial nuclear electric power generating plants. The code has not been tested through experiments on the quite different conditions of research reactors, which normally operate at near atmospheric pressure and temperatures generally well below 100 C.

When the code was applied to the LOCA analysis of MURR, the results for the targeted 30 MW operating level for the proposed upgraded reactor power indicated that fuel temperatures would approach within a few hundred degrees F of the level that would result in cladding damage. Because the code had never been "benchmark tested" under similar conditions, it was proposed that an appropriate benchmark experiment be conducted to check the code. This work has been sponsored by DOE during the last three years, since concerns similar to MURR's were found for the high power test reactors owned by DOE.

When initially conducting the benchmark test, it was found that degassing of the dissolved gas in the water resulted in enhanced heat transfer at much lower temperatures than where nucleate boiling would be expected to occur. Hence, enhanced heat transfer resulted compared to code predictions. The issue of degassified water is real for these research reactors, for most operate at several atmospheres pressure. In the case of MURR, the solubility of the water for air at operating temperature and pressure is several times higher than for the fill conditions at room temperature and atmospheric pressure. Yet during a LOCA, boiling temperatures will be reached quickly, preceded by complete degassing of the water moving through the fuel plates. The effect is one of enhanced heat removal capability initially, compared to what is predicted by RELAP. There is also some resistance to the flow reversal that occurs, resulting in upward natural convection flow, but this is a much smaller overall effect on peak temperature than is the enhanced heat transfer.

Comparisons between RELAP and a benchmark experiment involving a single channel (with the same width as between MURR fuel plates) are shown in Figure 1. RELAP currently has no way of accounting for the degassing, hence underpredicts the heat removal and overestimates the peak cladding temperature. Furthermore, it even under-estimates the heat transfer in single phase laminar flow. However, once true boiling commences and dissolved gas is no longer present, the RELAP code appears to give an accurate prediction of temperature changes, even under those of pulsed boiling in the narrow fuel element channels. The problem with laminar flow conditions is somewhat under the input control of the user, since different hydraulic diameters may be selected. However, the incorporating of dissolved gas effects would require computer code source program changes. The answer to the initial concerns about the safety analysis results is that RELAP had significantly overpredicted the peak fuel temperatures for the LOCA at MURR. Work is continuing on a related project in attempts to find a way of encoding the dissolved gases into the RELAP computer code.

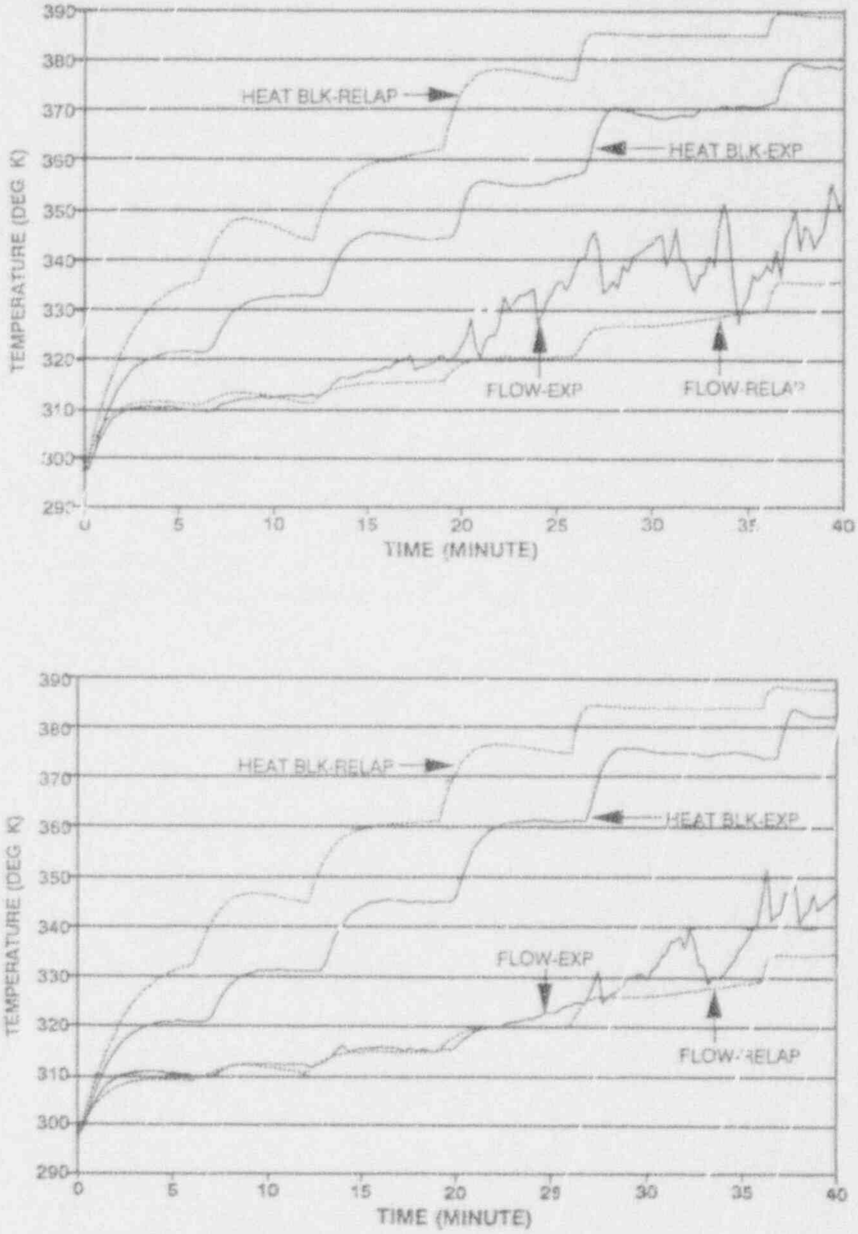


Fig. 1 Temperature comparisons between RELAP and measurements in the 2.032 mm (0.080 inch) heated water channel. Discrepancy occurs initially with laminar flow, and is worse for water with dissolved gas as the gas comes out of solution. The degasified water appears to give better agreement once nucleate boiling commences, indicating true turbulent flow conditions.

TIME DEPENDENT FLUX CHANGES IN MURR

Mohammed Salih, Jay Kunze (MU Nuclear Engineering) and Kiratadas Kutikkad (MURR)

The MURR is a reactor with a substantially different neutron energy spectrum in the reflector compared to that in the core, and the central flux trap also provides significant differences. The prompt neutron lifetime differs substantially between these parts of the reactor. Though several related studies and measurements had been made on MURR around 1970 (two PhD dissertation topics), the results were largely inconclusive concerning the ability of existing computer codes to analyze adequately neutron flux and power changes with time during rapid positive or negative transients.

This question is being reexamined because of the issues related to shutdown heat removal following a loss of coolant accident (LOCA). At such a time much occurs in the first second, including a rapid change in the flux levels and flux distribution throughout the reactor. The critical period for heat removal from the fuel plates is the moment that flow stops and reversal occurs to the upward convective flow direction. This happens within the first two seconds.

These studies of the flux distribution changes in short time periods following rapid control rod insertion are being reexamined using newer or more applicable computer codes than were available in the 1970 studies. Where appropriate, the results will replace the use of point kinetics analysis for evaluating safety characteristics of MURR.

REACTOR PLANT OPERATION DISPLAY SYSTEM and NUCLEAR PLANT RADIOLOGICAL DISPLAY SYSTEM

Mohammed Enani, Jay Kunze (MU Nuclear Engineering) and Kiratadas Kutikkad (MURR)

Modern computer systems can provide most helpful graphical displays of conditions of a complex operating system such as a reactor. These systems can also be used as training simulators to represent hypothetical emergency response conditions for an operating reactor plant (or of other complex systems). Incorporation of graphical displays and simulators for operator training has not been implemented at MURR to the extent that it has at the commercial power reactors throughout the US. Although the implementation of these systems on MURR may not be considered necessary (for safety and cost reasons), the advantages in ease of understanding current conditions and for providing simulator training are significant, albeit somewhat intangible, benefits.

Attempts to program such useful graphical display and analysis systems are now underway, primarily as a student research project. One generally new developmental application will be to use a nodal core analysis method to display fuel reloading effects, such as power distribution.

EXPERT SYSTEM FOR THE MURR CONTROL ROOM

Gin-Weigh Wu and Jay F. Kunze (MU Nuclear Engineering), Kiratadas Kutikkad and Walt Meyer (MURR)

Modern computer technology, especially desktop microcomputers, permit the fast accumulation of many data points, storage of these to a hard disk, and on-line evaluation of trends in these data points. This application is designated an "expert system," i.e., a computer monitor of a complex operational system, providing on-line evaluation according to an engineer-supplied analytical method. Such a system would not substitute for an operator, but would alert the operator to development of off-normal conditions. With the vast number of data points to be monitored, the computer expert system can process much more data than can the operator's eyes and brain, and can analyze more readily these data for trends and subtle changes, usually alerting the operator to them long before a conventional trip set point would give an alarm.

Only recently such systems are being applied to commercial power reactors, where there are hundreds of important data channels, any one of which might give an indication of potential plant shutdown. A brief shutdown of a 1200 MW(e) commercial nuclear power plant is costly, typically requiring purchase of about $1 million of replacement power. A brief shutdown at MURR is not nearly as expensive, but costly nevertheless in terms of irradiation and experiment interruptions, and stress on plant components.

An expert system is planned for installation in the Control Room at MURR. The connections to the control system must guarantee that the computer can provide no feedback signal into the control system, and in no way affect the control system readings or response. The several dozen channels being monitored must be recorded at a rapid rate, at least once a second, and as much as 1000 times per second for some, so that Fourier analysis of the signal can be performed. The data acquisition system now being assembled has a capability of 20,000 data points per second, and the computer to manipulate the data will have a capability of about 10 MIPS (million instructions per second). Installation in or adjacent to the Control Room is anticipated in spring 1993.

NON-DESTRUCTIVE EVALUATION GROUP

PERSONNEL

William H. Miller, Group Leader, Professor of Nuclear Engineering, Director of Energy Systems and Resources Program

Kiratadas Kutikkad, Research Scientist (Adjunct Assistant Professor, Nuclear Engineering)

The non-destructive evaluation group is a portion of the Nuclear Engineering program at the MURR Center. It encompasses a variety of experimental techniques –other than neutron scattering techniques–which can be used for non-destructive analysis. Current projects include: 1) the measurement of hydrogen content in materials of interest; 2) the relocation of the prompt neutron activation analysis facility; 3) the development of a depth profiling facility; and 4) maintenance and utilization of the filtered beams on beamport F.

With respect to hydrogen measurements, the modified notched neutron spectrum technique has been refined to give an experimental uncertainty of 0.4 weight ppm in metals, or an absolute mass sensitivity of less than 10 micrograms. The technique has now been applied to the measurement of hydrogen in steel, in Ti/Al alloys and in porous silicon. Work has been proposed to measure deuterium in metals (in support of neutron scattering work) and water in archaeological samples.

The prompt neutron activation analysis facility has been removed from beamport A in preparation for the installation of TRIAX there. Work is now underway to place this facility behind the scattering experiments on beamport D. The designs of several other PGNAA facilities have been obtained from other institutions in order to optimize the new design. Improvements in shielding design and detector placement are expected to provide modest improvements in sensitivity compared to the previous system. This new facility is also being designed to allow the addition of a depth profiling facility in the future.

A DSP-BASED, SMART ANALOG-TO-DIGITAL CONVERTOR FOR NUCLEAR RADIATION DETECTORS

William H. Miller (MU Nuclear Engineering and MURR) and Ron R. Berliner (MURR)

An innovative, digital signal processor (DSP) based system is being developed for analyzing and processing pulses produced by radiation detectors. This system will provide more accurate analysis of pulses from all radiation spectroscopy systems including those used for health physics instrumentation, environmental monitoring, and personnel monitoring. It is designed to replace pulse-type, Wilkenson and successive approximation analog-to-digital convertors (ADCs) that are currently used for pulse height analysis of detector pulses. Energy information will be derived by digital integration of the detector pulse, leading to a more accurate result. In addition, a DSP-based system will be capable of advanced features (e.g., pulse validation, pulse shape analysis, dynamic threshold determination, and noise reduction) while using fewer and simpler electronics for increased reliability, reduced electronic noise and less expense.

Progress to date includes the development of a wire-wrap, prototype circuit that accepts pulses from the detector, triggers ADC conversion based upon a digital threshold, stores the result in a first in, first out (FIFO) buffer, and provides an interface to a computer for data transfer and ultimate data analysis. This system is currently running at a 10 MHz sampling rate and is shown in Figure 1. It is being prepared for implementation on a printed circuit board.

Software development has also been initiated. Cross-correlation functions for pulse shape discrimination have been tested in both DSP and 486 PC environments. These functions provide excellent pulse shape discrimination capabilities. Preliminary tests also suggest that a combined pulse height and pulse shape analysis can be performed at rates of 10,000 counts per second, which is adequate for most detector applications.

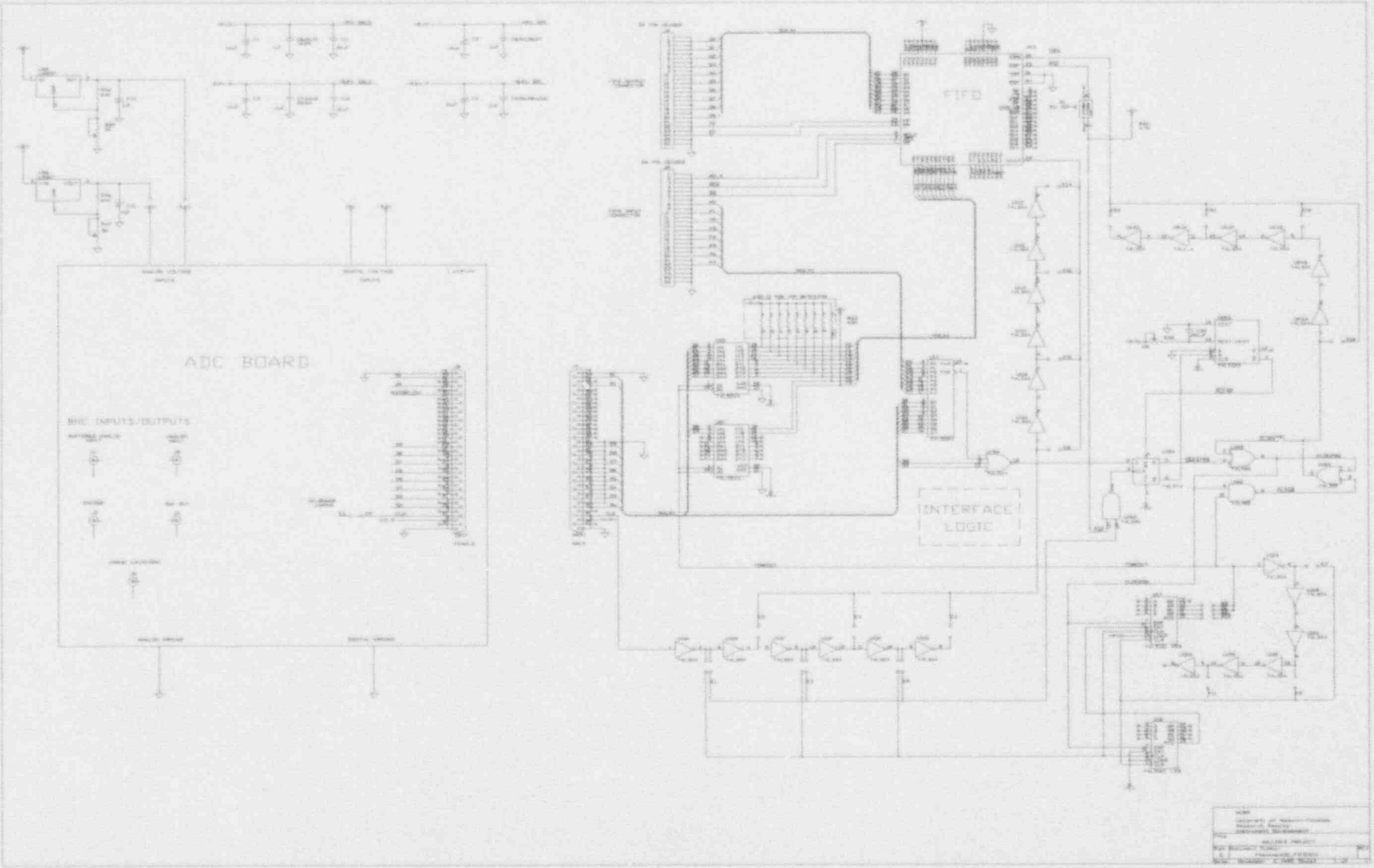


Fig. 1 Circuit diagram for the DSP nuclear detector pulse processor system

THE MODIFIED NOTCHED NEUTRON SPECTRUM TECHNIQUE FOR THE DETERMINATION OF HYDROGEN

William H. Miller (MU Nuclear Engineering and MURR)

The non-destructive determination of hydrogen content is important to many different disciplines. Unfortunately, techniques that accurately and nondestructively measure hydrogen content at parts-per-million (ppm) levels have not been readily available. One possible hydrogen measuring technique is the "modified notched neutron spectrum technique" that has been developed on beamport F at MURR. It is based upon a technique that was suggested approximately 25 years ago and is currently in use at EG&G Idaho, but has been refined here at MURR to increase the sensitivity by approximately a factor of 20. The resulting sensitivity is 0.4 weight-parts-per-million (wppm) of hydrogen in metals, or an absolute sensitivity of 10 micrograms.

The notched neutron spectrum technique uses a filtered spectrum that has been depleted of neutrons in the epithermal energy range by absorption at resonances. The technique then measures the extent to which these resonance regions are repopulated by moderation of the neutrons through hydrogen scattering interactions. By improving the geometry of activation foils that detect this moderation, the sensitivity of the technique is greatly increased.

The experimental technique utilizes the F beamtube. The beam is filtered by 0.1 cm of cadmium and 0.64 cm of indium. The sample holder holds the sample of interest between two activation foils that have an inner diameter of 4.4 cm and an outer diameter of 9.5 cm. These foils, which are a 0.1 cm thick alloy of 0.5% indium in lead, are activated for a minimum of 6 hours. They are then counted for approximately 3 hours between two opposing NaI detectors creating nearly 100% counting efficiency.

Several different types of samples containing hydrogen have been studied using this new technique. Figure 1 shows typical data for hydrogen in steel. As can be seen, accurate results below 1.0 wppm are possible. Samples of Ti/Al alloy have been loaded with large amounts of hydrogen (on the order of 2% hydrogen by weight or 20,000 wppm) and measured. Experimental measurements have been compared with hydrogen weight gain measurements and the results have agreed to within 4%. It was also found that the technique is linear over the range from 20,000 wppm (for Ti/Al) to 0.6 wppm (for steel). Finally, hydrogen content in porous silicon has been measured at concentrations down to 40 micrograms.

This technique has utility for a wide variety of applications in which the amount of hydrogen content must be determined. The measurement process is straightforward, is nondestructive, can be applied to radioactive samples, and provides a new analytical tool for determining hydrogen content.

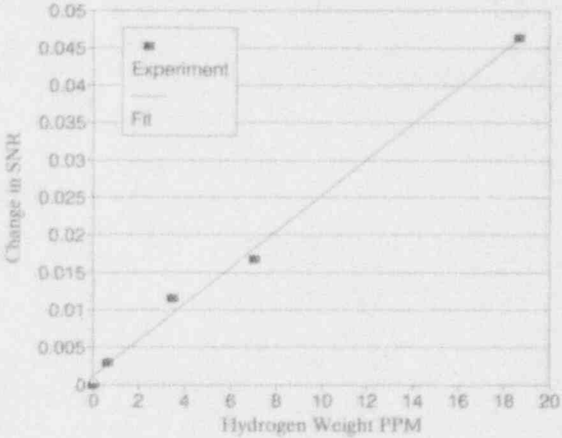


Fig. 1 Changes in foil signal-to-noise ratio (SNR) vs. hydrogen

UNIVERSITY REACTOR SHARING PROGRAM

UNIVERSITY REACTOR SHARING

The US Department of Energy has provided financial support for MURR's Reactor Sharing Program since 1980. During the annual reporting period completed early in FY92 DOE provided $73,000 to support the program and MURR has matched this with $33,507. DOE started this program to provide increased availability of university nuclear reactor facilities to non-reactor-owning colleges and universities (user institutions). MURR, with our unique facility, also provides support to institutions with research reactors that do not have the capabilities to perform the required experiments. In FY92, 39 research projects involving 99 faculty and 41 students at the 31 institutions listed below were supported:

Arizona State University
Auburn University
Cal State University-Fullerton
Dartmouth College
Franklin and Marshall College
Harvard University
Iowa State University
Johns Hopkins University
Louisiana State University
New Mexico Institute of Mining & Technology
Oregon State University
Princeton University
Purdue University
Ripon College
Stephens College
University of Arizona
University of Arkansas
University of California-Berkeley
University of California-Los Angeles
University of Chicago
University of Cincinnati
University of Conneticut
University of Hawaii
University of Kentucky
University of Massachusetts
University of Medicine and Dentistry of New Jersey
University of North Dakota
University of Rochester
Utah State University
Vanderbilt University
Washington University (St. Louis, MO)

Projects involved work in many fields including anthropology, archaeology, animal science, analytical epidemiology-nutrition, crystallography, geology, materials science, physics, cosmo-chemistry, biochemistry-nutrition, nuclear analysis development, genetics and human nutrition. A list of some of the publications resulting from Reactor Sharing projects at MURR follows.

PUBLICATIONS RESULTING FROM REACTOR SHARING PROJECTS

Davis, A.M.; MacPherson, G.J.; Clayton, R.N.; Mayeda, T.K.; Sylvester, P.J.; Grossman, L.; Hinton, R.W.; Laughlin, J.R., Melt Solidification and Late-Stage Evaporation in the Evolution of a FUN Inclusion from the Vigarano C_3V Chondrite, *Geochimica et Cosmochimica Acta* (submitted)

Haas, J.R.; Haskin, L.A., Compositional Variations Among Whole-Rock Fragments of the L6 Chondrite Bruderheim, *Meteoritics* (submitted)

Korotev, R.L., Geochemical Stratigraphy of Two Regolith Cores from the Central Highlands of the Moon, *Proceedings of Twenty-First Lunar and Planetary Science Conference*, Lunar and Planetary Institute, Houston, TX, March 12-16, 1990 (submitted)

Owens, B.E.; Dymek, R.F., Fe-Ti-P-Rich Rocks and Massif Anorthosite: Problems of Interpretation Illustrated from the Labrieville and St. Urbain Plutons, Quebec, *Contributions to Mineralogy and Petrology* (submitted)

Owens, B.E.; Dymek, R.F., Petrological Constraints on the Origin of Anorthosite of Extreme Composition: The Labrieville Massif (LBV), Quebec, *Journal of Geology* (submitted)

Shelton, M.E.; Green, M.A.; Mathias, C.J.; Welch, M.J.; Bergmann, S.R.; James, H.L.; Weinheimer, C.J., Assessment of Regional Myocardial and Renal Blood Flow Using Copper-PTSM and Positron Emission Tomography, *Circulation* (submitted)

Smith, M.S.; Dymek, R.F.; Chadwick, B., Petrogenesis of Archaen Malene Supracrustal Rocks, NW Buksefjordfen Region, West Greenland: Geochemical Evidence for Highly Evolved Archaean Crust, *Precambrian Research* (submitted)

Smith, M.S.; Dymek, R.F.; Schneiderman, J.S., Implications of Trace Element Geochemistry for the Origin of Cordierite-Anthophyllite Rocks from Orijarvi, SW Finland, *Journal of Geology* (submitted)

Sylvester, P.J.; Ward, B.J.; Grossman, L., Chemical Compositions of Siderophile Element-Rich Opaque Assemblages in an Allende Inclusion, *Geochimica et Cosmochimica Acta* (submitted)

Wang, M-S.; Lipschutz, M.E., Consortium Study of MacAlpine Hills 88105 Lunar Meteorite, *Geochimica et Cosmochimica Acta* (in preparation)

Windsom, K.E.; Seifert, K.E.; Anderson, R.R., Studies of the Precambrian Geology of Iowa: Part 1. The Otter Creek Layered Igneous Complex, *Journal of Iowa Academy of Science* **98**(4), 170 (1991)

Windsom, K.E.; Seifert, K.E.; Anderson, R.R., Studies of the Precambrian Geology of Iowa: Part 2. The Matlock Keratophyre, *Journal of Iowa Academy of Science* **98**(4), 178 (1991)

EDUCATIONAL ACTIVITIES

July 1991 - June 1992

- 12 faculty, 48 graduate students and 68 undergraduate students were provided financial assistance by MURR
- 6 doctoral and 6 master's degrees were granted for research at MURR
- 3,043 people from 32 countries toured MURR

NSF RESEARCH EXPERIENCES FOR UNDERGRADUATES AT MURR

For the fourth consecutive year, MURR was selected by the National Science Foundation (NSF) as a site for a summer "Research Experience for Undergraduates" program. Eleven students representing seven universities participated and seventeen MU faculty and Reactor Center staff served as mentors to provide research guidance and consultation for the students. Each student worked with one or two mentors to design and carry out an individual research project. In addition, a dozen other faculty and reactor staff members joined the mentors in providing 35 hours of lecture/discussion/training sessions for the students. MURR has now trained 44 future scientists in "Scientific Applications Involving Neutrons" under the sponsorship of the NSF. The program is currently funded for three years and will be offered again during the summers of 1993 and 1994. A list of student projects is given at the end of this section.

Although copper is known to be an essential trace element, a recommended daily allowance of this element is yet to be determined. Copper is bound to many important enzymes, and transport proteins are involved in binding this trace element and facilitating absorption. Maggie Stammeyer undertook a project to study the binding of copper to proteins. She worked to develop a model system to study how various parameters affect the binding of copper to known proteins, so that unknown copper-binding proteins could be characterized. The parameters investigated included pH, buffers, reducing agents, and the influence of other trace elements such as zinc and iron on copper binding. Known proteins (24) were blotted to nitrocellulose with a slot-blot apparatus, subjected to varied conditions, probed with high specific activity Cu-64 (300 Ci/mg Cu), and autoradiographed. The results of one study are shown in Figure 1. The binding of Cu-64 was protein dependent, with greatest binding by carbonic anhydrase (D1) and metallothionein (G2). Preincubation with nonradioactive copper reduced Cu-64 binding by all proteins, as indicated in Figure 1(b). The Cu-64 binding by certain proteins was reduced by preincubation with zinc, particularly superoxide dismutase (F3). This technique was developed for studying copper-binding proteins using high specific activity Cu-64, and it appears to offer many advantages in the investigation of copper metabolism.

Neutron diffraction can be used to obtain information about the arrangement of molecules on surfaces and about changes which may occur during heating or cooling of the surface. Douglas Seman used a computer modeling method called molecular dynamics to predict the behavior of n-propanol molecules absorbed on a graphite surface (Figure 2). As with the shorter alcohols studied previously, hydrogen-bonding (the affinity of a hydroxyl hydrogen atom on one molecule for the oxygen atom of another molecule) appears to be the dominant interaction in the system. The molecular dynamics method uses the computer to generate all possible atom motions for a given strength of molecule-surface and molecule-molecule interactions. When the model correctly predicts the behavior of the surface layer of atoms (i.e., the properties agree

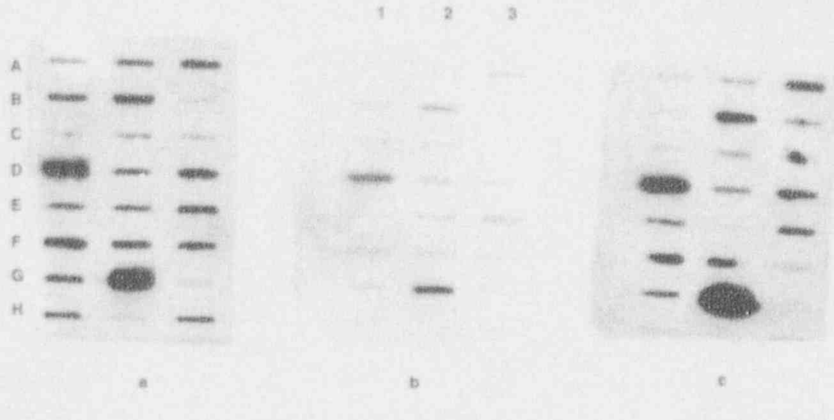


Fig. 1 Scans of 3 autoradiographs. Figure 1(a) is the control, with no additional treatment other than probing with Cu-64. The blot displayed in Figure 1(b) was preincubated (30 min) with nonradioactive copper (10 µg/100 ml) while the blot in Figure 1(c) was preincubated (30 min) with nonradioactive zinc (10 µg/100 ml).

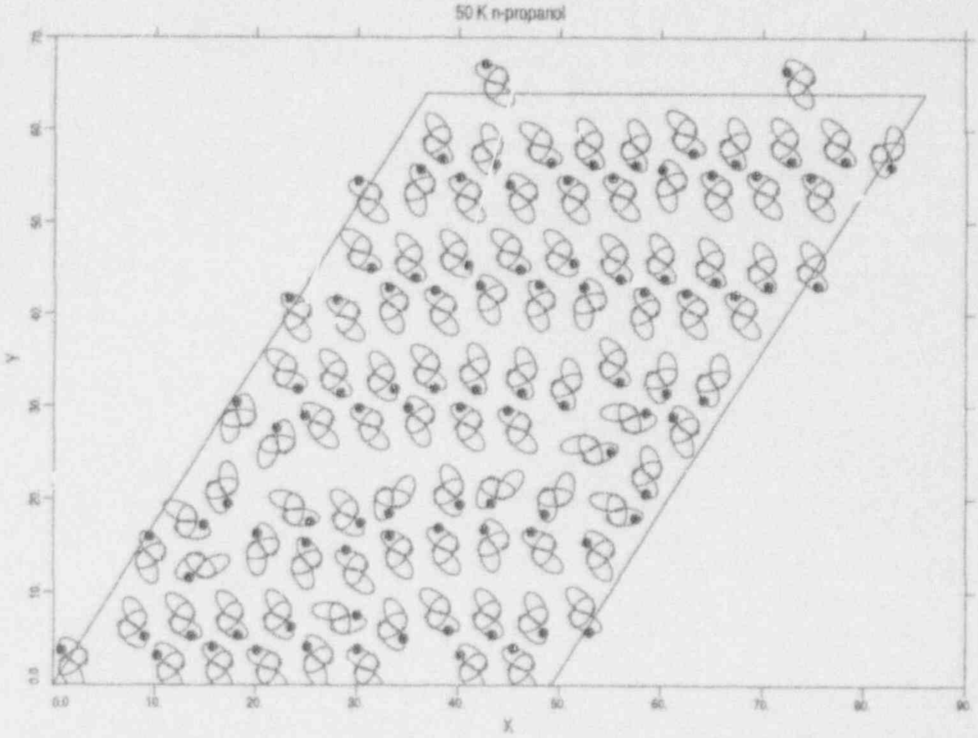


Fig. 2 A schematic view of a surface layer of n-propanol molecules on graphite as predicted by molecular dynamics. The ellipses are drawn to illustrate the outline of the molecular backbone in a space-filling manner. Solid circles indicate hydrogen atoms responsible for hydrogen bonding.

with those derived from neutron scattering measurements), the intermolecular forces have been adequately described. The model can then predict other properties of surface layers and properties of bulk (3-dimensional) solids and liquids which are not easily accessible by experimental methods.

When silicon is used as an anode in an electrolytic cell with an HF solution as the electrolyte, a chemical reaction occurs on the surface of the silicon. The resulting material, apparently a residual skeleton from partial dissolution of the silicon, exhibits substantially altered physical and chemical properties. This "porous silicon" is obviously altered in color, it exhibits photoluminescence and it promises to have interesting electrical, chemical and quantum properties. David Gilliam's summer project was to synthesize porous silicon and to use neutron and X-ray diffraction techniques to gain information about its structure. The results of David's diffraction experiments (Figure 3) show that porous silicon has lost most of the long-range structural order that the original material possessed and has incorporated substantial amounts of hydrogen. Further studies to investigate the catalytic properties of porous silicon are planned.

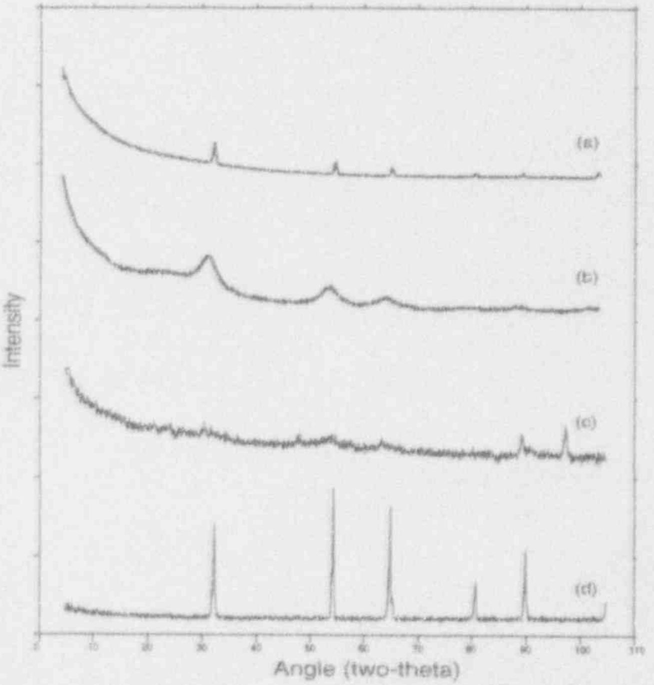


Fig. 3 Diffraction patterns of silicon powder for neutrons (a) and X-rays (d) compared with porous silicon diffraction of neutrons (b) and X-rays (c). The two high-angle peaks in (b) result from scattering by boron nitride in the sample cell.

Dale Witte's project called for engineering design and data analysis skills to be applied to residual stress measurements of welds in the Advanced Solid Rocket Motor for the NASA space shuttle. The Electronic and Mechanical Properties group at MURR is studying a scaled-down model of one of the cylindrical sections which will be welded together to make the reusable rocket motor. Recovery of the motor from the ocean for reuse is feasible only if the problem of stress corrosion cracking (initiated by chemical reaction of stressed steel with salts in ocean water) can be eliminated. The actual cylinder sections (12 ft diameter by 10 ft tall, 0.6" thick high-strength steel) were far too large to be handled and the 230 pound model (3 ft diameter by 1 ft tall – also 0.6" thick) presented difficulties as well. Dale worked with several engineering professionals to program a computerized robot arm assembly which held the heavy model in precise orientations in a small beam of neutrons from the reactor (Figure 4). Diffraction from the beam was analyzed to determine stresses at various points in the vicinity of the weld. Calculation of the stress tensor at each point required investigation from at least six different directions using a diffractometer designed to work with much smaller samples. Dale's calculations permitted the group to make the necessary measurements for accurate stress determinations (Figure 5). The model will be heat-treated and remeasured to verify that stresses associated with the welding process have been removed. Dale also helped analyze a flat plate model of the weld and developed computer programs to aid in data analysis.

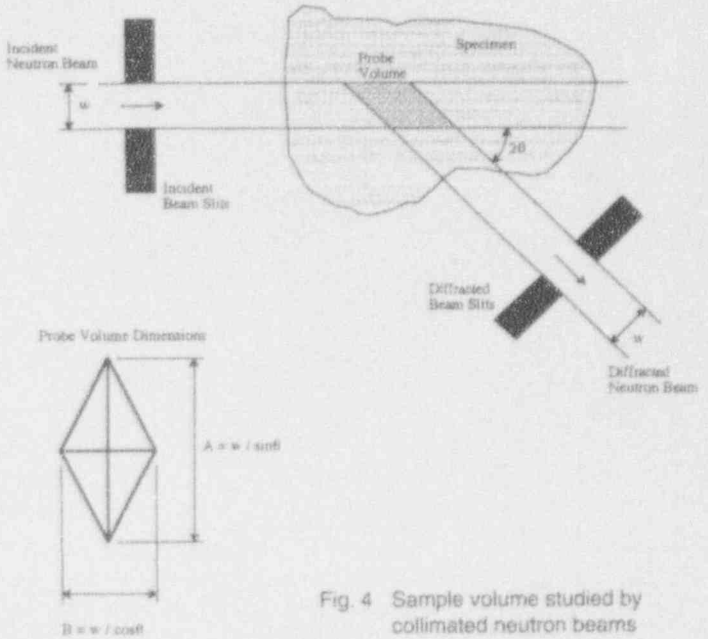


Fig. 4 Sample volume studied by collimated neutron beams

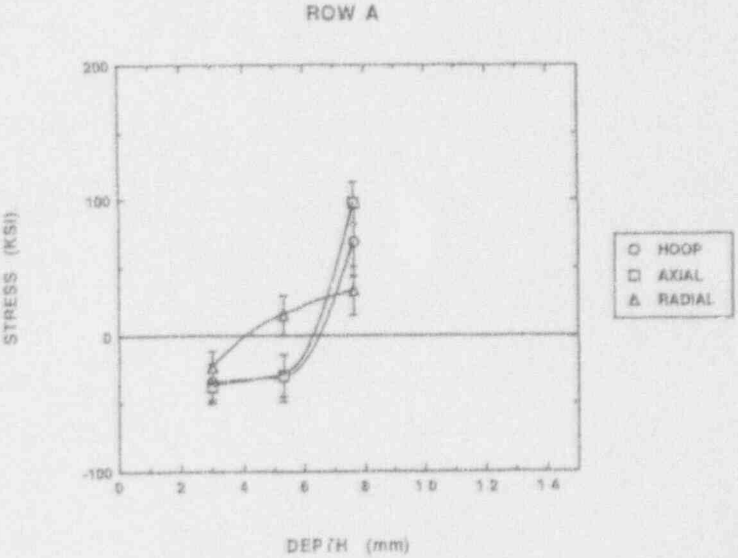


Fig. 5 Variations of stresses with depth in sample. Note that radial stresses are relatively constant but axial and hoop stresses change sharply between 6 and 8 mm in depth.

1992 Summer REU Program
Student Projects

Patience Bennett (MU Electrical and Computer Engineering), *"Automation of a Sample Changer for a Neutron Beam Hydrogen Spectrometer"* Mentor - Dr. W. Miller, MU Nuclear Engineering

Janice Bequette (Central Missouri State University Industrial Safety), *"An Inventory of Potentially Hazardous Chemicals at MURR"* Mentor - Dr. S. Langhorst, MURR Health Physics

Albert Crook (St. Louis University Math and Physics), *"Computer Simulation of Electrochemical Concentration Gradients"* Mentors - Dr. T. Storvik (MU Chemical Engineering) and Dr. L. Krueger, MURR Actinide Chemistry Group

David Gilliam (Lincoln University Physics and Math), *"Structural Measurements of Porous Silicon with Neutron Diffraction"* Mentor - Dr. B. Heuser, MURR Nanostructures and Excitations Group

Joseph Kim (Northwestern University Chemistry), *"Toenails as an Indicator of Arsenic and Selenium in Humans - Determination by Neutron Activation Analysis"* Mentor - Dr. J.S. Morris, MURR Nuclear Analysis Program

Kent Riley (University of Michigan Nuclear Engineering), *"Development of a Remote Processing Technique for Radiopharmaceutical Production"* Mentors - Dr. A. Ketring and Dr. G. Ehrhardt, MURR Radiopharmaceutical Group

Doug Seman (University of Michigan Engineering and Physics), *"A Study of Molecular Dynamics of n-propanol Physiosorbed on Graphite using Computer Simulation"* Mentor - Dr. K. Herwig, MURR Nanostructures and Excitations Group

Maggie Stammeyer (MU Chemical Engineering), *"A Procedure for the Study of Trace Metal Binding to Proteins using High Specific Activity Cu-64"* Mentors - Dr. T. Chaudhuri and Dr. K. Zinn, MURR Analytic Epidemiology, Nutrition and Immunology Group

David Walters (University of Maryland Chemistry), *"Chemical Characterization of Obsidian Sources in the American Southwest and Northern Mexico"* Mentors - Dr. M. Glascock and Dr. H. Neff, MURR Nuclear Archaeometry and Geochemistry Group

Dale Witte (MU Mechanical & Aerospace Engineering), *"Characterization of Residual Stresses in a Sub-scale Cylinder of the Advanced Solid Rocket Motor"* Mentors - Dr. A. Krawitz and Dr. A. Winholtz, MU Mechanical & Aerospace Engineering and MURR Electronic and Mechanical Properties Group

Kim Woods (MU Electrical Engineering), *"Automation and Control of a 4-circle Neutron Diffractometer"* Mentors - Dr. H. Yasar and Dr. F. Ross, MURR Magnetic and Crystal Structures Group

COURSE LECTURES AND LABS PRESENTED BY FULL-TIME STAFF

July 1991 - June 1992

DATE	LECTURER	COURSE TITLE AND SUBJECT OF LECTURE
07/91	R. Berliner	NSF REU: Neutron Powder Diffraction
07/91	T. Chaudhuri	NSF REU: New Radiotherapies for Cancer Treatment
07/91	T. Chaudhuri	NSF REU: Areas of Fundamentals Related to Monoclonal Antibodies in Nuclear Medicine and Role of Surface Antigens in the Detection and Treatment of Ovarian and Other Cancers
07/91	G.J. Ehrhardt	NSF REU: Radioisotope Generators
07/91	G.J. Ehrhardt	NSF REU: Use of β-Emitting Microspheres in Radiotherapy
07/91	J.W. Farmer	NSF REU: Radiation and Defects in Solids
07/91	M.D. Glascock	NSF REU: Nuclear Analysis Methods
07/91	B.J. Heuser	NSF REU: Small Angle Neutron Scattering
07/91	H. Kaiser	NSF REU: Neutron Interferometry
07/91	A.R. Ketring	NSF REU: Radiopharmaceutical Chemistry
07/91	C.L. Krueger	NSF REU: Actinide Research at the MURR
07/91	J.S. Morris	Columbia College, Introduction to Radiochemistry: Fundamentals of Radiochemistry
07/91	J.S. Morris	NSF REU: Nuclear Applications in the Life Sciences
07/91	J.S. Morris V.L. Spate M.M. Mason C.K. Baskett C.L. Reams	Missouri Scholars Academy Laboratory: NAA Laboratory
07/91	H. Neff	NSF REU: Nuclear Archaeology
07/91	F.K. Ross	NSF REU: Introduction to Neutron Scattering
07/91	W.B. Yelon	NSF REU: Introduction to Gamma-Ray Scattering

COURSE LECTURES AND LABS

DATE	LECTURER	COURSE TITLE AND SUBJECT OF LECTURE
07/91	K.R. Zinn	NSF REU: Isotope Applications in Biochemistry and Nutrition
08/91	J.J. Rhyne	NSF REU: Magnetism and Neutron Scattering
08/91	G.J. Ehrhardt	Chem 429, Nuclear Medicine and Radiology: Accelerator Production of Radioisotopes
09/91	G.J. Ehrhardt	Chem 429, Nuclear Medicine and Radiology: Particle Accelerators and Nuclear Reactions Using Charged Particles
09/91	G.J. Ehrhardt	Chem 429, Nuclear Medicine and Radiology: Radioisotope Generators
10/91	G.J. Ehrhardt	Chem 429, Nuclear Medicine and Radiology: Nuclear Medicine Applications of Radioactive Microspheres
10/91	A.D. Krawitz	Stephens College, History of Western Art I: Scientific Methods in Art History
10/91	K.R. Zinn	Biochemistry 410, Seminar: Identification and Comparisons of Selenoproteins in Rat Brain and Mouse Neuroblastoma Cells
11/91	M.D. Glascock	NE 409, Interactions of Radiation with Matter: Instruction on Neutron Activation Analysis
12/91	J.S. Morris	Chem 312, Instrumental Methods of Analysis: Instrumental Analysis
01/92	J.R. Schuh	NE 404, Reactor Lab: Indoctrination and General Health Physics
02/92	G.J. Ehrhardt	NE 406, Clinical Research and Applied Medical Physics: Radiotherapeutic Nuclear Medicine Research at MURR
02/92	M.D. Glascock	NE 404, Reactor Lab: Neutron Activation Analysis
02/92	M.D. Glascock	Anthro 349, Artifacts and Data Analysis: Nuclear Techniques Applied to the Study of Archaeological Materials
02/92	J.R. Schuh	NE 471, Radiation Protection: Indoctrination and General Health Physics

DATE	LECTURER	COURSE TITLE AND SUBJECT OF LECTURE
03/92	W.A. Meyer A.R. Schoone N.E. Tritschler	NE 404, Reactor Lab: Reactivity Measurements
03/92	J.S. Morris	Lincoln U, NAA Laboratory: Instrumental Analysis
03/92	J.S. Morris	Chem 361, Introduction to Radiochemistry: Introduction to Radiochemistry
03/92	J.S. Morris	Central Missouri State U, Chem 390: Instrumental NAA
03/92	H. Neff	Anthro 349, Artifacts and Data Analysis: Compositional Analysis of Pottery
04/92	J.S. Morris	Chem 12, General Chemistry: Nuclear Analysis
04/92	K.R. Zinn	Biochemistry 410, Seminar: Selenium Metabolism in the Brain
06/92	T. Chaudhuri	NSF REU: New Radiotherapies for Cancer Treatment
06/92	S.L. Gunn	NSF REU: Industrial Utilization of Research Reactors
06/92	M.D. Glascock	NSF REU: NAA to Source Archaeological Obsidian
06/92	M.D. Glascock	NSF REU: Isotopes and Radiation, Neutrons and Nuclear Reactions
06/92	K. Kutikkad	NSF REU: Fission and Nuclear Reactors
06/92	S.M. Langhorst	NSF REU: General Radiation Safety
06/92	W.A. Meyer Jr	NSF REU: MURR: Past and Future
06/92	J.S. Morris	NSF REU: Radioactive Decay, NAA, Gamma-Ray Spectroscopy
06/92	G.K. Moum	NSF REU: The MURR Computer Network
06/92	A.R. Schoone	NSF REU: MURR Today
06/92	K.R. Zinn	NSF REU: Radionuclides in Biochemistry

COURSES AND LABS TAUGHT

STAFF MEMBER	COURSE TITLE AND NUMBER
R.M. Brugger J.F. Kunze	NE 403, Applied Topics in MP/HP (s)
J.W. Farmer	ECE 124, Circuit Theory I (f)
R.A. Hultsch	Columbia College Physics (f)
A.D. Krawitz	MAE 344, Composite Materials (w)
J.F. Kunze	NE 315, Energy Systems & Resources (w) NE 409, Interaction of Radiation with Matter (f) NE 435, Physics Diagnostics Radiology I (f)
J.F. Kunze S.M. Langhorst	NE 471, Radiation Protection (w)
J.F. Kunze W.H. Miller P.K. Lee S.M. Langhorst	NE 403, Applied Topics in MP/HP (f)
K. Kutikkad J.F. Kunze	NE 402, Nuclear Fuel Cycle (w)
S.K. Loyalka	NE 411, Nuclear Reactor Theory I (w) NE 432, Nuclear Thermalhydraulics/Safety (f)
W.H. Miller	NE 346, Introduction to Nuclear Reactor Engineering I (f) NE 391, Nuclear Radiation Detection (f) NE 421, Nuclear Pulse Analysis (w) NE 429, Radiation Dosimetry (w)
W.H. Miller S.M. Langhorst	NE 301, Nuclear Science and Engineering for Secondary Teachers (s)

BY MURR STAFF AND RESEARCH INVESTIGATORS

STAFF MEMBER	COURSE TITLE AND NUMBER
J.J. Rhyne S.A. Werner	Physics 478, Magnetism and Magnetic Materials (w)
E.O. Schlemper F.K. Ross C.L. Barnes	Chem 401, Introduction to X-Ray Diffraction (s)
G. Schupp	Physics 176, University Physics (f) Physics 450, Research (s) Physics 450, Research (f)
E.O. Schlemper	Chemistry 12, General Chemistry (f) Chemistry 12, General Chemistry (w)
P.R. Sharp	Chemistry 12, General Chemistry (w) Chemistry 411, Dragonmetallics (f)
H. Taub	Physics 312, Introduction to Thermodynamics (w) Physics 314, Mechanics (f)
W.A. Volkert	NE 328, Introduction to Radiation Biology (f)
W.A. Volkert A.R. Ketring G.J. Ehrhardt K.V. Katti S. Jurisson	Radiology 400, Radiopharmaceutical Chemistry (w)
R.A. Winholtz	MAE 224, Engineering Materials I (f) MAE 224, Engineering Materials I (w)
S.A. Werner	Physics 176, University Physics (f) Physics 450, Research (s) Physics 450, Research (f)

DEGREES COMPLETED

STUDENT DATE	DEGREE DEPARTMENT	ADVISORS/COMMITTEE MEMBERS	THESIS
University of Missouri System			
K-Y. Cheng December 1991	MS NE Medical Physics	R.M. Brugger G.J. Ehrhardt S.M. Langhorst	A Study of New Seeds for Brachy Therapy
W. Dai May 1992	MS Nuclear Engineering	J.F. Kunze S.K. Loyalka R.A. Hultsch	Dissolved Air Effects in Narrow Vertical Channels Experimental Verification Related to MURR
W. Jia December 1991	MS Chemistry	E.O. Schlemper G.J. Ehrhardt K.R. Zinn	A Study of Rhenium Labeled HSA Microspheres as a Radiation Synovectomy Agent
L. Lang May 1992	PhD Chemistry	D.E. Troutner E.O. Schlemper R.N. Loeppky S. Jurisson J.S. Morris W.A. Volkert	Radiolabeling Proteins with Bifunctional Chelating Agents
M. McNally May 1992	MS Chemical Engineering	T.S. Storvick D.G. Retzloff P.R. Sharp	Preliminary Process Design for Pyrochemical Processing of Purex Waste
J.D. Nurrenbern December 1991	MS NE Medical Physics	J.F. Kunze W.H. Miller P.K. Lee	Investigation of Radionuclide Release from Shipping Cask Surfaces
L. Pellett December 1991	PhD Food Science	D.T. Gordon K.R. Zinn R.A. Sunde R. Dowdy T. Veum	The Interaction Between Dietary Iron and Copper and Nutrition Affecting Ceruloplasmin Metabolism, Cu-67 Retention, and Macrophage Production of O_2 and H_2O_2

BY MURR RESEARCH STUDENTS

STUDENT DATE	DEGREE DEPARTMENT	ADVISORS/COMMITTEE MEMBERS	THESIS
K.J. Riggle May 1992	PhD Nuclear Engineering	W.H. Miller J.S. Morris M.D. Glascock	Environmental Monitoring for U and Np Using ENAA
S-K.C. Wang May 1992	PhD Physics	H. Taub H. White F.K. Ross S. Satpathy M. Greenlief	Diffraction Studies of the Multilayer Structure of Simple Physisorbed Films
J. White December 1991	MS Ceramic Engineering, UMR	D.E. Day R.F. Brown G.J. Ehrhardt Microspheres	Properties and Manufacturing Techniques of Human Serum Albumin

Other Universities

STUDENT DATE	DEGREE DEPARTMENT	ADVISORS/COMMITTEE MEMBERS	THESIS
C.L. Chen August 1991	PhD Toxicology Oregon State U	P.D. Whanger K.R. Zinn	The Effect of Vitamin B_{12} on Selenium and Arsenic Metabolism
B. Saini-Eidukat December 1991	PhD Geology and Geophysics, University of Minnesota	P. Weiblen M.D. Glascock	Platinum Group Elements in Anorthositic Rocks of the Duluth Complex, Minnesota: Petrogenetic and Economic Implications

GRADUATE STUDENTS FUNDED BY MURR

NAME	POSITION	CAMPUS/DEPARTMENT
Agarwal, S.R.	Student Assistant Professional	Computer Science
Bahadori, D.	Student Assistant Technical	Electrical and Computer Engineering
Barnes, K.J.	Graduate Research Assistant	Physics and Astronomy
Bradford, D.	Graduate Research Assistant	Physics and Astronomy
Chao, W-W.	Graduate Research Assistant	Nuclear Engineering
Cheng, T-P.	Graduate Research Assistant	Chemistry
Clothier, R.	Post Doctoral Fellow	Physics and Astronomy
Cogswell, J.W.	Student Assistant Professional	Anthropology
Date, S.V.	Graduate Research Assistant	Radiochemistry
Ding, X.	Graduate Research Assistant	Physics and Astronomy
Elam, J.M.	Graduate Research Assistant	Anthropology
Greenfield, M.C.	Student Assistant Technical	Nuclear Engineering
Greenwood, S.	Graduate Research Assistant	UMR Materials Research
Griffin Jr, J.L.	Graduate Fellow	Nuclear Engineering
Gulati, P.	Student Assistant Professional	Computer Science
Hamacher, K.A.	Graduate Fellow	Physics and Astronomy
Hayashida, F.M.	Student Assistant Professional	U of Michigan Anthropology
Hirtz, G.J.	Student Assistant Professional	Nuclear Engineering
Hoard, R.J.	Student Assistant Professional	Anthropology
Hu, Z.	Graduate Fellow/Grad Res Asst	Chemistry
Isnard, O.	Graduate Fellow	U of Grenoble Physics/Materials Science
Iyer, R.R.	Graduate Research Assistant	Computer Science
Jia, W.	Graduate Research Assistant	Chemistry
Jimenez, H.N.	Graduate Research Assistant	Chemistry

NAME	POSITION	CAMPUS/DEPARTMENT
Kulasekere, R.	Graduate Research Assistant	UMR Physics
Kulasekere, K.C.	Student Assistant Professional	Nuclear Engineering
Lai, C-J.	Graduate Fellow	Nuclear Engineering
Lee, W-T.	Graduate Fellow	Physics and Astronomy
Liang, Q.	Graduate Research Assistant	Chemistry
Ma, D.	Graduate Fellow	Chemistry
Mann, J.T.	Student Assistant Professional	Undeclared
Manning, G.	Graduate Research Assistant	Physics and Astronomy
McIntyre, A.L.	Graduate Research Assistant	UMR Materials Research
Messmer, J.P.	Student Assistant Technical	Physics and Astronomy
Reams, C.L.	Student Assistant Technical	Political Science
Rong, X.	Graduate Research Assistant	Nuclear Engineering
Salem, J.D.	Graduate Fellow	Physics and Astronomy
Salih, M.S.A.R.	Graduate Research Assistant	Nuclear Engineering
Su, Z.	Graduate Research Assistant	Physics and Astronomy
Tang, Y.Y.	Graduate Research Assistant	Physics and Astronomy
Wagoner, R.A.	Research Assistant	Purdue U Physics
Wang, M.	Graduate Research Assistant	Chemistry
Wang, N.	Graduate Research Assistant	Chemistry
Winslow, N.A.	Graduate Research Assistant	Physics and Astronomy
Wu, G-W.	Graduate Research Assistant	Nuclear Engineering
Wu, Z.	Student Assistant Professional	Nuclear Engineering
Xie, H.	Graduate Research Assistant	Physics and Astronomy
Yin, H.	Student Assistant Professional	Industrial Engineering

UNDERGRADUATE STUDENTS FUNDED BY MURR

NAME	POSITION	CAMPUS/DEPARTMENT
Adams, C.G.	Student Assistant Technical	Civil Engineering & Geological Sciences
Bell, B.J.	Student Assistant Professional	Washington U Pre-Medicine
Bennett, P.A.	Student Assistant Professional	Electrical and Computer Engineering
Bentley, R.L.	Student Assistant Clerical	Education
Benton, T.	Clerk	Stephens College Business
Bequette, J.M.	Student Assistant Technical	Central Missouri State U Industrial Safety & Safety Management
Biere, N.N.	Student Assistant Clerical	Psychology
Billings, B.J.	Student Assistant Service	Central Missouri State U Animal Technology
Bormann, L.M.	Student Assistant Professional	Northeast Missouri State U Chemistry
Brewer, M.D.	Student Assistant Technical	Geological Sciences
Condra, C.W.	Student Assistant Technical	Business and Public Administration
Crook, A.G.	Student Assistant Professional	St. Louis U Math and Physics
Cruz, J.	Student Assistant Technical	General Studies
Dinger, R.	Student Assistant Technical	Journalism
Elwell, W.S.	Student Assistant Technical	Geological Sciences
Eubanks, W.A.	Student Assistant Technical	Electrical and Computer Engineering
Gazaway, T.L.	Student Assistant Clerical	Biological Sciences
German, G.E.	Student Assistant Technical	Forestry, Fisheries and Wilflife
Gilliam, D.L.	Student Assistant Professional	Lincoln U Physics/Math
Gottschalk, S.E.	Student Assistant Technical	Arts and Sciences Undeclared
Gross, J.B.	Student Assistant Professional	Loras College Chemistry
Hakimi, N.M.	Student Assistant Technical	Undeclared
Hayes, W.W.	Student Assistant Technical	Food Science and Nutrition
Hilburn, D.N.	Student Assistant Clerical	Human and Environmental Sciences
Hinderks, B.A.	Student Assistant Clerical	Chemical Engineering
Hinkebein, K.W.	Electronics Tech II	Law
Hodges, S.J.	Student Assistant Professional	Stephens College Math/Computer Science
Hogan, C.S.	Student Assistant Technical	Mechanical and Aerospace Engineering
Hosch, M.D.	Student Assistant Professional	Central College Physics
Jalbuena, R.	Student Assistant Technical	Electrical Engineering
Johnson, T.N.	Student Assistant Professional	Southeast Missouri State U Sociology/Anthropology
Jokerst, S.F.	Student Assistant Technical	Electrical Engineering
Khanna, V.	Student Assistant Professional	Computer Science

NAME	POSITION	CAMPUS/DEPARTMENT
Kim, J.	Student Assistant Professional	Northwestern U Chemistry
Kruessel, T.M.	Student Assistant Clerical	Psychology
Lane, D.G.	Student Assistant Technical	Chemistry/Biological Sciences/History
Law, H-M.R.	Student Assistant Technical	Physics and Astronomy
Love, J.	Student Assistant Technical	Chemical Engineering
Luginsland, J.W.	Student Assistant Professional	U of Michigan Nuclear Engineering
Mills, D.S.	Student Assistant Technical	Chemical Engineering
Noren, E.	Student Assistant Professional	Kansas State U Math/Science
Piechowski, M.F.	Student Assistant Technical	Geological Sciences
Radke, D.L.	Student Assistant Technical	Electrical Engineering
Riley, K.J.	Student Assistant Professional	U of Michigan Nuclear Engineering
Roberts, C.J.	Student Assistant Technical	Agricultural Economics
Santee, J.L.	Student Assistant Technical	Business
Scott III, J.L.	Student Assistant Professional	U of Notre Dame Computer Science
Sellers, S.D.	Student Assistant Technical	Electrical Engineering
Seman, D.J.	Student Assistant Professional	U of Michigan Nuclear Engineering
Stammeyer, M.M.	Student Assistant Professional	Chemical Engineering
Stephens, L.S.	Student Assistant Technical	Liberal Arts
Stich, T.A.	Student Assistant Technical	Chemistry/Pre-medicine
Stith, B.K.	Student Assistant Technical	History
Stith, P.S.	Student Assistant Technical	Chemical Engineering
Stryker, K.S.	Student Assistant Technical	Art History and Archaeology
Stuck, J.T.	Student Assistant Technical	Electrical and Computer Engineering
Thomas, B.L.	Student Assistant Technical	Mechanical Engineering
Thomas, D.C.T.	Student Assistant Technical	Chemical and Environmental Engineering
Tones, K.K.	Student Assistant Clerical	Arts and Sciences
Volkert, E.W.	Student Assistant Technical	Pre-Med
Wald, M.A.	Student Assistant Technical	Mechanical Engineering
Wallman, A.T.	Student Assistant Technical	Industrial Engineering
Walters, D.B.	Student Assistant Professional	U of Maryland Chemistry
Warren, B.D.	Student Assistant Technical	Accounting
Watring, T.S.	Student Assistant Technical	Mechanical Engineering
Williams, M.L.	Student Assistant Technical	Electrical Engineering/Civil Engineering/Math
Witte, D.A.	Student Assistant Technical	Mechanical Engineering
Woods-Bowen, K.Y.	Student Assistant Professional	Electrical Engineering

MURR TOURS

July 1991 - June 1992

University of Missouri Affiliation	**486**
Secondary Schools (High School, Jr. High, etc.)	**1256**
Elementary Schools	**291**
Other Universities	**300**
Walk-In	**309**
Professional Organizations	**401**
TOTAL	**3,043**

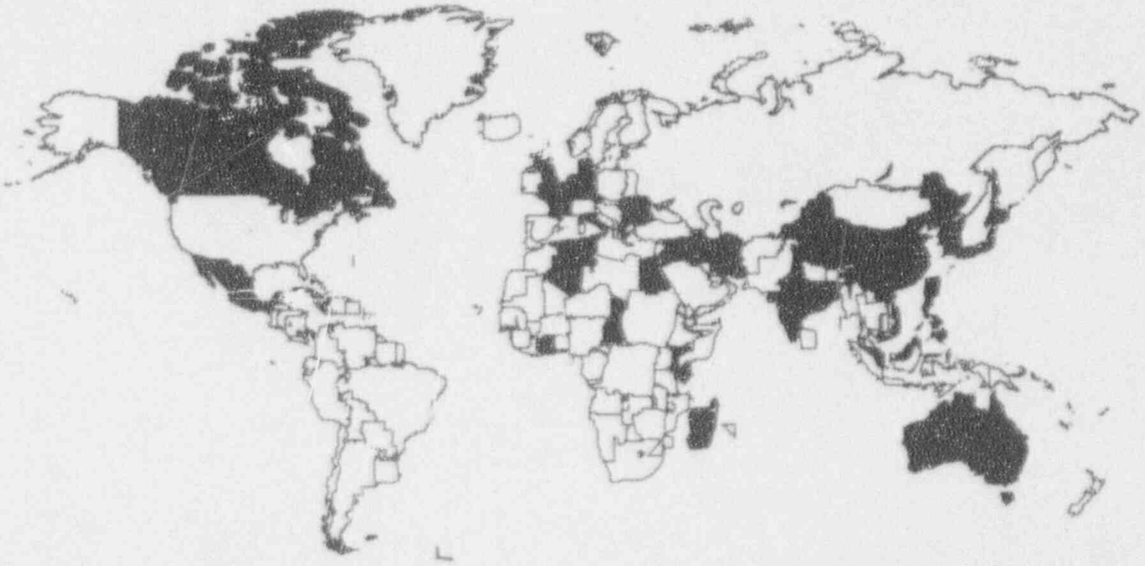
Special groups who toured the reactor facility this year include 28 members of the Missouri Valley Chapter of the Society of Nuclear Medicine and 11South Korean delegates.

Once again MURR hosted a series of tours for Parents Weekend (28 participants), MU's Summer Welcome Program (61), St. Louis Partnership schools (240 students) and the Missouri Scholars Academy (260 students).

The MURR Video was loaned to 16 Missouri high schools, 5 elementary schools and two departments of the University of Missouri, two other universities as well as six professional organizations.

INTERNATIONAL VISITORS

July 1991 - June 1992



ALGERIA
AUSTRALIA
AUSTRIA
CANADA
CHAD
DENMARK
DJIBOUTI
EGYPT
ENGLAND
FRANCE
GERMANY
GREECE
INDIA
IRAN
IRAQ
ITALY
IVORY COAST
JAMAICA
JAPAN
KENYA
KOREA
MADAGASCAR
MALAYSIA
MEXICO
PEOPLE'S REPUBLIC OF CHINA
PHILIPPINES
ROMANIA
TAIWAN
TUNISIA
UNITED KINGDOM
VIET NAM
YUGOSLAVIA

MURR OPERATIONS GROUPS

REACTOR OPERATIONS

REACTOR UPGRADE

Walter A. Meyer Jr, Reactor Manager
Anthony R. Schoone, Reactor Operations Engineer
Roland A. Hultsch, Reactor Physicist
Charles M. Anderson, Shift Supervisor
Barry C. Bezenek, Shift Supervisor
Gregory F. Gunn, Shift Supervisor
Nolan E. Tritschler, Shift Supervisor
Leslie P. Foyto, Senior Reactor Operator
John L. Fruits, Senior Reactor Operator
Robert A. Hudson II, Senior Reactor Operator
Vernon L. Jones, Senior Reactor Operator
J. Michael Kilfoil, Senior Reactor Operator
Michael L. Wallis, Senior Reactor Operator
Timothy P. Warner, Senior Reactor Operator
Paul J. Muren, Reactor Operator
Philip C. Neel, Reactor Operator
William O. Oldham, Reactor Operator
Michael L. Randolph, Reactor Operator
Robert E. Walker, Reactor Operator
Viola W. Sharp, Senior Secretary
Rebecca J. Brooks, Senior Secretary (since June 1992)

Reactor Operations staff focussed their efforts on maximizing the safe operation of the reactor to produce neutrons for MURR's broad-based programs in research, education and service. The staff continued to pursue the upgrade of reactor instrumentation and control equipment as well as reactor auxiliary equipment, to help ensure long-term reliability for producing neutrons. From July 1, 1991 through June 30, 1992, the reactor operated at full power 90% of all available hours. There were 18 unscheduled shutdowns during the year. A challenging maintenance outage to replace the original primary isolation valves was completed in two stages, the first lasting about 24 hours and the second 12.

INSTRUMENTATION AND CONTROL SYSTEM UPGRADES

Progress continues on the upgrade of reactor instrumentation and control systems. A DOE grant for University Reserach Reactor (URR) Instrumentation upgrades helped finance replacement of MURR's Nuclear Instrumentation System (NIS). Except for the wide range linear drawer, all of the components of the Gamma Metrics design NIS have been delivered. These components are being tested, with a projected installation in late 1992. A second instrumentation grant for DOE will help cover replacement of the existing control rod drive mechanisms, with bids going out in September 1992.

REACTOR AUXILIARY SYSTEM UPGRADES

Significant upgrades made this past year to reactor auxiliary systems should enhance the reactor's availability. The reactor valve operating system was improved by the addition of a new air compressor dedicated to the valve operating system, backed up by the facility air compressor. Installation of a new reverse osmosis (RO) unit with over twice the capacity of the unit it replaced has enhanced our ability to produce reactor grade water for reactor and pool water makeup, as well as provide facility DI water.

Operations staff are planning additional changes to the DI system in FY93. Splitting the reactor makeup water and the facility DI water systems will provide an important benefit: the ability to pump the pool to low levels more quickly for deep pool maintenance.

STAFF RECOGNITION

The twenty members of Reactor Operations on staff at the beginning of FY92 represent 194 years of service, an average of nearly 10 years per person. MURR longevity figures are shown at right.

The cumulative experience of our Operations staff, coupled with the impressive longevity of the majority of our members, provides a very strong team. MURR's Reactor Operations staff is the heart of the Center, intent on producing neutrons for the wide range of multidisciplinary research, education and service programs that make the MURR Center our nation's premier university research reactor.

MURR Reactor Operations Longevity

Chuck Anderson	15	years
Barry Bezenek	23	years
Les Foyto	4	years
John Fruits	4	years
Greg Gunn	11	years
Rob Hudson	5	years
Rolly Hultsch	8	years
Vern Jones	14	years
Mike Kilfoil	12	years
Walt Meyer	17	years
Paul Muren	1	year
Phil Neel	5	years
Willie Oldham	1	year
Mike Randolph	6	years
Tony Schoone	10	years
Vi Sharp	14	years
[After nearly 21 years of dedicated service to the University of Missouri, Vi announced her early retirement, effective August 31, 1992.]		
Nolan Tritschler	22	years
Rob Walker	2	years
Mike Wallis	8	years
Tim Warner	12	years

REACTOR OPERATIONS* SUMMARY

July 1991 - June 1992

MURR operated 24 hours per day, 7 days per week (averaging approximately 152 hours per week). MURR operated at full power 90% of all available hours in FY92.

	FY92	TOTAL
Total Hours Operated	7,938	165,304
Total Hours at Full Power	7,874	162,442
Total Integrated Power (MWD)	3,283	61,822

- There were 56 scheduled shutdowns for maintenance, refueling and sample changes in the flux trap.

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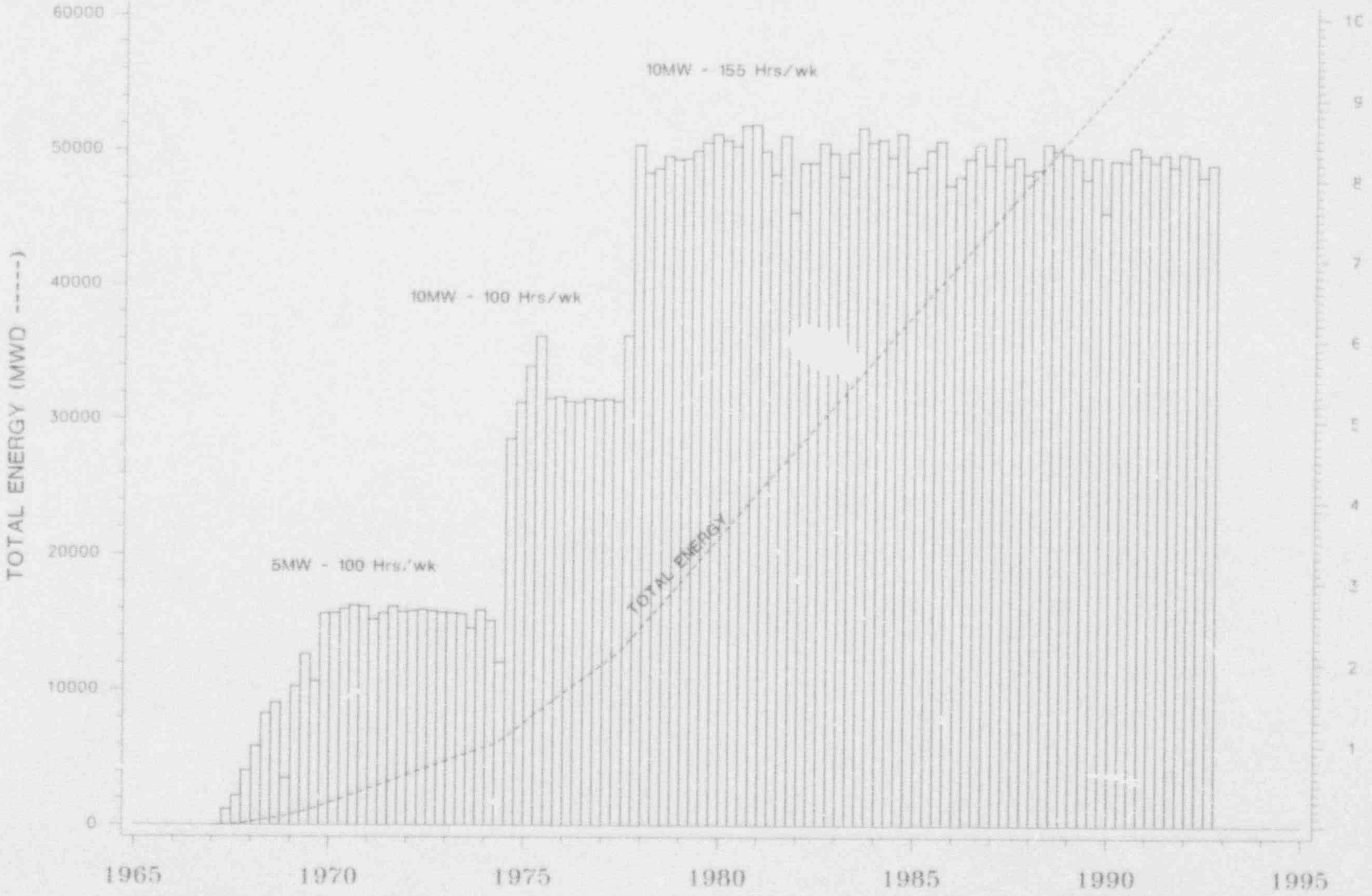
*For details see "Reactor Operations Annual Report 1991-1992"

OPERATING EXPERIENCE
UNIVERSITY of MISSOURI RESEARCH REACTOR



UPGRADE GROUP

PERSONNEL

J. Charles McKibben, Group Leader (acting)
Kiratadas Kutikkad, Research Scientist (Adjunct Assistant Professor, Nuclear Engineering)
Walter A. Meyer Jr, Reactor Manager
Chester B. Edwards Jr, Facilities Manager
Susan M. Langhorst, Health Physics Manager (Assistant Professor, Nuclear Engineering)
Roland A. Hultsch, Reactor Physicist
Jay F. Kunze, Senior Research Investigator (MURR) and Chair and Professor of MU Nuclear Engineering
William H. Miller, Senior Research Investigator, Group Leader (MURR) and Professor of MU Nuclear Engineering, Director of Energy Systems and Resources Program
Olawale B. Oladiran, Staff Engineer

The MURR Center presents to MU the opportunity and the obligation to become a leading university in nuclear related fields. The development and advancement of industries and laboratories in these areas of science and engineering as well as national projects such as the Advanced Neutron Source depend upon the availability of highly qualified and well trained personnel. Over the past 24 years, MURR has progressed through a series of upgrades that has increased greatly its versatility for research, and efforts to expand and adapt to meet the growing needs of research and technology continue. During 1985-1989, a major three-part plan was drafted encompassing:

- an addition to the research laboratory building
- an increase in the reactor power level
- a cold neutron source for enhanced beam research opportunities

The proposed expansion to the research building will more than double the laboratory space. Other aspects of the upgrade include the addition of new research programs, instrumentation and equipment, improved reactor systems and a new fuel design. Work to support this effort is spread throughout various MURR groups and involves many faculty associates.

BUILDING ADDITION With the evolution of reactor programs and MURR's administrative transfer to MU from the University of Missouri System in 1989, the need for an expanded research laboratory building clearly emerged as the highest priority. The reactor (as a neutron source) is not the limiting entity; it is fully capable of providing greatly expanded radioisotope production, sample irradiations and beam research opportunities whenever the necessary laboratory facilities become available.

Last year, an extensive study of space needs for reactor-based research involved more than 100 faculty and staff in an in-depth planning process. This group comprised the respondees to an extensive space need and program questionnaire sent to over 20 academic departments, and the laboratory needs identified in the survey process were developed into laboratory profile sheets. A planning group reviewed and consolidated the original space requests into a reduced set of a total additional gross square footage of 109,439 and estimated project cost of approximately $22.4 M.

The conceptual design of the first phase of the building addition was completed by Sverdrup Corporation of St. Louis, MO, with the final report to be presented in FY93. This addition will add 81,500 GSF to the building and has a project cost of $15.25 million. The addition, which includes the highest priority items as recommended by the Reactor Research Advisory Subcommittee, has two parts: a 54,500 GSF addition on the north side of the building with laboratories, a guide hall, support shops, offices, etc.; and a 27,000 GSF south side addition with laboratories, hot cell, glove boxes, shipping and receiving, support shops, offices, etc.

POWER UPGRADE The Upgrade Group is actively pursuing the plan to nearly triple the power level of MURR from the "administrative" 10 MW power limit to a technical limit. To facilitate the power upgrade, a new fuel loading was designed for the plate-type fuel elements with denser maximum fissile material loading and a more uniform power density. The new fuel elements will reduce significantly the fuel cycle costs by achieving a higher power history per element compared to the current fuel elements.

While NRC approval for operating the new fuel at the current 10 MW power level was obtained in August 1990, fabrication and testing of the new fuel are dependent upon startup funding from DOE. This funding is anticipated within the 1993 federal fiscal year.

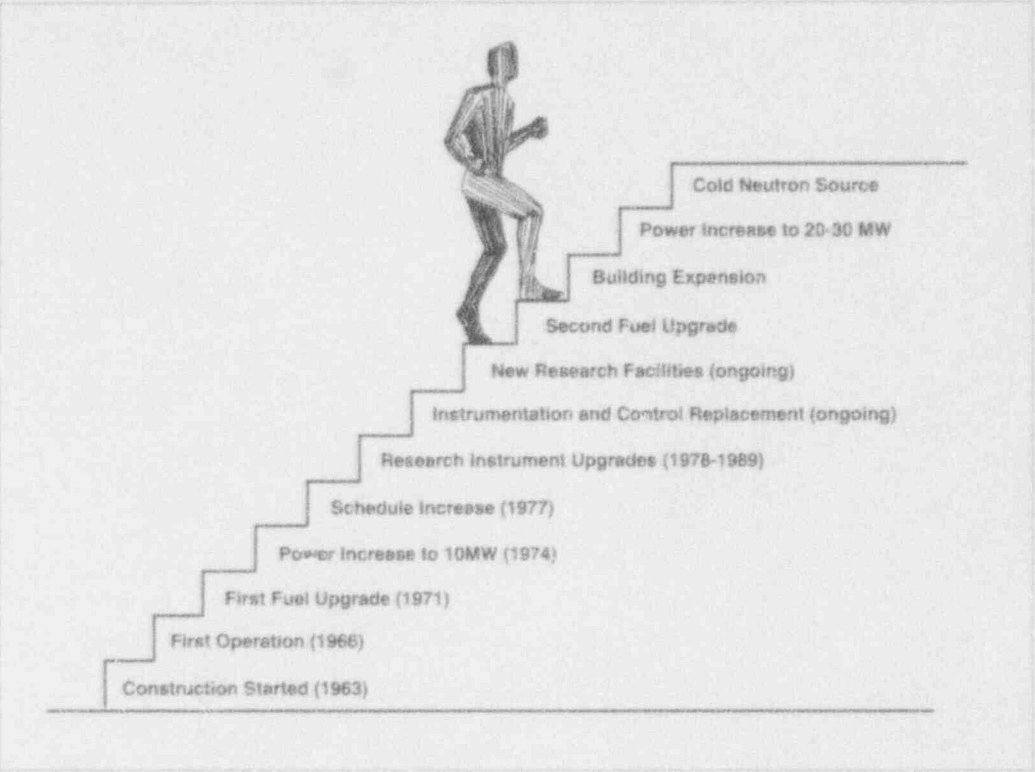
Many facets of the power upgrade effort are done in close cooperation with MU Nuclear Engineering. Efforts are made to interest graduate students in many of these projects, thus affording students valuable reactor experience. Currently six students (five at the PhD level) are involved in various upgrade related projects. Some of the areas where we have made significant progress are listed below:

- A loop for simulating the thermal-hydraulic conditions inside the MURR core was developed in association with MU's Nuclear Engineering Program to benchmark results from the NRC-approved transient analysis computer program RELAP for low power, low pressure and low temperature reactors. With the help of this loop, we have managed to establish that the results obtained using RELAP are quite conservative (see **REACTOR NUCLEAR ENGINEERING PROGRAM** section).

- MURR Loss of Flow (LOF) and Loss of Coolant Accidents (LOCA) have been analyzed for 20, 25 and 30 MW power levels using RELAP. These analyses indicate that MURR can be taken safely to at least the 25 MW power level.

- Early on in the efforts to upgrade the power of the MURR, it was realized that one of the areas where considerable effort has to be put is in the environmental impact of the power upgrade, including the effects of the trace amounts of radioactivity that are released from the MURR due to routine operation. [These minute amounts of radioactivity are diluted and released at a higher elevation to make its impact on the environment negligible.] To have a better handle on the environmental transport of radioactivity, the outdated wind measurement system at MURR was replaced by a state-of-the-art system capable of providing information that can be fed directly into a computer. Studies were undertaken to compare various computer programs capable of estimating the effect of meteorological conditions and terrain information on routine release. After considerable review, the NRC-approved computer program XOQDOQ was selected and installed at MURR. As a part of the power upgrade request, NRC mandates MURR to do an analysis of the worst-case accidental radioactive release situation. A similar computer program review is underway to choose a program that handles such calculations.

- Another area of on-going work is the neutronics (reactor physics) analysis of the new core at higher power levels. Plans are to perform experiments to obtain basic



reactor physics data such as neutron lifetime and the reactor response to reactivity transients. Information collected from these investigations will be used to benchmark some of the computer programs that are being developed by graduate students specifically for analyzing MURR transients.

- While trying to put together a new Safety Analysis Report (SAR) for upgrading the power of MURR, it became apparent that there is no NRC-mandated standard format for SARs for research reactors. We are in the process of drafting such an SAR format applicable to all research reactors, with an estimated submittal time for NRC review and approval in 1993.
- Other planned improvements include implementation of expert systems and graphical interfaces to help the operators and to assist them in better identifying the onset of abnormal operating conditions (see **REACTOR NUCLEAR ENGINEERING PROGRAM** section).

SUPPORT ACTIVITIES As part of the efforts to reduce the volume of LLW generated by the MURR Center, the Upgrade group assisted the Reactor Operations group in the redesign of sample irradiation cans. Monte Carlo analysis techniques were used to determine the effect of replacing cadmium with boron as the thermal neutron shield. The revised can eliminated fully the production of radioactive Cd (a mixed waste).

We also assisted the Service Applications group in improving and documenting their methodology for preparing the safety analysis reports of new samples before being irradiated in the reactor. The Upgrade group maintains several state-of-the-art reactor analysis computer programs for predicting many important parameters such as the reactivity effects of new samples, heat generation rates and heat transfer rates in samples that have never been irradiated before.

MURR SUPPORT GROUPS

FACILITIES OPERATIONS

HEALTH PHYSICS

COMPUTER DEVELOPMENT

INSTRUMENT DEVELOPMENT

FACILITIES OPERATIONS

Chester B. Edwards Jr, Facilities Manager
Olawale B. Oladiran, Staff Engineer
C. Bradley McCracken, Staff Engineer
David G. Nickolaus, Senior Drafting Technician
Electronics Shop:
  Thomas H. Seeger, Chief Electronics Technician
  Joe D. Baskett, Research Electronics Technician
  Mark Richardson, Electronics Technician (since January 1992)
Machine Shop:
  Mac L. Evans, Chief Research Engineering Technician
  Kenneth W. Beamer, Senior Research Engineering Technician
  Ronald R. Kitch, Senior Research Engineering Technician
  Charles L. Kribbs, Senior Research Engineering Technician
Leah Moreau, Senior Secretary (since November 1991)

Facilities Operations staff provide and coordinate technical service on design, construction, installation, modification and calibration of reactor, research and service equipment for MURR staff, faculty users and students. The Electronics Shop and Machine Shop deliver support to meet routine reactor maintenance and technical needs as well as research program requirements. In addition, the shops support the reactor services group with construction and welding of sample cans, special tools and fixtures, and with hot cell manipulators maintenance. Drafting Services provides a variety of services including design drafting of research instrumentation and laboratory facilities; graphics for publication in various technical journals; posters to display scientific research; and various other projects.

Special projects during the year include:

- engineering support and collaboration with Costar on the equipment upgrade of the new Mark III irradiator case, web drive and gas system. The Mark III irradiator equipment development was completed and tested, with the exception of the gas system and control systems. The project was placed on indefinite hold with all equipment packaged and stored by Costar.
- Fire sprinklers were installed in the 111 area outside the Alpha Laboratory in compliance with the NRC Administrative Judge's Order.
- Cell Culture Laboratory (room 215) base construction was completed, with specialized equipment and laboratory HVAC controls to be installed, tested and placed into service in FY93.
- The conceptual design of the building addition was completed by Sverdrup Corporation of St. Louis, MO, with the final report and rendering to be presented in FY93. The distribution of new and existing space assignments locates the programs handling multiple curie quantities of radioactive materials in hot cells and glove boxes in the south addition; programs handling millicurie to curie quantities of radioactive materials in the existing laboratory building; and programs handling microcurie to millicurie quantities of radioactive materials in the north side. The building addition on both the north and south sides matches the existing building basement and grade elevations plus each has a second level. The addition

results in 81,500 GSF, with approximately 54,500 GSF on the north side housing laboratories, a guide hall, support shops, offices and associated mechanical spaces; and 27,000 GSF on the south side housing laboratories, hot cells, glove boxes, shipping and receiving, support shops and associated mechanical spaces. The reduced conceptual design is a $15.25 million FY93 building as recommended by the Research Reactor Advisory Subcommittee (RRAS).

- A third temporary office building (TOB-3), 1960 sq ft, was ordered for the south side of the laboratory building west of the loading dock.
- SANS vacuum chamber and carriage design and bid documents were completed. A Request for Proposals (RFP) was submitted to a number of fabricators and is due back in early FY93.
- Design of a new topaz counting machine with 48 canals operating linerally is well underway. The objective of the new system is independent discrete counting of each canal.
- Two new sample rotators were designed, constructed and installed for in-pool topaz irradiation holders and 4" and 5" silicon.
- Fixture design was started for the residual sStress equipment to handle NASA's 1/4 scale Challenger booster rocket cylinder for evaluation of the weld areas.
- Auxiliary annunciator panel was installed in the reactor Control Room to accommodate Costar equipment and other program needs.
- Secondary water cooling system was upgraded and replaced with pH conductivity monitoring equipment for piping corrosion control.
- New dedicated reactor valve-op air compressor was installed and valve-op system removed from facility compressed air system.
- Two hot cell manipulators received from Rockwell International as excess property were modified and installed.
- New facility exhaust stack radiation air monitor from NMC Corporation was received and installed.
- Partial shipment of new reactor nuclear instrument channels equipment from Gamma Metrics was received, with testing and installation expected in FY93.
- Atomic spectroscopy instrument was installed in laboratory 218.
- TRIAX instrument was received from Oak Ridge National Laboratory. Drum, air cylinder, and gear mount restoration were completed, with installation to be completed in FY93.
- Cooling coil was added to pump room at cooling tower basement to protect operating equipment in the area.
- New weather instrument was installed and verified operable in lab 213.

REACTOR HEALTH PHYSICS

Susan M. Langhorst, Manager (Assistant Professor, Nuclear Engineering)
John P. Ernst, Assistant Manager and Health Physicist
James R. Schuh, Health Physicist
Ray W. Stevens, Senior Health Physics Technician
Joseph W. DeMers, Senior Health Physics Technician (left September 1991)
Rex G. Ayers, Health Physics Technician (since August 1991)
W. Derek Pickett, Health Physics Technician (since January 1992)
Andrea J. Shipp, Health Physics Technician (upgraded from HP Trainee April 1992)
Leslie M. Powell, Senior Secretary (upgraded from Secretary June 1992)

The primary responsibility of the Health Physics (HP) group is the radiation safety program to support the multi-faceted research, service and educational programs conducted at MURR. Health physics coverage of these programs presents a unique challenge--to ensure radiation safety with regard to personnel training, personnel dosimetry, reactor operations, processing of radioisotopes, radiochemical research development for ultimate use in radiopharmaceuticals, transportation, system design, environmental assessment, emergency response, radioactive material control and accountability, contamination assessment and control, radwaste management, and facility/materials licensing.

BROAD SCOPE LICENSE RENEWAL

On February 27, 1992, the University of Missouri System (UM) applied for renewal of its Broad Scope License (License No. 24-00513-32). The existing Broad Scope License had established the *administrative* structure for Type A materials licenses at the University as follows: a Central Radiation Safety Committee (CRSC) and one Radiation Safety Officer (RSO). The CRSC and RSO are responsible for reviewing and approving the use of radioactive materials at the four campuses under all the University's materials licenses except for the Exempt Release License. The materials licenses are:

- Source and Special Nuclear Materials License, No. SNM-247
- Cobalt Irradiator at MU campus, License No. 24-00513-33
- MURR Cobalt-60 Irradiation Facility, License No. 24-00513-34
- MU Hospital Co-60 Teletherapy Unit, License No. 24-00513-35
- MURR Exempt Release License No. 24-00513-36E

Each campus and the MURR Center have been responsible for the *operational* function of these licenses at their respective sites. For MURR, this operational responsibility has been with the Isotope Use Subcommittee (to the Reactor Advisory Committee) and with the MURR HP group.

In recent years, the NRC has been focussing on improving and strengthening administrative controls defined for materials licenses. To improve the effectiveness of its radiation safety programs, the University requested in its Broad Scope License renewal that both *licensing* and *operational* responsibilities be established within the respective committees and health physics units by issuance of separate radioactive materials licenses for each campus and for the MURR Center. Defining administrative controls consistent and compatible with those already in place under the MURR reactor license provides the most effective and logical control of radioactive materials at MURR. The NRC is currently reviewing the University's renewal application, and we are in the process of answering some of their questions concerning our reorganization and how we plan to conduct our radiation safety programs.

WASTE DISPOSAL

ADCO Services, Inc. has continued to act as our institutional waste broker. Through ADCO, we disposed of 472.5 cubic feet of LSA material generated at MURR.

ALARA

The ALARA program implemented to improve the review and documentation of these efforts has continued to develop. As stated in the MURR FY91 Annual Report, the challenge of instituting a more meaningful ALARA program at MURR has been the organizing of appropriate ALARA review levels that initiate review and documentation for a wide range of reasonable personnel doses. Established investigational levels are shown in Table 1.

The average monthly whole body deep doses in each group for FY92 are indicated in Table 2. Reorganization of research groups did occur in April 1992, but the ALARA review program structure remained constant for FY92 pending renewal of the contract for personnel dosimetry services.

TRAINING

Increased efforts to provide supplementary training for individuals working at the MURR Center have resulted in the development and/or presentation of the following special lectures and videos:

- Radiation Fundamentals
- HP Instrument Training
- General Safety (video)
- Chemical Safety (videos)
- History of Radioactivity
- Personnel Monitoring
- Radiobiology
- Special Subjects for Custodial Personnel
- Biological Effects
- Radioactive Material Shipping
- Radioactive Decay

Efforts continued on making the new stack monitoring system operational (Nuclear Measurements Corporation Model RAK-22ABIB-P.6, Continuous Air Monitor System). Equipment and programming problems have delayed final testing and our acceptance of the system as MURR's primary stack monitoring system. In December 1991, a new access to the MURR containment building roof was made in the North tower. This new access port allows easier entrance to that area for inspection of the roof and systems and instrumentation located in the area. Previous access was either through the facility stack exhaust system, which was normally limited to times of reactor shutdown, or by portable ladder from the laboratory roof.

TABLE 1 INVESTIGATIONAL LEVELS
(mrem per month)

	LEVEL I[*]	LEVEL II[c]
Whole body; head and trunk; active blood forming organs; lens of eyes; or gonads	> 30 above and 33% of average monthly dose[†]	> 50 above and 50% of average monthly dose,[†] or >180
Hands and forearms (shallow dose)	> 1000	> 3000
Skin of whole body[*] (shallow dose)	> 300	> 600

[*] personnel dose noted
[c] report required assessing personnel dose
[†] average monthly dose based upon previous four quarters
[*] applicable for significant quantities of beta emitters

TABLE 2 FY92 AVERAGE MONTHLY WHOLE BODY DEEP DOSE

GROUP	DOSE
Computer Development Director's Office Nuclear Engineering Radiation Effects	minimal
Facilities Management Instrument Development Nuclear Analysis Program Neutron and Gamma-Ray Scattering Radioisotope Applications	10 mrem
Reactor Services	30 mrem
Health Physics	50 mrem
Operations	80 mrem

COMPUTER DEVELOPMENT

Gene K. Moum, Senior Research Scientist (Adjunct Assistant Professor of Computer Science)
Stephen P. Fox, Computer Programmer/Analyst II
W. Michael Shell, Computer Programmer/Analyst II (left September 1991)
Chao Wang, Computer Programmer/Analyst II (since October 1991)

The day-to-day operation of the MURR Center has become increasingly aided by computers. The variety of applications range from business oriented spreadsheets to computer aided design, from research data reduction and analysis to desktop publishing. There are currently over 140 user accessible computers in daily use at the Center. These are both workstations and multi-user computers. This total does not include the dozens of dedicated computers that control spectrometer motors, monitor background radiation, control computer network traffic and do data acquisition.

The Computer Development group is responsible for the overall support and operation of computers and computing facilities at MURR. We provide, support and maintain the Local Area Network (LAN) within the Center that provides the connectivity necessary for researchers to access their instruments and data, workgroups to access common databases, and staff to communicate and exchange files electronically.

The LAN spans four buildings with five wiring closets and 28 active network control devices. Connection to the LAN is provided from every room in the building via 15 miles of MURR-installed cabling.

The Computer Development group also supports the central computing facility for the Center. This consists of a pair of MicroVAX III+ computers that provide support for data reduction and analysis, word processing, spreadsheets and statistical analysis.

Computer Development is involved in several long range projects:

- a new irradiation monitoring system for the silicon program
- a new generation control system for the new gemstone counting machine
- a new generation of control computer system for the neutron and gamma-ray spectrometers for the several programs on the beamport floor.

The Computer Development group generally supports two or three students in technical positions. We are currently supporting our second Master's candidate in cooperation with MU's computer science department.

INSTRUMENT DEVELOPMENT GROUP

Ronald R. Berliner, Senior Research Scientist (Adjunct Associate Professor of Physics and Adjunct Associate Professor of Electrical and Computer Engineering)
Don Bryan, Computer Engineer
Douglas Charlton, Computer Engineer

The mission of the Instrument Development Group is the design and fabrication of electronic instruments for research. The group has concentrated on the development of position sensitive neutron detectors, and their support software for neutron diffraction applications, the fabrication of computer interfaces for neutron diffraction instruments and the development of special purpose micro-controllers for process control applications.

These three areas of activity have dominated the activities of the group for the past year. The renewal of neutron diffraction resources at MURR and the construction of several new instruments has required the ID group to devote substantial resources to the fabrication of spectrometer computer interfaces. Four new interfaces have been constructed and a total of 25 stepper motor controllers and 6 Timer-Counters have been fabricated. The group continues to be responsible for the maintenance of the high resolution powder diffractometer position sensitive detector system. Several new capabilities and hardware features have been added to the system in the course of the last year. The instrument has completed 650 runs during the period July 1, 1991 - June 30, 1992. Fortunately, it has required little maintenance during the year.

R. Berliner was on leave at the Australian Nuclear Science and Technology Organization during the period March 1992 - July 1992 but several projects initiated prior to that time were approved or funded during that period. A grant for the construction of a position sensitive detector for the residual stress spectrometer was funded and work on the definition of the analog and digital subsystems for that instrument are underway. The group has also undertaken the development of a new process control micro-controller using the MC68HC16 micro-processor. Finally, the group is participating in the development of new techniques for nuclear signal processing as part of its effort for the DOE funded Smart-ADC grant.

A PEAK-DETECTOR ADC

D. Charlton, T. Stuck and R. Berliner

Research on the development of a conceptually new analog-to-digital converter (ADC) for nuclear pulse-height analysis is in progress. The design utilizes new techniques for determining the time and magnitude of the occurrence of a nuclear detector pulse such as those from proportional counters. The occurrence of a peak is detected by differentiating the input pulse and using the zero-crossing to signal the pulse peak. The peak amplitude is captured and stored by an operational amplifier (op amp) peak-detector circuit, which uses a high-speed op amp for the input stage and a low bias current op amp for the output stage. The stored peak value is converted to digital form by a fast-conversion-time, 12-bit ADC integrated circuit. This is in contrast to the "standard" Wilkinson pulse-height-analyzer design that has been in use for many years. The major virtue of the new design is its simplicity as the complex analog-to-digital portion of the circuit is encapsulated in a commercial single-chip off-the-shelf device. Figure 1 is a block diagram of the new pulse height analyzer.

The circuit has been tested and performs the desired function. Tests are under way to measure its differential and integral linearity for a comparison with data obtained for existing Wilkinson ADC designs. Additional new features are being incorporated into the new ADC. Tests of the in-circuit performance of a digitally-adjustable potentiometer (E2POT) for use in both the Wilkinson-ADC designs and the new ADC. This digitally adjustable potentiometer would replace many of the manually-adjustable potentiometers used in the electronics that service the neutron position sensitive detector systems.

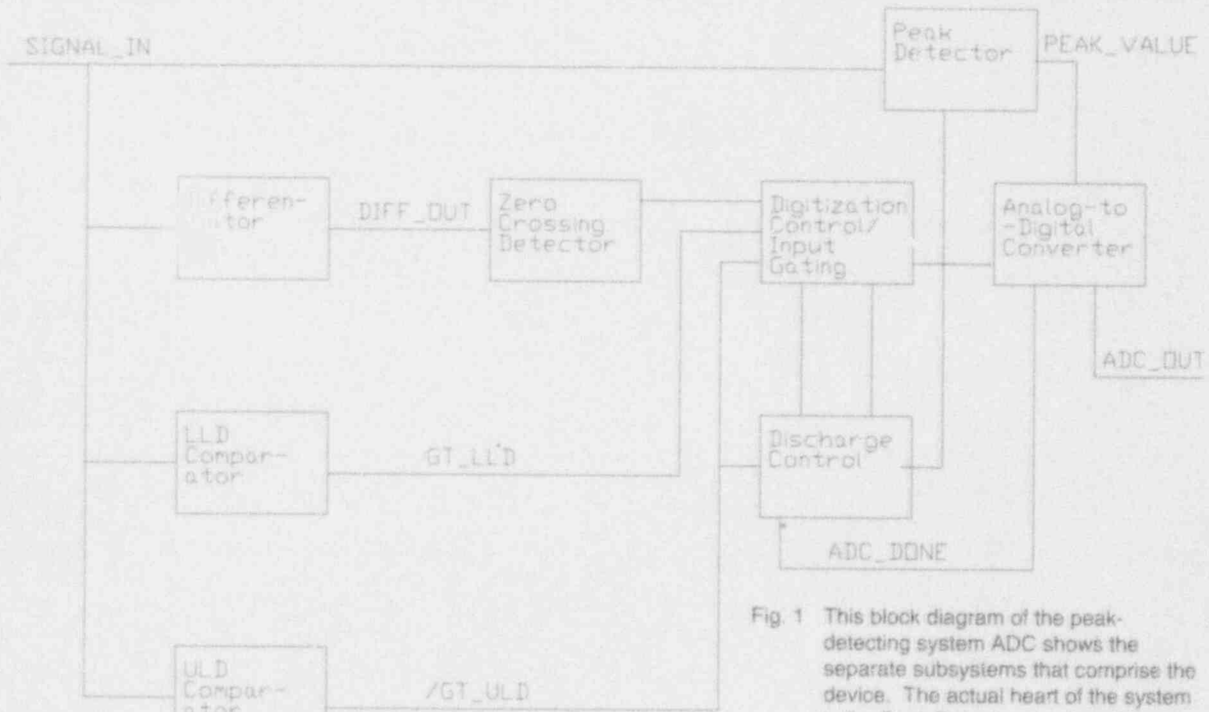


Fig. 1 This block diagram of the peak-detecting system ADC shows the separate subsystems that comprise the device. The actual heart of the system is the Peak Detector circuit and the analog-to-digital (ADC) subsystems.

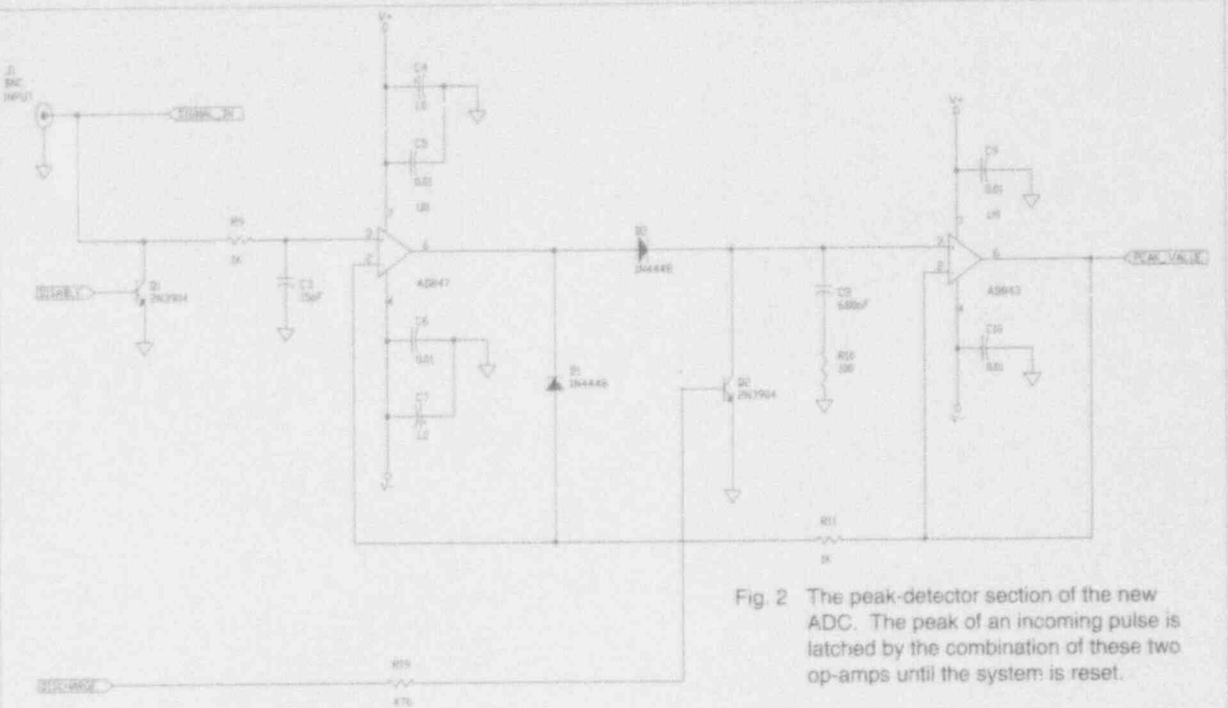


Fig. 2 The peak-detector section of the new ADC. The peak of an incoming pulse is latched by the combination of these two op-amps until the system is reset.

A NEW PROCESS CONTROL MICRO-CONTROLLER

D. Bryan, D. Charlton, W. Eubanks, T. Stuck, R. Berliner

Design and development of a process control system using the Motorola MC68HC16 micro-controller is underway. The micro-processor that forms the heart of the system is a high speed 16-bit CMOS device with on chip peripherals such as a multiplexed 10-bit analog-to-digital converter (ADC), a general purpose timer (GPT), 1024 words of high speed static random access memory (SRAM), and a queued serial communications module (QSM).

The process controller design utilizes most aspects of the HC16 by incorporating an RS-232 serial interface into the QSM, which allows the process controller to control and communicate with other RS-232 devices, using the GPT to accumulate nuclear detector pulses, allowing the controller to service detector systems, and having 22 general purpose input-output (I/O) lines arranged as two 8-bit parallel ports and one 6-bit parallel port for sensor input or actuator output. The I/O lines can also be software configured for specific tasks according to their application requirements. Drivers have been added to the micro-processor's address and data buses so that off-board peripherials can be memory mapped, simplifying software design for a variety of external devices.

The new controller communicates with its host computer via the IEEE-488 communications interface, standard with all other Instrument Development spectrometer computer interfaces. It has a capacity for up to 1M-bytes of Read-Only-Memory (ROM) program space and 128 k-bytes of Random Access Memory (RAM) data space in addition to the 1k-byte high speed on-chip RAM.

Software development for this process controller will be in C and Assembly language, allowing compact yet portable, modular code. Extensive applications for this controller can be developed due to its large program and data space and the flexible design of the hardware.

A representation of the completed system is shown in Figures 1 and 2. The device as configured is set up for one detector input and for input and output to both IEEE-488 and RS-232 communications.

Fig.1 A block diagram of the MC68HC16 process controller. The large box in the center is the 1" x 1" microprocessor chip itself.

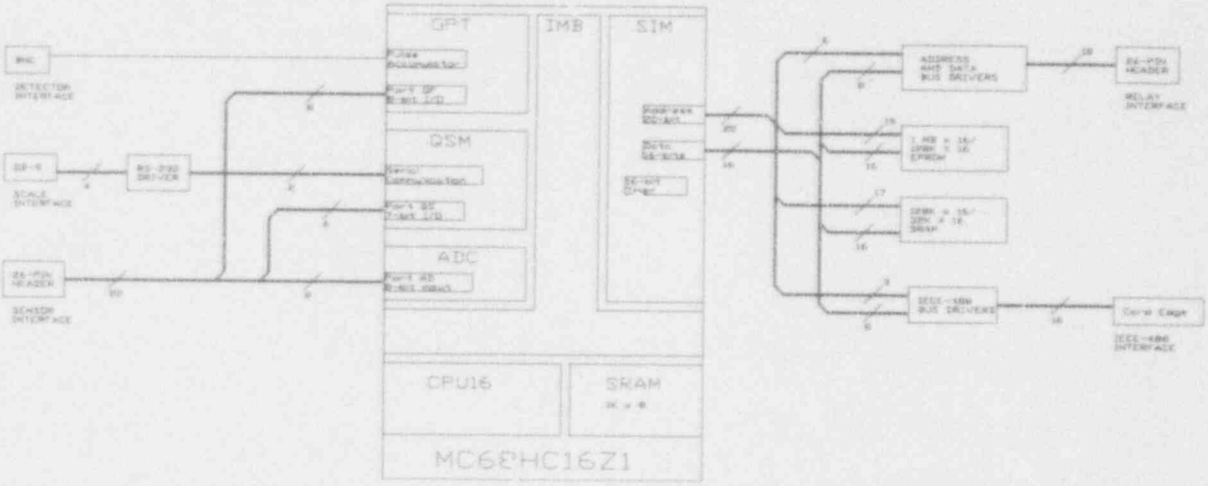
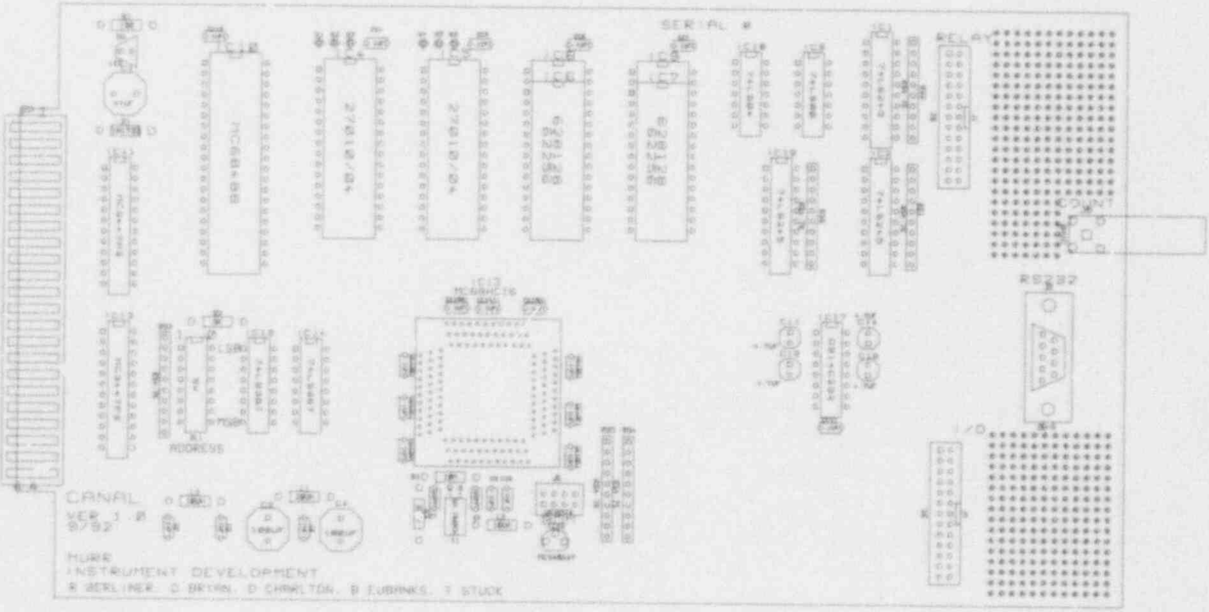


Fig. 2 The process controller circuit board layout



SERVICE APPLICATIONS

July 1991 - June 1992

- Supplied 1,574 shipments of 50 different isotopes

William F. Reilly, Assistant Director, Technical Applications
Stephen L. Gunn, Reactor Services Engineer
Martha A. Carter-Tritschler, Supervisor, Reactor Services
Wei Jia, Reactor Services Project Specialist (since June 1992)
Leonard H. Manson III, Reactor Services Project Specialist
Paul D. Miller, Reactor Services Project Specialist
James P. Lanigan, Senior Research Laboratory Technician (promoted/transferred from Radiopharmaceutical group August 1991)
Timothy G. Arnold, Research Laboratory Technician (since November 1991)
Coralie T. Benson, Research Laboratory Technician
J. Eddie Brooks, Research Laboratory Technician (since May 1992)
David A. Crawford, Research Laboratory Technician (hired as Laboratory Assistant August 1991; promoted March 1992)
Melissa M. Gibbs, Research Laboratory Technician (since August 1991)
I. Joseph Henke, Research Laboratory Technician
William D. Howell, Research Laboratory Technician
G. Lawrence Jesse, Research Laboratory Technician
Stacy S. Lanigan, Research Laboratory Technician (left July 1991)
Zelly Les, Research Laboratory Technician (left July 1991)
William A, Martin, Research Laboratory Technician (since August 1991)
Vannak Phouk, Research Laboratory Technician
Kathleen D. Pierson, Research Laboratory Technician (promoted from Laboratory Assistant December 1991)
Matthew R. Sanford, Research Laboratory Technician (promoted from Laboratory Assistant September 1991)
Janice A. Caldera, Laboratory Assistant (left August 1991)
Todd E. Gould, Laboratory Assistant (left August 1991)
Khim Long, Laboratory Assistant
Timothy I. Morris, Laboratory Assistant (since August 1991)
Thi T. Tran, Laboratory Assistant
Jeffrey E. Valenti, Laboratory Assistant (left August 1991)
Kathryn A. Harbour, Secretary
Susan A. Cable, Senior Clerk Typist

The Service Applications group provides the interface between experimenters and the reactor. The group evaluates experiments to ensure that all safety requirements are met, prepares samples for irradiation, processes irradiated materials, and packages and ships materials after irradiation. This group also handles the processing of production isotopes, silicon and gemstones that produce the bulk of revenue for research work. During FY92, Service Applications performed work for 140 industrial customers, 42 universities and four government agencies.

TABLE 1 IRRADIATION POSITIONS

POSITION	CAN DIAMETER (inches)	MAXIMUM SAMPLE DIAMETER (inches)	MAXIMUM SAMPLE LENGTH (inches)	MAXIMUM THERMAL FLUX (n/cm^2-sec)
Flux Trap	1.125 .400	1.00 0.275	3.75 1.00	5×10^{14} 5×10^{14}
Row 1	1.125	1.00	3.75	8×10^{13}
Row 2	2.350	2.00	10.00	5×10^{13}
Row 3	3.350	3.00	10.00	3×10^{13}
Row 4	4.350	4.00	10.00	6×10^{12}
Row 5	5.350	5.00	10.00	1×10^{12}

SERVICE IRRADIATIONS The Service Applications group performs work for both University and non-University users. The two major areas are the reflector, which is accessible any time for inserting or removing samples, and the flux trap, which is accessible once a week during scheduled maintenance shutdowns. All irradiations for experimenters outside the University are charged by a standard charge schedule. Unfunded researchers at other Universities may apply for DOE funded Reactor Sharing support.

Two one-inch diameter pneumatic tubes permit samples to be positioned rapidly in high flux reflector positions for short irradiations and then returned for neutron activation analysis. Specialized work in lower flux positions includes neutron radiography and membrane filter production. MURR also has a cobalt facility for intense gamma irradiations.

ISOTOPES MURR has been active in finding new uses for radioisotopes. Certain radioisotopes, such as P-32 and S-35, are produced in large volume and sold by the Curie on a routine basis. Other isotopes are produced on demand as service irradiations. Figure 1 shows the number of type A and type B shipments of radioisotopes during FY92; Table 2 is a list of the isotopes shipped.

NTD SILICON The facility has been active in treating silicon by transmutation of Si to P since 1975. The current treatment capacity exceeds 40 tonnes per year at 1 ppb P. Plans are being made to double that capacity in 1993 due to strong worldwide demand.

GEMSTONES The service group operates a gemstone enhancement program to create blue topaz by treating white topaz with fast neutrons. The resultant blue topaz must be analyzed stone by stone to ensure compliance

TABLE 2 ISOTOPES SHIPPED

Ag-110m
Au-198
Br-82
Ca-45
Ca-47
Cd-109
Ce-141
Co-58
Co-60
Cr-51
Cs-134
Cu-64
Dy-159
Dy-165
Er-169
Eu-152
Fe-55
Fe-59
Gd-153
Gd-159
Ge-71
Hg-203
Ho-166
In-114m
Ir-192
Lu-177
Mn-56
Na-24
Os-191
P-32
Pd-103
Pr-142
Pt-191
Pt-193m
Pt-195m
Rb-86
Re-186
Rh-105
Ru-103
S-35
Sb-124
Sc-46
Se-75
Si-31
Sm-145
Sm-153
Sn-113
Sn-119m
Sr-85
Ta-182
Tb-161
Te-123m
Te-125m
Te-129
Tm-170
W-187
Y-90
Yb-169
Zn-67
Zr-95

with US NRC release criteria. The current capacity is about six million discrete analyses per year. The analysis program is also offered for material treated at other facilities.

SHIPPING The service group also operates hot cells and provides a complete shipping program to ensure that material can be returned to experimenters.

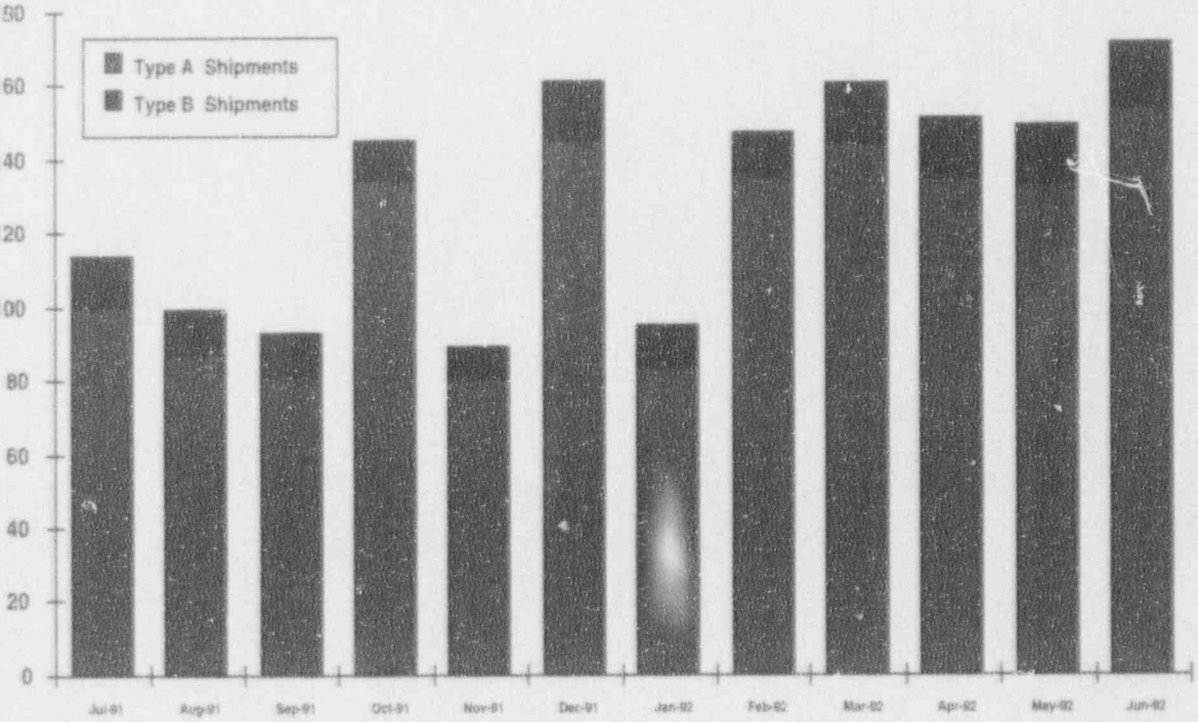


Fig. 1 Radioactive shipments from MURR.

SILICON ANALYSIS PROGRAM

Michael D. Glascock, Senior Research Scientist (Adjunct Assistant Professor, Nuclear Engineering)
Hector Neff, Research Scientist (Adjunct Assistant Professor, Anthropology)
Jeffrey R. Denison, Senior Research/Laboratory Technician

The Silicon Analysis Program was developed in the early 1980s by Dr. Michael D. Glascock. While the program was originally designed to satisfy the analytical needs of a rapidly growing semiconductor industry, the past decade has brought steady growth and a broadening of scope. In the beginning, the work performed mainly involved bulk impurities analysis for research and development divisions of a handful of US companies. Today, in addition to the R & D work, companies are using the Silicon Analysis Program to provide quality assurance analysis on materials such as silicon, high purity graphite, high purity quartz and ^{11}B. The program is internationally recognized and performs analysis for companies in Denmark, Japan and Italy as well as the United States. During FY92, the program performed work for 11 industrial customers and two universities. Figure 1 illustrates the FY92 sample volume.

The Silicon Analysis Program utilizes neutron activation analysis (NAA) to determine trace impurities in semiconductor and other high purity materials. NAA offers many advantages over other analytical techniques, such as:

- An ability to analyze simultaneously for approximately forty elements.
- An ability to detect accurately elemental concentrations in the ppb range and below.
- An ability to perform the analysis without destruction or loss of the sample.

Table 1 lists the elements determined and typical detection limits of a standard analysis of high purity silicon.

Several changes made to the standard procedures over the last year have improved the program significantly. The first was the replacement of the old graphite irradiation containers with high purity graphite containers. This not only reduced the sample contamination potential from graphite dust residue but also significantly reduced the activity level of the containers after irradiation. Second, a mandatory de-ionized water, ultrasonic cleaning procedure was added. This new procedure has proven to clean the samples more thoroughly than rinsing with DI water alone. Third, a short irradiation analysis service has been developed for non-silicon matrix samples. The short analysis can provide the customer with data on short-lived element isotopes such as Al, Cl, Mg, Mn and Ti.

Earlier this year the Silicon Analysis Program was awarded a $4,000 contract from Eagle-Picher, Inc. to perform a feasibility study to determine if high purity quartz irradiation containers held any advantages over the presently used high purity graphite containers. The results of the study indicated that no significant advantages in data quality, container activity level or container reusability exited. A disadvantage to using quartz over graphite is cost, with quartz irradiation containers being about 3 times more expensive than graphite containers. So, for the present, the Silicon Analysis Program will continue to use high purity graphite containers.

Future program improvements include the possible installation of a soft-walled clean room for pre-irradiation sample handling and an increased sample irradiation time to improve detection sensitivity.

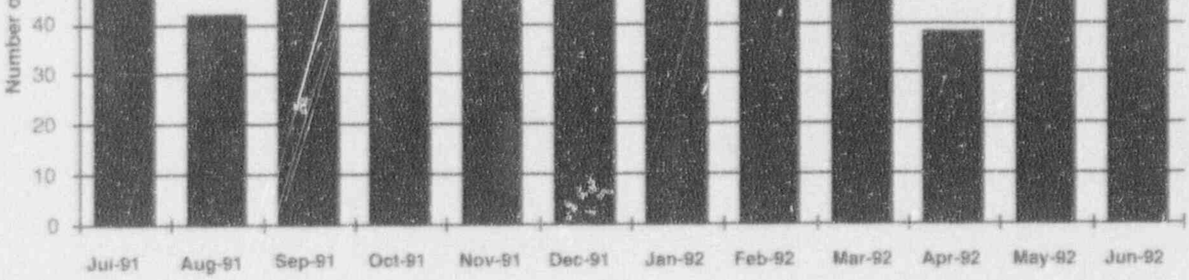


Fig. 1 Number of samples analyzed during FY92

Table 1 **Approximate detection limits for impurities in an undoped silicon specimen weighing 15 grams after a 40-hour irradiation in a thermal neutron flux of 2.5 x 1013 n/cm^2/s.***

Elements	Concentration PPB by weight	Atoms/cc of silicon	Element atoms per billion silicon atoms
Ag	1.5E-03	2E+10	4E-04
As	2.5E-03	5E+10	1E-03
Au	1.5E-05	1E+08	2E-06
Ba	1.0E-01	1E+12	2E-02
Br	3.0E-03	5E+10	1E-03
Ca	6.0E+01	2E+15	4E+01
Cd	2.5E-02	3E+11	5E-03
Ce	2.0E-02	2E+11	4E-03
Co	4.0E-04	2E+10	2E-04
Cr	2.0E-02	5E+11	1E-02
Cs	1.0E-04	1E+09	2E-05
Cu	3.0E-02	6E+11	1E-02
Eu	4.5E-04	4E+09	8E-05
Fe	8.0E-01	2E+13	4E-01
Ga	4.0E-03	8E+10	2E-03
Hf	1.5E-03	1E+10	2E-04
Hg	4.0E-03	3E+10	6E-04
In	8.0E-03	1E+11	2E-03
Ir	1.5E-05	1E+08	2E-06
K	5.5E-01	2E+13	4E-01
La	1.0E-03	1E+10	2E-04
Mo	1.5E-01	2E+12	4E-02
Na	3.5E-02	2E+12	4E-02
Ni	4.0E-01	1E+13	2E-01
Pt	7.0E+00	5E+13	1E+00
Rb	1.5E-02	3E+11	6E-03
Sb	2.0E-03	2E+10	4E-04
Sc	6.5E-05	2E+09	4E-05
Se	6.0E-03	1E+11	2E-03
Sn	2.0E-01	2E+12	4E-02
Sr	2.0E-01	3E+12	6E-02
Ta	2.5E-04	2E+09	4E-05
Tb	2.0E-04	2E+09	3E-05
Th	6.5E-04	4E+09	8E-05
Ti	2.0E+02	5E+15	1E-02
U	1.5E-03	1E+10	2E-04
W	1.5E-03	1E+10	2E-04
Yb	6.0E-03	5E+10	1E-03
Zn	4.5E-02	1E+12	2E-02
Zr	3.0E-01	5E+12	1E-01

*Determined by using a high-resolution HPGe detector to perform two measurements of 30 minutes after 2 days and 6 hours after 15 days, respectively.

In addition to the production of isotopes for MURR and MU research work, the MURR Radiopharmaceutical group (RPG) performs numerous irradiation and processing services for other researchers at MU, other universities and industry. Typical examples of such work include:

- Production and processing of the short-lived tracers K-42 and Na-24 for studies of essential hypertension by Professor Allan Jones and other scientists at the MU Physiology and Pharmacology Departments have now been turned over to MURR Service Applications after more than 15 years of continuous supply by RPG. RPG has supplied well over 2000 samples during this time, supporting substantial external grant money to the above departments.

- Fluxtrap irradiaton and processing of high-specific-activity Re-186 for radioimmunotherapy clinical trials in colon, lung and ovarian cancer patients sponsored by NeoRx Corporation in Seattle, St. Louis, Los Angeles, and Davis, CA have been a major effort, with treatment of 40-50 patients annually and 1-2 samples processed per week, each exceeding 1 Curie of Re-186. A patent is held jointly by MU and NeoRx on a novel rhenium reactor target material which has made the processing at MURR and NeoRx simple, safe, and convenient.

- Weekly production of rare-earth radioisotopes such as Ho-166, Lu-177, Sm-153, and Gd-159 for commercial medical research in radioimmunotherapy and for patient trials of Sm-153 EDTMP, a novel bone cancer pain palliation agent invented at MU and MURR. Sm-153 EDTMP currently is entering Phase III trials in US patients, supported by production at MURR and sponsored by the Dow Chemical Corporation.

- Production of Pt-195m for Professor Walter Wolf at UCLA for use in the preparation of Pt-195m cisplatin, an experimental radiodiagnostic agent tracking the response to cisplatin chemotherapy in human brain cancer patients. About 25 patients are treated each year.

- The MU Department of Chemistry and the Center for Radiological Research (CRR, which has been "spun off" from MURR) receive frequent shipments of Au-198, As-76, Pd-109, Rh-105, and Cd-115/In-115m, Dy-166/Ho-166, and W-188/Re-188 generators for their research in the biological applications of radioisotopes and for lab course work. Some of these isotopes, notably Rh-105, require substantial and complicated processing at MURR.

- Several irradiations producing isotopes of hafnium were performed for Professor Girit at Yale University for research in physics; University of Missouri physicists also occasionally receive shipments of Pd-103 and Pr-142 for research and calibration purposes. Preparation of Te-128 and Te-125m for Professors Boolchand and Bresser at the University of Cincinnati for Mössbauer studies has been done also.

- Irradiation of Pd-102 to produce Pd-103 commercially for use in TheraSeed™ implants for the treatment of prostate cancer in the US is performed at MURR. RPG has also transferred this duty to Service Applications, which involves 20-30 pneumatic tube irradiations of samples of Pd-102 each year to support the quality control program used by the manufacturer of TheraSeed,™ Theragenics Corporation in Atlanta, GA.

- Irradiation of hydroxyapatite (HA) microspheres for CeraMed Corporation in support of Mallinckrodt Medical's development of Ho-166 HA microspheres for rheumatoid arthritis radiotherapy by radiation synovectomy.

Nuclear Analysis Program RADIOISOTOPE PRODUCTION

The three-part mission of MURR–research, education and service–is often overlapping and interconnected. Many research efforts involve developing a tool, technique or technology that evolves into a service application, perhaps even is picked up by industry and carried out on a much larger scale. Nutritional research carried out in the Nuclear Analysis Program at MURR has in the past several years employed some high specific activity radioisotopes that are not commercially available. MURR is the nation's only supplier, allowing scientists at many institutions to conduct new investigations into the role of specific trace elements.

Cu-67/Cu-64 High specific activity Cu-67 and Cu-64 are produced on a monthly basis at MURR. These radionuclides are provided to researchers throughout the United States for experiments in nutrition and radiopharmaceutical evaluations at such institutions as Purdue, Washington University and the Human Nutrition Research Center (USDA-ARS, Grand Forks, ND).

Se-75 Researchers use this important tracer to study the nutritional significance of the essential trace element selenium and possible roles it may play in cancer prevention. The tracer was first investigated and used by MURR researchers in nutritional studies, and by the late 1980s MURR had become a national supplier. Se-75 requires a long, uninterrupted irradiation and chemical speciation following processing. Different chemical forms of Se-75 are being considered currently, since various chemical forms behave differently biologically; for example, some forms may be anti-cancer agents. Se-75 is supplied for a wide spectrum of selenium research to some 20 laboratories throughout the country, including the National Cancer Institute, the National Institutes of Health, Oregon State, Vanderbilt, the University of Rochester and Utah State. This past year shipment of Se-75 went international, accommodating researchers in Germany, Sweden, Great Britain and India.

APPENDICES

Publications

Patents

Papers Presented

Grants and Contracts

Professional Service Activities

Outside Participants

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PATENTS ISSUED OR FILED July 1991 - June 1992

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Day, D.E.; Ehrhardt, G.J., Composition and Method for Radiation Synovectomy of Arthritic Joints, Granted (US #5,039,326)

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Katti, K.V., Production of Trifluoracetates of Transition Metals and Lanthanides from the Water Catalyzed Reactions of Trifluoracetic Acid with the Metals, Disclosed

Katti, K.V.; Cavell, R.G., Carbonylation of Methanol Using a Novel Transition Metal Catalyst Precursor, applications filed in USA, Japan, Poland, Mexico and European Community (EPC)

Katti, K.V.; Volkert, W.A.; Ketring, A.R., New Multifunctional Ligands for Potential Use in the Design of Therapeutic or Diagnostic Radiopharmaceutical Imaging Agents, Submitted

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Miller, W.H.; Berliner, R., A DSP-Based Analog-to-Digital Convertor, Disclosed

Popovici, M.; Heuser, B.J.; Yelon, W.B., Neutron Monochromator for Small Angle Scattering, Disclosed

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Physics, Chemistry and Materials Science PRESENTATIONS

Invited

Farmer, J.W., Radiation Effects in Materials, Cooper Nuclear Power Group, Nuclear Engineering Seminar series, University of Missouri-Columbia, August 6, 1991

Herwig, K., Neutron Diffraction and Inelastic Scattering from a Two-Dimensional Hydrogen Bonded System: Ethanol on Graphite, Physics Condensed Matter Seminar series, University of Missouri-Columbia, November 20, 1991

Kaiser, H., Coherence Experiments with Thermal Neutrons, Physics Condensed Matter Seminar series, University of Missouri-Columbia, September 18, 1991

Kaiser, H., An Overview of Neutron Interferometry, Argonne National Laboratory-IPNS, Argonne, IL, December 20, 1991

Kaiser, H., Neutron Interferometry: A Unique Tool for Quantum Measurements, XIIth Moriond Workshop, Les Arcs, France, January 25 - February 1, 1992

Katti, K.V., New Tc-99m and Re-188 Complexes Derived from Multifunctional Phosphorus Hydrazides, Center for Radiological Research, Columbia, MO, August 10, 1991

Katti, K.V., New Directions in Ligand Design for Chemistry with the "Early and Late" Transition Metals, University of Missouri-Columbia Chemistry DyNAMITE® Seminar series, November 26, 1991

Katti, K.V., First Examples of Heteroatomic Chelation of 188ReO4-99mTcO4- and PdCl42- with a Phosphine-Phosphineoxide (or a phosphinimine) Bridged Bifunctional System, Ninth International Symposium on Radiopharmaceutical Chemistry, Paris, France, April 6-10, 1992

Katti, K.V., Linear, Cyclic and Polymeric Phosphazines as Building Blocks for New Tc-99m, Re-188 and Pd-109 Complexes, Anorganisch Chemisch Institut, University of Gottingem, West Germany, April 13, 1992

Katti, K.V., Biochemical and Catalytical Applications of Transition Metal Chemistry Derived from Novel Phosphorus-Nitrogen Systems, Anorganische Chemisch Laboratorium, University of Amsterdam, The Netherlands, April 16, 1992

Katti, K.V., Synthesis, Structure and Reactivity Aspects of New Multifunctional Phosphorus-Nitrogen Compounds, MURR, June 3, 1992

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Popovici, M., Focusing Techniques in Neutron (and SR) Crystal Diffractometry, Brookhaven National Laboratory, Upton, NY, March 27, 1992

Popovici, M., Focusing Techniques in Neutron Scattering - Elastic and Inelastic, Oak Ridge National Laboratory, Oak Ridge, TN, June 15, 1992

Rhyne, J.J., Magnetism in Rare Earth Metallic Superlattices, 39th Midwest Solid State Conference, Ames, IA, September 27-28, 1991

Rhyne, J.J., Neutron Scattering: A Window on Magnetic Phenomena, Physics Dept. Colloquium, Kansas State University, Lawrence, KS, December 5, 1991

Rhyne, J.J., Magnetic Exchange Interactions in Rare Earth Artificial Metallic Superlattices, Physics Dept. Colloquium, Washington University, St. Louis, MO, February 26, 1992

Rhyne, J.J., The MU Research Reactor (MURR) and Neutron Applications in Materials Science, Nuclear Engineering seminar series, University of Missouri-Columbia, March 31, 1992

Schupp, G., Mössbauer Scattering Studies with High-Intensity Sources, Physics Seminar, Tennessee Technical University, Cookeville, TN, September 24, 1991

Schupp, G., Mössbauer Scattering Studies with High-Intensity Sources, Physics Colloquium, University of Idaho, March 30, 1992

Schupp, G., Mössbauer Scattering Studies with High-Intensity Sources, Physics Colloquium, Boise State University, Boise, ID, April 3, 1992

Schupp, G., Mössbauer Scattering Studies with High-Intensity Sources, Physics Colloquium, Idaho State University, Pocatella, ID, April 6, 1992

Schupp, G., Mössbauer Scattering Studies with High-Intensity Sources, Physics Colloquium, Utah State University, Logan, UT, April 8, 1992

Taub, H., The Multilayer Structure and Growth of Simple Films Studied by Neutron and Synchrotron X-Ray Diffraction, Chemistry Department Seminar, University of California-Berkeley, November 1, 1991

Taub, H., The Multilayer Structure and Growth of Simple Films Studied by Neutron and Synchrotron X-Ray Diffraction, Physics Department seminar, Washington University, St. Louis, MO, February 3, 1992

Werner, S.A., Is Copper-Manganese a Spin Glass?, International Conference on Magnetism, ICM-91, Edinburgh, Scotland, September 4, 1991

Werner, S.A., Cu-Mn: The Prototype Spin Glass is Not a Spin Glass, Physics Seminar, University of Missouri-Kansas City, November 1, 1991

Werner, S.A., Spectral Filtering in Neutron Interferometry, Southeast Section of the American Physical Society meeting, Durham, NC, November 11, 1991

Werner, S.A., Neutron Interferometry: Quantum Interference on a Macroscopic Scale, Physics department seminar, University of Texas, Austin, January 22, 1992

Werner, S.A., Neutron Interferometry: Quantum Interference on a Macroscopic Scale, Physics department seminar, Arizona State University, Tempe, AZ, January 30, 1992

Werner, S.A., Is Cu-Mn a Spin Glass?, Physics department seminar, McGill University, Montreal, Canada. February 13, 1992

Werner, S.A., Neutron Interferometry-Quantum Interference on a Macroscopic Scale, Physics department seminar, McGill University, Montreal, Canada, February 14, 1992

Werner, S.A., Neutron Interferometry, Physics department seminar, Northeast Missouri State University, Kirksville, MO, April 1, 1992

Winholtz, R.A., Microstress in the Mechanical Behavior of Materials, Physics Condensed Matter Colloquium, University of Missouri-Columbia, October 9, 1991

Yelon, W.B., Structural Studies of Rare Earth-Iron Nitrides by Neutron Diffraction, Physics Department, Washington University, St. Louis, MO, March 2, 1992

Yelon, W.B., Neutron Diffraction Studies of Rare Earth Transition Metal Nitrides, INTERMAG'92 Conference, St. Louis, MO, April 12-15, 1992

Contributed

Ding, X.; Farmer, J.W., Lattice Parameter Changes in Neutron Irradiated YBa2Cu3O7, March meeting of the American Physical Society, Indianapolis, IN, March 16-20, 1992

Dozier, W.D.; Herwig, K.W.; Shinar, R.; Jia, H.; Shinar, J., Study of Short-Range Motion of Atomic Hydrogen in Amorphous Silicon by Neutron Reflectometry, 1992 Spring Meeting of the Materials Research Society (MRS), San Francisco, CA, April 26 - May 1, 1992

Dozier, W.D.; Shinar, R.; Jia, H.; Shinar, J., Neutron Reflectometry (NR) Study of Short-Range H Motion in a-Si:H, American Physical Society March meeting, Indianapolis, IN, March 16-20, 1992

Farmer, J.W., Neutron Diffraction and Defect Studies of High Tc Superconductors, Midwest Superconductivity Consortium, West Lafayette, IN, August 5, 1991

Farmer, J.W., Radiation Effects and Hardening, Lecture for Cooper Nuclear Station Visit, Columbia, MO, August 12, 1991

Farmer, J.W., Neutron Diffraction and Defect Studies of High Tc Superconductors, Midwest Sueprconductivity Consortium, Columbus, OH, January 8, 1992

Farmer, J.W.; Su, Z., Capture Kinetics of the Individual DX Center Levels, 16th International Conference on Defects in Semiconductors, Bethlehem, PA, July 21-25, 1991

Farmer, J.W.; Su, Z., Nonexponential Optical Ionization Kinetics of DX Centers in AlGaAs, March meeting of the American Physical Society, Indianapolis, IN, March 16-20, 1992

Hansen, F.Y.; Taub, H., Melting Mechanism in Monolayers of Flexible Rod-Shaped Molecules, March meeting of the American Physical Society, Indianapolis, IN, March 16-20, 1992

Herwig, K.W.; Trouw, F., The Melting Behavior of Ethanol Physisorbed on Graphite, APS March meeting, Indianapolis, IN, March 16-20, 1992

Heuser, B.J.; Spooner, S.; Wignall, G.; Yelon, W.B.; Yelon, A.; Petrova-Koch, V.; Boley, M.S.; Mobley, R.; Miller, W.H., Characterization of Electrochemically Etched Si, APS March meeting, Indianapolis, IN, March 16-20, 1992

Kirk, M.A.; Farmer, J.W., Flux Pinning by Neutron Irradiation Induced Defects, Gordon Conference on Superconductivity, Oxnard, CA, January 15, 1992

Mitchell, J.; Ross, F.K.; Cowan, D.L., A Structural Study of Radiation-Induced Damage in Topaz, American Crystallography Association Annual Meeting, Toledo, OH, July 21-26, 1991

Popovici, M.; Yelon, W.B., Design Relations for Synchrotron Radiation Curved Two-Crystal Monochromators, National Conference on Synchrotron Radiation Instrumentation, Baton Rouge, LA, October 28-31, 1991

Tang, Y.Y.; Yelon, W.B.; Heuser, B.J., Neutron Diffraction Studies of Hydrogen in Ti-Al Alloys, 1991 Gordon Conference on Hydrogen/Metal Systems, Tilton, NH, July 15-19, 1991

Trouw, F.; Herwig, K.W., The Melting Behavior of Ethanol Physisorbed on Graphite, American Physical Society meeting, Indianapolis, IN, March 16-20, 1992

Wagoner, R.A.; Mullen, J.G.; Schupp, G.; Yelon, W.B., Precision Measurements of Elastic Scattering from Alkali Halides, March meeting of the American Physical Society, Indianapolis, IN, March 16-20, 1992

Wang, S-K.; Dai, P.; Angot, T.; Ehrlich, S.N.; Larese, J.Z.; Taub, H., Synchrotron X-Ray Diffraction Study of the Multilayer Growth of Xe on Ag(111): Preliminary Results, March meeting of the American Physical Society, Indianapolis, IN, March 16-20, 1992

Xie, Q-Y.; Hu, Z.; Mitchell, J.; Schlemper, E.; Ross, F.K., Electron Density Analysis with X-Ray and Neutron Diffraction Data, American Crystallography Association Annual Meeting, Toledo, OH, July 21-26, 1991

Invited

Ehrhardt, G.J.; Volkert, W.A.; Ketring, A.R., Production of Isotopes at Nuclear Reactors, 4th International Symposium on the Synthesis and Applications of Isotopes and Isotopically Labelled Compounds, Toronto, Canada, September 3-7, 1991

Kunze, J.F., State of the Nuclear Engineering Program at the University of Missouri-Columbia, MU Nuclear Engineering Seminar Series, University of Missouri-Columbia, September 3, 1991

Loyalka, S.K., Risk Assessment, MU Nuclear Engineering Seminar Series, University of Missouri-Columbia, October 29, 1991

McKibben, J.C., Status of the TRUMP-S Litigation, Test, Research and Training Reactors meeting, Cambridge, MA, October 23-25, 1991

McKibben, J.C., The University of Missouri Research Reactor, IAEA Technical Committee Meeting on Research Reactor Problems of the 1990s, Dhaka, Bangladesh, June 22-26, 1992

McKibben, J.C.; Rhyne, J.J., Status of the MURR Upgrade, Test, Research and Training Reactors meeting, Cambridge, MA, October 23-25, 1991

McKibben, J.C.; Rhyne, J.J., Update on the University of Missouri-Columbia Research Reactor Upgrade, Winter 1991 American Nuclear Society meeting, San Francisco, CA, November 10-14, 1991

McKibben, J.C.; Rhyne, J.J., Update on the University of Missouri-Columbia Research Reactor Upgrade, Second Meeting of the International Group of Research Reactors (IGORR), Saclay, France, May 18-19, 1992

McNally, M.G., TRUMP-S Research, MU Chemical Engineering Seminar series, University of Missouri-Columbia, February 25, 1992

Meyer, W.A.; Hultsch, R.A., Status of Efforts to Obtain a Spent Fuel Shipping Cask, presented by J.C. McKibben, Test, Research and Training Reactors meeting, Cambridge, MA, October 23-25, 1991

Meyer, W.A.; Tritschler, N.E.; Hudson, R.A.; Muren, P.J., Randolph, M.L., Reactor Startup Demonstration, Summer 1992 Nuclear Science for Secondary Teachers Workshop, University of Missouri-Columbia, June 24, 1992

Riggle, K.J., Environmental Monitoring of Uranium and Neptunium Using Epithermal Neutron Activation Analysis, MU Nuclear Engineering Seminar, University of Missouri-Columbia, April 7, 1992

Storvick, T.S., Japanese Plan to Reprocess Nuclear Fuel, Summer 1992 Nuclear Science for Secondary Teachers Workshop, University of Missouri-Columbia Nuclear Engineering Department, June 24, 1992

Contributed

Kunze, J.F.; Smith, D.; Bennett, P.C., Contamination Clean Up and Subsequent Weeping Observation on Metal Shipping Cask Surface Materials, Annual Meeting of Health Physics Society, Washington, DC, July 25, 1991

Invited

Chaudhuri, T., Radioimmunodiagnosis and Treatment of Ovarian Cancer, American Society for Microbiology and Immunology Symposium, Columbia, MO, April 3, 1992

Chaudhuri, T., Radioimmunodiagnosis and Treatment of Ovarian Cancer, MURR, April 8, 1992

Ehrhardt, G.J.; Volkert, W.A.; Ketring, A.R., Production of Isotopes at Reactors, 4th International Symposium on the Synthesis and Applications of Isotopes and Isotopically Labeled Compounds, Toronto, Ontario, September 3-8, 1991

Ehrhardt, G.J., Nuclear Medicine Applications of Radioactive Microspheres, MU Chemical Engineering Department seminar, Columbia, MO, October 22, 1991

Ehrhardt, G.J., Applications of Radioactive Microspheres for Liver and Kidney Malignancies and Rheumatoid Arthritis Therapy, Brookhaven National Laboratory, Upton, NY, February 26, 1992

Ehrhardt, G.J., Recent Developments in Nuclear Medicine Research at the University of Missouri Research Reactor, High Country Nuclear Medicine Meeting, Vail, CO, March 16-20, 1992

Ehrhardt, G.J., Nuclear Medicine Radiotherapy with Labeled Microspheres or with Re-186/Re-188, MU Chemistry seminar series, University of Missouri-Columbia, June 5, 1992

Ehrhardt, G.J.; Curtis, R.L.; Lattimer, J.C.; Day, D.E.; Bartlett, B.; Quesenberry, J.; Ross, G.; Volkert, E.W.; Lanigan, J.P.; Evans-Blumer, M., Investigation of Pre-Operative Sterilization of Kidney Cancers Using Intra-Arterial Samarium-153 Microspheres, 39th Annual Meeting of the Society of Nuclear Medicine, Los Angeles, CA, June 9-12, 1992

Jimenez, H.N., Synthetic Pathways to Metal Labeled Steroids of High Receptor Binding Affinity for Therapy, MU Chemistry DyNAMITE® seminar series, University of Missouri-Columbia, March 31, 1992

Ketring, A.R., Radiotherapeutic Agents for the Treatment of Pain Associated with Metastatic Bone Cancer, Brookhaven National Laboratory Medical Department, Upton, NY, April 29, 1992

Ketring, A.R., Development of Therapeutic Bone Agents, MU Chemistry seminar series, University of Missouri-Columbia, June 25, 1992

Lang, L., Radiolabeling Proteins with Bifunctional Chelating Agents, MURR, May 4, 1992

Liang, Q., New Peroxide Process for W-188/Re-188 Gel Radioisotope Generators, Schlemper Research Group-University of Missouri-Columbia, January 29, 1992

Morris, J.S., Determination of F in Biological Samples via NAA, MU Chemistry DyNAMITE® seminar series, University of Missouri-Columbia, March 10, 1992

Morris, J.S.; Spate, V.L.; Baskett, C.K.; Mason, M.M.; Reams, C.L., Determination of Fluoride in Human Sera Via Cyclic Instrumental Neutron Activation Analysis (CINAA), Second International Symposium on Nuclear Analytical Chemistry (NAC-II), Toronto, Canada, June 3-5, 1992

Wang, M., Multifunctional Phosphorus Hydrazides as Potential Chelating Agents Diagnostically and Therapeutically Useful Radionuclides, MU Chemistry DyNAMITE* seminar series, University of Missouri-Columbia, March 31, 1992

Wang, N., Labeling of Protein Microspheres with Radiorhenium as a Radiation Synovectomy Agent, Schlemper Research Group-University of Missouri-Columbia, February 18, 1992

Contributed

Anderson, C.J.; Schwarz, S.W.; Rocque, P.A.; Connett, J.M.; Guo, L.W.; Philpott, G.W.; Zinn, K.R.; Meares, C.F.; Welch, C.J., Copper-Labeled Antibodies for PET Imaging, Ninth International Symposium on Radiopharmaceutical Chemistry, Paris, France, April 7-10, 1992 (abstract)

Baskett, C.K.; Spate, V.L.; Mason, M.M.; Reams, C.L.; Morris, J.S., The Determination of Selenium in Urine, Second International Symposium on Nuclear Analytical Chemistry (NAC-II), Toronto, Canada, June 3-5, 1992

Ehrhardt, G.J.; Curtis, R.L.; Lattimer, J.C.; Day, D.E.; Bartlett, B.R.; Quesenberry, J.T.; Ross, G.; Volkert, E.W.; Lanigan, J.P.; Evans-Blumer, M., Investigation of Pre-Operative Sterilization of Kidney Cancers Using Intra-Arterial Sm-153 Glass Microspheres in a Rabbit Model, High Country Nuclear Medicine Conference, Vail, CO, March 16-20, 1992

Ehrhardt, G.J.; Curtis, R.L.; Lattimer, J.C.; Day, D.E.; Bartlett, B.R.; Quesenberry, J.T.; Ross, G.; Volkert, E.W.; Lanigan, J.P.; Evans-Blumer, M., Investigation of Pre-Operative Sterilization of Kidney Cancers Using Intra-Arterial Sm-153 Microspheres, Society of Nuclear Medicine meeting, Los Angeles, CA, June 8-12, 1992 (poster)

Ehrhardt, G.J.; Vanderheyden, J-L.; Su, F-M.; Fritzberg, A.R., Experience with Aluminum Perrhenate Targets for Reactor Production of High Specific Activity Re-186 for Radioimmunotherapy, High Country Nuclear Medicine Meeting, Vail, CO, March 16-20, 1992 (poster)

Gordon, D.E.; Peluso, M.; Zinn, K.R.; Stoops, D.; Ratliff, V.; Trokey, D.E., Retention of Ca-47, Mn-54, Fe-59, Zn-65, and Se-75 in Rats Fed Diets Containing Different Types and Amounts of Dietary Fiber, Institute of Food Technologists annual meeting, New Orleans, LA, June 18-20, 1992

Hunter, D.J.; Willett, W.C.; Morris, J.S., Use of Toenail Clippings as Biomarkers of Trace Element Intake, First International Conference on Dietary Assessment Methods, University of Minnesota, Minneapolis, MN, June 10-13, 1992

Jimenez, H.; Singh, P.R.; Katti, K.V.; Ketring, A.R.; Volkert, W.A., New Approaches to Pd-109 Chelates and their Bioconjugates, 39th Annual Meeting of the Society of Nuclear Medicine, Los Angeles, CA, June 9-12, 1992

Kopicka, K.; Katti, K.V.; Ketring, A.R.; Volkert, W.A., Methylene Bridged Phosphineoxide-Phosphine as a Versatile Chelating Agent for Re-188, 39th Annual Meeting of the Society of Nuclear Medicine, Los Angeles, CA, June 9-12, 1992

Mason, M.M.; Reams, C.L.; Baskett, C.K.; Spate, V.L.; Morris, J.S.; Mills, S., Determination of Total and Bound Selenium in Sera Via INAA, Second International Symposium on Nuclear Analytical Chemistry (NAC-II), Toronto, Canada, June 3-5, 1992

Morris, J.S., The Missouri University Research Reactor Nuclear Analysis Program, Second International Symposium on Nuclear Analytical Chemistry (NAC-II), Toronto, Canada, June 3-5, 1992

Singh, P.R.; Katti, K.V.; Kopicka, K.; Katti, K.K.; Ketring, A.R.; Volket, W.A., New Bifunctional Framework Derived from Phosphorus Hydrazide Chelating Agents for Labeling Proteins with Tc-99m and Re-188, 39th Annual Meeting of the Society of Nuclear Medicine, Los Angeles, CA, June 9-12, 1992

Singh, P.R.; Volkert, W.A.; Ketring, A.R.; Troutner, D.E.; Katti, K.K.; Katti, K.V., A New Chelating Heterodifunctional Phosphorus-Nitrogen Ligand for Radiolabeling of Proteins, Ninth International Symposium on Radiopharmaceutical Chemistry, Paris, France, April 6-10, 1992

Spate, V.L., Applications of the University of Missouri Research Reactor and Food Preservation Using the Co-60 Gamma Facility, Rock Bridge Elementary School, Columbia, MO, February 26, 1992

Volkert, W.A.; Katti, K.V.; Corlija, M.; Meyer, K.; Franklin, T.; Singh, P.R.; Ketring, A.R.; Holmes, R.A., Small Molecule Neutral-Lipophilic Tc-99m Agents Derived from Functionalized Phosphinimines, 39th Annual Meeting of the Society of Nuclear Medicine, Los Angeles, CA, June 9-12, 1992

Volkert, W.A.; Singh, P.R.; Katti, K.K.; Ketring, A.R.; Katti, K.V., A New and Versatile Multifunctional Phosphorus Hydrazide Ligand for Formulating Neutral-Lipophilic Complexes of Tc-99m, Re-186 and Pd-109 Radionuclides, *Ninth International Symposium on Radiopharmaceutical Chemistry*, Paris, France, April 6-10, 1992

Wang, M.F.; Ge, Y.W.; Kopicka, K.; Katti, K.V.; Singh, P.R.; Katti, K.K.; Volkert, W.A.; Ketring, A.R.; Meyer, K.; Corlija, M.; Hoffman, T.J.; Holmes, R.A., New Tc-99m Radiopharmaceuticals Built on Multifunctional Phosphorus Hydrazides, 39th Annual Meeting of the Society of Nuclear Medicine, Los Angeles, CA, June 9-12, 1992

Zinn, K.R.; Chaudhuri, T.; Trokey, D.E.; Sunde, R.; O'Dell, B., Identification and Comparisons of Selenoproteins in the Brain and Mouse Neuroblastoma Cells, FASEB meeting, Atlanta, GA, April 19-28, 1992

Invited

Glascock, M.D., Recent Advances in the Investigation of New World Obsidians, Archaeometry and Technology of Pre-Columbian Materials session of the International Symposium on Archaeometry, Los Angeles, CA, March 21-28, 1992

Hayashida, F., Inka Provincial Administration: A Ceramic Perspective, MURR, January 28, 1992

Contributed

Bove, F.J., El Proyecto Paryjuyu: Resultados Preliminarios y Implicaciones, 5th Annual Symposium on Guatemalan Archaeology, Guatemala City, Guatemala, July 23-27, 1991

Elam, J.M.; Glascock, M.D.; Neff, H., Source Identification and Hydration Dating of Obsidian Artifacts from the Valley of Oaxaca, Mexico, International Symposium on Archaeometry, Los Angeles, CA, March 21-28, 1992

Ericson, J.E.; Glascock, M.D., Obsidian. Flow Differentiation by INAA, Coso, California, Society for American Archaeology Conference (SAA), Pittsburgh, PA, April 8-12, 1992 (poster)

Glascock, M.D., Recent Advances in the Investigation of New World Obsidian, International Archaeometry Symposium, Los Angeles, CA, March 23-27, 1992

Glascock, M.D.; Ericson, J.E., Chemical Characterization of Obsidian Flows and Domes of the Coso Volcanic Field, China Lake, California, 28th International Symposium on Archaeometry, Los Angeles, CA, March 23-27, 1992

Glascock, M.D.; Ericson, J.E., Chemical Characterization of Obsidian Flows and Domes of the Coso Volcanic Field, China Lake, California, Society for American Archaeology Conference (SAA), Pittsburgh, PA, April 8-12, 1992

Glascock, M.D.; Neff, H.; Stryker, K.S.; Johnson, T.N., Sourcing Archaeological Obsidian by an Abbreviated NAA Procedure, 2nd International Symposium on Nuclear Analytical Chemistry, University of Toronto, Canada, June 3-5, 1992 (poster)

Johnson, T., Sourcing and Dating of Obsidian Artifacts from Pacific Coastal Guatemala, Missouri Academy of Science Annual Meeting, Rolla, MO, April 24, 1992

Neff, H., A Role for Sourcing in Evolutionary Archaeology, Society for American Archaeology Conference (SAA), Pittsburgh, PA, April 8-12, 1992

Sisson, E.B.; Neff, H.; Bishop, R.L., Regional Sub-Styles and Provenience of Polychrome 'Laca' with Codex-Style Motifs, 47th International Congress of Americanists, New Orleans, LA, July 8, 1991

Stryker, K.S., A Chemical Investigation of Mississippian Pottery from Moundville, Alabama Using Neutron Activation Analysis, Missouri Academy of Science Annual Meeting, Rolla, MO, April 24, 1992

GRANTS AND CONTRACTS

PRINCIPAL INVESTIGATORS, TITLE	AGENCY/AMOUNT	PERIOD
Benfer, R.A.; Elam, J.M., Source Analysis and Hydration Dating of Post-Formative Period Obsidian Artifacts from the Valley of Oaxaca, Mexico	NSF $8,480	07/91-12/91
Brugger, R.M.; Kunze, J.F., Investigation of Reactions and Moderators for an Accelerator Based Epithermal Neutron Source for Boron Neutron Capture Therapy	DOE $143,700	10/90-09/92
Ehrhardt, G.J., Radiorhenium for Radioimmunotherapy	NeoRx Corp. $525,354	10/89-06/93
Ehrhardt, G.J.; Ketring, A.R., Further Development of the W-188/Re-188 Gel Generator for Radiotherapy	Mallinckrodt Medical, Inc. $39,587	06/91-05/92
Ehrhardt, G.J.; Zinn, K.R.; Day, D.E., Radiation Synovectomy with Re-186 and Re-188 Labeled Protein Microsphere Conjugates	Mallinckrodt Medical $99,876	06/90-05/92
Glascock, M.D., The Archaeometry Laboratory at MURR	NSF $300,000	01/92-12/94
Glascock, M.D., Center for Archaeometric Research at the University of Missouri	NSF $109,977	01/91-12/91
Heuser, B.J., A Novel Approach in the Study of Defects in Metals	MU Research Council $1,478	06/92-12/92
Kaiser, H.; Werner, S.A., US-Austria Cooperative Research: Comparison of Interferometric Measuring Methods (Physics and Materials Research)	NSF $12,200	03/88-08/91
Ketring, A.R., Development of Facilities for Supply of Holmium-166 (^{166}Ho) and Samarium-153 (^{153}Sm)	Mallinckrodt Medical, Inc. $70,000	06/92-05/93
Ketring, A.R.; Katti, K.V.; Volkert, W.A., Development of New MRI Contrast Agents and Radiopharmaceuticals	Mallinckrodt Medical, Inc. $307,815	06/92-05/94

* () is amount to MURR

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PRINCIPAL INVESTIGATORS, TITLE	AGENCY/AMOUNT	PERIOD
Ketring, A.R.; Stringham, L.M., Supply of ^{105}Rh, ^{166}Ho, ^{153}Sm and ^{153}Sm-EDTMP	Dow Chemical Company $702,969	05/88-10/92
Kunze, J.F., National Academy for Nuclear Training Fellowships in Nuclear Engineering, 1990-91	Institute of Nuclear Power Operations $10,000	08/90-07/91
Kunze, J.F.; Alger, D.M.; Blanchard, P.; Brugger, R.M.; Langhorst, S.M., US Department of Energy Radioactive Waste Management Fellowship Program, Participatory Status Renewal for 1990-91 through 1994-95	Oak Ridge Associated Universities $ varies/approved	09/90-08/95
Kunze, J.F.; Alger, D.M.; Blanchard, P.; Brugger, R.M.; Langhorst, S.M.; Lee, P.K.; Loyalka, S.K.; Manahan, S.; Miller, W.H.; Peyton, R.L.; Prelas, M.A., US Department of Energy Radioactive Waste Management Fellowship for Michael Chung	Oak Ridge Associated Universities $18,400	09/90-08/91
Kunze, J.F.; Glascock, M.D., pH Effect of Decontamination of Metal Surfaces	Sandia National Laboratories $26,997	08/91-02/92
Kunze, J.F.; Langhorst, S.M.; Glascock, M.D.; Meyer Jr, W.A., Irradiate and Evaluate Lithium-Aluminide Targets	Argonne National Laboratory $8,902	07/91-01/92
Laporte, R.; Morris, J.S., Selenium in Prospectively Collected Human Sera and Cancer Risk University of Minnesota	NIH $533,000 (24,000)*	01/89-12/92
Loyalka, S.K.; Anderson, H.D.; Keck, B.D.; Chan, P.C.; Krawitz, A.D., Research Experiences for Undergraduates in Particulate Systems Engineering, Summer 1990	NSF $40,023	03/90-09/91
Maxon, H.; Ehrhardt, G.J., Efficacious Skeletal Imaging Radiopharmaceuticals University of Cincinnati	NIH $1,175,537 (12,000)*	04/87-03/92
McKibben, J.C.; Meyer Jr, W.A.; Edwards Jr, C.B.; Langhorst, S.M.; Morris, J.S., MURR FY1991 Upgrade of Equipment and Instrumentation	DOE $225,000	09/90-09/92

* () is amount to MURR

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PRINCIPAL INVESTIGATORS, TITLE	AGENCY/AMOUNT	PERIOD
Miller, W.H., Determination of Prompt Neutron Emissions for Actinide Waste Isotopes	ORNL subcontract through Texas A&M University $7,000	05/92-02/93
Miller, W.H., National Academy for Nuclear Training Fellowships in Health Physics, 1991-92	Institute of Nuclear Power Operations $10,000	08/91-07/92
Miller, W.H.; Banerji, S.K.; Blanchard, P.E.; Carson, W.; Kunze, J.F.; Langhorst, S.M.; Loyalka, S.K.; Manahan, S.; Morris, J.S.; Peyton, R.L.; Mossman, L.J.; Carroll, S.A., US Department of Energy Environmental Restoration and Waste Management Fellowships, Participatory Status for 1990-91 through 1994-95	Oak Ridge Associated Universities $ varies/approved	07/90-06/95
Miller, W.H.; Berliner, R., A DSP-Based, Smart ADC for Nuclear Radiation Detectors	DOE/ER Special Research Grant Program $164,000	04/92-04/94
Miller, W.H.; Kunze, J.F.; Banerji, S.K., The Determination of Radioisotope Levels in Municipal and Industrial Sewage Sludge	Callaway Nuclear Power Plant (Union Electric Company) $60,000	09/91-08/93
Morris, J.S., Analytic Epidemiology Support via Neutron Activation Analysis: Determination of Selenium in Human Serum	Microbiological Assoc. Inc. $94,723	10/90-09/91
Morris, J.S., Reactor Sharing	DOE $70,000	09/91-08/92
Morris, J.S., Reactor Sharing	DOE $70,000	09/90-08/91
Morris, J.S., Trace Elements and Human Health	NIH $376,969	07/89-12/93
Morris, J.S.; Glascock, M.D.; Farmer, J.W.; Berliner, R.R.; Zinn, K.R.; Kaiser, H.; Ehrhardt, G.J.; Ketring, A.R.	NSF $82,000	04/90-10/92

* () is amount to MURR

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PRINCIPAL INVESTIGATORS, TITLE	AGENCY/AMOUNT	PERIOD
Neff, H., Archaeological Research in Pacific Coastal Guatemala	MU Faculty Research Council $893.50	02/92-03/92
Neff, H.; Glascock, M.D., Prehistoric Chronology of the Oaxaca Valley	MU Weldon Spring Fund Competition 1990-91 $20,480	06/91-05/92
Ponnamperuma, C.; Gehrke, C.W.; Caldwell, W.; Carle, G.; Desiderio, D.; Eisenstark, A.; Freeman, D.; Gibson, E.; Hare, E.; Hobish, M.; Knoll, A.; Kumar, S.; Kuo, K.; McEntire, J.; Morris, J.S.; Smith, M.; Stalling, D.L.; Zumwalt, R., A Lunar-Based Chemical Analysis Laboratory (LBCAL)	NASA $350,000 (7,500)*	10/89-09/92
Rhyne, J.J.; Berliner, R.R.; Worcester, D.L., Upgrade of the University of Missouri Small-Angle Neutron Scattering Spectrometer Instrument	NSF Division of Materials Research $182,000	05/90-10/91
Rhyne, J.J.; Werner, S.A.; Stassis, C.; In collaboration with Yelon, W.B.; Taub, H.; Ross, F.K.; Kaiser, H.; Berliner, R.; Goldman, A.I.; Zaretsky, J.L., Development of a Triple-Axis Neutron Spectrometer at MURR	NSF $225,000	06/91-03/93
Ross, F.K., HgI_2 Crystal Perfection Studies	EG&G Energy Measurements Santa Barbara Operations $25,000	08/91-09/92
Ross, F.K.; Berliner, R.; Chaudhuri, T.; Ehrhardt, G.J.; Farmer, J.W.; Glascock, M.D.; Ketring, A.R.; Morris, J.S.; Neff, H.; Yelon, W.B.; Zinn, K.R., Scientific Applications of Neutrons at a University Research Reactor	NSF $165,000	02/92-07/95
Ross, F.K., Subcontract to "An Orientable High Angular Resolution Array of Gamma Ray Detectors"	University of Florida/ US Department of the Navy No. N00014-89-J-3030 $40,540	07/90-04/93

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* () is amount to MURR

GRANTS AND CONTRACTS

PRINCIPAL INVESTIGATORS, TITLE	AGENCY/AMOUNT	PERIOD
Storvick, T.S.; Krueger, C.L.; Sharp, P.; McKibben, J.C.; Langhorst, S.M., Engineering, Chemistry and MURR Program Support of the Rockwell International TRUMP-S Project	Rockwell International Rocketdyne Division $519,312 (273,487)*	04/92-03/93
Storvick, T.S.; Viswanath, D.S.; Krueger, C.L.; Sharp, P.; McKibben, J.C.; Langhorst, S.M.; Retzloff, D.G., Engineering, Chemistry and MURR Program Support of the Rockwell International TRUMP-S Project	Rockwell International Rocketdyne Division $1,383,367 (928,597)*	01/90-03/92
Taub, H., Phase Transitions in Surface Films, NATO Advanced Study Institute, Erice, Italy	NATO $60,890	11/89-08/91
Taub, H., Studies of the Structure, Phase, Transitions and Dynamics of Adsorbed Films	NSF $258,500	09/90-02/94
Taub, H.; Hansen, F.Y., Molecular Dynamics Simulations of Hydrocarbon Molecules Adsorbed on a Graphite Substrate	200 Service Units Pittsburgh Supercomputing Center	04/91-04/92
Taub, H.; Schmidt, P.W., Acquisition of a High-Intensity X-Ray Diffractometer	NSF Materials Research Instrumentation Program $152,700	09/90-01/93
Troutner, D.E.; Schlemper, E.O.; Ketring, A.R., Bifunctional Chelates of ^{105}RH and ^{199}Au as Potential Radiotherapeutic Agent	DOE $573,285	06/89-06/93
Volkert, W.A.; Ketring, A.R.; Kuntz, R.R.; Feldbush, T.L.; Mitchel, E., Production of Radiolabeled Monoclonal Antibody Conjugates by Photoaffinity Labeling	DOE $532,974	07/89-10/92
Werner, S.A.; Kaiser, H., Neutron Interferometry and Neutron Schrödinger Wave Optics	NSF, Physics Division $816,000	01/89-05/95
Winholtz, R.A.; Krawitz, A.D., Characterization of Residual Stresses in a Subscale Cylinder of the Advanced Solid Rocket Motor	Babcock & Wilcox $90,337.66	05/92-10/92

* () is amount to MURR

PRINCIPAL INVESTIGATORS, TITLE	AGENCY/AMOUNT	PERIOD
Winholtz, R.A., Single Crystal and Grain Interaction Stresses Measured with Neutron Diffraction	NSF $94,041	06/92-05/95
Winholtz, R.A., Summer Research Fellowship	MU Research Council $3,779--**declined**	06/92-08/92
Yelon, W.B., Neutron Scattering of Titanium-Aluminum Alloys	McDonnell Douglas $30,000	02/91-02/92
Yelon, W.B., Neutron Supermirrors	Ovonic Synthetic Materials Company, Inc. $20,000	12/90-06/92
Yelon, W.B.; Farmer, J.W.; Cowan, D.L.; Ross, F.K., Neutron Diffraction and Defect Studies of High T_c Superconductors	Midwest Superconductivity Consortium (MISCON) $249,417	07/90-07/92
Yelon, W.B.; Popovici, M., Optimized Optics for Heat-Loaded Double Monochromators	Argonne National Laboratory Advanced Photo Source $26,294	10/91-09/92
Yelon, W.B.; Schupp, G., Inelastic Scattering in Condensed Matter with High Intensity Mössbauer Radiation	DOE $138,694	03/91-12/92
Yelon, W.B.; Schupp, G., Multi-Beam Mössbauer Scattering Facility	DOE $74,065	10/91-09/93
Zinn, K.R., Drug Evaluation Study, Phases I-IV	Oread Laboratories $79,000	07/89-12/92

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PROFESSIONAL SERVICE ACTIVITIES

National and International Committee Participation

DOE Nuclear Medicine Project Review, Review Team, May, 1992, Bethesda, MD G.J. Ehrhardt, member

Co-Moderator for Scientific Paper Session #84, Radiopharmaceutical Chemistry: Proteins/Antibodies III, 39th Annual Society of Nuclear Medicine meeting, Los Angeles, CA, June 9-12, 1992, A.R. Ketring, member; Scientific Program Committee, Proteins/Antibodies Category, member

Executive Committee-American Nuclear Society, Central-Eastern Missouri Section, K. Kutikkad, member

American Board of Health Physics, Panel of Examiners (Part 2), Susan M. Langhorst, member

IAEA Technical Committee Meeting on Research Reactor Problems of the 1990s, Dhaka, Bangladesh, June 22-26, 1992, J.C. McKibben, member, the USA representative

International Group of Research Reactors (IGORR) Organizing Committee, J.C. McKibben, member

Test, Research, and Training Reactors (TRTR) National Organization Executive Committee, J.C. McKibben, chair (1991-1992), member

Professional Engineering Examination Committee, American Nuclear Society, W.H. Miller, member

Accreditation Board for Engineering and Technology, Ad Hoc Visitor for American Nuclear Society, W.H. Miller

Scientific Committee, Louis Neel Laboratory, C.N.R.S., Grenoble, France, J.J. Rhyne, member

Advisory Committee, Los Alamos Neutron Scattering Center, J.J. Rhyne, member

Journal of Magnetism and Magnetic Materials, J.J. Rhyne, Advisory Editor

Neutron Special Interest Group (of the American Crystallographic Association), F.K. Ross, Chairman, 1989, member

PROFESSIONAL SERVICE ACTIVITIES

Chemistry Participating Research Team at the High Flux Beam Reactor, Brookhaven National Laboratory, H. Taub, member

Executive Committee Midwest Analytical Team for Reseach Instrumentation of X-Rays, H. Taub, member

Executive Committee Midwest Universities Group to establish a Collaborative Access Team at the Advanced Photon Source, Argonne National Laboratory, H. Taub, member

National Science Foundation New Young Investigator Award Selection Panel, H. Taub, member

Board of Assessment, Physics Laboratory, National Institute of Standards and Technology, S.A. Werner, member

Intermag '92 (St. Louis, MO, April 13-16, 1992), Publication Committee, W.B. Yelon, chair; Program Committee, member; Steering Committee, member

Magnetism and Magnetic Materials Conference, Publication Committee, W.B. Yelon, chair, Steering Committee, member

University Standing Committee Participation

UM Central Radiation Safety Committee, G.J. Ehrhardt, J.S. Morris, and J.J. Rhyne, members

University Award Committees-Chancellor's Award Committee for Outstanding Faculty Research and Creative Activity, Spring 1992, M.D. Glascock, member

MU Honorary Degrees Committee, A.D. Krawitz, member

MU MAE Promotion and Tenure Committee, A.D. Krawitz, chair

MU Graduate Faculty Senate Procedures Committee, S.M. Langhorst, chair

UM Research Board, S.A. Werner, member

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Governmental Agencies

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Dozier, W.D.
Flesher, S.
Frischherz, M.C.
Hettinger, J.
Katz, J.J.
Kirk, M.A.
Liu, J.Z.
Michalski, T.J.
Trouw, F.R.
Vandervoort, K.G.
Vleck, B.M.
Welp, U.
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Ehrlich, S.N.
Larese, J.Z.
Cumberland Science Museum
Thieme, M.S.
EG&G Energy Measurements
James, C.
Naison, D.
Van den Berg, L.
Foundation for Ancient Research & Mormon Studies
Stocker, T.
Iowa Geological Survey
Anderson, R.R.
Jet Propulsion Laboratory
Vandersande, J.W.
Museum of Northern Arizona
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National Institute of Standards and Technology
Borchers, J.A.
Casella, R.C.
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Industrial Agencies

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Central Research Institute of Electric Power Industry (CRIEPI), Japan
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IBM, Munich, Germany
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Peluso, M.
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Stich, T.A.
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Su, Z.M.
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Taub, H.
Temenak, J.
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Tones, K.K.
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Wang, S-K.
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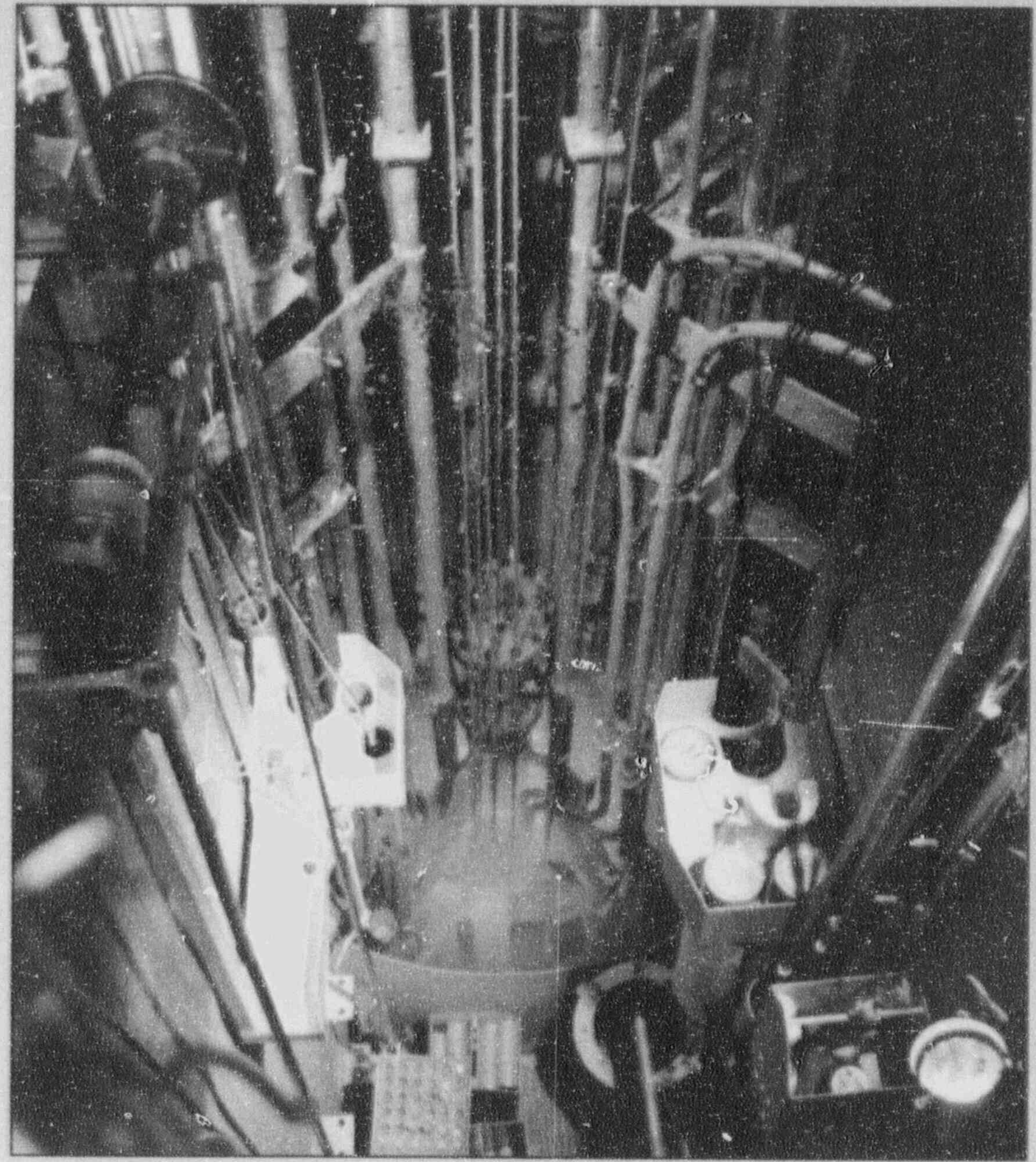
- University of Massachusetts
- University of Michigan
- University of Missouri-Columbia
- University of Missouri-KC
- University of Missouri-Rolla
- University of North Carolina
- University of Oklahoma
- University of Rochester Medical Center
- University of South Florida
- University of Southern California
- University of Texas M.D. Anderson Cancer Center
- University of Utah
- University of Virginia
- University of Washington
- Utah State University
- Vanderbilt University Med Center
- Washington University
- Zynaxis Cell Science, Inc.



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