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Aerosol Release and Transport Program Quarterly Progress Report for January-March 1982

R. E. Adams

M. L. Tobias

Prepared for the U.S. Nuclear Regulatory Commission
Office of Nuclear Regulatory Research
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AEROSOL RELEASE AND TRANSPORT PROGRAM QUARTERLY
PROGRESS REPORT FOR JANUARY-MARCH 1982

R. E. Adams M. L. Tobias

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FOREWORD

This report summarizes progress under the Aerosol Release and Transport (ART) Program [sponsored by the Division of Accident Evaluation of the Nuclear Regulatory Commission's (NRC's) Office of Nuclear Regulatory Research] for the period January-March 1982.

Work on this program was initially reported as Volume III of a four-volume series entitled *Quarterly Progress Report on Reactor Safety Programs Sponsored by the NRC Division of Reactor Safety Research*. Prior reports of this series are

| <u>Report No.</u> | <u>Period covered</u> |
|-------------------|-----------------------|
| ORNL/TM-4655 | April-June 1974 |
| ORNL/TM-4729 | July-September 1974 |
| ORNL/TM-4805 | October-December 1974 |
| ORNL/TM-4914 | January-March 1975 |
| ORNL/TM-5021 | April-June 1975 |

Beginning with the report covering the period July-September 1975 through the report for the period July-September 1981, work under this program was reported as *LMFBR Aerosol Release and Transport Program Quarterly Progress Report*. Prior reports under this title are

| <u>Report No.</u> | <u>Period covered</u> |
|-------------------|-----------------------|
| ORNL/NUREG/TM-8 | July-September 1975 |
| ORNL/NUREG/TM-9 | October-December 1975 |
| ORNL/NUREG/TM-35 | January-March 1976 |
| ORNL/NUREG/TM-59 | April-June 1976 |
| ORNL/NUREG/TM-75 | July-September 1976 |
| ORNL/NUREG/TM-90 | October-December 1976 |
| ORNL/NUREG/TM-113 | January-March 1977 |
| ORNL/NUREG/TM-142 | April-June 1977 |
| ORNL/NUREG/TM-173 | July-September 1977 |
| ORNL/NUREG/TM-193 | October-December 1977 |
| ORNL/NUREG/TM-213 | January-March 1978 |
| ORNL/NUREG/TM-244 | April-June 1978 |
| ORNL/NUREG/TM-276 | July-September 1978 |
| ORNL/NUREG/TM-318 | October-December 1978 |
| ORNL/NUREG/TM-329 | January-March 1979 |
| ORNL/NUREG/TM-354 | April-June 1979 |
| ORNL/NUREG/TM-376 | July-September 1979 |
| ORNL/NUREG/TM-391 | October-December 1979 |
| ORNL/NUREG/TM-416 | January-March 1980 |
| ORNL/NUREG/TM-417 | April-June 1980 |
| ORNL/TM-5806 | July-September 1980 |
| ORNL/TM-7884 | October-December 1980 |
| ORNL/TM-7946 | January-March 1981 |
| ORNL/TM-7974 | April-June 1981 |
| ORNL/TM-8149 | July-September 1981 |
| ORNL/TM-8307 | October-December 1981 |

Beginning with the report covering the period October–December 1981, work under the program is being reported as *Aerosol Release and Transport Quarterly Progress Report*. Copies of all these reports are available from the Technical Information Center, Oak Ridge, Tennessee 37830.

SUMMARY

M. L. Tobias

The Aerosol Release and Transport Program at Oak Ridge National Laboratory is designed to investigate the release, transport, and behavior of aerosols that may carry radionuclides originating from a severe accident resulting in core melting. Aspects of the program apply to both light-water reactors and liquid-metal fast breeder reactors. The experimental programs are being conducted in the Fuel Aerosol Simulant Test (FAST) Facility [which also includes the Containment Research Installation-III (CRI-III) vessel], the Nuclear Safety Pilot Plant (NSPP) Facility, and the CRI-II Facility. The analytical efforts are designed to support the experiments and to provide an independent assessment of the safety margins that exist for the estimation of the radiological consequences of a core meltdown accident.

During this period, the FAST Facility was placed in a standby state until funding resumes.

Results from experiment No. 502 in the NSPP are reported. Operational details from experiment No. 503 are described. Experiment No. 502 continues the study of the effect of steam on an Fe_2O_3 aerosol. A peak steam pressure of 0.27 MPa (absolute) was attained at a temperature of 399 K. The maximum Fe_2O_3 aerosol concentration measured was $1.7 \mu\text{g}/\text{cm}^3$, or about twice that reached in experiment No. 501. Aerodynamic mass median diameters were $1.4 \mu\text{m}$ at 22 min and $0.9 \mu\text{m}$ at 107 min. Scanning electron microscope pictures show spherical particles resembling those seen in U_3O_8 experiments with steam, but the primary particles are much smaller.

In preparation for NSPP run No. 503, tests were conducted to determine plasma torch operating parameters appropriate for generating large quantities of aerosol. Test 503 was conducted late in this quarter, and results will be reported later.

In the core-melt experiments at the CRI-II Facility, an earlier experiment on the vaporization of silver alloy from tube bundle material to simulate the behavior of control rod components was repeated. In this test, the silver alloy content was increased to 10% of the Zircaloy weight, and the total mass was doubled. Silver and indium vaporization was found to be much reduced. Fission product release tests (nonradioactive) were carried out using strontium, barium, and cerium oxides added to powdered uranium oxide in Zircaloy capsules. High vaporizations were noted, suggesting a reducing interaction with zirconium. In other tests, Zircaloy fuel tubes were loaded with metal powders of molybdenum, ruthenium, and tellurium, blended with uranium oxide along with low and high weight fractions of stainless steel. None of the fission product additives appear to have moved out of the furnace zone onto trapping filters. Practically all the material found on them came from the steel additive and the zirconium.

In the analytical program, a model of the moisture balance in the NSPP was constructed to account for mass addition through steam injection, vapor absorption in the air, and vapor depletion by wall condensation and

rainout. The model has been tested in NSPP run No. 093, a steam-only run. Close agreement (3.8%) was found between the formulation used for flow metering and the observed rate of condensation.

GLOSSARY OF ACRONYMS

| | |
|-------------------|---------------------------------------|
| ACER | Annular Core Research Reactor |
| AMMD | aerodynamic mass median diameter |
| ART | Aerosol Release and Transport |
| BET | Brunauer-Emmet-Teller method |
| DWR | boiling-water reactor |
| CDA | core-disruptive accident |
| CDV | capacitor discharge vaporization |
| CRBR | Clinch River Breeder Reactor |
| CRI | Containment Research Installation |
| CSTF | Containment Systems Test Facility |
| FAST | Fuel Aerosol Simulant Test |
| GSD | geometric standard deviation |
| H ₂ DA | hypothetical core-disruptive accident |
| LANL | Los Alamos National Laboratory |
| LMFBR | Liquid-Metal Fast Breeder Reactor |
| LWR | light-water reactor |
| NRC | Nuclear Regulatory Commission |
| NSPP | Nuclear Safety Pilot Plant |
| ORNL | Oak Ridge National Laboratory |
| PSL | polystyrene latex |
| PT | plasma torch |
| PWR | pressurized-water reactor |
| RF | radio frequency |
| SEM | scanning electron microscope |
| TEM | transmission electron microscope |
| TMI | Three Mile Island |

AEROSOL RELEASE AND TRANSPORT PROGRAM QUARTERLY
PROGRESS REPORT FOR JANUARY-MARCH 1982

R. E. Adams M. L. Tobias

ABSTRACT

This report summarizes progress for the Aerosol Release and Transport Program sponsored by the Nuclear Regulatory Commission's Office of Nuclear Regulatory Research, Division of Accident Evaluation, for the period January-March 1982. Topics discussed include (1) the source term experimental program in the Fuel Aerosol Simulant Facility; (2) Fe_2O_3 in steam (light-water reactor accident) aerosol experiments in the Nuclear Safety Pilot Plant (NSPP); (3) core-melt experiments in the Containment Research Installation-II Facility, including studies of the behavior of fission product simulant elements as well as control rod silver alloy components; and (4) analytical modeling of the moisture balance in steam experiments in the NSPP.

1. INTRODUCTION

The Aerosol Release and Transport (ART) Program at Oak Ridge National Laboratory (ORNL), sponsored by the Nuclear Regulatory Commission's Office of Nuclear Regulatory Research, Division of Accident Evaluation, is a safety program concerned with aerosol release and transport. The program's scope includes aerosol release from fuel, transport to and release from primary containment boundaries, and behavior within containments. The overall goal of the program is to provide the analytical methods and experimental data necessary to assess the quantity and transient behavior of radioactive aerosols released from reactor cores as a result of postulated events of varying severity up to and including accidents resulting in core melting.

The program is divided into several related experimental and analytical activities as summarized below:

1. studies related to hypothetical liquid-metal fast breeder reactor (LMFBR) core-disruptive accidents (CDAs) that involve fuel interactions, expansion, and thermal behavior within the sodium pool as the resultant fuel-vapor bubble is produced and transported through the sodium to the cover-gas region;
2. development of apparatus to investigate the characteristics and transport behavior of materials vaporized from molten fuel;
3. study of the characteristics and behavior of fuel-simulant aerosols in several small vessels; and
4. production and study of aerosols in the Nuclear Safety Pilot Plant (NSPP) for the validation of models, with particular emphasis on the

behavior of mixtures of nuclear aerosol species relevant to both light-water reactor (LWR) and LMFBR systems.

Varying levels of effort are anticipated within these categories, with analytical models accompanying the experimental work. The analytical requirements fall into four categories: (1) fuel response to high rates of energy deposition, (2) fuel-bubble dynamic behavior and transport characteristics under sodium, (3) release of aerosols and associated simulant fission products from heated and melting fuel, and (4) dynamic aerosol behavior at high concentrations in the bubble and containment atmospheres.

An attempt will be made to consolidate and present the analyses and data in a manner that will facilitate direct assessment of the radiological hazard associated with arbitrary hypothetical accident scenarios.

2. EXPERIMENTAL PROGRAM

2.1 Source Term Experiments in FAST/CRI-III

A. L. Wright A. M. Smith

The Fuel Aerosol Simulant Tests (FAST) and the Containment Research Installation-III (CRI-III) tests are performed by using the capacitor discharge vaporization (CDV) technique to place UO_2 fuel samples into high-energy states that could be produced in LMFBR hypothetical core disruptive accidents (HCDAs). The primary goal for the FAST/CRI-III test program is to use the experimental results as a data base for developing analytical models that could be used to predict fuel transport through the coolant in severe accidents.

Because of lack of program funding, work in the FAST/CRI-III program was halted this quarter and for the rest of FY-1982. The facility was placed in a "standby" state so that if project funding is resumed in FY-1983 (as expected), the work could be restarted efficiently. Project staff were reassigned to other projects.

2.2 Secondary Containment Aerosol Studies in the NSPP

R. E. Adams R. F. Benson
M. T. Hurst

2.2.1 Introduction

Studies relating to the behavior of aerosols released under LWR accident conditions into secondary containment environments were continued. Three tests have been conducted involving Fe_2O_3 aerosol in a steam environment. Results from Run 501 were reported previously;¹ results from Run 502 are contained in this report. Because Run 503 was conducted late during the quarter, only operational details can be reported at this time.

2.2.2 LWR aerosol experiment No. 502

Experiment No. 502 was the second in a series of tests using Fe_2O_3 as a simulant for aerosols emanating from molten core support and structure materials. The purpose of the test was to observe the influence of condensing steam on an aerosol of Fe_2O_3 at a mass concentration greater than that produced in Run 501 ($0.9 \mu\text{g}/\text{cm}^3$).

To prepare the test atmosphere, steam was introduced into the vessel, which was initially at 0.032 MPa (absolute), to bring the vessel atmosphere (air) to an average temperature of 388 K and a pressure of 0.218 MPa (absolute). This step required about 1 h. At this point, the rate of steam introduction was reduced, and the accumulated steam condensate was removed to a holding vessel. The Fe_2O_3 aerosol generation was then started, and introduction into the steam environment continued for 17 min.

Steam injection at low rates was maintained for 6 h to balance steam losses caused by wall condensation and rainout. Problems with the steam injection control valve resulted in the production of a peak steam temperature and pressure higher than anticipated. At 2 h after the start of aerosol generation, the peak vessel atmosphere pressure was 0.307 MPa (absolute) and the temperature was 399 K; at 6 h, the pressure was 0.270 MPa (absolute) and the temperature was 393 K. Steam injection was terminated at 6 h and the vessel allowed to cool for the next 18 h.

Aerosol mass concentration. The maximum average Fe_2O_3 aerosol mass measured was $1.7 \mu\text{g}/\text{cm}^3$ at 4.3 min after termination of aerosol generation. This mass concentration was greater than that achieved in Run 501 by about a factor of 2 but still was not nearly as large as desired. Operation of the fan-mixer produced a fairly homogeneous mixture of aerosol and steam, as evidenced by Fig. 1, which contains the results from six of the seven individual aerosol mass samplers. The high temperature and pressure caused some operational problems with the aerosol mass samplers. No usable samples were obtained with sampler 151 (upper region of the vessel), only one sample was obtained with sampler 154 (lower region of the vessel), and only two samples were obtained with sampler 157 (about mid-plane of the vessel).

The behavior of an Fe_2O_3 aerosol in these first two tests appears quite similar. On the whole, there is some difference in behavior between U_3O_8 and Fe_2O_3 aerosols in a steam environment. This difference in behavior may be caused by a difference in aerosol concentration; no U_3O_8 aerosol tests in steam were conducted in the 1- to $2\text{-}\mu\text{g}/\text{cm}^3$ concentration range. The third Fe_2O_3 aerosol test (No. 503) will be a further attempt to produce Fe_2O_3 aerosol in a large concentration so that a better comparison can be made of the behavior of these two aerosols.

Aerosol particle size. The aerodynamic mass median diameter (AMMD) of the Fe_2O_3 aerosol was measured both by a cascade impactor and a centrifuge sampler. The "wet" aerosol was dried by dilution with dry air before introduction to the samplers. The AMMD of the "dried" Fe_2O_3 was $1.4 \mu\text{m}$ ($\sigma_g = 1.6$) at 22 min as measured by the centrifuge sampler; at 107 min, the AMMD was $0.9 \mu\text{m}$ ($\sigma_g = 3.3$) as measured by the cascade impactor. Scanning electron microscope (SEM) pictures of the dried aerosol show spherical clumps of particles quite similar in appearance to dried U_3O_8 aerosol. The most notable difference is in the size of the primary particles; those of the Fe_2O_3 aerosol are much smaller than those of the U_3O_8 aerosol.

Aerosol distribution. At the termination of the test (24 h), the approximate aerosol distribution, as determined by the total fallout and total plateout samplers and the final filter samples, was as follows: aerosol settled onto the floor of the vessel, 79%; aerosol plated onto the internal surfaces, 21%; and aerosol still suspended in the vessel atmosphere, nil.

2.2.3 LWR aerosol experiment No. 503

Experience in generation of Fe_2O_3 aerosol in Tests 501 and 502 shows that operating parameters for the plasma torch (PT) aerosol generator, which produces high concentrations of U_3O_8 aerosol, are not necessarily those that will produce large quantities of Fe_2O_3 aerosol. Tests were

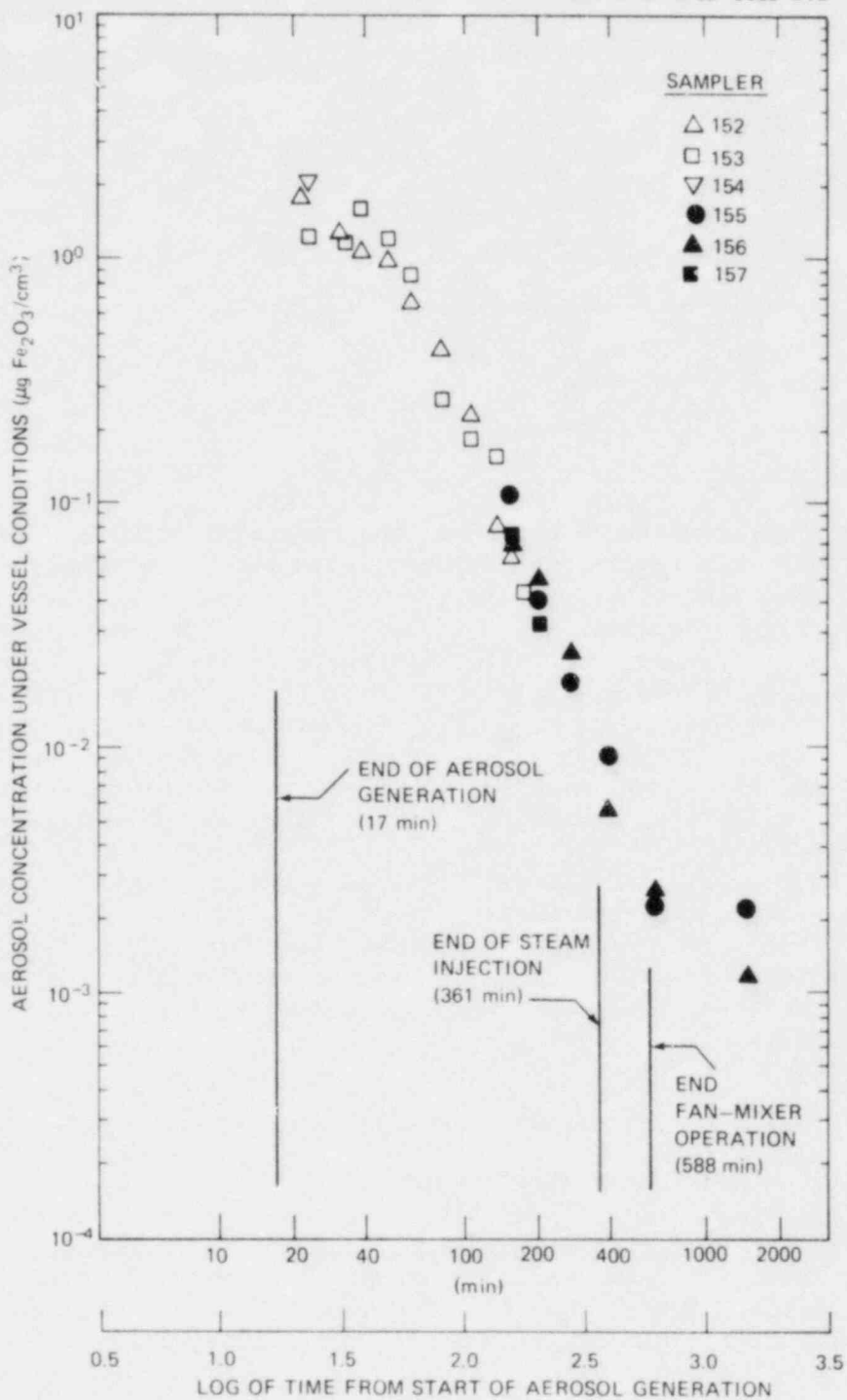


Fig. 1. Fe_2O_3 aerosol mass concentration as a function of time for Run 502.

conducted in a dry atmosphere to determine the optimum parameters for efficient generation of Fe_2O_3 aerosol. These new operating parameters were used in generating aerosol for Test 503. The basic steps in the test were the same as for Test 502 except that the temperature and pressure achieved in the vessel prior to aerosol generation were somewhat lower.

Results from this test will be included in the next report of this series.

2.3 Basic Aerosol Experiments in CRI-II

G. W. Parker G. E. Creek
A. L. Sutton, Jr.

2.3.1 Introduction

This program of basic experiments is carried out in the 5-m³ test vessel, CRI-II, as part of the ART Program. Vapor-condensation aerosols likely to comprise mixtures of importance in various nuclear accidents are systematically produced in 100-g quantities and then characterized. Generation techniques until recently were based on powdered metal-oxygen combustion in a dc plasma arc or, in the case of sodium, by ignition of the liquid metal in a high-pressure spray. These processes, of course, produced only the oxide forms. Recently, because of the current interest in a limited number of pure metallic aerosols, the PT has been adapted to produce vapor-condensation aerosols of the more volatile metals without oxidation.

In the program of basic aerosol experiments, there will be continuing investigations of the fundamental properties of the single-component aerosols, including those of tin, silver, cadmium, and indium for the pressurized-water reactor (PWR); tin and boron for the boiling-water reactor (BWR); and calcium, aluminum, and silicon for the concrete basemat interaction products for core meltdown.

2.3.2 Characterization of the aerosols of silver and tin metals

Experiments conducted recently in the parallel program to characterize core-melt aerosols have indicated the important contribution to the total aerosol mass of the PWR control rod alloy metals silver, cadmium, and indium and of the tin component of the Zircaloy fuel cladding in both types of LWRs. Because it appeared to be the most volatile component of the PWR control rods, cadmium was selected initially for characterization of its aerosol forms. The results were reported in the previous progress report.¹

Silver and tin were the next two metallic aerosols to be generated and characterized by essentially the same method as that described previously for cadmium. Of these two, silver was more readily vaporized in the argon-hydrogen dc plasma than was tin; however, while not all measurements on the tin aerosol are complete, both the size distribution (AMMD) and the

geometric standard deviation (σ_g) have been derived for both from the usual spiral centrifuge measurements illustrated in Figs. 2 and 3. The AMMD values are above $6 \mu\text{m}$, which suggests a relatively large primary particle size and also a relatively low average number of primary particles per agglomerate. The actual value of the number of primary particles per agglomerate was calculated by means of (1) the instantaneous number count of agglomerates per unit volume, (2) the actual mass concentration ($\mu\text{g}/\text{cm}^3$), and (3) the size of an average primary particle, obtained from the measure of the total surface area (m^2/g). A value of about 800 particles was calculated for silver compared with only 200 for cadmium. Data for tin are not complete; however, it is expected to fall near the cadmium value. A transmission photomicrograph of the silver aerosol is shown in Fig. 4, where only a small amount of crystallization is evident.

Another metallic aerosol shown to be a significant contributor to the total released aerosol mass for both types of LWRs is manganese, a minor component (1.5 to 2.0% by mass) of stainless steel. In the next series of basic aerosol experiments involving core component metals, manganese powder is expected to be vaporized at a higher mass concentration than usual because the dc power on the torch has been increased from 40 to nearly 80 kW.

2.3.3 Aerosol generator development

Renewed interest in the possible initiation of large-scale LWR accident aerosol release and transport experiments at various European research centers has resulted in an appraisal of the possibility of redesigning and enlarging the ORNL-type of PT aerosol generator for adaptation and use in these experiments.

Two new concepts for obtaining higher PT aerosol generation efficiencies are being considered, including (1) the substitution of hydrogen as the metal powder carrier gas (instead of argon), thereby furnishing a significant energy boost through combustion of both metal and hydrogen, and (2) increasing the enthalpy of the plasma gas by increasing the dc power up to the limit of the capability of the present electrode cooling system.

An order of magnitude increase in both the physical size of the torch and in the rate of aerosol generation is also being evaluated in cooperation with a manufacturer (Technology Application Services Corporation) of specialty multimegawatt plasma devices.

The interest in aerosol generation on a large scale is divided into two systems (1) for the direct vaporization of a "fissium" or fission product simulant mixture, usually limited to a few elements like strontium, cesium, iodine, and tellurium, and (2) for the vaporization of the heavy aerosol formers including control rod silver alloy components and structural material such as tin, manganese, chromium, and iron. Our program will try to find a suitable process for each of these systems.

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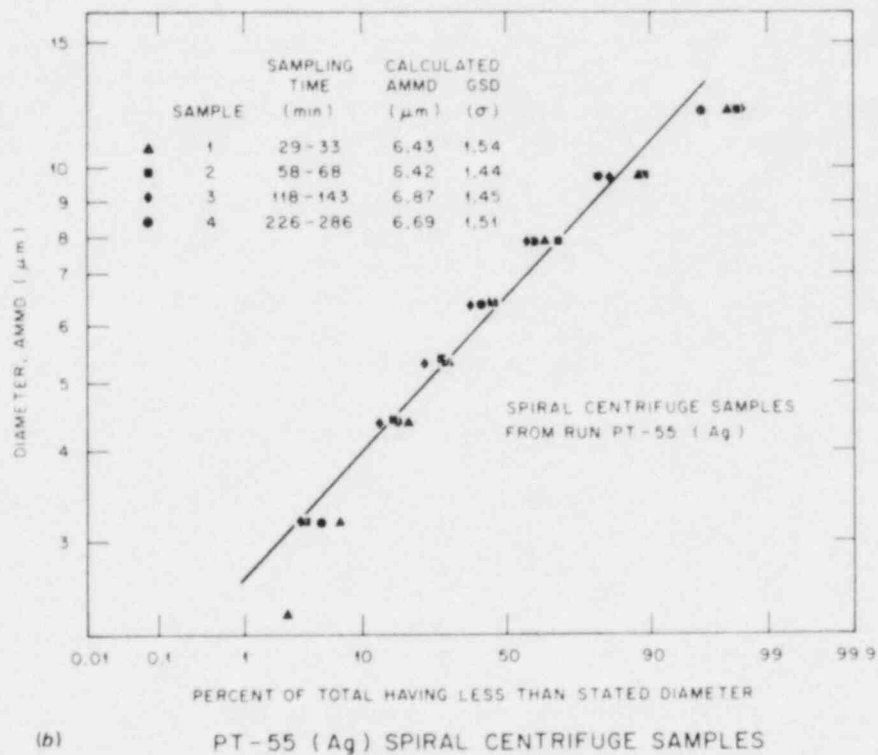
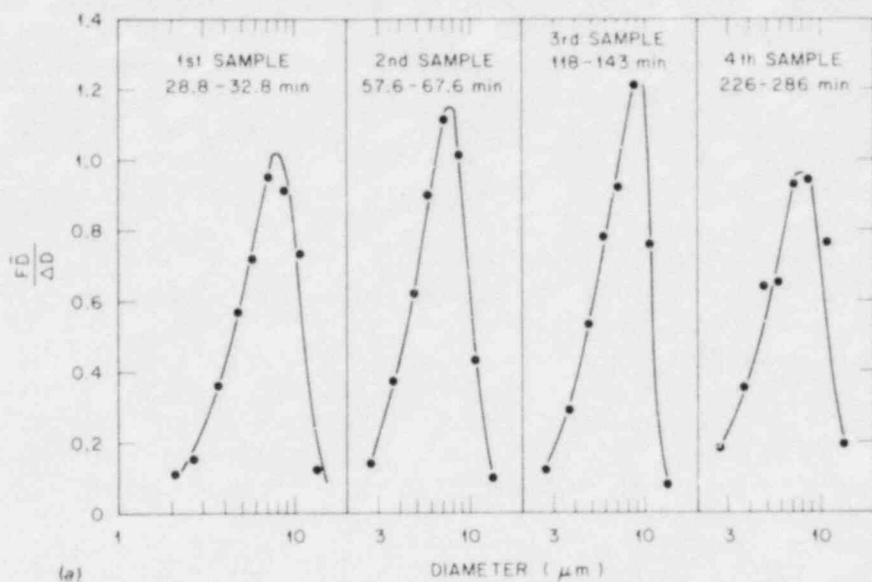


Fig. 2. Fractional size distribution of silver metal aerosol showing nearly constant agglomerate size. (a) By fractional method^{2,3} and (b) by log probability method.

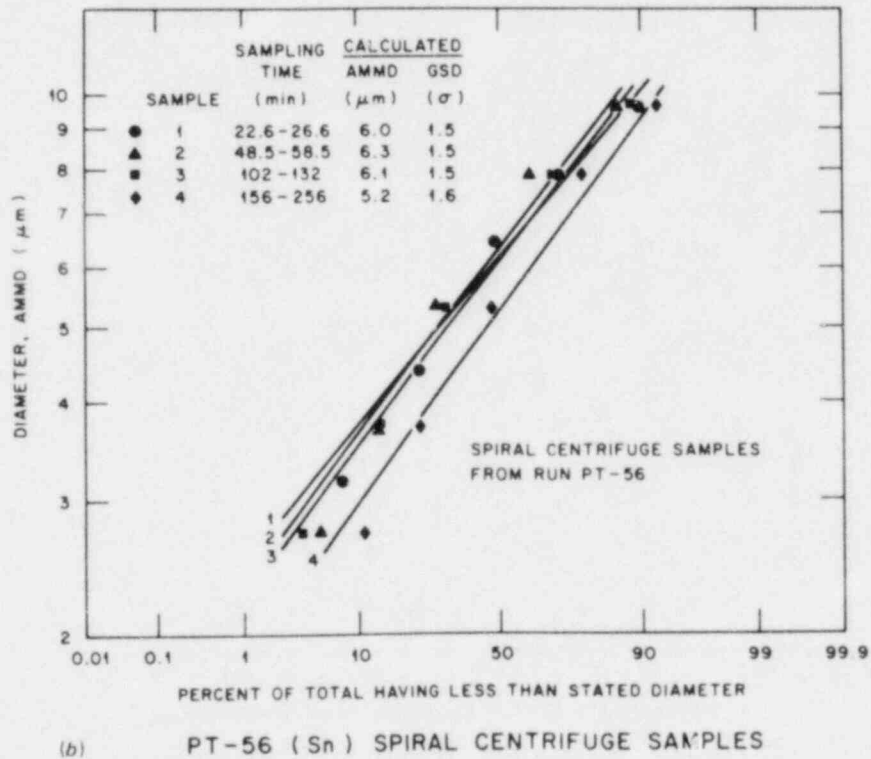
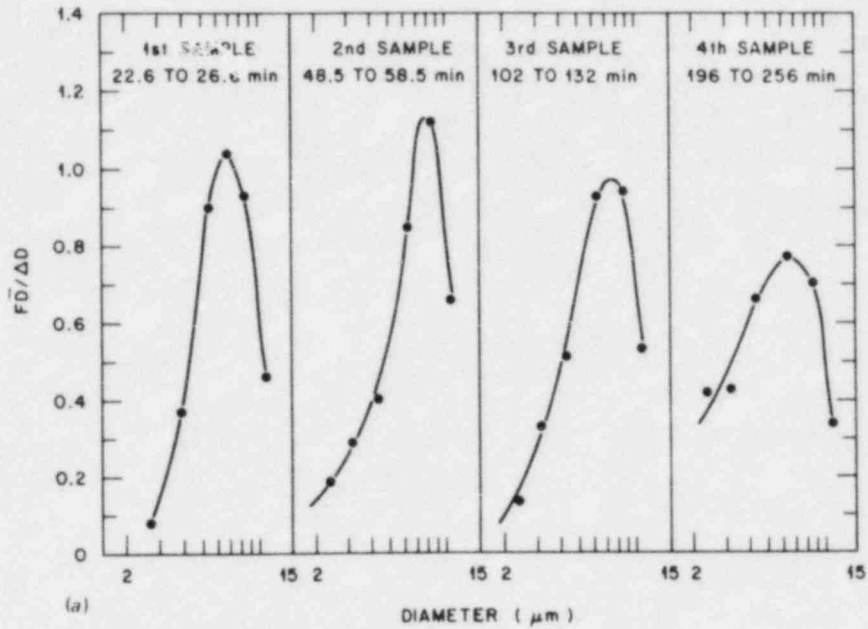


Fig. 3. Fractional size distribution of tin metal aerosol showing slightly decreasing agglomerate size. (a) By fractional method^{2,3} and (b) by log probability method.

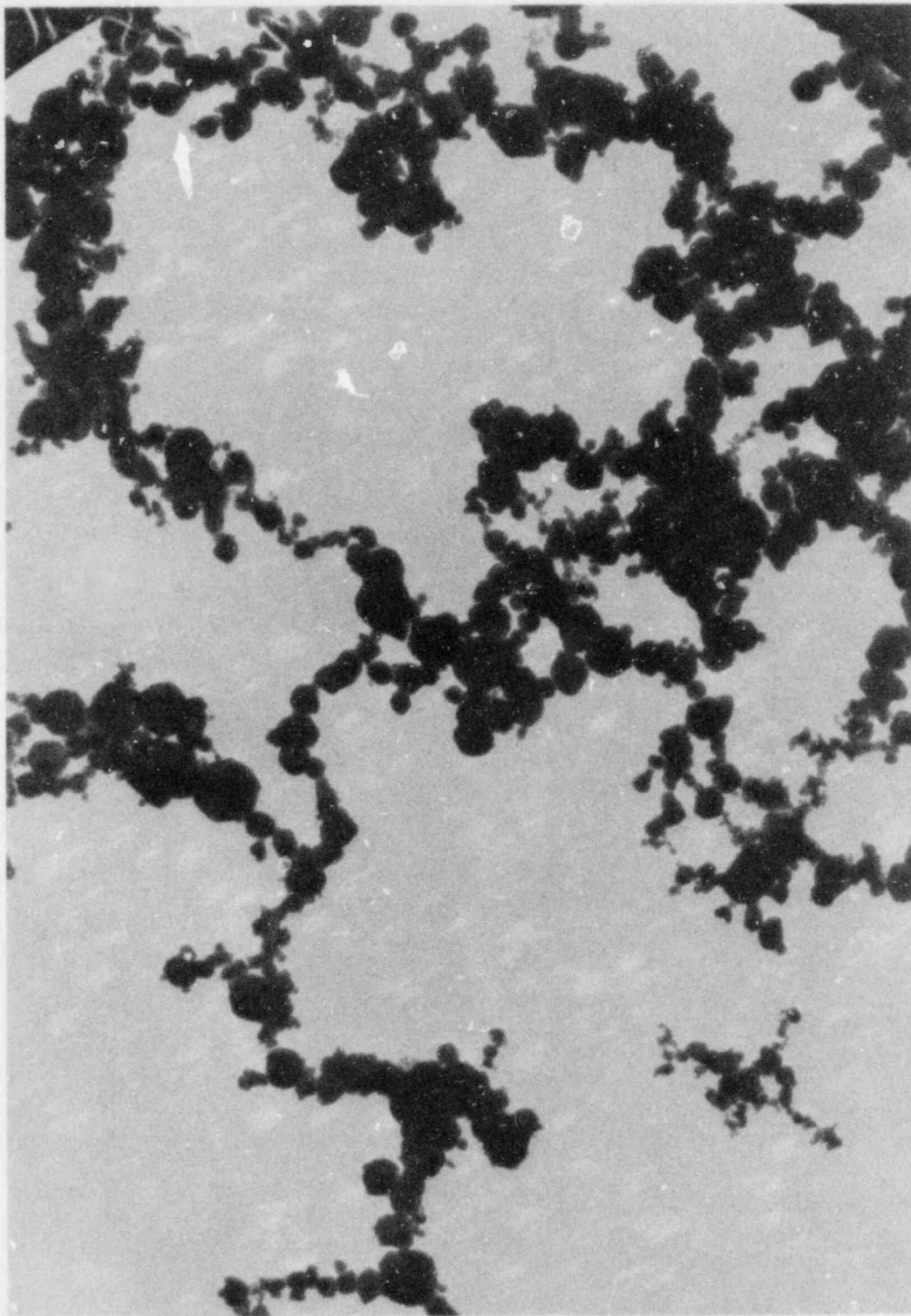


Fig. 4. Silver metal aerosol agglomerates showing mainly spherical particles (1 cm = 0.2 μ m).

2.4 Core-Melt Aerosol Release and Transport

G. W. Parker G. E. Creek
A. L. Sutton, Jr.

2.4.1 Introduction

The core-melt ART experiments are intended to address phenomena associated with LWR Class IX accidents, particularly the postulated large-scale vaporization of fission products, core components, and structural materials. To attain the desired high temperature and melting rates, the radio frequency (RF) induction melting of Zircaloy-clad fuel pins in pre-sintered powdered oxide shells of UO_2 or ZrO_2 , the "skull-melting" technique, has been chosen as the basis of the experimental system. A wide variety of core simulating charge mixtures may be used in this method, and fission product tracers may also be employed. Additionally, apparatus scale-up appears to be reasonably straightforward.

Most of the previous work has dealt with the PWR silver alloy control rod interaction with the Zircaloy cladding and the extensive vaporization of cadmium and silver at temperatures as low as $1400^\circ C$, where the stainless steel sleeve containing the alloy appears to rupture. The cladding is therefore extensively wetted by the silver through formation of low-melting silver-zirconium alloy. Upon further temperature increase, the cladding is melted off the UO_2 but leaves the pellets in a free-standing mode, presenting the appearance of being wetted by a UO_2 -Zr solid solution or eutectic. Further heating in steam to about $2400^\circ C$ produces a liquid phase containing both Zr and ZrO_2 in addition to UO_2 . The silver-zirconium phase is not distinguishable from the eutectic mixture. Free metallic uranium has been reported to be formed at the Zr- UO_2 interface; however, no effort has been made to identify it in this program.

2.4.2 Vaporization of control rod silver alloy components

In the previous progress report,¹ results were reported from core-melt test No. 16, which consisted of a short tube bundle with a total weight of about 0.75 kg, with the addition of an amount of silver alloy to the extent of only 6.7 wt % of the zircaloy. The extensive vaporization of cadmium (53%) and to a lesser extent silver (6.1%) and indium (5.4%) was enough to show that these materials would contribute about 200 kg of total aerosol mass from a complete core melt of a full-size reactor if the same fractional releases were to occur.

In a repeat experiment, Run No. 18, a longer fuel bundle weighing about 1.5 kg total mass was used, and the silver alloy was increased to the more nearly correct proportions of 10% of the Zircaloy weight (Table 1). Results similar to test No. 16 were obtained for the vaporization of cadmium (47%); however, the vaporization of both silver and indium (~0.5%) was reduced, perhaps by the extra mass of Zircaloy, by about a factor of 10. The detailed results of the material balance analysis are given in Tables 2, 3, and 4. Note that the extra length of the Zircaloy fuel capsules caused a significant part of the bundle to be outside the influence of the RF induction field; therefore, this part was not heated except by

Table 1. Core-melt Test 18^a

| | |
|-------------------------|--------------------------------|
| UO ₂ | 966 g |
| Zry-4 (348.7 g Zr) | 354 g |
| Ag, Cd, In, Sn | 35.3 g |
| H ₂ O added | 121 mL |
| H ₂ released | 151 L (STP) = 88% Zr oxidation |
| Total time heated | 29 min |

^aA 7-in. tube bundle.

Table 2. CM-18 filter paper analysis

| Sample No. | Maximum temperature (°C) | Release (mg) | Weight analysis (mg) | | | | |
|------------|--------------------------|--------------|----------------------|------|-----|----|-----|
| | | | Cd | Ag | In | Sn | U |
| 18-A | 1600 | 535 | 532 | 2.9 | <1 | <1 | 0.0 |
| 18-B | 1800 | 87 | 82.3 | 4.5 | <1 | <1 | 0.0 |
| 18-C | 2200 | 13 | 2.3 | 10.6 | 1 | <1 | 0.0 |
| 18-D | 2400 | 22 | <1 | 17.1 | 4.9 | <1 | <1 |

Table 3. CM-18 furnace deposited aerosols

| Sample No. | Temperature (°C) | Release (mg) | Weight analysis (mg) | | | | |
|------------|------------------|--------------|----------------------|------|-----|----|-----|
| | | | Cd | Ag | In | Sn | U |
| 18-A | 1600 | 675 | 670 | 2.9 | 1.9 | <1 | 0.0 |
| 18-B | 1800 | 201 | 197 | 2.9 | 1.4 | <1 | 0.0 |
| 18-C | 2200 | 36 | 5.7 | 28.0 | 2.7 | <1 | 0.0 |
| 18-D | 2400 | 23 | <1 | 18.8 | 4.1 | <1 | 0.0 |

Table 4. Total aerosol release

| Element | Mass released (g) | Mass inventory (%) |
|---------|----------------------|------------------------|
| Cd | 1.49 | 44.9 |
| Ag | 0.09 | 0.34 |
| In | 0.02 | 0.47 |
| Sn | 0.004 | 0.07 ^a |
| U | 3 x 10 ⁻⁵ | 3.5 x 10 ⁻⁶ |

^aBased on tin in silver alloy and Zry-4.

conduction until the fuel bundle gradually collapsed into a uniform molten mass. This delay could have led to more extensive alloying of the silver-zirconium with stainless steel that would lead to a lower vapor pressure corresponding to a lower mole fraction of solute.

2.5 Fission Product Behavior in the Core-Melt Environment

2.5.1 Behavior of strontium, barium, and cerium

The initial fission product release test in the core-melt experiments was conducted with the simulant alkaline earth oxides BaO and SrO added to powdered UO₂ along with cerium as a rare earth surrogate in the form of CeO₂. The powdered mixture was first sintered at 1400°C to somewhat simulate real reactor fuel; however, because no facilities were available for pelletizing the fuel, it was loaded at "tap density" into Zircaloy capsules in the loose powder form. Our choice of the oxide form for the alkaline earth fission product additives is consistent with most estimates that are based on chemical thermodynamics and oxygen potentials in the LWR fuel system. One such analysis of the postulated chemical states of the fission product elements divided into groups is that given by Lindemer.⁴ In an unpublished diagram (Fig. 5), he also gives the expected form for the rare earth elements, including cerium.

This melting experiment (CM-19) (Table 5) was conducted in a step-wise heat-up method beginning with a 5-min period at 1800°C, then 8 min at 2200°, and finally 4.5 min at 2400°C. Hydrogen was measured from each heating interval. The total amount of hydrogen (100 L) is approximately that expected for 100% zirconium metal-water reaction.

The vaporization of both barium and strontium was much higher (9 and 14%, respectively) than could be accounted for except by the reducing interaction with molten zirconium. A similar reducing interaction with stainless steel and Inconel has been reported by Powers⁵ at Sandia.

In a future experiment, we plan to substitute an all-stainless-steel cladding system to measure the relative reducing effectiveness of the two metal systems.

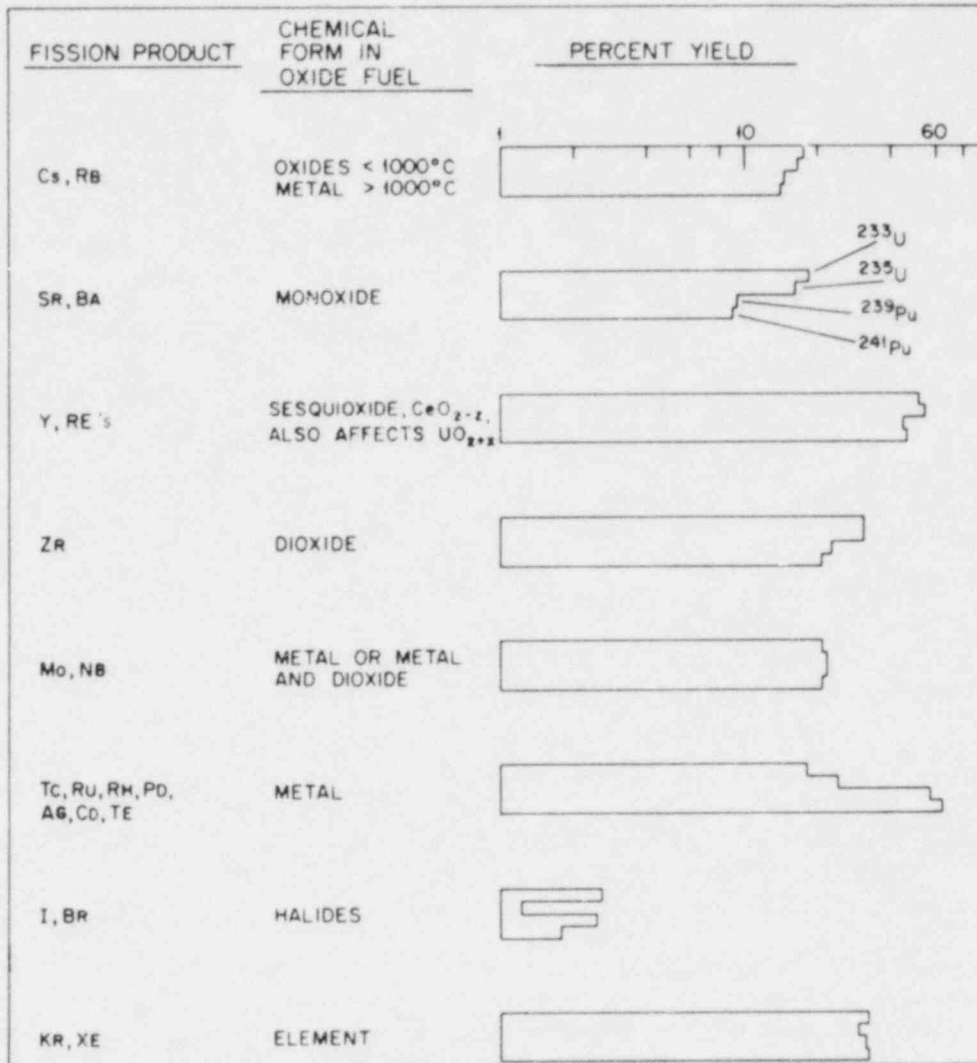


Fig. 5. Predicted chemical state of fission elements in LWR fuel according to Lindemer. (Source: T. B. Lindemer, ORNL, private communication to George Parker, ORNL, May 1982.)

2.5.2 Behavior of molybdenum, ruthenium, and tellurium

Two additional tests were conducted as part of the core-melt aerosol release series involving fission product simulant additives. The fuel mixture for both tests contained appropriate amounts of molybdenum, ruthenium, and tellurium as metal powder blended with UO₂. Stainless steel in the form of spacer plates was added in one test to give a low steel composition (17% by weight); the other test had a high steel composition (67% by weight). Both mixtures were presintered at 1200–1400°C before being loaded into the Zircaloy fuel tubes.

Table 5. Core-melt experiment CM-19

| Element | Weight of each element (g) | Release to the filters (%) | | | |
|-----------------|----------------------------|----------------------------|----------------------|----------------------|----------------------|
| | | 1st Heat (1800°C) | 2nd Heat (2200°C) | 3rd Heat (2400°C) | Total release |
| UO ₂ | 501.35 | 6 x 10 ⁻⁶ | 6 x 10 ⁻⁶ | | 1 x 10 ⁻⁵ |
| Zr | 178.3 | | | 1 x 10 ⁻³ | 1 x 10 ⁻³ |
| Fe | 12.11 | | | | 0.0 |
| Cr | 2.99 | | | | 0.0 |
| Ni | 1.32 | | | | 0.0 |
| Sn | 2.72 | 2 x 10 ⁻² | 0.17 | 5 x 10 ⁻² | 0.23 |
| Mn | 0.16 | | | | ^a |
| Sr | 0.086 | 5.4 | 5.1 | 0.77 | 11.3 ^b |
| Ba | 0.13 | 2.9 | 3.1 | 1.3 | 7.3 ^b |
| Ce | 0.19 | | | | 0.0 |

^aTo be determined.

^bThis does not include addition to the total from furnace wash: Sr = 1.4%, Ba = 2.0%.

The test procedure involved RF heating in steam of the fuel element bundle to melting in three steps: (1) ambient temperature to 1800°C, (2) 1800 to 2200°C, and (3) 2200°C to melting, which occurred at 2400–2500°C. Each heating step required ~8 min. Aerosol material carried from the furnace region during each heating step was trapped on high-efficiency aerosol filters. Details on only one of these experiments, CM-21, are given in Table 6. The appearance of the fuel residues of several experiments is shown in Fig. 6.

Complete analysis (by x-ray fluorescence) has been accomplished only on that aerosol material contained on the filters (Fig. 7), and the results that follow pertain only to that material carried out of the furnace zone to the filters. The results from the two tests were quite similar and show that almost none of the fission product additives were transported to the aerosol filters. Practically all of the aerosol material found on the filters came from the steel additive and from the Zircaloy fuel tubes. During the first heating step to 1800°C, only a small amount of the molybdenum additive (about 0.14%), and then only from the high steel content mixture, was found on the filter. During the heating step from 1800 to 2200°C, about 1% of the tin inventory (from the Zircaloy), 0.5–1% of the manganese (from the steel), and only traces of uranium, zirconium, iron, and chromium (0.02–0.003%) appeared on the filter. Similar

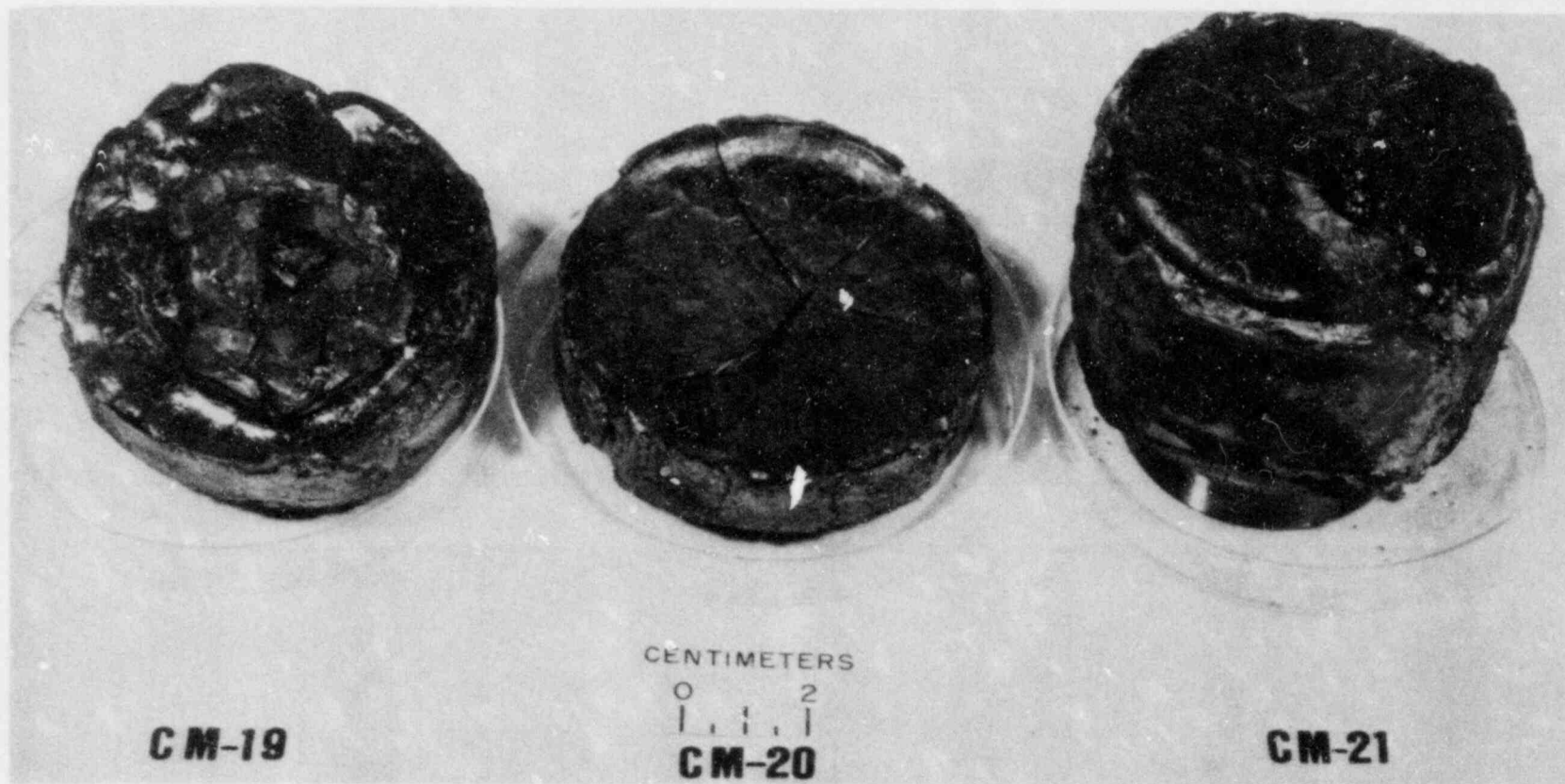
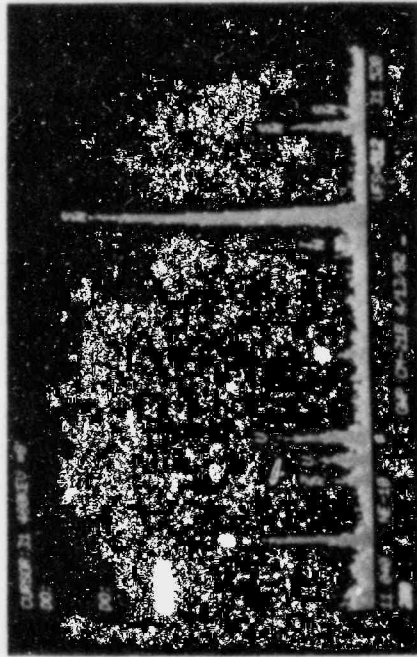
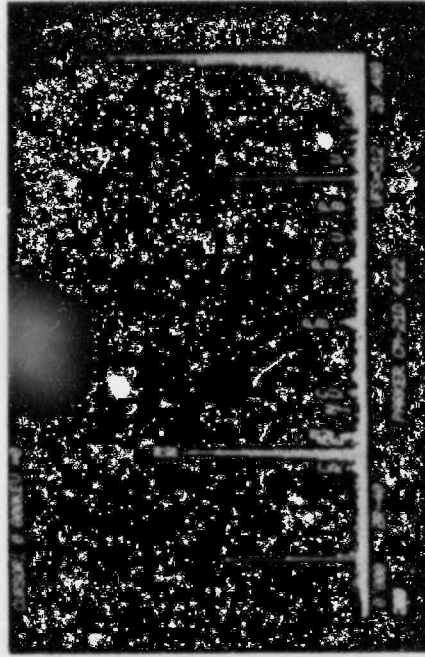


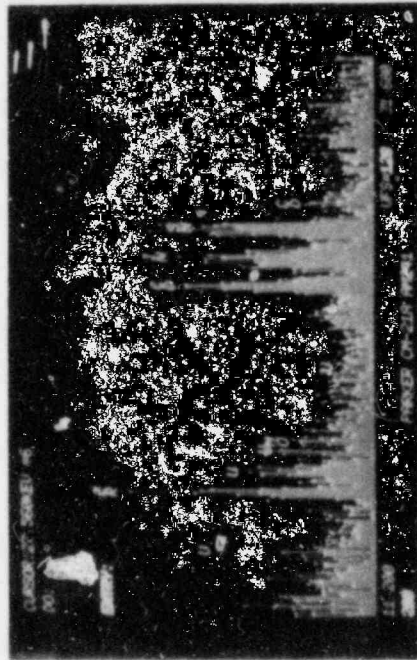
Fig. 6. Core-melt residue from Tests CM-19, CM-20, and CM-21 after heating to 2400°C.



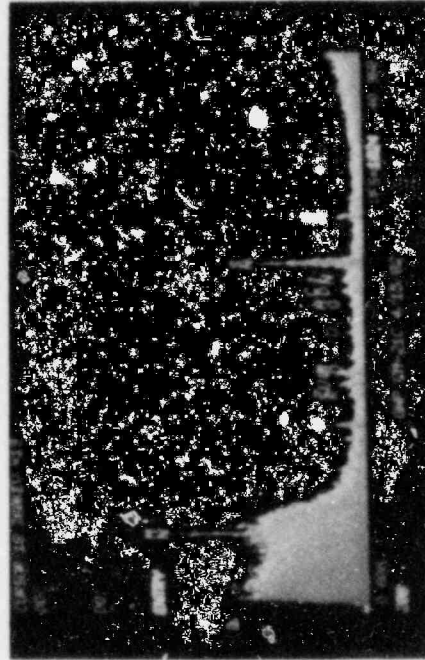
FILTER CM-21B



FILTER CM-21D



FILTER CM-21A



FILTER CM-21C

Fig. 7. X-ray fluorescence analysis of core-melt aerosols.

Table 6. Core-melt experiment CM-21

| Element | Weight of each element (g) | Release ^a (%) | | | | |
|-----------------|----------------------------|--------------------------|----------------------|----------------------|----------------------|----------------------|
| | | 1st Heat (1600°C) | 2nd Heat (1800°C) | 3rd Heat (2200°C) | 4th Heat (2400°C) | Total release |
| UO ₂ | 625.0 | 5 x 10 ⁻⁶ | 2 x 10 ⁻³ | 3 x 10 ⁻⁴ | 8 x 10 ⁻⁶ | 2 x 10 ⁻³ |
| Zr | 178.3 | 1 x 10 ⁻⁵ | 9 x 10 ⁻⁴ | 3 x 10 ⁻⁴ | | 1 x 10 ⁻³ |
| Fe | 70 | | | 9 x 10 ⁻⁴ | 9 x 10 ⁻⁵ | 1 x 10 ⁻³ |
| Cr | 17.00 | | | 6 x 10 ⁻² | 7 x 10 ⁻³ | 7 x 10 ⁻² |
| Ni | 7.7 | | | | | 0 |
| Sn | 2.7 | 0.17 | 1.6 | 1.5 | | 3.27 |
| Mn | 0.96 | | | 0.56 | 1.7 | 2.3 |
| Mo | 0.33 | 0.14 | | | | 0.14 |
| Ru | 0.43 | | | | | 0 |
| Te | 0.06 | | | | | 0 |

^aAll releases doubled to account for expected furnace plateau (see text).

release percentages were noted on the final heating step from 2200°C to melting. The total amount of the tin inventory released and found on the filter was about 3%, and about 1.5% of the manganese was released. It is anticipated that similar amounts of tin and manganese will be found deposited on the furnace walls. In Table 6, we have arbitrarily doubled the filter analysis to project the result to the expected total release. These values seem to be lower than those reported in the recent literature.

The possibility of having lost the tellurium additive during the 1200°C sintering step is being considered; consequently, a duplicate core-melt test will be performed with nonsintered fuel material containing tellurium additive. The further possibility of a volatile gaseous form of tellurium (H₂Te) being generated will be evaluated by the addition of an activated charcoal adsorption bed downstream from the aerosol filter.

3. ANALYTICAL PROGRAM

John Petrykowski

3.1 Calculating the Moisture Balance of the NSPP Vessel for Wet Aerosol Tests

3.1.1 Introduction

Recent tests in the NSPP vessel, conducted to determine the effects of steam environments on the behavior of dry aerosols, identified the need to develop an accurate model of the vessel's moisture balance. A moisture-laden atmosphere may dominate the aerosol behavior, so it is important to develop a simple, reliable moisture balance model that accounts for

1. mass addition through steam injection,
2. vapor absorption in air,
3. vapor depletion caused by wall condensation, and
4. rainout.

If all other mechanisms are negligible, including steam condensation on suspended primary-aerosol particles, the moisture balance can be expressed as

$$M_R = M_S - M_C - M_A, \quad (1)$$

where M_R , M_S , M_C , and M_A are the masses of water caused by rainout, injection, wall condensation, and air absorption, respectively. The terms on the right-hand side can be measured directly or inferred from experimental data. The rainout of airborne water droplets can then be calculated from Eq. (1). The remainder of this report is devoted to a discussion of simple models for calculating M_S , M_C , and M_A .

3.1.2 Steam injection

Steam was injected into the NSPP vessel through a short, 3/4-in.-diam full-flow pipe. Initial attempts at metering the injection rate using a one-dimensional Fanno-flow model were unsuccessful because the small pressure drops in the short pipe could not be measured accurately. At this point, a small metering orifice was installed at the outlet of the pipe. In this configuration, a simple one-dimensional isentropic flow model was used to calculate mass flow rates from pressure drop data. The orifice is a more reliable flowmeter than the straight pipe because, for a given flow, the pressure drop is larger and can be measured with less precision, and the flow rate calculation is noniterative.

The one-dimensional isentropic flow of a gas passing through a single restriction is given by

$$w = P_0 A (P/P_0)^{1/K} \{2K[1 - (P/P_0)^{(K-1)/K}]/[RT_0(K-1)]\}^{1/2} \quad (2)$$

if the flow is choked, or by

$$w = P_0 A [2/(K+1)]^{(K+1)/(K-1)} [K/RT_0]^{1/2} \quad (3)$$

if the flow is not choked, where

w = mass flow rate,

P_0 = upstream stagnation pressure,

P = exit-plane pressure,

T_0 = upstream stagnation temperature,

A = throat area of orifice,

K = specific heat ratio,

R = specific gas constant = universal gas constant/molecular weight.

The flow is choked if the pressure ratio is less than the critical value,

$$(P/P_0)_C = [2/(K+1)]^{K/(K-1)}, \quad (4)$$

where the specific heat ratio varies between 1.13 for saturated steam and 1.3 for superheated steam.

This model was tested in NSPP Run 093, a steam-only run, by integrating the appropriate equation with respect to time and comparing it with the total condensate that had collected in the vessel catch tank from the beginning of injection to the end of tank cooldown (a 24-hr interval). It should be mentioned that any net change in relative humidity has been ignored and would be negligible compared with the quantity of steam injected. Comparison of the mass of steam injected with the mass of steam condensed shows the following values in close agreement:

$$M_{S_{total}} = \int_{t_i}^{t_f} w dt = 220.2 \text{ kg}, \quad (5)$$

$$M_{CT} = (M_C + M_R)_{total} = 212.1 \text{ kg}, \quad (6)$$

$$\text{Error} = \frac{M_{S_{\text{total}}} - M_{CT}}{M_{CT}} \times 100\% = 3.8\% . \quad (7)$$

In this context, steam condensation includes condensation at the wall, M_C , and rainout, M_R .

3.1.3 Water vapor content of the vessel atmosphere

A convenient measure of the water vapor content of a nonreacting mixture is the relative humidity ϕ defined by

$$\phi = P_w / P_{\text{sat}} , \quad (8)$$

where P_w is the partial pressure of water vapor and P_{sat} is the saturation pressure of water at the mixture temperature. In the NSPP vessel, the mixture is essentially air and water. The mass of water can be determined if the relative humidity is known. It is

$$M_w = \phi P_{\text{sat}} V / RT , \quad (9)$$

where V is the vessel volume and T is the mixture temperature.

The relative humidity can be measured directly from a moisture sample reading or indirectly using Eq. (8). The indirect method assumes that the water vapor partial pressure is known. Fortunately, this quantity can be determined by measuring the change in the vessel pressure caused by steam injection. If the atmosphere is initially dry, the partial pressure of water is

$$P_w = P - P_i (T/T_i) , \quad (10)$$

where P_i and T_i represent the initial pressure and temperature in the vessel. When the vessel atmosphere is initially wet, the total relative humidity is

$$\phi = \phi_o + [P - P_i (T/T_i)] / P_{\text{sat}} , \quad (11)$$

where ϕ_o is the initial relative humidity.

A direct method for measuring the water vapor content of air uses a moisture absorbent pack. A sampled volume of air is drawn through the absorbent pack and into an evacuated chamber. The pack absorbs the moisture,

and its increased weight is recorded. The pressure and temperature of the dried air are also recorded. As a function of these variables, the relative humidity is given by

$$\phi = \frac{m_w (P/P_{\text{sat}})}{[m_w + \hat{M}_w (PV/RT)_A]} , \quad (12)$$

where

m_w = mass of moisture in sample,

\hat{M}_w = molecular weight of water,

P_A = pressure of dried air,

T_A = temperature of dried air,

V_A = chamber volume,

R = universal gas constant.

The advantage of this method is that it is independent of the initial relative humidity.

3.1.4 Vapor condensation at the wall

An important moisture removal mechanism in the NSPP vessel is vapor condensation at the vessel wall. Condensation can be measured using a wall-condensation sampler. This sampler, a hexagonal plate mounted flush against the vessel wall, collects moisture that condenses on the defined surface and channels the condensate into a collection tank for volume measurement. If the wall condensation is spatially uniform, the sampled volume can be scaled to give the total wall-condensate volume. The total wall-condensate mass M_c is given by

$$M_c = (SF) \rho V_c , \quad (13)$$

where

SF = scale factor = $\frac{\text{wall area of vessel}}{\text{surface area of sampler}}$,

ρ = density of condensate,

V_c = condensate sample volume.

REFERENCES

1. R. E. Adams and M. L. Tobias, *Aerosol Release and Transport Program Quart. Prog. Rep. for October-December 1981*, NUREG/CR-2299, Vol. 4 (ORNL/TM-8307).
2. T. S. Kress and M. L. Tobias, *LMFBR Aerosol Release and Transport Program Quart. Prog. Rep. for October-December 1981* (NUREG/CR-2299, Vol. 3), ORNL/TM-8149, p. 13.
3. J. E. Smith and M. L. Jordan, "Mathematical and Graphical Interpretation of the Log-Normal Law for Particle Size Distribution Analysis," *J. Colloid Sci.* 19, 549-59 (1964).
4. T. B. Lindemer, Oak Ridge National Laboratory, private communication to George Parker, Oak Ridge National Laboratory, May 1982.
5. Letter from Dana A. Powers, Sandia, to R. Sherry, ORNL, Apr. 19, 1982; Subject: Marviken Experiment Working Group 3.

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