U. S. NUCLEAR REGULATORY COMMISSION
OFFICE OF INSPECTION AND ENFORCEMENT

REGION I

Report No. 50-184/79-05

Docket No. 50-184

License No. TR-5   Priority --   Category E

Licensee: U.S. Department of Commerce

National Bureau of Standards

Washington, D.C. 20234

Facility Name: NBS Reactor

Inspection At: Gaithersburg, Maryland

Inspection Conducted: August 28-31, September 1, 4 and 5, and October 23-26, 1979

Inspectors: _K. E. Plumlee_ _1/21/80_
K. E. Plumlee, Radiation Specialist   date

_____   _____
                                          date

_____   _____
                                          date

Approved by: _H. H. Archer, acting_ _1/22/80_
              , Chief, Radiation Support Section,   date
FF&MS Branch

Inspection Summary:

Inspection on August 28-31, September 1, 4 & 5, and October 23-26, 1979
(Report No. 50-184/79-05)

Areas Inspected: Routine, unannounced inspection by a regional based inspector of research reactor operations and surveillance, requalification training, radiation protection program, and radioactive waste management. Upon arrival a tour was conducted to permit observation of operations. This inspection involved 75 hours on site by one NRC regional based inspector.

Results: Of the five areas inspected no items of noncompliance were identified in four areas. One item of noncompliance was identified in one area (Infraction - Failure to conduct adequate surveys as necessary to identify and post six radioactive materials areas, and meet the requirements of 20.301, Paragraph 4).

Region I Form 167
(August 1979)

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DETAILS

1. Persons Contacted

*Dr. R. Carter, Director, Reactor Radiation Division
 Mr. P. Cassidy, Health Physicist
*Mr. T. Hobbs, Supervisor of Reactor Health Physics
*Dr. T. Raby, Chief, Reactor Operations Section
 Mr. C. Rowe, Health Physicist
*Mr. J. Torrence, Deputy Chief, Reactor Operations Section
 Dr. A. Schwebel, Chief, NBS Health Physics Section; Radiation
   Safety Officer
 Mr. J. Shubiak, Health Physicist
 Dr. R. Zeisler, Acting Chief of Analytical Chemistry

*Denotes those present during exit interviews, 4:15 p.m., September 5,
1979, and 4:00 p.m., October 26, 1979.

2. Licensee Action on Previously Identified Enforcement Items

(Closed) Item of Noncompliance (184/78-04-01): Hood face flow velocities
less than 100 fpm. Measurements on this inspection showed that the face
flows were > 100 fpm and inserts were provided in two unused hoods to
limit the loss of head in the ventilation system.

3. Radiation Surveys and Sampling

   a. Confirmatory Survey

      Part of the routine inspection effort was to survey the reactor
      building and the surrounding area, shown in Attachment 1, Reactor
      Building site, to verify licensee compliance with the requirements of
      10 CFR 20.201 "Surveys", 10 CFR 20.203 "Caution signs, labels, signals
      and controls," and 10 CFR 20.301 "Waste disposal."

      The routine survey was commenced near the reactor building, on
      August 28, 1979. This identified first some radioactive "hot spots"
      at places shown as G_f, G_1 - G_9, and x on Attachment 1. Survey on the
      highest part of the roof identified R_s. Further survey inside the
      fenced enclosure, which is a blacktopped area except for concrete
      structures such as the ramp and the loading docks, identified a
      general area of contamination near and beneath the heat exchanger,
      which was known to be internally contaminated. It was unclear
      whether the areas indicated in Attachment 1 near either end of the heat
      exchanger were joined or separate. Figures and tables in this report
      were excerpted, and modified as necessary, from the attached licensee

report "Report on the Nature and Source of Radioactive Material Found at the NBS Site," dated October 26, 1979, (Attachment 5). The maximum contact dose rate determination was 2 to 3 mrem/hr in the grass, at G_f; 10 mrem/hr on the roof, at R_s; and 50 mrem/hr on the blacktop at several points near either end of the heat exchanger. The heat exchanger and the area Y were part of a posted, roped-off radiation area within the fenced enclosure. No licensee survey records indicated the presence of radioactive contamination at these places.

The inspector observed that the maximum concentration of hot spots was near the primary outlet of the heat exchanger, indicated on Attachment 2, and the shape of area Y (Attachment 1) was similar to that of a liquid spill. No liquid was visible. The licensee later drained about 70 gallons of non-contaminated water from the secondary inlet of the heat exchanger but found no water in the primary outlet, which was the lowest point in the primary system. The licensee's evaluation is that the contamination was released from the heat exchanger, after it was placed there in 1974.

The inspector and the licensee promptly commenced an urgent non-routine survey effort to identify any other contamination in the environs. Surveys of additional areas on the roof identified R_v and R_f. Surveys on the ground identified hot spots at G_p and on the south lawn as well as a second location on the front lawn, shown on Attachment 3. Surveys to identify any drainage path of radioactive contamination out through the storm drain identified contamination in the holding basin and at and beyond the outfall, which was greater than 400 feet from the storm drain opening shown on Attachment 1.

The Reactor Health Physics and the Reactor Operations personnel stated that they had not monitored this outfall because they did not know of it previously. Records showed that routine monitoring had been performed of the secondary cooling system blowdown and of an outfall that received part of the roof runoff.

The inspector reviewed the NBS drawings of the drainage system. Although part of the drain line for the storm drain shown on Attachment 1 was indicated on the drawing the outfall was off the sheet and there was no indication of how or where the line terminated.

The licensee has prepared sketches such as Attachment 3, for future use, and has a monitoring schedule for this outfall. The outfall is checked at least weekly or after each heavy rain, if more frequent, at this time.

b. <u>Followup Survey of the Results of the Cleanup</u>

The licensee removed each of the hot spots including one that was in a crack between the ramp and the building. This was covered and sealed.

The inspector conducted a followup survey on October 26, 1979. Except in the vicinity of the heat exchanger, which had been moved to the side of the waste annex, and in the vicinity of the waste annex which also contained radioactive materials, no hot spots greater than 0.1 mrem/hr on contact were identified.

The heat exchanger survey indicated 10 mrem/hr on contact.

The heat exchanger was shipped to the Barnwell, South Carolina waste burial site during early November 1979.

The waste annex was surveyed before the cleanup and this indicated a maximum of 0.4 mrem/hr on contact at any wall, on the outside.

c. <u>Quantities of Radioactive Materials Cleaned Up</u>

This report uses the terminology, tables and figures, insofar as this is feasible, that the licensee provided in "Report on the Nature and Source of Radioactive Material Found at the NBS Reactor Site," dated October 26, 1979. A single particle containing all of the radioactivity from the sample was isolated in several examples, and several particles were analyzed. The results indicated the activity was primarily Co-60 with a small percentage contribution due to Fe-55.

The licensee measured the quantities of Co-60 removed from the various areas as follows (the percentages of the total are the inspector's numbers). No other gamma emitting isotope was identified by analyses of samples by the licensee and by DOE, Idaho Operations Laboratory. The licensee measured the Fe-55 specific activity at about 1% of the Co-60 specific activity by measuring the x-radiation associated with electron capture by Fe-55, in one particle.

Location	Microcuries	% of Total
Area Y, and nearby	~ 2,000	~ 87
Roof	132	6
Grass Areas	119	5
Drain System and Drainage Area	60	2 1/2

The spatial distribution of these samples is shown on Attachment 3. The reported minimum detectable activity (MDA) is 4E-5 µCi/g for these analyses (4E-2 pCi/g shown on Attachment 3). Additional data is listed in Tables I & II of Attachment 4.

The licensee indicated on the shipping papers that 45 mCi of Co-60 was present in the heat exchanger as shipped to the burial site, however because of measuring problems with such an object, this is an approximation.

4. Regulatory Considerations

a. Radioactive Materials Area Posting Requirements

10 CFR 20.201 "Surveys" states: "(a) As used in the regulations in this part, "survey" means an evaluation of the radiation hazards incident to the production, use, release, disposal, or presence of radioactive materials or other sources of radiation under a specific set of conditions. When appropriate, such evaluation includes a physical survey of the location of materials and equipment, and measurements of levels of radiation or concentrations of radioactive material present. (b) Each licensee shall make or cause to be made such surveys as may be necessary for him to comply with the regulations in this part."

10 CFR 20.203(e)(1) "Caution signs, labels, signals and controls -additional requirements" states: "Each area or room in which licensed material is used or stored and which contains any radioactive material (other than natural uranium or thorium) in an amount exceeding 10 times the quantity of such material specified in Appendix C of this part shall be conspicuously posted with a sign or signs bearing the radiation caution symbol and the words:

Caution
Radioactive Material(s)

By reference to 10 CFR 20 Appendix C and to Figure 3 and Table I of this report it appears that the following six areas, as found, were required to be posted in order to comply with the above in that each area contained in excess of 10 µCi of Co-60. The inspector noted that the licensee was unaware of the presence of the radioactive material and failed to post these areas prior to the inspection.

Area	Quantity of Co-60(µCi)
Front Lawn	11.8
North Lawn	22.2
Rear Lawn	82
Roof	132
Holding Basin	24
Outfall	32.1

The inspector identified the above as examples of failures to make such surveys as may have been necessary in order to comply with the requirements of 10 CFR 20.203(e). (184/79-05-01)

The licensee had established full compliance by removal of the radioactive materials before the completion of the inspection. Corrective measures to prevent recurrence have not been reviewed.

b. Waste disposal

10 CFR 20.301 "WASTE DISPOSAL - General requirement" states: "No licensee shall dispose of licensed material except:

(1) By transfer to an authorized recipient...

(2) As authorized pursuant to § 20.302 or

(3) As provided in § 20.303, or 20.304, applicable respectively to the disposal of licensed material by release into sanitary sewerage systems or burial in soil, or in § 20.106 (Radioactivity in effluents to unrestricted area)."

By reference to the preceding paragraphs of this report it appears that a non-exempt quantity of licensed material was inadvertently released, contrary to the above requirements. This appears to be the result of an inadequate evaluation of the radiation hazards incident to the outdoor storage of the used reactor heat exchanger since 1974; as well as, the result of inadequate surveys to detect the leakage of radioactive materials from the heat exchanger and to detect the spread of radioactive materials to the lawn, roof and the storm drain system and outfall area.

The inspector identified the above as examples of failures to make such surveys as may have been necessary in order to comply with the requirements of 10 CFR 20.301. (184/79-05-01)

The licensee removed the contamination and established full compliance by the completion of the inspection. Corrective actions to prevent recurrence have not been reviewed.

5. Procedural Considerations

Part of the inspection effort was to determine compliance with the requirements of the Technical Specifications. Section 7.4 "Procedures" states: "Written procedures shall be provided and utilized for the following: ... d. Radiation and radioactive contamination control..."

Procedures maintained pursuant to the above requirements state that:

(HP 2.7.1) all radioactive materials outside of Building 235 shall be contained;

(HP 2.7.7) contaminated areas shall be posted as Contamination Control Zones;

(HP 3.1) periodic radiation surveys will be conducted in those areas where radiation exists;

(HP 3.2.6) periodic smear sampling and counting will be conducted in those areas where radioactive contamination exists.

The inspector noted that conservative action levels were contained in these procedures so as to achieve timely implementation of the regulations referenced in Paragraph 4, for example the determination of a surface smear exceeding 200 dpm/100 cm^2 ($\sim$ 0.1 μCi/100 cm^2) of beta-gamma radio-activity required followup if the area was not a posted Contamination Control Zone.

Based on the description in the proceeding paragraphs, it appears that, contrary to the above requirements, on August 28, 1979, there were:

a.   A two millicurie quantity of Co-60 outside of Building 235 that was not contained. Survey records of this area did not identify this material, including a survey performed on September 28, 1979.

b.   Six contaminated areas each containing in excess of 10 microcuries of Co-60 that were not posted as Contamination Control Zones.

c.   Six contaminated areas each containing in excess of 10 microcuries of Co-60 that were not known to the licensee because of inadequate surveys to determine the presence of such quantities of Co-60.

The inspector discussed the above items with licensee representatives and indicated that apparent licensee failure to adequately utilize procedures established for the control of radiation and radioactive contamination appeared to have prevented the timely discovery of the above contaminated areas, thereby resulting in the apparent Item of Noncompliance discussed in paragraph 4. The inspector indicated licensee adherence to radiation and contamination control procedures would be reviewed during a subsequent inspection in conjunction with the above apparent Item of Noncompliance.

6.   Fuel Storage and Transfer Facilities

The inspector observed the facilities maintained for the storage of new fuel and for the storage, cropping, and transfer of spent fuel into shipping containers for removal from the facility. The inspector surveyed those areas.

The licensee representative stated that radioactive materials posting will be maintained in fuel storage areas wherever fuel is present.

No problems were identified.

7. Surveillance Tests

Part of the inspection effort was to review the records of selected surveillance tests that are required by the Technical Specifications (TS) including: confinement testing (TS 5.1), primary system relief valve testing (TS 5.2), reactor shim and control rod testing (TS 5.4), various engineered safety system tests (TS 5.5), primary coolant monitor tests and sampling (TS 5.6), emergency system surveillance (TS 5.7), radiation monitor surveillance (TS 5.8), and environmental monitoring (TS 5.10).

The inspector noted that the licensee maintained a system of scheduling and auditing to assure the timely completion of surveillance tests.

No overdue tests were identified.

8. Personnel Monitoring Program

a. External Exposures

Part of the inspection effort was to observe the personnel monitoring practices and to review the exposure records for the year 1978 and the year 1979 up to August. Less than half of the badge exposures were taken to be reactor-related exposures and the remainder were accelerator-related or other exposures. Records are maintained of whole body and extremity doses. Neutron dosimeters are provided to several individuals. Form NRC-5s are maintained.

Review of these records did not identify any omissions, numeric errors or overexposures.

Review of the licensee's reports to the NRC did not identify any errors in personnel exposure information.

b. Internal Exposures

The inspector reviewed records of urinalyses and whole body counts and observed the controls maintained by the licensee to limit the uptake of radioactive materials.

The inspector reviewed the records of thirty-six individuals involved in the cleanup of radioactive contamination who had received subsequent whole body counts. Personnel who performed groundskeeping activities

in the areas where contamination was found were also being identified
and included in the bioassay program. No uptakes of radioactive
materials were identified.

No uptakes of radioactive materials were identified by review of
selected records for 1978 and 1979, and the licensee representative
stated there were no detectable uptakes indicated by any of the
whole body counts performed during this period.

Review of selected licensee records of bioassays showed that exposures
were calculated based on the indicated uptake of tritium by the
individual, and added to his dosimetry results in examples where
tritium was detected, none of these additions being as great as 10
mrem whole body dose due to tritium uptake by any individual for a
single week or 100 mrem for a calendar quarter.

9. Procedures

Review of the availability of procedures and the administrative control
of procedures did not identify any problems in either the reactor operations
or the radiation protection procedures. The inspector noted that the
licensee's review and auditing of procedures was documented.

Paragraph 5 documented an apparent problem involving radiation and radio-
active material control procedures.

10. Effluent and Radwaste Controls

a. Gaseous and liquid process effluents

The inspector reviewed selected records of gaseous and liquid effluent
releases over the period January 1 - September 30, 1979, to determine
compliance with requirements of 10 CFR 20.106 "Radioactivity in
effluents to unrestricted areas," 10 CFR 20.301 "Waste disposal - general
requirements," and 10 CFR 20.303 "Disposal by release into sanitary
sewerage systems."

The inspector noted that none of these records indicated that any
releases were made that exceeded any regulatory limit.

b. Solid radwaste

The inspector surveyed the waste annex and observed the waste handling
equipment.

One drum of compacted radwaste was upended and then righted and
opened to check for any liquid. The contents appeared to be dry.

The review of radioactive waste shipping record #9055, dated December 29, 1978, showed it contained the original instead of the decayed source strength, 4.8 Ci instead of 0.3 Ci of Ce-144. This material was possessed under Byproduct Materials License No. 08-566-5.

The licensee had previously identified the need for correction but had not yet corrected the shipping record (which was corrected during the inspection).

The follow up to Bulletin 79-19 is scheduled on the next routine inspection of reactor radiation protection. This will include a review of procedures and quality assurance maintained by the licensee in shipping radioactive materials.

11. Reactor Status

The licensee had discovered a shim control element was rubbing and the reactor was shutdown prior to the inspection. The licensee evaluation indicated a high probability that the shim element had distorted or swollen and required replacement. This was not accomplished during the inspection.

The reactor remained shutdown and defueled during the inspection.

The licensee stated that the outage might exceed 4 months and the startup will be observed by an individual qualified to requalify operators at that time (Chief or Deputy Chief Nuclear Engineer).

12. Operator Training and Requalification

Review of operator training schedules and examination scores indicated that the licensed Reactor Operators and Senior Reactor Operators had completed the requirements scheduled during March, 1979 and none were overdue at this time.

13. Staff Changes

a. Operating Personnel

One operator terminated during September, 1979, and his replacement was not announced by the completion of the inspection.

b. Health Physics Personnel

One health physicist was transferred from reactor health physics to LINAC health physics. A previous health physics employee was rehired as a health physicist for the reactor.

The titles of the Section Chief and the Reactor Health Physics Supervisor were changed to Supervisory Health Physicists.
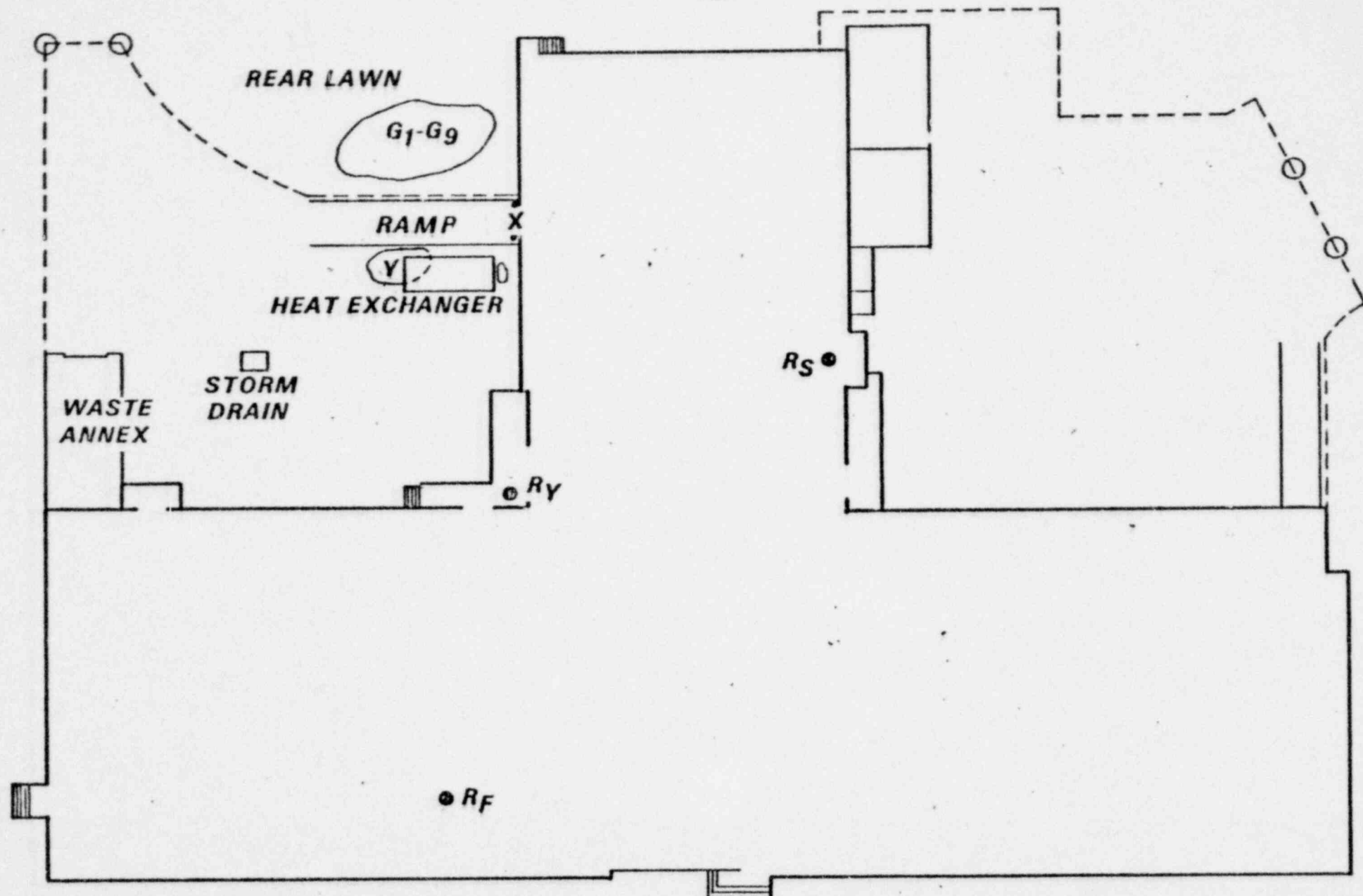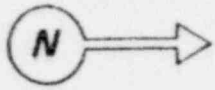
No items of noncompliance were identified involving organization and staffing.

14. Exit Interview

The inspector met with the licensee representatives (denoted in Paragraph 1) at the conclusion of the inspection.

The inspector reviewed the scope and findings of the inspection.

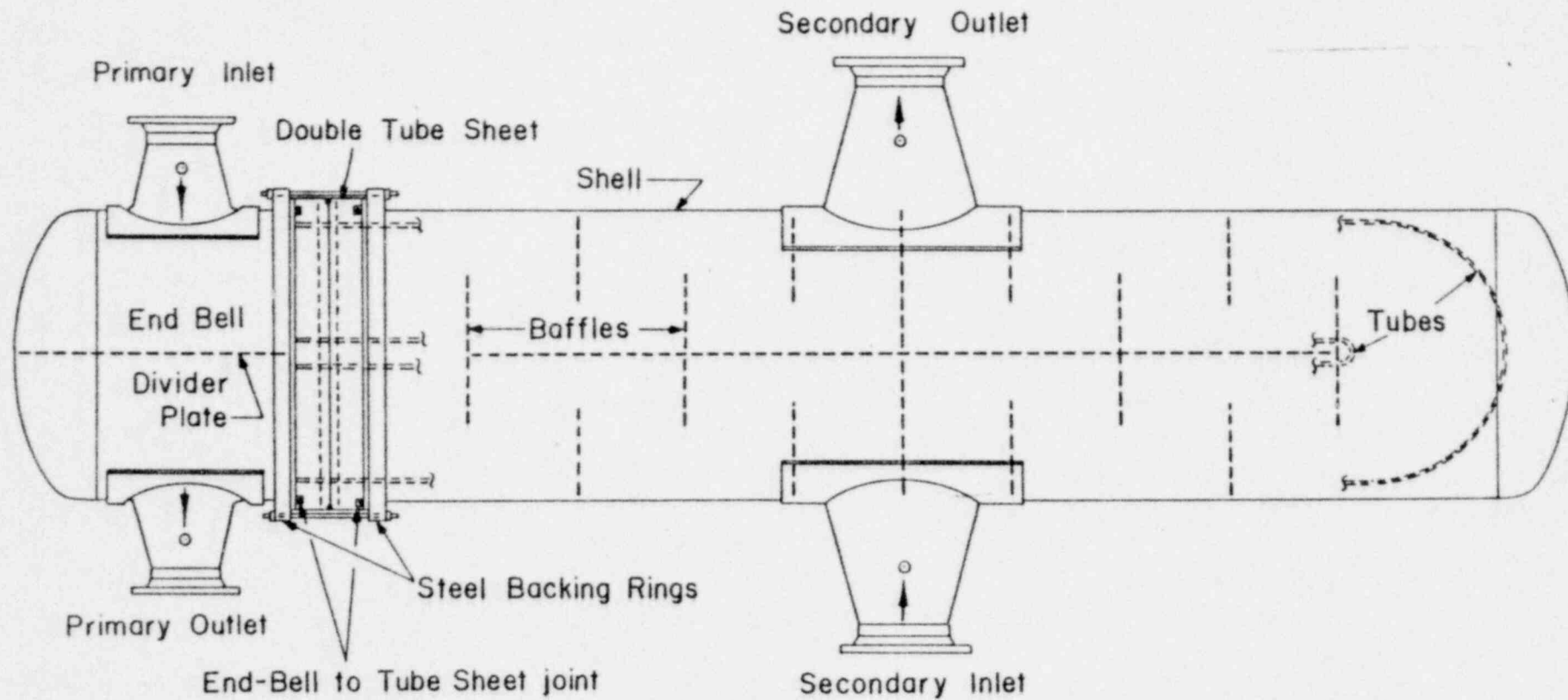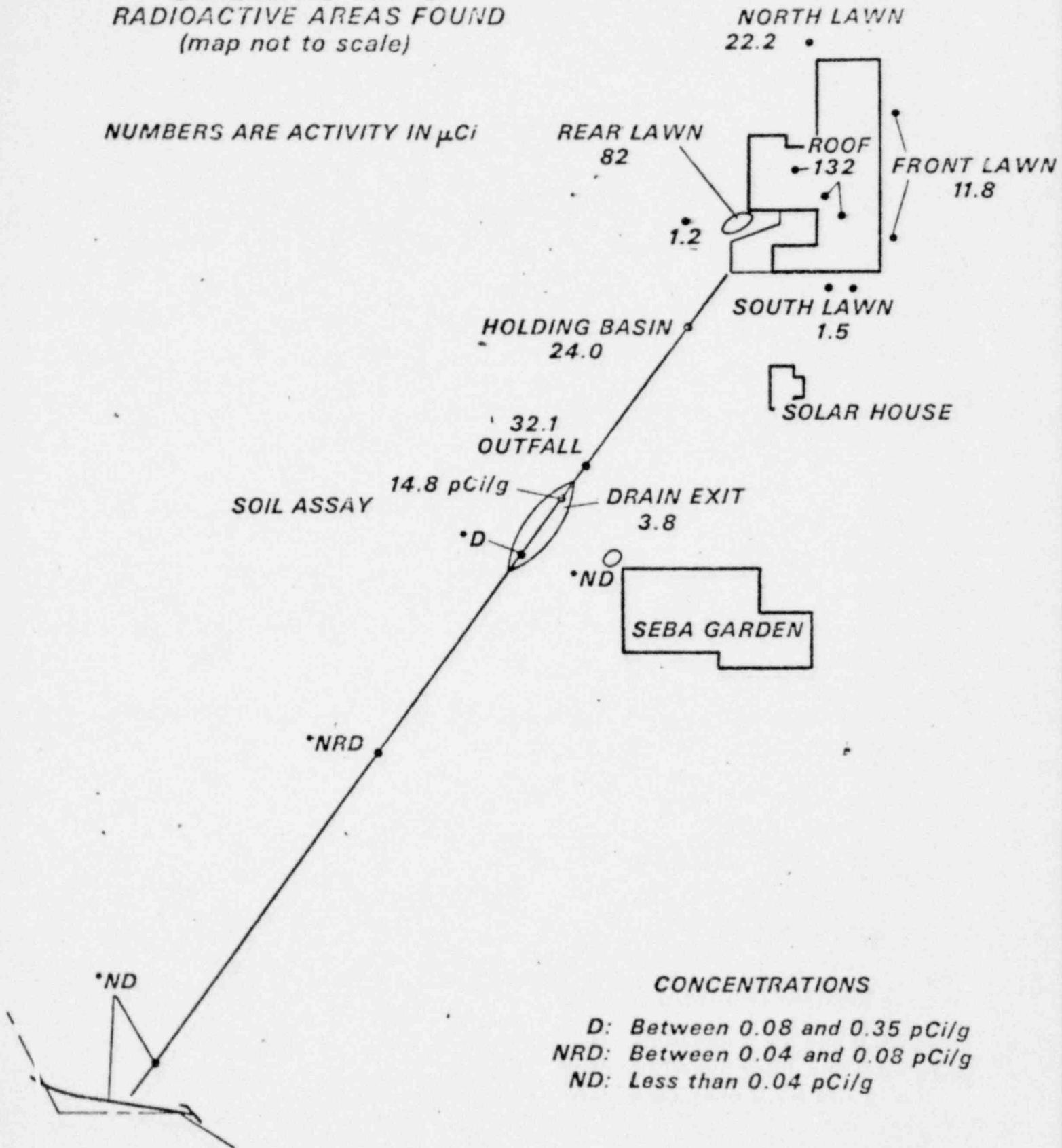The licensee's report dated October 26, 1979, was reviewed.

REAR LAWN

G_1-G_9

RAMP

X

Y

HEAT EXCHANGER

STORM
DRAIN

WASTE
ANNEX

R_Y

N

R_S

R_F

G_F

G

REACTOR BUILDING SITE
ATTACHMENT 1

Primary Inlet

Double Tube Sheet

Secondary Outlet

Shell

End Bell

Baffles

Tubes

Divider Plate

Primary Outlet

Steel Backing Rings

End-Bell to Tube Sheet joint

Secondary Inlet

ATTACHMENT 2

SCHEMATIC MAP OF
RADIOACTIVE AREAS FOUND
(map not to scale)

NUMBERS ARE ACTIVITY IN μCi

NORTH LAWN
22.2

REAR LAWN
82

ROOF
132

FRONT LAWN
11.8

1.2

HOLDING BASIN
24.0

SOUTH LAWN
1.5

SOLAR HOUSE

32.1
OUTFALL

14.8 pCi/g

SOIL ASSAY

•D

DRAIN EXIT
3.8

•ND

SEBA GARDEN

•NRD

•ND

CONCENTRATIONS

D: Between 0.08 and 0.35 pCi/g
NRD: Between 0.04 and 0.08 pCi/g
ND: Less than 0.04 pCi/g

ATTACHMENT 3

Table I

Detailed Assay Results: Active Samples

Exterior to Enclosed Area

Sample	Activity, μ Ci	σ (%)	Sample	Activity, μ Ci	σ (%)
G_F	11.5	2.6	D_{10}^A	2.4	
G_1	2.7		D_{10}^B	0.03	14.5
G_2	12.4	2.5	D_{10}^C	0.2	4.8
G_3	24.5	2.0	D_{10}^D	0.17	5.6
G_4	3.7	4.4	D_{10}^E	0.05	11.0
G_5	0.9	8.8	D_{10}^F	0.12	6.5
G_6	4.1	4.2	D_{10}^G	0.14	6.0
G_7	0.2	4.5	D_{10}^H	0.08	7.8
G_8	1.4	7.1	D_{10}^I	0.1	6.7
G_9	22.3	2.0	D_{10}^J	0.04	11.9
G_N	22.2	1.8	D_{10}^K	0.09	7.4
R_F	8.7		D_{10}^L	0.04	10.7
$R_1 y$	28.4	1.8	D_{10}^M	0.03	13.5
R_S	1.9		D_{10}^N	0.10	7.0
R_S^2	47.0	1.5	D_{10}^O	0.2	4.9
R_S^3	46.4		D_{10}^P	0.008	27.4
D_1	10.0				
D_2	24.0	1.9			
D_3	0.8	9.1			
D_4	3.3	4.6			
D_5	18.0	2.2			

Standard deviation expresses only counting uncertainty

Table II

Activity of "Beyond Drain Exit" Samples

(Units are pCi/g)

Sample	^{137}Cs	^{40}K	^{60}Co
D_{11}^{A}	1.72 ± 0.066	0.77 ± 0.0045	14.8 ± 0.23
D_{11}^{B}	0.474 ± 0.028	1.3 ± 0.056	D
D_{11}^{C}	0.559 ± 0.031	1.52 ± 0.061	NRD
D_{11}^{D}	0.263 ± 0.015	2.27 ± 0.048	ND
D_{11}^{E}	NRD	1.68 ± 0.045	ND
Outfall Grass	D	1.38 ± 0.084	NRD
SEBA Garden	0.286 ± 0.023	2.45 ± 0.067	ND

Notes:

Standard deviation includes only counting statistics.

ND = Not detected

NRD = Not reliably detected

D = Can be reliably reported as "detected", but quantification to 10% is not possible

Copy 5
10/26/79

Report on the Nature and Source of

Radioactive Material Found at the

NBS Reactor Site

Prepared by the

Ad Hoc Committee on Radioactive Material

Table of Contents

Chapter 1

Results of the Survey to Find

and Remove Radioactive Material

<u>Results of the Survey to Find</u>
<u>and Remove Radioactive Material</u>

I.   <u>Introduction</u>

On August 28, 1979, an inspector from the Nuclear Regulatory Commission (NRC) came to NBS on a routine, unannounced inspection of the NBS Reactor (NBSR) Health Physics. As part of the inspection, he conducted a routine survey of the area around the NBSR. The inspector's survey instrument was a Ludlum model 16 survey meter (0 to 500 cpm, ranges x 1,000, x 100, x 10, x 1; he was on the x 10 scale, or 0 to 5000 cpm, on the recollection of NBS personnel, and the ambient background was about mid scale) with a Ludlum model 44-2 probe, which has a 1" x 1" NaI(Tℓ) detector. This survey meter probe, thus, is sensitive only to photons above about 100 keV; it reads counts per minute and would not read in mR/hr unless it was calibrated with the same photon energy as that measured. Using this sensitive detector he found a spot of radioactivity on the lawn at the front of the reactor. In cooperation with NBS personnel, he subsequently found other spots of radiation in the rear of the reactor adjacent to a fence enclosing a controlled area in which a heat exchanger that was removed from the reactor in 1974 was stored, and a more extended spot under and beside the inactive heat exchanger. Further investigation led to the detection of two more spots on the roof of the reactor building. Samples taken from these radioactive spots were taken into the NBS laboratories and assayed with analytical equipment (see Sec. 2.3 for a description of the equipment used). All the radioactivity was shown to come from ^{60}Co. As a result of these initial finds, a thorough survey of the NBS grounds was begun immediately, along with other investigations.

On August 31, Dr. Ernest Ambler, NBS, Director, appointed an <u>ad hoc</u> committee consisting of R. S. Carter, A. Schwebel, K. Bell, and E. Passaglia (chairman) to:

1)   "Track the complete survey which is now in progress so as to insure that such a survey includes a careful examination of the Bureau's grounds, relevant equipment and the shoes of those NBS employees who may have walked in the area of the nuclear reactor.

2)   ..."undertake a complete review of the Bureau's safety and operating procedures with respect to ionizing radiation in general and radioactivity in particular...and to suggest...revisions...as may be needed...to prevent future occurrences of this nature."

Implicit in Dr. Ambler's assignment is a directive to find the origin of the radioactive material, and to find how it got to where it did. Accordingly, the ad-hoc committee took the following to be its tasks:

1)   Carry out a survey of the total Bureau grounds to define the limits of the spread of the radioactive material and to clear up any radioactivity found.

2)   To seek the source of the radioactive material.

3)   To determine how the material got to where it was found.

4)   To recommend procedures to prevent any recurrence.

This Chapter is the report on the first of these talks. tasks

2.   The Surveys Carried Out

2.1   Introduction

It is important to recognize that three surveys were carried out. These were

1)   A site survey. This was made to define the limits of the radio-activity and to determine if any hazard was posed to the workers of NBS and the citizens of the surrounding areas. This survey was completed by September 4, 1979. All radioactive spots found in this survey were removed and cleaned up by August 31, 1979.

2)   A survey of the streams external to the NBS site. This was carried out with an inspector from the Maryland Department of Health, and will be called the "Off Site Survey." This survey was carried out on September 7, 1979. The objectives were the same as in (1).

3)   A monitoring survey of the area immediately surrounding the reactor building. This survey was completed on September 11, 1979, and all radioactive spots were removed by that date. This was carried out with very sensitive instruments (see below, Sec. 2.3) and had two purposes: (a) to discover any extremely low-level radioactive spots that might have been missed in the site survey and (b) to assure that no further radioactive material was spread during investigations carried out in the enclosed area.

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This report will describe the results of all three of these surveys.

2.2 Definitions

Four terms will be used in describing the results of the survey. They are:

1) "Area". This denotes the general region of the survey, e.g., "reactor roof," "storm drain," etc.

2) "Spot". This denotes a place where radioactive ^{60}Co was found. It might denote a very localized region or a more extended region. In every case, the activity was dug up and removed to a safe storage area.

3) "Sample". This denotes a sample (usually a clump of dirt, mud, or sand), removed from a spot. A spot might yield one or more samples.

4) "Particles". The cause of the ^{60}Co radiation was small metallic particles (from fractions of a microgram to as high as 300 micrograms). Hence "particles" refers to radioactive particles recovered from the samples (often a tedious process). A "sample" might contain one or more particles.

2.3 Equipment

This section describes the instruments used in the survey, and those used in assaying the level of radioactivity in the samples taken to the laboratory.

The survey instruments used were of several types. The most sensitive instrument used was a Nuclear Enterprises Model MARK IV Scintillometer, which uses a 3" x 3" NaI(Tℓ) detector. This was used for the monitoring survey. It has a low range of 0 to 0.025 mR/hr, with additional ranges of 0 to 0.1, 0.25, 1.0, and 2.5 mR/hr. A variable time constant switch is provided; for all the surveys, it was used on the most rapid, i.e., 0.25 seconds. This gave an indication which fluctuated rapidly, but quickly showed any increase in ambient backgrounds. An estimate of the minimum detectable activity is 20 percent above ambient, which was approximately 0.005 mR/hr. There is no audio output with this instrument.

Other survey instruments included 1" x 1" NaI(Tℓ) probes and thin end-window G-M probes. The scintillators were less useful as no audio output was available and the sensitivity was no greater than with the inspector's detector. The G-M devices were used primarily for pinpointing radioactive spots when a more sensitive, but less directional instrument showed a high reading. The G-M meters were useful in the initial stages of the survey, as the radioactive spots were enough above ambient background that responses

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from the G-M instrument were positive. The scintillator and the G-M probes were used with survey meters that have a scale of 0 to 0.2 mR/hr., with ranges of x 1, x 10, and x 100. Ambient background with these was also 0.005 mR/hr., with wide fluctuations, since both are event counters.

All surveys were conducted by experienced Health Physics or Reactor Operations personnel.

The assays were carried out on equipment of the Analytical Chemistry Activation Analysis Group by personnel of that Group for particle assays and Health Physics personnel for sample assays. Various detectors and multi-channel analyzers (MCA) were used for qualifications and quantification of activity. The detectors were, for nearly all assays, Ge(Li) from 63 cc to 80 cc. The MCA's included several older Nuclear-data units, including the Health Physics Model ND4410, and the multi-user minicomputer-based ND 6600. The Low Energy Photon Spectrometer (LEPS) was used for typifying metal particles, and for screening those which could profitably be sent to the X-ray Fluorescence group. The 63 cc Ge(Li) and ND-3, a model ND 100 MCA, were used for quantifying the ^{60}Co activity in a sample. For samples of greater than about 1 µCi, an aluminum plate was mounted at 1.43 m above the open top detector shield. For samples of between 0.01 µCi and 1 µCi, a distance of 0.32 m separated detector and sample. Total activity per sample was measured, as sample geometries varied. Lower level samples were counted directly on the detector in a plastic bottle, with activity per unit mass reported. The minimum activity which can be detected, with a 95 percent confidence level, is 0.00008 µCi for soil samples and 0.00005 µCi for water samples. This limit is based on the work of Lloyd Currie, as reported in Analytical Chemistry, 40, 586 (1968).

2.4 Site Survey

2.4.1 Description of the Reactor Site

In order to make clear the topography of the area and the location of the radioactivity found, it is necessary to give a description of the reactor site. A plan view of the reactor building and the immediately adjacent site is shown in Fig. 1. To the rear of the building and at the left on the diagram is a fenced in area restricted to authorized personnel. In the south east corner of this area is an entrance to the annex to the reactor building used to store low-level radioactive waste from a'l of NBS prior to shipping. This area contains the heat exchanger removed from the reactor in 1974. It was stored beside a ramp that leads into the reactor building. This area will be referred to as

the "Enclosed Area." This Enclosed Area is paved with concrete and asphalt and contains a storm drain. The pipe from this storm drain runs in a south-westerly direction. Approximately 60 feet from the enclosed area(150' from the storm drain), there is an access vent to this storm drain. This will be called the "Holding Basin." Approximately 375' from the enclosed area, the storm drain exits onto the NBS field. This will be called the "Outfall." The NBS site boundary is approximately 310 yards from the Outfall. About 10 yards from the site boundary there is a marshy area and a small stream. A topographic drawing of this site is shown in Fig. 2. An aspect of the survey was to test the hypothesis that birds were the mechanism of transfer. The Committee believes that the results of this survey have discounted this theory.

2.4.2   Site Survey Strategy

The objective of the site survey was to define the limits of the radioactive material and to determine if any hazard was posed to the workers of NBS and the citizens of the surrounding areas. With this in mind, the following areas were checked. Each will be described in later sections.

1)   The area immediately adjacent to the reactor building, including the enclosed area and the lawns around the reactor building.

2)   The Holding Basin, the Outfall, and the area between the Outfall and the site boundary.

3)   Lawn equipment and storage areas for equipment.

4)   Shoes of NBS plant workers who may have had access to the area around the reactor building.

5)   The total length of the NBS site boundary fence.

6)   The solar house, including interior and bird nesting area.

7)   Building 245 (Radiation Physics) including roof and interior.

8)   Trees south of South Drive.

9)   SEBA garden

10)   Lawn immediately adjacent to all NBS buildings, building roofs, and all trees near buildings.

11)   The grove of woods north and north-east of the reactor building.

12)   Engineering Mechanics high bay area.

2.4.3   Results

For clarity of discussion, the results section will be separated into those areas that gave positive results and those that gave negative results.

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2.4.4 Positive Results Areas

All the areas that gave positive results are shown in Table 1. This table shows the areas, the number of radioactive spots in the area, the number of samples taken, and (when available) the number of particles from the samples. It is to be noted that all positive results were obtained from three locations:

1) The Enclosed Area.
2) The immed.ate vicinity of the reactor building and its roof.
3) The Holding Basin, the Outfall, and the draining area for about 150' beyond the Outfall.

All the samples external to the Enclosed Area were assayed in the laboratory. Details are in Table II. The samples from the Enclosed Area were not assayed, except for three particles.

While it is not possible to draw any general conclusions about radiation levels because of absorption by surrcunding material, for the worst case (i.e., in air) one microcurie corresponds to a radiation level of 0.00132 mR/hr at one meter (0.014 mR/hr at one foot).

2.4.4.1 Reactor Lawns

These are shown in Fig. 1. The front lawn of the reactor showed one spot (GF on map) (the original one found by the NRC inspector); the rear lawn adjacent to the enclosed area showed 9 spots (G_1-G_9); and one single spot was found on the north lawn (GN). Exhaustive survey as part of the monitoring survey found eleven more very low-level spots in the lawns. This will be described in the section under "monitoring survey."

2.4.4.2 Reactor Roof

The reactor roof yielded three spots of radioactivity. These are shown in Fig. 1. One of these spots (R_S, near the reactor stack) was broken into three samples, yielding a total of five samples. One particle was separated from R_F, R_S^1, and R_S^3. Each contained one particle. The other samples have not been separated.

2.4.4.3 Holding Basin

This yielded one spot and one sample. This sample has not been separated for particles.

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2.4.4.4 Outfall

This area and the next merit discussion as to their topography. Immediately at the point where the underground pipe that drains the Enclosed Area exits onto the NBS field, there is a pool approximately six feet in diameter. The area of this pool is denoted the "Outfall". This area yielded four spots and four samples. One of them (D_1) was separated to yield one particle.

2.4.4.5 Drain Exit and Beyond

Diametrically opposite to the exit of the pipe described in the previous section, and across the small pool, the terrain rises slightly then falls away gradually to the site fence approximately 310 yards away. For a distance of about 150' from the Outfall a region of increased activity was found in a shallow channel several feet wide. This is denoted the "Drain Exit Spot". At sixteen points in this Spot, the activity was somewhat higher than in the immediately adjacent vicinity. These yielded samples D_{10}^A through D_{10}^P. One of these (D_{10}^A) was separated to yield a particle. The total activity of radioactive cobalt in these samples was low, being only 3.8 μ Ci.

In addition to these more active samples, seven more samples were taken for laboratory assay. One (D_{11}^A) was 50 feet from the Outfall (i.e., from the beginning of the drain exit spot) and the other (D_{11}^B) 130' from the Outfall (i.e., twenty feet from the end of the spot). These samples did not show any increased activity over the immediate vicinity and were taken in order to estimate the general level of radioactive ^{60}Co of the spot and how it varied with distance from the Outfall. Midway between the Outfall and the site boundary (i.e., about 150 yards from the Outfall) sample D_{11}^C was taken, and at the edge of the stream (about 20 yards from the site boundary) sample D_{11}^D was taken. A sample of sediment from the stream was taken and denoted D_{11}^E. These D_{11} samples are collectively called "Beyond Drain Exit" despite the fact that two of them were from the Drain Exit Spot.

One sample of grass was taken adjacent to the Outfall, and a soil sample was taken from the corner of the SEBA garden nearest to the Drain Exit Spot. The activity of all these samples is discussed in Sec. 2.4.4.7.

2.4.4.6 Enclosed Area

The Enclosed Area contains the heat exchanger that was removed from the reactor in 1974 and a large number of concrete blocks. A rope approximately 10 feet from the heat exchanger defined the 2.5 mR/hr. line. The radiation

came principally from the heat exchanger, and this masked that coming from the spot beneath it. All these materials were exhaustively tested. The heat exchanger was exhaustively swabbed, and all the concrete blocks were surveyed. The only spot of radioactivity that was found was under and adjacent to the front end of the heat exchanger. This spot was about 6' x 25'. A large amount of sand was in this spot. All the sand and radioactive material was removed and put into barrels. The heat exchanger was then moved, and a large number of pieces of asphalt that showed activity were chipped out of the ground and stored in a barrel. The total activity of the samples recovered has been determined to be 2 ± 0.25 mCi. The Enclosed Area is and has been restricted to authorized personnel. All isolated spots of activity have been removed. There is, at yet, a general area of asphalt beside the ramp which shows up to four times background with no definable hot spots.

2.4.4.7 Samples Beyond Drain Exit

As described in Sec. 2.4.4.5, these are samples D_{11}^A through D_{11}^E, the sample of grass at the Outfall, and a soil sample at the corner of the SEBA garden. The activity of these samples followed a consistent pattern. The results are given in Table III. The first sample, D_{11}^A, was 50' from the outfall. It showed an activity of 14.8 ± 0.23 pCi/g. Radioactivity from ^{60}Co could be detected in sample D_{11}^B, (130' from the Outfall) but its level was not high enough to permit quantification with an accuracy of $\pm 10\%$*. Radioactivity from ^{60}Co in sample D_{11}^C (150 yards from the Outfall) could not be reliably detected. The final two samples (D_{11}^D and D_{11}^E) showed no detectable level of ^{60}Co activity.

The sample of grass taken from the Outfall showed no reliably detectable ^{60}Co activity, and the soil sample from the edge of the SEBA garden showed no detectable activity.

The significance of these results merits some discussion. First there is an obvious decrease with distance. While at the beginning of the Drain Exit Spot the ^{60}Co activity is nine times the average ^{40}K activity, toward the end of the spot it has decreased to about 10% of the ^{40}K, and near the site boundary is a maximum of 1/50 the ^{40}K level. Second, it is instructive to compare these concentration with the maximum permissible concentrations (MPC) regulations for

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*See Table III for the quantitative definition of the terms "detected", "not reliably detected" and "not detected".

^{60}Co of the NRC. Regulations for MPC do not exist for soil. For water they are 50 pCi/ml for soluble cobalt and 30 pCi/ml for insoluble. Equating one ml. of water with one gram of soil, the activity of D_{11}^A, the most active sample, has a concentration of about 1/3 the MPC for ^{60}Co in water. The minimum detectable concentration is about 1/1000 the MPC concentration. Samples D_{11}^D and D_{11}^E are at or below this minimum detectable concentration.

2.4.5   Negative Result Areas

All areas were checked with various of the instruments described in Sec. 2.3. All the surveys were conducted by experienced personnel. A positive result (namely, the location of a radioactive spot) was taken to be when the reading on the survey meter in use rose to at least 20 percent above background and then decreased to background as the detector was passed over the spot.

The lawn equipment survey was made on all the lawn equipment of NBS, and on the shoes of all NBS plant workers who might have had an opportunity to work on the grounds near the reactor. The NBS site fence was surveyed by two teams of two workers each. One team went clockwise for a full circuit, and one team counter clockwise. In each team, one man was on the outside of the fence and one on the inside. The solar house and building 245 were thoroughly checked inside and outside, paying special attention to bird nesting areas. The area under all the trees south of South Drive were checked, as was the entire grove of woods north and north east of the NBSR. The areas around all NBS buildings and their roofs were checked, as were the areas under all trees near the buildings. The SEBA garden was checked, and the road exterior to the NBS site at the gates. Particular attention was paid to the drainage area from the active spot from beyond the Outfall to the site boundary. From this area, water, soil, and drain sediment samples were taken for laboratory analysis. (See section 2.4.4.7 for results). A survey was made of the high bay area in the Engineering Mechanics Building, since trucks used for shipping spent fuel stop in this area to have fire shields removed.

Particular mention should be made of the fact that in the site survey, all radioactive spots around the NBSR building were limited to 35' of the building. (During the monitoring survey, one low level spot was found 75' from the building, as discussed in the next section.) A thorough survey to somewhat beyond 200' of the reactor building did not locate any further radioactivity in this area, except, of course, that associated with the drain system for the Enclosed Area.

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Three samples of soil were taken for laboratory assay from the region beyond the rise South and East of the NBSR. These three samples (denoted O_1, O_2 and O_3) showed no ^{60}Co at the minimum detection level of 0.040 pCi/gm.

2.5 The Monitoring Survey

The results of the site survey described in the previous section were completed on September 4, 1979. However, since extensive work was being carried out in the Enclosed Area (primarily associated with moving and opening of the heat exchanger), and to assure that any radioactive spots that might have been missed during the site survey were found, the area around the NBSR was continuously monitored. This was done with the first of the instruments described in Section 2.3, which is considerably more sensitive than any of the other survey meters used in the site survey. Use of this instrument detected eleven more spots of radiation, plus some residual radiation at two of the spots previously found in the site survey. The assay results from these spots (one sample per spot only was necessary) are given in Table IV. Seven ($G_{12}^1 - G_{12}^9$, including two at previously found spots G_2 and G_5) of these new finds were on the rear lawn near the reactor near the previous spots in this area, two were on the south lawn (G_{13}^2 and G_{13}^3), one on the front lawn (G_{12}^F), and one across the road from the fenced Enclosed Area (G_{13}^1). This spot was 75' from the reactor, which is the farthest of any spot except those associated with the Storm Drain (see Sec. 2.4.4.3, to 2.4.4.5 and 2.4.4.7). As can be seen from Table IV, these spots were very low level, the maximum being only 2.1 µCi. The total amount in these spots is 13 µCi.

Two random samples of soil were taken from the reactor lawn behind and in front of the NBSR. The results are discussed in Sec. 2.8.

2.6 Sample Assay Results

The detailed results of the assay of the samples exterior to the enclosed area are given in Tables II and V. The total activity of the "lawn" samples is 118.9 µCi including those from the monitoring survey. For the roof samples it is 132.4 µCi, and for the drain samples it is 59.9 µCi. Hence the total activity outside the enclosed area is 311 µCi. The most intensely radioactive spot was R_S^2 on the roof, with an activity of 47.0 µCi. On the ground, the most active was G_3, from the lawn behind the reactor, with an activity of 24.5 µCi. This most active spot would produce a maximum radiation level of 0.34 mR/hr at one foot for the source in air. The actual level measured from the spot in the ground was about 0.2 mR/hr.

The description, analysis, and source of the particles recovered from the samples is the subject of the second chapter of this report. The isolation of particles from the samples is a laborious and tedious task. As a result only a small number of particles have been isolated. These are indicated in Table 1. The mass of the particles varies from fractions of a microgram to 300 µg.

2.7 Off Site Survey

The off site survey was conducted in cooperation with an inspector from the Maryland Department of Health. Two samples during this survey were taken on site, one each on the front and rear lawns of the NBSR, but not, of course, including any spots. The other samples were as follows:

Water Samples (each 500 ml.)
> East inlet to Izaak Walton Pond
> West inlet to Izaak Walton Pond
> Pavilion in Izaak Walton Park
> Exterior water sample from caretakers house
> The well of Mr. George Lees (at his request)
> Exit of NBS stream

Sediment Samples
> East and West inlets to Izaak Walton Pond
> NBS stream outside of site boundary.

While the site survey was being conducted, Mr. George Lees, who owns a house near the NBS site, approached the NBS employees carrying out the survey. He requested that his well water be tested. This request was, of course, honored.

All the water samples showed no detectable ^{60}Co activity at a level of at least a factor of three below the limit of detection of the equipment used, which is 0.1 pCi/ml at a 95% confidence level. The sediment samples showed no detectable activity with the limits shown in Table III. The NBSR front lawn sample showed no detectable ^{60}Co; the rear lawn had a detectable but no quantifiable amount of ^{60}Co.

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3. Conclusions

The radioactive material found on the Bureau grounds was determined to be of such a level and dispersion that it did not represent a health or safety hazard to the general public or to NBS employees. No radioactive material (beyond that occurring naturally) was found off the Bureau grounds. On the Bureau grounds, the most extreme area of activity found was still 780 feet from the site boundary, and concentration of material at this point was not high enough to permit a measurement of activity. More specifically, the conclusions drawn from the survey results are best described with reference to the schematic map shown in Fig. 3. This fi, shows the only sites of radioactive ^{60}Co found. It is evident from this map that these sites are immediately adjacent to the reactor building, at the holding basin, and at the exit of the storm drain. No other sites of ^{60}Co radioactivity have been found during the exhaustive survey conducted. This pattern indicates that the Enclosed Area was the source of radioactive material found outside that area. The drain from the Enclosed Area provides a natural means for this material to be transported from that area to the Holding Basin, the Outfall, and the area immediately beyond. The remainder of the radioactive materials was found in the immediate vicinity of the reactor building. The evidence therefore clearly indicates that the radioactive material was restricted to the immediate vicinity of the Enclosed Area, and several other areas that are connected to this one by natural means and in no other areas. There are two mechanisms that could account for the local spread of radioactive material from the enclosed areas. These are local swirling winds (which could lift particles onto the roof) or tracking by feet, or both. The Enclosed Area, as can be seen in Fig. 1, is a courtyard with open sides facing the south and west. The prevailing strong winds in this area are from the west, and this courtyard forms a natural trap for them. Swirling winds are in fact often observed in this area. The sand associated with the Enclosed Area spot (see Sect. 2.4.4.6) is in a location next to the loading ramp that forms a quiet nook in these swirling winds, and where loose material is expected to be deposited. No other evident mechanisms beyond wind and tracking are available to explain the localized spread of radioactive material.

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Chapter 2

The Analysis and Source of the Particles

The Analysis and Source of the Particles

I. Introduction

The objective of this phase of the investigation was to determine the source of the particles found in the survey described in the previous chapter. More specifically, the objective was to determine if the particles were formed in the reactor or had some other origin. The evidence on the location of the particles available from the survey, namely that they seemed to have been distributed from the spot under the heat exchanger by natural drainage and tracking and/or wind made it plausible to hypothesize that they were released from the heat exchanger when it was removed from service in 1974. However, the nature of the winds in the Enclosed Area is such that loose material collects at the location where the heat exchanger was stored. Indeed, the sand found under the heat exchanger very likely collected there by just this mechanism. Hence another (but less likely) hypothesis was that they were somehow released into the Enclosed Area from some other source, and then were collected under the heat exchanger by winds. Possible other sources considered were ^{60}Co radiation sources, or one of the trucks that came into the Enclosed Area to pick up radioactive waste. For this reason it became important to determine if the particles could have been formed in the reactor.

In order to make this determination it was, at a minimum, necessary to do the following:

a) Analyze the particles thoroughly as to composition and morphology.

b) Find components in the primary stream of the reactor that have the same composition as the particles. Because cobalt alloys are commonly used for corrosion and wear resistant surfaces in bearings, pumps and valves, attention was focused on these components in the reactor.

c) Develop at least a plausible explanation of how particles are formed from these components, and check this explanation if possible. A point of departure was the well-known fact that particles of various sizes and shapes are formed during the wear of metals, and the nature of the particles can be an indication of the nature of the wear process.

d) Determine the specific activity of the cobalt to see if it could have been irradiated to that level in the reactor core. At the same time, the activity of any other isotopes (e.g., Cr and Fe) when compared to the ^{60}Co activity, could give an indication of the age of the particles, i.e., the time since activation.

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In addition to this, strongly corroborative (if not conclusive) evidence would be available if particles could be located within the heat exchanger and/or the reactor and shown to be of the same nature and type as particles found outside.

With this in mind, the primary side of the heat exchanger was opened by removing the head (Fig. 4). Extremely fine, almost colloidal material showing ^{60}Co activity was found. Three particles were separated from this colloidal material. Two were found at the flange connecting the head to the body of the heat exchanger. One of these, found at the seven o'clock position when facing the heat exchanger was labelled HEX 7-1. In addition, two particles were found in the water of the pool directly under the reactor core. This pool is used to store the spent fuel elements prior to shipping. These particles were analyzed along with a number of representative particles from outside the heat exchanger. The analysis of these particles, analysis of pump components, and the specific activity of the particles are discussed in subsequent sections.

2. Chemical and Metallurgical Analysis

2.1 Particle Analysis

The particles analyzed and their source are shown in Table VI. As can be seen from this table, particles were obtained from samples from each of the principal areas in which radioactive spots were found: the Enclosed Area, the reactor lawn, the reactor roof, the Outfall and the Drain Exit Spot. In addition, the particle from the heat exchanger and one of the particles from the spent fuel element storage pool were analyzed.

The Report of Analysis of these particles is attached as Appendix A. A discussion of these results is given below in Sect. 2.3.

2.2 Wear Rings

Water is circulated through the primary circuit of the reactor by three centrifugal pumps. The impeller of these pumps contains a ring approximately nine inches in diameter whose convex surface mates against a concave surface consisting of another wear ring on the pump housing. The purpose of this assembly and pair of rings is to seal the pump chamber and to prevent water from flowing back into the intake side of the pump. This is not a normal load-bearing assembly; in normal operation there is a clearance of somewhat less than 1mm between these two surfaces. The pump specifications call for

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the impeller wear ring to be coated with a hard facing material, but not the
wear ring on the housing. Such an approach is common practice in wearing
situations. The manufacturer of these pumps (Allis-Chalmers) notified us
that the surface of the impeller wear ring was coated with a cobalt base wear
alloy by a flame spray process. Allis-Chalmers does not manufacture the wear
ring. Our pumps are 15 years old and the records of Allis-Chalmers do not
identify the supplier of the wear ring.

When the reactor was constructed, and during the testing phase while
still containing light water, one of these circulating pumps began running
rough, and was incipiently binding. It was determined that the piping to
which the pump was connected was not accurately aligned, causing the binding.
The pump was removed from service, and when disassembled it was found that
the wear rings were indeed heavily worn. This impeller wear ring was preserved,
but the mating wear ring was discarded. This series of events occurred in 1967.
Spare wear rings were purchased along with the pumps.

The worn and spare wear rings were extensively investigated. A photograph
of the worn impeller wear ring is shown in Fig. 5. The worn surface can be
readily seen. This surface is shown at higher magnification in Fig. 6. Other
pictures are given in Appendix A.

Sections were made of the worn wear ring and of a new wear ring. These
cross sections were extensively analyzed, and the analysis is reported in
Appendix A. A discussion of these results will be given below in Section 2.3.
Here we present some metallurgical observations on the wear ring.

Cross sections of both the new and the worn ring at a magnification of
6X are shown in Fig. 7. From these it can be seen that the wear surface
itself was deposited in a groove approximately 1mm deep in the body of the
wear ring. The worn impeller wear ring, as described in Appendix A, has a
composition that is high in cobalt, chromium and nickel, with regions of a
cobalt-chromium type of alloy. The Co-Cr-Ni phase has a composition very
close to Stellite alloy SF 6, while the other phase has a composition
corresponding to Stellite 157. Both these alloys are made for flame spraying.

*Stellite type alloys are frequently employed in wearing situations

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The microstructure of the spare impeller wear ring is unusual and bears out the results on the worn impeller wear ring. The compositions as described in Appendix A again are very nearly those of SF-6 (darker phase) and Stellite 157 (lighter phase). The dark spots are porosity. It appears that the ring was formed by flame spraying two different Stellite alloys, probably during the same operation, one powder from one side and the other powder from the other side. An expert on flame spraying contacted at the Stellite Corporation said he had never heard of this being done, and knew of no reason why it should be done. He ventured the opinion that the amount of porosity is excessive.

Although the wear mating ring from the housing was discarded a new one purchased at the same time as the pump was available. Analysis showed it to be 316 stainless steel with no coating on any surface.

Examination of the cross section of the worn impeller wear ring at high magnification (Fig. 8) leads to an interesting observation. There is a thin (10-20 μm) surface layer that appears to be a different material from the base material. It in fact appears to be material transferred from the opposing ring surface, as would occur during wear. This surface layer was analyzed extensively, and the results are given in Appendix B. The composition at a representative location is 39% Fe, 13.1% Co, 18.5% Cr and 25.7% Ni. The significance of these results is discussed below in Section 2.3.

There is also a very thin composite layer on the back side of the impeller wear ring. An analysis of this layer is given in Appendix A, Table 5. This layer is probably designed to prevent fretting or seizing between the wear ring and the surface on which it is mounted. It has no bearing on this investigation.

2.3 Discussion

The chemical analysis of the particles and the wear impeller ring as reported in Appendix A, and the metallurgical observations as presented in Section 3 of this chapter lead to the following set of observations:
1) The hard facing of the wear ring consists of stellite 157 (66% Co, 21% Cr, 4%W, 2% Fe, no Ni) and very likely stellite SF-6 (19% Cr, 14.5% Ni, 7.5% W, 55% Co). Because the latter is a multiphase alloy, the composition varies markedly from point to point. Significantly, the Fe content of all portions of this hard facing is very low (about 2%).

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2) The surface of the worn impeller wear ring is very variable in composition (Table 4, Appendix A; Table 1, Appendix B; Sec. 2.2 of this chapter). Significantly, this surface contains iron in significant amounts (up to 40%) along with Cr, Ni and Co as the principal constituents. The iron cannot have come from the alloys used to make the wear ring hard facing, since they contain low concentrations of iron ($\sim$ 2%).

3) Fig. 8 in Sec. 2.2 very clearly shows a layer on the surface of the wear ring that appears quite separate from the matrix. Indeed, in the process of putting hardness indentations in this layer, it separated by fracture from the matrix. This is clearly seen in Fig. 1, Appendix B. It is high in iron content, and contains Cr, Ni and Co as the main alloying constituents. (See Table 4, Appendix A, Table 1, Appendix B).

4) All the particles except D_{10}^A are hetereogeneous. They consist of a rather large flake of stainless steel with cobalt rich surface inclusions on one side. These inclusions have the composition of stellite 157, (lines E, Table 1, Appendix A) very nearly stellite SF-6 (see table II, line F, Appendix A), and the composition of the worn wear ring surface layer (in Appendix A, compare Table 1, line C, D, and Table 2, line C, D, with Table 4, lines A-H), noting that this layer is of variable composition. These particles also contain Al in sometimes significant quantities, and often as oxide rather than Al metal.

5) Particle D_{10}^A is a homogeneous particle with a composition corresponding to the surface of the worn impeller wear ring and spots on particles S_1 and HEX 7-1 when account is taken of the variability in these compositions.

To account for this series of observations, the committee fully subscribes to the conclusions given by the analysts in Appendix A. It further suggests the following as a reasonable explanation of the source of the particles. As the pump from which the impeller wear ring was recovered began to fail, severe abrasion between it and the mating surface began to occur. This severe abrasion between the wear surface and its mating 316 stainless steel surface caused the surface layer seen in Fig. 8. This severe abrasion caused surface alloying giving the very variable Fe-Co-Cr-Ni alloy found on the wear surface, and on regions of particles S_1 and HEX 7-1, and particle D_{10}^A. The particles were very likely formed as follows. Particle D_{10}^A, the very small homogeneous particle with the composition of the surface alloyed layer, very likely chipped off from the surface. As seen from Fig. 1, Appendix B, this

surface layer can break off easily. Particle D_{10}^A has the approximate dimensions of this surface layer. The remainder of the particles are hetereogeneous, consisting of a stainless steel flake with occlusions on one side consisting of material of compositions found in the worn impeller wear ring. These heteregeneous particles could have been formed in two different ways. The first way is that during the final stages of the failure of the pump, significant flakes of stainless steel were abraded from the mating surface. This clearly would form particles with the observed morphology, although the particles are rather large (HEX 7-1 is about 2mm long; see Fig. 4, Appendix A). An alternative explanation is as follows. During machining in manufacture, particles and small burrs are left on the machined surfaces. Such particles are often found in wear debris (A. W. Ruff, private communication). Any such particles or dislodged burrs ca. .ed through the region between the impeller wear ring and its mating wear ring could form heterogeneous particles with the morphology observed, including the scratches on the "back" side of the particles (cf Appendix A, page 2). The committee feels that probably both of these mechanisms operated.

These particles were then lodged in the reactor and activated by neutron radiation for varying lengths of time and in locations of varying neutron flux, as described in the next Section.

The above explanation explains all but the aluminum content of the particles. While this must be somewhat more hypothetical, the committee believes that there are at least two reasonable explanations for the high aluminum content of the particles. First, the piping of the reactor is aluminum, giving many possibilities for aluminum particles or corrosion products. Second, the corrosion of holes in the heat exchanger would have produced a significant amount of corrosion product which could deposit on the particles to provide the observed aluminum content. As already noted, this is very often the form of aluminum oxide. Third, a number of traverses of the particles through the tubes of the heat exchanger and the reactor piping could have provided the abrasion necessary to coat the particles with aluminum as observed.

The committee feels that the normal wear process of these wear rings would have caused the very fine, almost colloidal material found in the heat exchanger, and that the relatively large particles found in the heat exchanger and outside it were caused by the severe wear conditions that caused the failure of the wear ring.

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The next Section discusses the evidence from the activity of the particles.

3. Investigation of the Radioactivity of the Particles

3.1 Introduction

In order to determine whether or not the particles could have been formed in the reactor, it is necessary to determine their specific activity. The distribution of neutron flux and the maximum neutron flux in the reactor are known. If the particles were indeed released when the heat exchanger was removed, then they would have been in the reactor flux for a maximum of 1210 days. In addition, they would have decayed for five years (the heat exchanger was removed in August 1974). Hence the decay time (the "age") of the particles cannot be less than five years. To carry out this analysis it is necessary to measure the ^{60}Co specific activity, (for the formation) and that of one other isotope (for the age). For the second, ^{55}Fe was used.

To carry out this analysis, six particles were analyzed to determine their specific activity. They were G1, G2, 1F, D1, HEX 7-1, and D_{10}^A. Their γ-ray activities were entirely from decay of the 5.27 year half-life ^{60}Co isotope. Low energy x-rays are emitted subsequent to electron capture in the iron-55 isotope with a 2.7 year half-life, and other low-energy x-rays are generated by the fluorescence of other elements in the particle by the cobalt decay radiation. Only the ^{60}Co activity from the particles is significant from a radiation safety point of view.

3.2 Cobalt-60 Specific Activity

The activity of the six samples was determined using a Ge-Li detector γ-ray spectrometer calibrated with a standard cobalt source by the Activation Analysis Group in the Center for Analytical Chemistry and reported in Appendix C. The results are shown in the third column of Table VII. The mass of cobalt in each of the six particles was estimated from the electron microprobe analysis but could not be determined accurately because of the limited range of the probe electrons and the heterogeneous nature of most of the particles. Only D_{10}^A appears to be homogeneous and even there only half of the volume could be probed. (See Appendix A). Therefore, the cobalt mass was determined by neutron activation analysis. (See Appendix C). The facilities at Oak Ridge National Laboratory were used because the NBSR was shut down. The high activity already present from the decay of the ground state of ^{60}Co made it

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impractical to use the activation of ^{59}Co to the ground state of ^{60}Co to determine the mass of ^{59}Co present. Fortunately, however, about half of the neutrons absorbed by ^{59}Co activate an isomeric state of ^{60}Co which decays to the ground state with 10.5 minute half-life emitting a low energy γ-ray. This low energy γ-ray can be detected by a thin germanium detector which is relatively insensitive to the much higher energy γ-ray from the ground state of ^{60}Co. Thus, accurate determinations of the ^{59}Co present in each particle could be made and are given in the second column of Table VII. The specific activity of the cobalt has been calculated from the data in columns two and three. It is given in column four in ~~micro~~curies per gram of ^{59}Co and in column five in ~~micro~~curies per gram of total cobalt.

It is also possible to determine the ratio of the mass of ^{60}Co to ^{59}Co (N_{60}/N_{59}) (and hence the specific activity) directly because ^{60}Co itself can be activated to ^{61}Co which decays with a 99 minute half-life emitting a low energy γ-ray that can also be detected by the low energy detector. The specific activity in ~~micro~~curies per gram of total cobalt based on these direct measurements was determined for four of the particles and is shown in column six. The agreement is quite good except for D_{10}^A which is not visible to the naked eye because it is so small. This made it difficult to locate properly in the counting system resulting in a significant uncertainty in the value for the mass of ^{59}Co. Consequently, the value for the specific activity of D_{10}^A given in column six is more reliable because it relies only on ratios and uncertainties such as sample location in the counting system cancel out.

If the particles were removed from the reactor along with the heat exchanger, they must have been out of the neutron radiation field for at least five years. Their maximum time in the reactor flux would have been 1210 days. The ^{60}Co specific activity expressed as curies per gram of cobalt can be calculated for varying particle histories in the reactor, and for times out of the core. Two cases will be calculated. The appropriate equation is:

$$\frac{N_{60}}{N_{59}} = \sigma_{59} \, \phi\tau \, \frac{1-e^{-\lambda'\tau}}{\lambda'\tau} \, e^{-\lambda_{60}T}$$

τ = Irradiation time in reactor

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T = Decay time since removal from radiation field

σ_{59} = Neutron activation across section of ^{59}Co

ϕ = Neutron flux

λ_{60} = Decay constant for ^{60}Co

$\lambda' = \lambda_{60} - \sigma_{59} \phi + \sigma_{60} \phi$

σ_{60} = Neutron absorption across section for ^{60}Co

$$\text{Specific Activity} = \frac{1150}{1 + \frac{N_{60}}{N_{59}}} \cdot \frac{N_{60}}{N_{59}} \quad Ci/g$$

Case 1: Maximum activity that could be generated. This assumes that the particle was in the maximum flux in the reactor for the full 1210 days, and released when the heat exchanger was removed (five years ago; ^{60}Co half life of 5.27 years used for convenience).

$\phi = 1.9 \times 10^{14}$ n/cm^2-s

τ = 1210 days at 10 MW = 1.045×10^8 s.

T = 5.27 yrs (^{60}Co half-life so $e^{-\lambda_{60}T} = 1/2$)

Then $\dfrac{N_{60}}{N_{59}} = .312$

and Specific Activity = 342 Ci/g

Case 2: Particle circulated with primary cooling water until trapped in heat exchanger just before heat exchanger removal.

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$$\phi = 4 \times 10^{12} \, n/cm^2 s$$

$$\tau = 1210 \text{ days}$$

$$T = 5.27 \text{ yrs.}$$

Then $\dfrac{N_{60}}{N_{59}} = .00645$

and specific activity = 7.1 Ci/g

The experimentally determined activities are shown graphically in Figure 9 which also includes markers indicating the two cases evaluated above.

The above comparisons assume a decay time of about five years. (The Co-60 half-life of 5.27 years was selected for convenience in the above calculations).

3.3 Iron 55 Activity and the Age of the Particle

If the decay time were less than five years, the particles could not have been removed with the heat exchanger which was removed in August 1974. Therefore it is important to determine the decay time if possible. This gives the time the particle was out of the flux. This can be done if the specific activity of two different isotopes with appropriate half-lives can be determined. All the particles contain iron. One of the iron isotopes, Fe-54, activates to Fe-55 which decays with a 2.7 year half-life. This is a convenient half-life compared to 5.27 year Co-60 half-life. The Fe-55 decay, however, does not emit a γ-ray but decays by electron capture emitting a low energy x-ray. The specific activity requires knowledge not only of the activity but also of the mass of iron. Although all the particles contain iron, only one is homogeneous enough to provide a reliable iron mass determination. This is particle D_{10}^A. The micro-probe analysis of half its volume showed it to be homogeneous and this was confirmed by the neutron activation analysis of the ^{59}Co which agreed with the micro-probe results. Therefore, D_{10}^A was counted by the Nuclear Radiation Division and the ^{55}Fe activity determined. (See Appendix D). This was combined with the iron mass determination to give the specific activity in terms of the ratio of the number of ^{55}Fe nuclei to the ^{54}Fe nuclei (N_{55}/N_{54}). If we define:

$$R_{Co} = \frac{N_{60}}{N_{59}} \quad \text{and} \quad R_{Fe} = \frac{N_{55}}{N_{54}}$$

then the decay time T can be determined from the equation:

$$T = \frac{1}{\lambda_{55} - \lambda_{60}} \ln \left\{ \frac{\sigma_{54}}{\sigma_{59}} \frac{R_{Co}(T)}{R_{Fe}(T)} \frac{\dfrac{1 - e^{-\lambda'_{55}\tau}}{\lambda'_{55}\tau}}{\dfrac{1 - e^{-\lambda'_{60}\tau}}{\lambda'_{60}\tau}} \right\}$$

where the various symbols have the same meaning as given previously.

The term in the square brackets is a slowly varying function of τ and ϕ so the equation for T must be iterated with the equations for

the ratio of $\dfrac{N_{60}}{N_{59}}$ and $\dfrac{N_{55}}{N_{54}}$ to obtain a consistent set of T, τ and ϕ. When this

is done using the measured values of $R_{Co}(T)$ and $R_{Fe}(T)$, one obtains
T=4.5 years* with an estimated standard deviation of 15% and $\phi\tau =$
1.3×10^{22} n/cm^2

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*To determine this number exactly, the neutron absorption cross section of
Fe-55 which appears in the expression $\lambda'_{55} = \lambda_{55} - \sigma_{54}\phi + \sigma_{55}\phi$ is needed.
This cross section has not been measured, but an estimate can be made. The
^{55}Fe isotope is produced at ORNL. If the σ_{55} cross section were large enough
to effect the production rate, it would have been noticed. The cross sections
for all the other iron isotopes are small, being of the order of two barns.
Therefore, σ_{55} was assumed to be similar to the cross sections for the other
iron isotopes and taken equal to σ_{54} for convenience. The calculations are
not sensitive to the exact value.

3.4 Discussion

The age of particle D_{10}^A and the time at which the heat exchanger was removed are well within the one standard deviation error estimate assigned to the age of the particle. The fluence ($\phi\tau$) experienced by D_{10}^A is less than the maximum available (2.0 x 10^{22} n/cm^2-s) and the decay age shows that the particle left the radiation field at about the time the reactor was shut down for the heat exchanger removal.

Figure 9 shows that all of the particles could have been generated by irradiation in the NBSR. The two with the lowest specific activity, Dl and G2, could have been irradiated while circulating with the primary cooling water. The others must have been retained for various periods of time in higher flux regions of the reactor.

The large variation of the specific activities and the decay age of D_{10}^A show that the particles could not have resulted from the disintegration of a radioactive source left in the heat exchanger.

4. Conclusions

The investigations carried out demonstrate the following:

a) The compositions found on the particles is the same as that of various regions on the worn impeller wear ring.

b) The composition and morphology of the particles found outside is the same as one found in the heat exchanger and one found in the pool water.

c) The morphology of the particles shows that they could have been formed by the wear process that the wear ring underwent.

d) The flux in the reactor and the residence time in the flux can account for the specific activity of the particles.

e) The age of the one particle on which it was possible to measure the age is consistent with the time the heat exchanger has been removed from the reactor.

f) The wide distribution in particle specific activity shows that they did not come from a ^{60}Co radiation source.

The Committee considers this to be overwhelming evidence that the particles were formed in the reactor by the mechanisms described in this Chapter.

Chapter 3

Possibilities for Particle Release

Possibilities for Particle Release

I. Introduction

As described in the previous Chapter, the evidence that the radioactive particles were formed in the reactor is convincing. Any other origin for the particles is so exceedingly unlikely that its probability is negligible. Hence, since the particles were formed in the reactor, it becomes important to try to determine how they were released. Because they were found under the heat exchanger, and because the time that particle D_{10}^A was out of the radiation field corresponds closely to the time the heat exchanger has been removed from the reactor, the most likely time for the release of the particles was when the heat exchanger was removed. The only other components that are removed from the reactor primary system which might contain significant amounts of particulate activity are the fuel elements and the pre-filters in the water purification system. The fuel elements are sealed in a shipping cask before placing on the shipping truck. The cask and truck are carefully surveyed before leaving the building. Although the loaded shipping truck passes through the enclosed area it does not stop there. It does, however, stop at the Engineering Mechanics Building where a crane is used to place a fire shield over the cask. No trace of activity has ever been found in or near the Engineering Mechanics Building. The filters, after being allowed to decay for a long period of time are packaged and removed directly to the waste storage building. They are never stored in the Enclosed Area, although they pass through it on the way to storage.

Hence, it is most likely that the particles were released during the removal of the heat exchanger. It becomes important, therefore, to try to reconstruct the events that occurred during the removal of the heat exchanger. Any indication of how the particles might have been released can give an indication that changes in procedures might be necessary for the future.

2. Heat Exchanger Removal

2.1 Background

Heat exchangers serve to cool the water that moderates the reactor core. In the NBSR, the moderating water is heavy water (D_2O) of 99.6% purity. This increases the efficiency of the reactor as compared to light water moderated reactors. The D_2O is pumped through the reactor core and through the primary side of the heat exchanger. Ordinary water (H_2O) is pumped through the secondary side and then to a cooling tower in a closed loop, and is never subject to the neutron flux of the reactor.

As shown schematically in Fig. 4 of Chapter 2, the D_2O is pumped through the more than 1000 tubes of the heat exchanger, which are in turn cooled on their outside by ordinary (light) water. In 1971 the first of 15 tubes developed leaks with the last one occurring in 1973. The reason for the leaks was mostly due to mechanical failure where vibration caused the tubes to rub against the baffles. There was one leak that could be positively traced to corrosion. As a result of these leaks, small amounts of heavy water were transferred from the primary to the secondary systems. After the first leak, the end bell was removed, so that the tube could be plugged and the heat exchanger thoroughly tested including eddy-current measurements of tube wall thickness. The tests however, were limited in that they could not provide indication of the condition of the tubes in the vicinity of

baffles. The leaky tube plus 32 other suspect tubes were plugged by fitting tight aluminum plugs into their ends. The heat exchanger is constructed totally of aluminum. Subsequently during the period 1971-1973, 13 more leaks involving 14 tubes developed. These plus 34 other suspect tubes were also plugged. In all 81 tubes including 15 identified leakers were plugged from 1971 to 1973.

Because of these problems, a new heat exchanger, this one constructed of stainless steel, was ordered. The leaky aluminum heat exchanger was removed in August of 1974, and replaced by the new stainless steel heat exchanger.

Removal of heat exchangers from a reactor is an uncommon operation. The removal of a major piece of equipment such as this is a very complex task that needs to be handled with great care. First, the D_2O in the primary loop needs to be preserved in an uncontaminated condition. Second, the level of radioactivity present (principally in the form of tritium oxide and radioactive particulate matter) is high and every precaution must be taken to insure the protection of workers and to prevent any unauthorized release of radioactive material to the environment.

2.2 The Removal

The actual removal of the heat exchanger was accomplished by personnel of the NBS Reactor Operations Division, with the help of private contractors. All of the radiation health and safety aspects were under the supervision of Health Physics personnel.

As part of its investigation, the Ad Hoc Committee attempted to recon-struct the actual series of events that occurred during the removal operation by interviewing the actual personnel involved. The sequence of events was gone over extensively, and a recapitulation is as follows.

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In the period from 8/15/74 to 8/22/74, the heat exchanger (HE) was disconnected from the reactor system and readied for removal from the building. This necessitated draining all the D_2O from the HE and blowing each tube several times to remove remaining traces of D_2O. The plugged tubes were not drained or blown out. Great care was taken to minimize exposure to tritium, especially when the flanges that connected the HE to the system were opened. Finally, on 8/22/74, the HE was wiped down thoroughly, and all the openings in the primary and secondary sides were sealed by bolting steel plates over them. It is to be noted that the primary head bell (see Fig. 4) was not removed during any of these operations.

While there were possibilities for radioactive particles to be released from the HE during these operations, the very stringent monitoring conducted makes their undetected release extremely unlikely. Moreover, even if a release had occurred, the particles would have been released in the building, and not outside, where they were found. The Committee concludes that release did not occur during this phase of the operation.

On 8/23/74 the completely sealed and wiped HE was moved to its final location beside the ramp. A rope defining the 2.5 mR/hr isodose line was put into place.

Because the HE was completely sealed and had been thoroughly wiped, the Committee concludes that release of particles did not occur during this phase of the operation. This is further substantiated by the fact that no contamination was found along the path travelled by the HE from the interior of the building to its location beside the ramp.

During the period from 9/4/74 to 9/27/74, the HE was thoroughly flushed with water. This flushing was done to remove as much tritium oxide as possible from the HE. Under continuous monitoring to assure that the

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amount of activity released was well below the maximum permissible concentration, the flush water was drained down the storm sewer.

On 9/27/74 the flushing was discontinued because personnel had to be used to connect the new heat exchanger. On 10/22/74 flushing was re-commenced and concluded on 10/25/74.

Because of the continuous monitoring of the effluent, and because no HE orifices were opened other than the small ones to which the flushing hoses were attached, the Committee concludes that particle release did not occur during these flushing operations.

On 10/31/74, the new heat exchanger was found to rattle. It was decided that it might be necessary to use the old HE temporarily, and hence it was readied for possible re-installation. To do this, it was necessary to remove all traces of light water from the primary side to prevent contamination of the D_2O. To do this, on 11/6/74, the covers to the access ports were removed and the primary tubes blown. All water was vacuumed out.

There was concern that some more tubes might be leaking. Therefore, on 11/7/74 the HE was hydrotested. The secondary side was filled with water and raised to a pressure of 110 PSIG. Five tubes were found to leak or were suspect, and these had to be plugged in order to be able to use the HE.

To plug the tubes, the end bell was removed from HE on 11/9/74. This was the only time the end bell was removed during the HE removal operation. On 11/11/74 the newly discovered leaking tubes were plugged, and on 11/14/74 the end bell was replaced and HE covered with polyethylene.

The new heat exchanger was subsequently fixed, so the HE did not have to be used. It was left with the primary side completely sealed and stored in its location until 9/5/79.

On that date, the end bell was again removed to look for particles, as described in Chapter 2, Sec. 2. When it was opened 15 of the plugs on the tubes were found to have fallen out, and there was evidence in the form of stains in the bottom of the end bell that water had been released from them, although there was no water in the end-bell. These tubes were those that had been originally plugged, not those that were plugged in the operation just described. Some or all of these tubes could have been filled with water by it being forced into them through their leaks during the hydro-testing on 11/7/74. About half of the tubes from which the plugs had fallen out were known leakers and the others could have developed leaks during the pressure testing. Very likely the plugs were forced out by freezing of the water.

Since these tubes were plugged, they were not blown out when the HE was prepared for possible reuse, and hence may have contained some particles, which could then have drained out along with the water in them. By the nature of the configuration, this water would have drained into the primary outlet port, (see Fig. 4) which was capped with a steel plate. These may have leaked at some time (perhaps due to freezing water) since water was not found in it when opened. The water would have drained, in fact, under HE in the spot where radioactive material was found. The possibility of this occurring with no radioactive particles remaining in the outlet port is very remote, but cannot be completely discounted. This possibility is made even more remote by the fact that subsequent swabbing and blowing of selected tubes that had remained plugged revealed no significant amount of loose contamination and no particles.

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Discussion and Conclusions

The removal of the HE from the reactor and all the operations performed on it as described in the previous section were performed under carefully laid out procedures. Unknown release of radioactive particles would have been very unlikely. Of all the operations carried out, the Committee feels that two are the most likely for particle release. The first is the removal of the end bell. The only time this occurred after reactor shut-down for the HE removal (with the exception of our investigation) was on 11/9/74 when it was removed to plug the leaking tubes after hydrotesting. The joint where the end-bell joins the body of the HE forms a natural crevice for the collection of particles, and two were found there during this investigation. This joint was wiped when the end bell was removed, but particles could have fallen out and have been missed, because of the level of activity emanating from the HE as a whole. The possibility is remote, but must be considered. The other possibility for release is along with the water that drained from the tubes that had their plugs fall out as previously described. The Committee considers these two as being the most likely of the possibilities for release, with the release at the time of removal of the end bell the more likely of the two.

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Tables I-VII

Table I

Areas with Positive Results

Area	Number of Spots	Number of Samples and Designation	Total Activity, μ Ci	Number of Particles from Samples
Front lawn of reactor	1	1, G_F	11.5	One present
Rear Lawn of reactor	9	9, G_1-G_9	72.2	One in G_1;[**] others not separated
North lawn of reactor	1	1, G_N	22.2	One present
Roof of Reactor Building	3	5; R_F, R_Y R_S^1, R_S^2, R_S^3	132.4	One in R_F;[**] One in R_S^1; One in R_S^3; R_Y, R_S^2 not separated
Holding Basin	1	1; D_2	24.0	Not separated
Outfall	4	4; D_1, D_3, D_4, D_5	32.1	One in D_1;[**] Others not separated
*Drain Exit	1 Extended	16; D_{10}^A-D_{10}^C; D_{10}^E-D_{10}^R	3.80	One in D_{10}^A;[**] Others not separated
Enclosed Area	1	Two barrels of sand; numerous asphalt chips	2 ± 0.25 mCi	Four removed: S_1^{**}, S_2, S_3,[**] S_4. Many others present
+Background samples; Beyond drain exit	NA	D_{11}, A through E --	See text. Sec. 2.4.4.7	See text. Sec. 2.4.4.
+Grass Sample of Outfall	NA		"	"
+SEBA Garden Soil	NA	--	"	"

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*This is one extended spot of low level activity extending to about 150 feet from the outfall.

+These were taken between the end of the previous area and the site boundary. They are discussed in Section 2.4.4.7 on "Samples Beyond Drain Exit".

**Particles on which analysis was performed.

Table II

Detailed Assay Results: Active Samples

Exterior to Enclosed Area

Sample	Activity, μ Ci	σ (%)	Sample	Activity, μ Ci	σ (%)
G_F	11.5	2.6	D_{10}^A	2.4	
G_1	2.7		D_{10}^B	0.03	14.5
G_2	12.4	2.5	D_{10}^C	0.2	4.8
G_3	24.5	2.0	D_{10}^D	0.17	5.6
G_4	3.7	4.4	D_{10}^E	0.05	11.0
G_5	0.9	8.8	D_{10}^F	0.12	6.5
G_6	4.1	4.2	D_{10}^G	0.14	6.0
G_7	0.2	4.5	D_{10}^H	0.08	7.8
G_8	1.4	7.1	D_{10}^I	0.1	6.7
G_9	22.3	2.0	D_{10}^J	0.04	11.9
G_N	22.2	1.8	D_{10}^K	0.09	7.4
R_F	8.7		D_{10}^L	0.04	10.7
$R_1 y$	28.4	1.8	D_{10}^M	0.03	13.5
R_S	1.9		D_{10}^N	0.10	7.0
R_S^2	47.0	1.5	D_{10}^O	0.2	4.9
R_S^3	46.4		D_{10}^P	0.008	27.4
D_1	10.0				
D_2	24.0	1.9			
D_3	0.8	9.1			
D_4	3.3	4.6			
D_5	18.0	2.2			

Standard deviation expresses only counting uncertainty

Table III (Cont'd)

	Marginal Detection (ND is less than this)	Reliable Detection (D is greater than this)	Minimum Necessary to quantify 10% error
^{137}Cs	0.032	0.066	0.253
^{40}K	0.032	0.069	0.333
^{60}Co	0.040	0.083	0.354

Note:

NRD indicates a level between the first and second columns

D indicates a level between the second and third columns

Table III

Activity of "Beyond Drain Exit" Samples

(Units are pCi/g)

Sample	^{137}Cs	^{40}K	^{60}Co
D_{11}^A	1.72 ± 0.066	0.77 ± 0.0045	14.8 ± 0.23
D_{11}^B	0.474 ± 0.028	1.3 ± 0.056	D
D_{11}^C	0.559 ± 0.031	1.52 ± 0.061	NRD
D_{11}^D	0.263 ± 0.015	2.27 ± 0.048	ND
D_{11}^E	NRD	1.68 ± 0.045	ND
Outfall Grass	D	1.38 ± 0.084	NRD
SEBA Garden	0.286 ± 0.023	2.45 ± 0.067	ND

Notes:

Standard deviation includes only counting statistics.

ND = Not detected

NRD = Not reliably detected

D   = Can be reliably reported as "detected", but quantification to 10% is not possib

Table IV

Areas Showing Negative Results

Area	Date Completed	Results
Lawn Equipment	9/1/79	Negative
Workshoes	9/4/79	"
Area to 200' from Reactor	8/31/79	"
Site Fence	9/3/79	"
Solar house, including interior and bird nesting area	9/2/79	"
Building 245, including interior	9/2/79	"
Trees South of South Drive	9/2/79	"
Groves N and NW of Reactor	9/4/79	"
All NBS Buildings, including roofs and trees	9/3/79	"
SEBA Garden	9/1/79	"
Road Outside of Fence at Exits	8/30/79	"
Area 150' beyond drain exit to site boundary (see Table 1)	9/1/79	"
High Bay Area in Engineering Mechanics Building	9/3/79	"
*Three samples over rise North and East Reactor	9/3/79	"

* These were soil samples taken far away from NBSR. The results showed no ^{60}Co at a detection sensitivity of 0.040 pCi/gm (see Sec. 2.4.5 and Table III).

Table V

Activity of Samples in Monitoring Survey

Sample	Activity, μ Ci	σ (%)
G_{12}^1	0.7	2.7
G_{12}^2	1.6	2.0
G_{12}^3	0.7	2.8
G_{12}^4	2.1	1.8
G_{12}^5	1.2	2.2
G_{12}^6	1.6	2.0
G_{12}^7	0.6	3.0
G_{12}^8	0.2	4.9
G_{12}^9	1.3	2.2
G_{12}^F	0.3	4.0
G_{13}^1	1.2	2.2
G_{13}^2	1.2	2.2
G_{13}^3	0.3	4.1

Standard deviation expresses only counting uncertainty

TABLE VI

Particles Analyzed

Particle Identification	Source
S_1	Enclosed Area; Sand under heat exchanger
S_3	" " "
G_1	Rear Lawn of Reactor
R_F	Reactor Roof
D_1	Outfall
D_{10}^A	Drain Exit Spot
HEX 7-1	Interior of Heat Exchanger
POOL-2	Spent Element Storage Pool

Table VII

Particle Specific Activities *

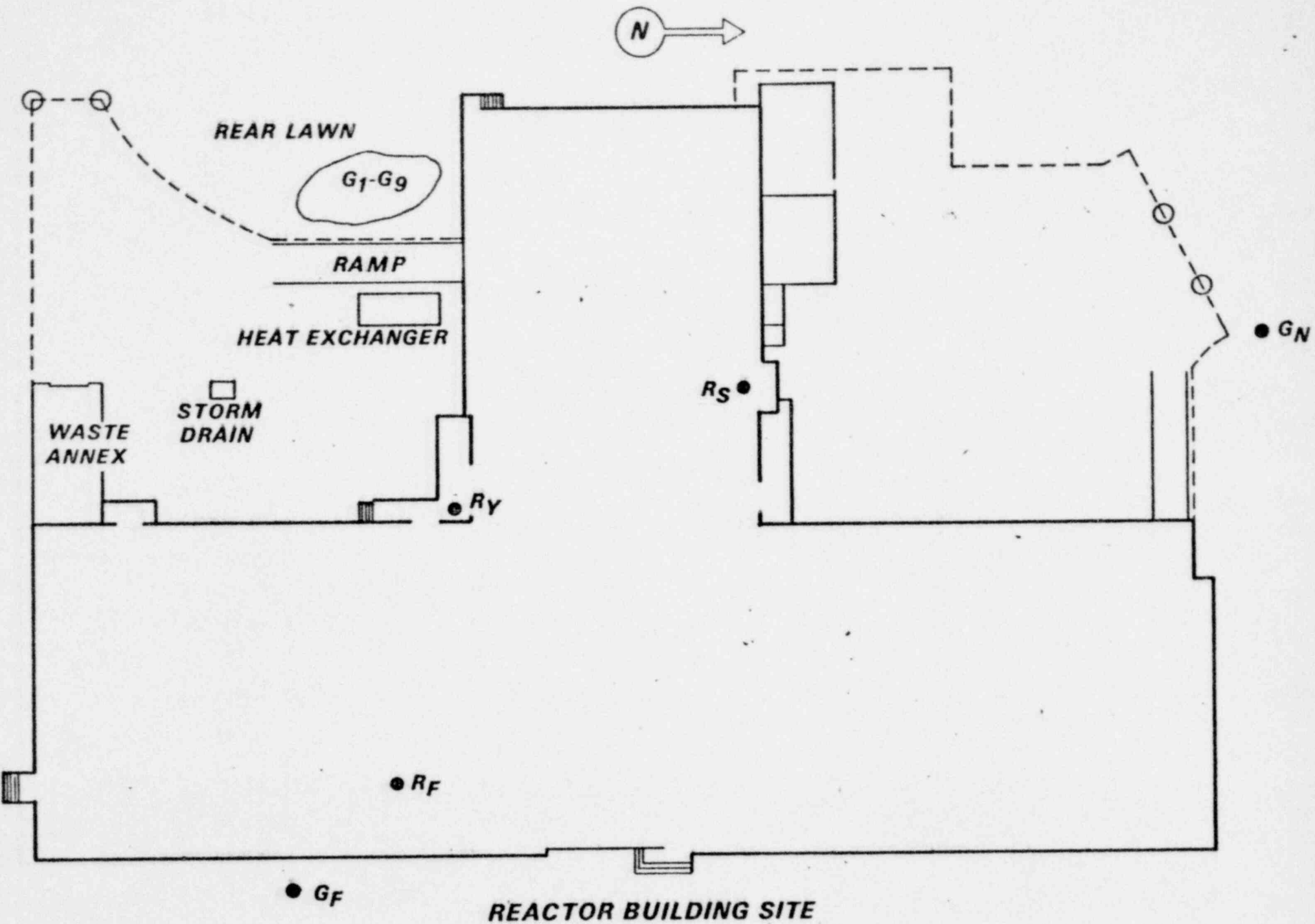| Particle	Wt. ^{59}Co (µg)	^{60}Co Activity (µCi)	Sp. Activity (Ci/g(59))	Sp. Activity (Ci/g(Co))	Sp. Activity (From Activation Ratios) (Ci/g(Co))
G2	14.0	12.8	0.91	0.91	--
D1	6.95	9.3	1.34	1.34	1.36
Rf	.88	9.0	10.2	10.1	10.3
G1	.48	5.58	11.6	11.5	11.5
HEX 7-1	13.1	257	19.3	19.3	--
D_{10}^{A}	.0092	2.70†	293	234	259

* Based on data contained in Appendix C

† Based on the average of the results from Appendix C and Appendix D.

Figures 1-9

Figure Captions

Fig. 1   A plan view of the reactor building and adjacent area showing where radioactive spots were found in the site survey.

Fig. 2   A general map of the reactor building and surrounding area out to the nearest site boundary, showing the location of various areas discussed in the text.

Fig. 3   A schematic map showing the location of all areas where radioactive ^{60}Co material was found.

Fig. 4   A schematic diagram of the cross section of the heat exchanger.

Fig. 5   A section of the worn wear ring. The diameter of the ring is nine inches.

Fig. 6   A view of the surface of the worn wear ring. Note the severe wear X4.

Fig. 7   A cross section view of the worn wear ring (left) and an unused wear-ring (right), showing the hard-facing inset. In each case, two sections of the wear ring are shown face to face. X6.

Fig. 8   Two views of the cross section of the worn wear ring, showing a deposited layer on the surface. This layer is variable in composition but contains a high concentration of iron (up to 40%). The matrix material is multi-phase, but contains a low concentration of iron ($\sim$2%). X500 upper, X1000 lower.

Fig. 9   Distribution of specific activity of six particles. Arrows indicate specific activity corresponding to the maximum achievable for a particle circulating with the primary water and for a particle residing in the maximum flux region of the reactor.
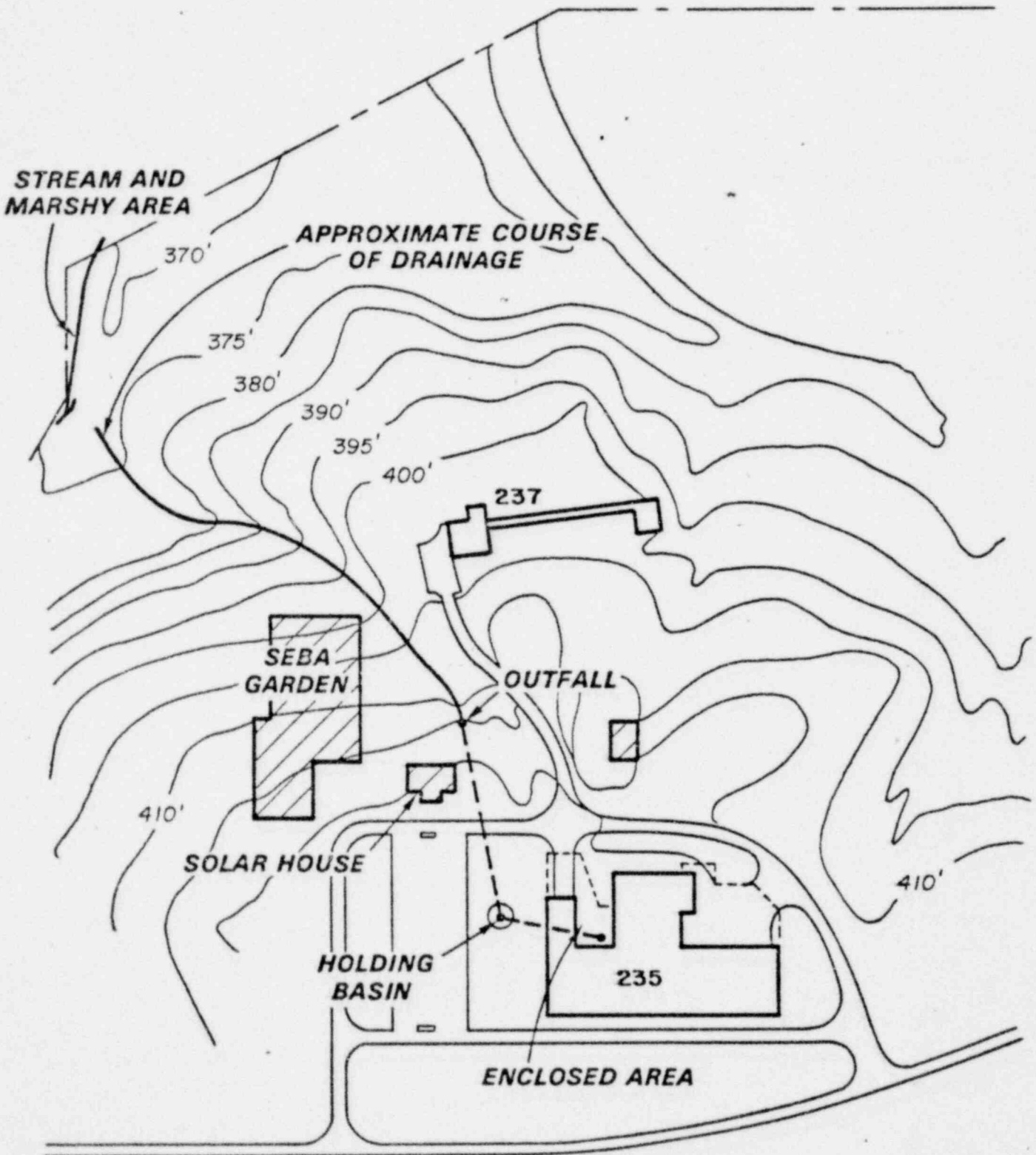
REACTOR BUILDING SITE

Figure 1

STREAM AND MARSHY AREA

APPROXIMATE COURSE OF DRAINAGE

370'

375'

380'

390'

395'

400'

237

SEBA GARDEN

OUTFALL

410'

SOLAR HOUSE

410'

HOLDING BASIN

235

ENCLOSED AREA

Figure 2

SCHEMATIC MAP OF
RADIOACTIVE AREAS FOUND
(map not to scale)
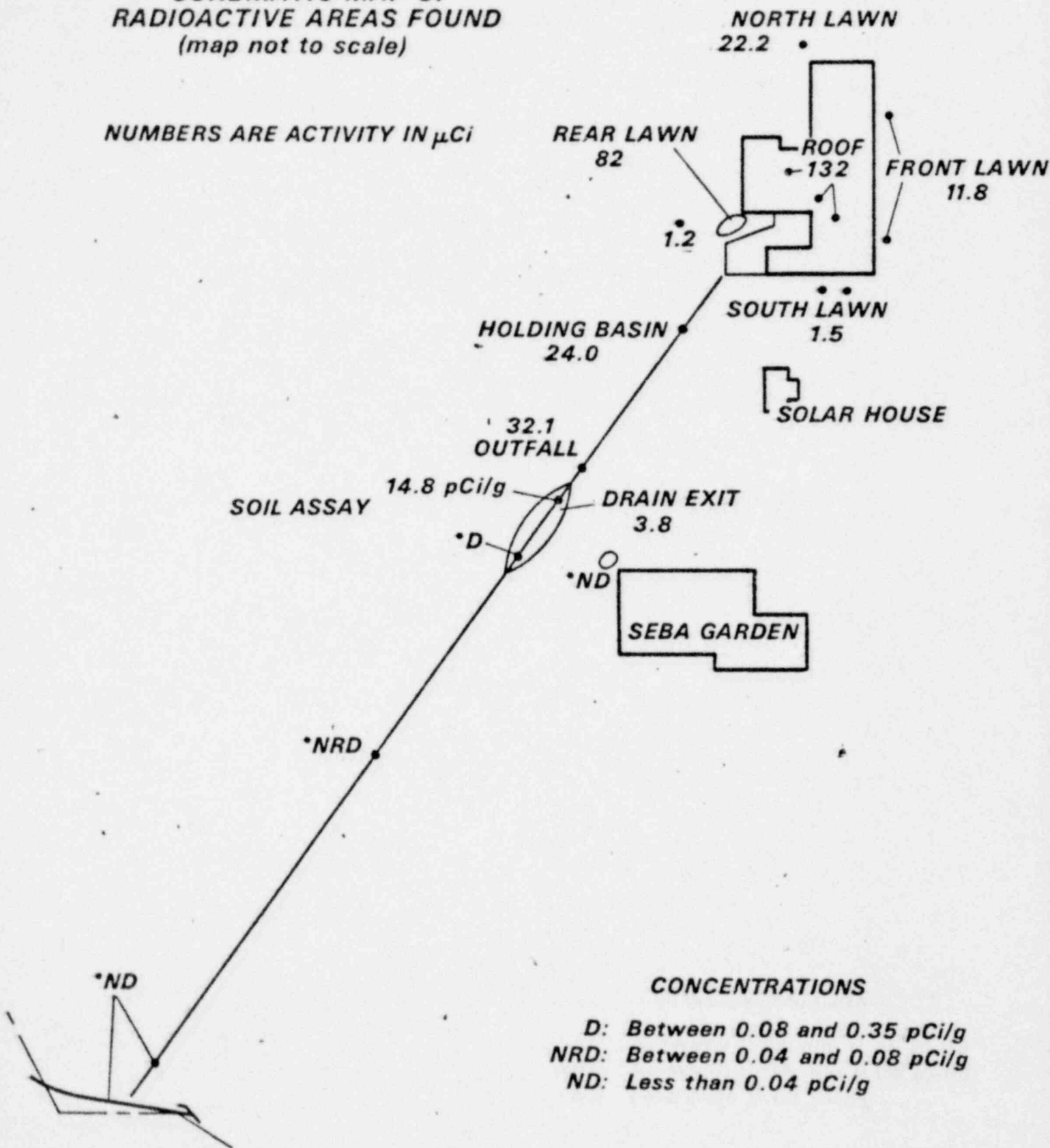
NORTH LAWN
22.2 •

NUMBERS ARE ACTIVITY IN μCi

REAR LAWN
82

ROOF
←132

FRONT LAWN
11.8

1.2

HOLDING BASIN
24.0

SOUTH LAWN
1.5

SOLAR HOUSE

32.1
OUTFALL

14.8 pCi/g

DRAIN EXIT
3.8

SOIL ASSAY

•D

•ND

SEBA GARDEN

•NRD

•ND

CONCENTRATIONS

D: Between 0.08 and 0.35 pCi/g
NRD: Between 0.04 and 0.08 pCi/g
ND: Less than 0.04 pCi/g

Figure 3

Fig. 4

Primary Inlet

Double Tube Sheet

Secondary Outlet

Shell

End Bell

Baffles

Tubes

Divider Plate

Steel Backing Rings

Primary Outlet

End-Bell to Tube Sheet joint

Secondary Inlet
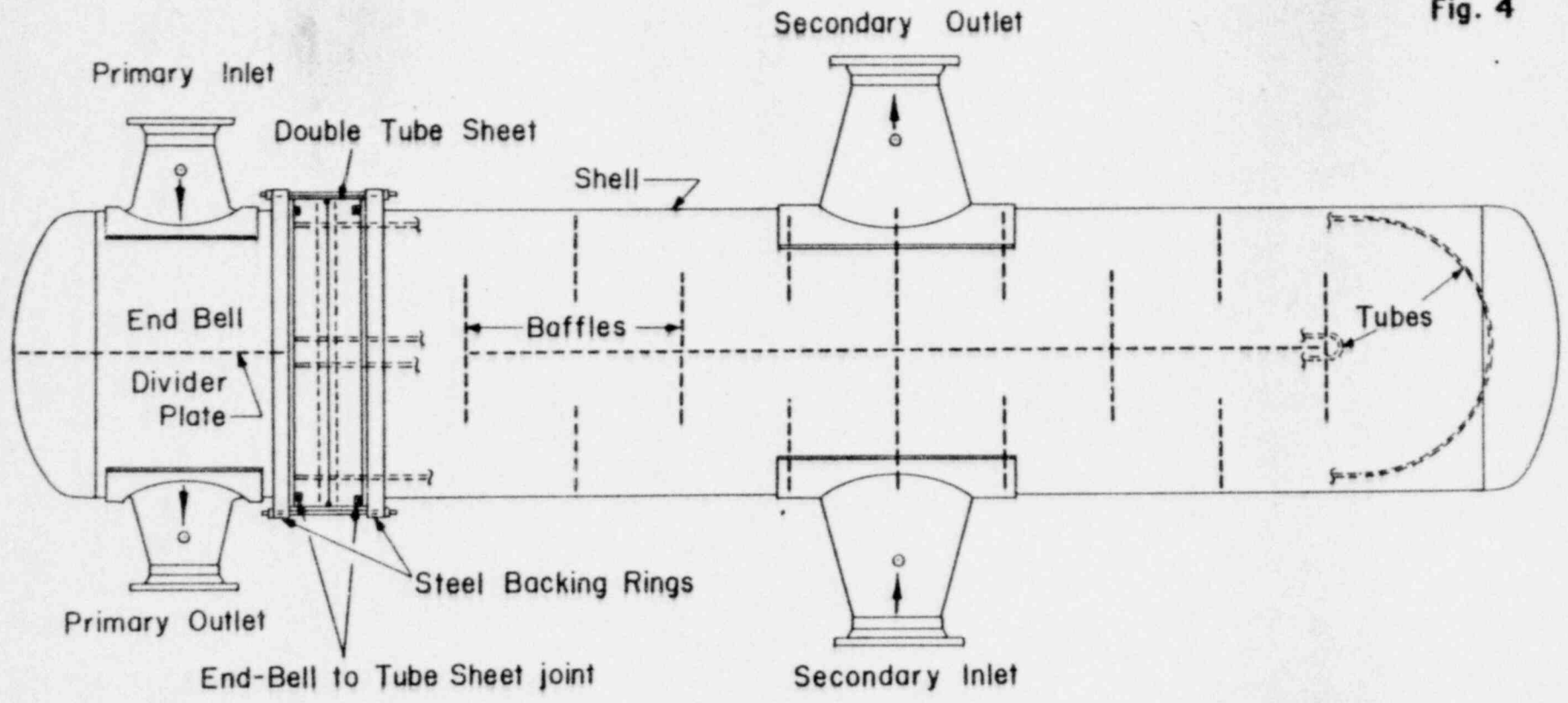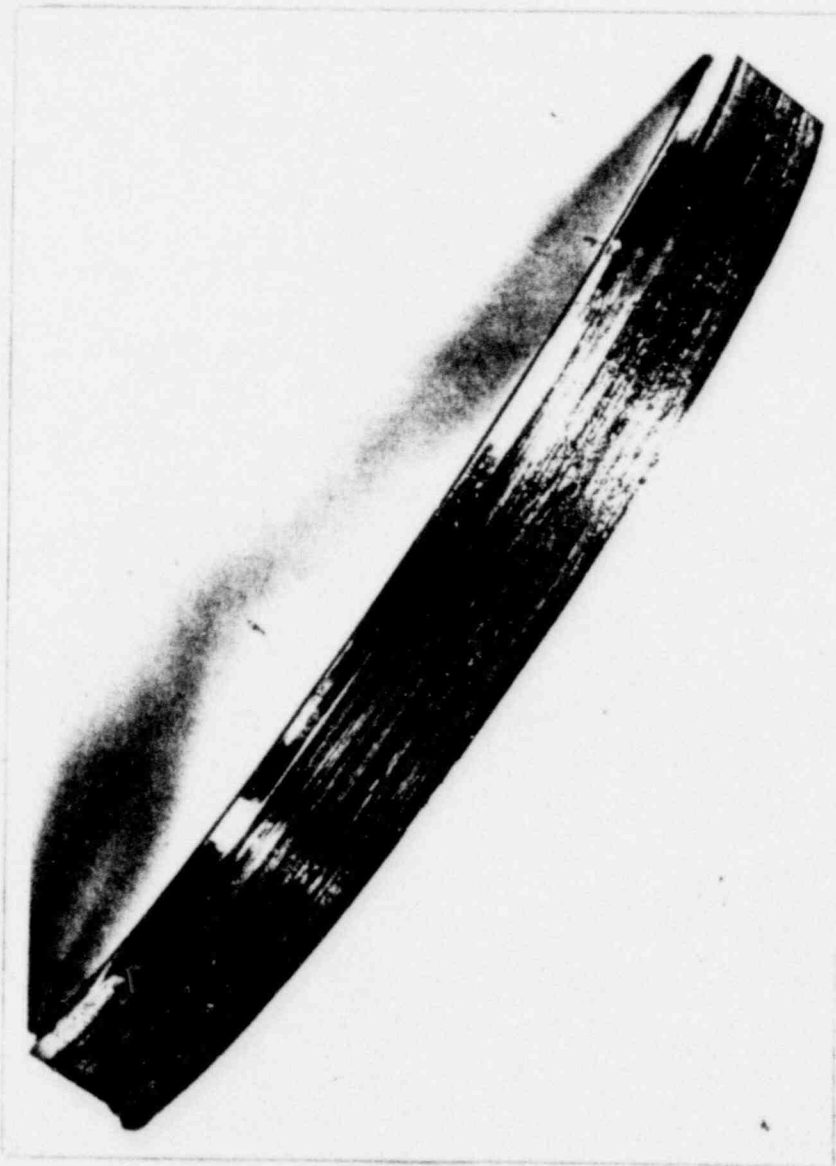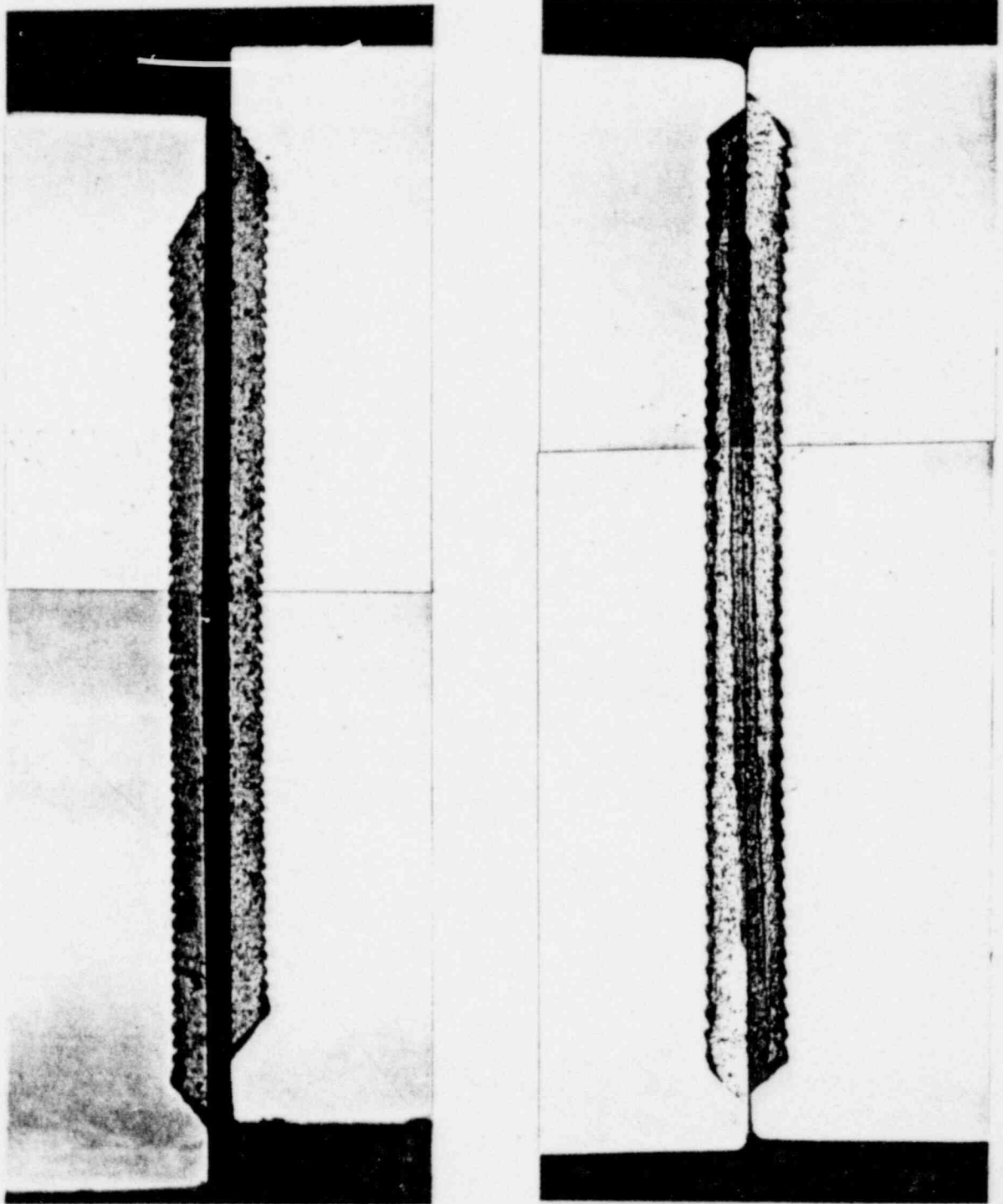
Figure 5.

Figure 6.
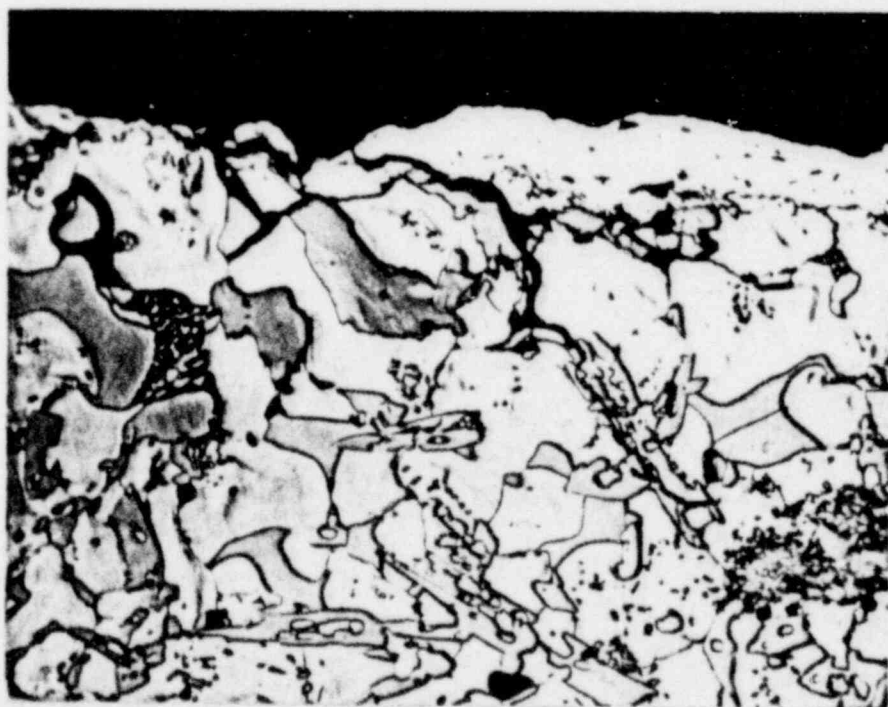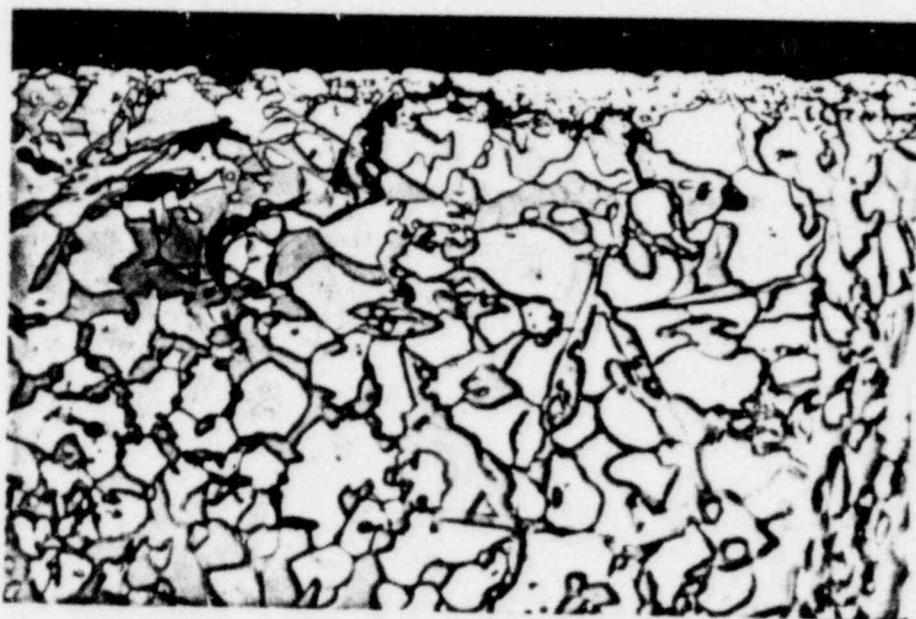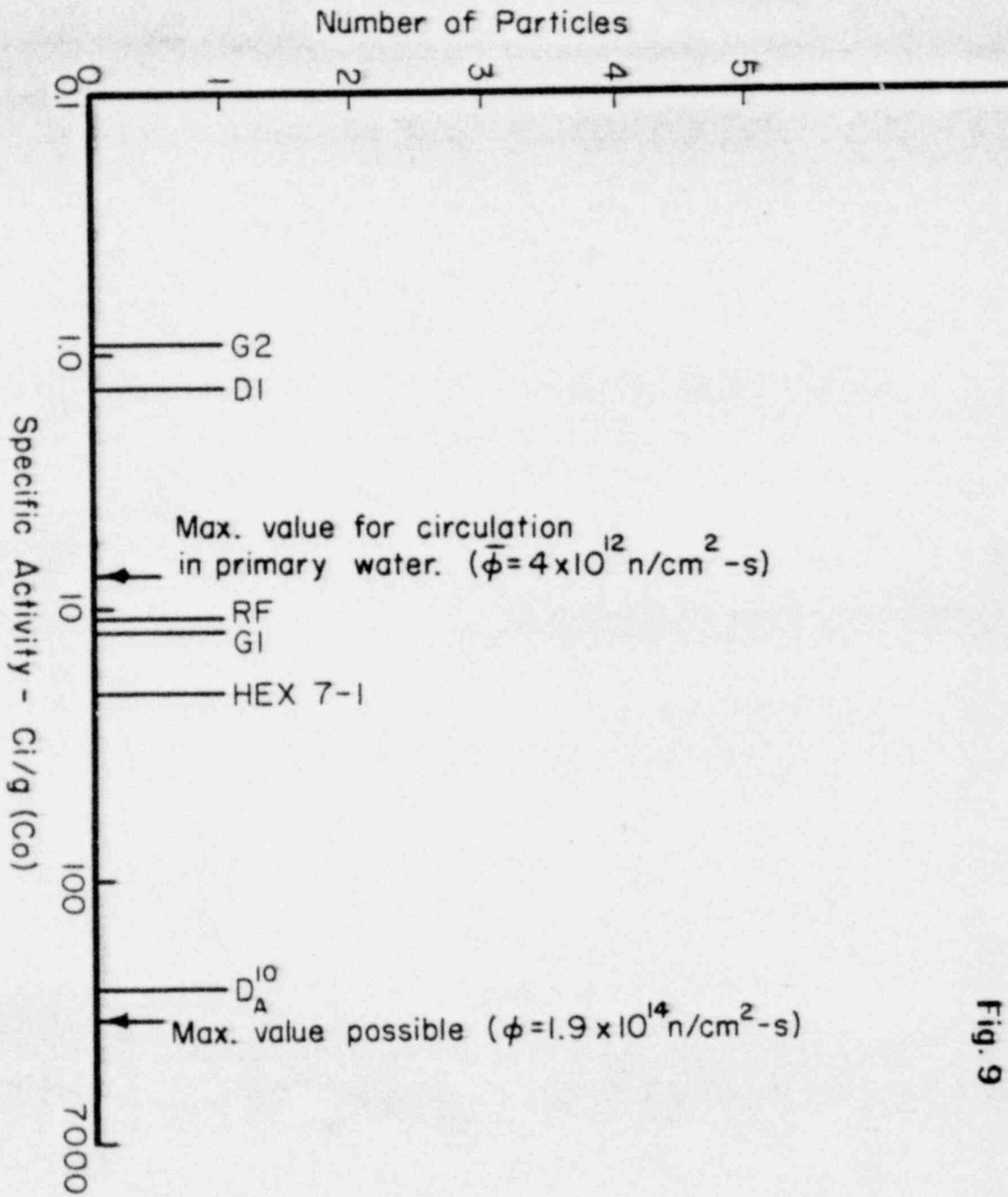
Figure 7.

Figure 8.

Fig. 9

Appendix A

U.S. DEPARTMENT OF COMMERCE
NATIONAL BUREAU OF STANDARDS
WASHINGTON, D.C. 20234

REPORT OF ANALYSIS

Particles From NBS Reactor Site

by

Scanning Electron Microscopy and X-ray Microanalysis

Submitted by: Elio Passaglia, Center for Materials Research
Laboratory No: 553-39045
Date Submitted: August 30, 1979
Analysts: R. L. Myklebust, D. E. Newbury, J. A. Small, E. B. Steel

Request: Determine the composition and structure of certain radioactive particles recovered from the NBS reactor site. Cobalt is of particular interest.

Description of Method and Results: The Cameca electron probe micro-analyzer equipped with an energy dispersive x-ray spectrometer was employed for the analysis. The x-ray spectral data were reduced to compositional values through the application of the NBS theoretical matrix correction procedure FRAME C. For the analysis of rough surfaces, the NBS special matrix correction procedure, FRAME P, which has recently been developed, was employed. Because of the uncertainties in the analysis of rough surfaces, the estimated errors in the analysis values must be presumed to be as high as ±10% relative. Note that only elements with atomic numbers of eleven (sodium) or greater are detected.

The following samples were received from the NBS reactor division: S-1, G-1, RF, D-1, HEX7-1, S-3, POOL 2, D10A, and pump corings (PUMP 1,2,3). An abraded wear ring and new wear ring from the system pumps were also provided. The particles were mounted for analysis in carbon dag on a carbon substrate. All particles were analyzed in the as-received condition. In addition, particles S-1 and HEX7-1 were examined after cleaning by ultrasonic agitation in an alcohol-acetone mixture (10:1). Particles S-1, HEX7-1, and D10A were analyzed on both sides, which required cleaning to remove carbon particles adsorbed from the carbon dag. Metallographic cross sections of the wear rings were analyzed.

The analysis of the particles reveals two distinct classes: Class 1 (samples S-1, G-1, RF, D-1, HEX7-1, S-3, and POOL 2) consists of heterogeneous particles which contain regions of aluminum, stainless steel (approximately Fe - 18Cr - 10Ni), and cobalt-chromium regions. Class 2 (sample D10A) contains a single particle which is nearly homogeneous and which consists of a stainless steel (Fe - 18Cr - 15Ni) with about 13% cobalt. The details of the analyses of these classes

are as follows: <u>Class 1</u>: In the as-received condition, all particles in this class produce x-ray spectra which indicate the presence of aluminum and stainless steel (Fe-Cr-Ni) in significant amounts, (10% or more) and cobalt as a minor element (<10%). The particles were found to be heterogeneous, with the composition varying markedly from point to point. Morphologically, the particles appear to be flakes, with the thickness dimension typically about 10% of the length or width.

Two samples, S-1 and HEX7-1 were chosen for detailed examination. Micrographs of these particles [e.g., Figures 1(a) and 1(b)] reveal that the particles have distinct structures on their "front" and "back" sides. The "back" side has distinct linear features which appear to be scratches. The "front" side has no such features, appearing instead rough and irregular.

Detailed x-ray microanalysis (Table 1) of the back surface of S-1 reveals that the dark areas "A" in Figure 1(a) contain principally aluminum and iron, while the bright areas "B" consist of stainless steel.

The "front" side of S-1, Figure 1(b), contains similar Al-Fe ("C") and stainless steel ("D") regions but the microanalyses (Table 1) reveal that a significant cobalt content is also observed in these regions. A cobalt x-ray area scan of the front surface of S-1, Figure 2, reveals the presence of several high cobalt content regions, as well as a general cobalt distribution. Close examination of the largest region, Figures 3(a), (b), reveals a flat, approximately triangular particle. X-ray microanalysis at three points (Table 1, E1, E2, E3) indicates that this cobalt-rich particle is nearly homogeneous in composition and consists of cobalt (65 wt%) - chromium (21%) - tungsten (3.3%) with virtually no nickel. The particle is surrounded by an aluminum-rich matrix. Note that the stainless steel regions contain about 10% cobalt which is in marked contrast with the stainless steel on the back surface. Other cobalt-rich particles in Figure 3(b) gave similar compositional results. The cobalt-rich particle E (designated S1-SP5B) was subsequently extracted mechanically for specific radioactivity measurements. During the cutting operation, particle S-1 fractured. Analysis of the fresh edges of the S-1 fragments indicated that the interior of particle S-1 was stainless steel.

Sample HEX7-1 in class 1 was also studied extensively. This particle also had distinctly different morphology on the front and back surfaces, Figures 4(a), (b), with very little cobalt found on the back side in either the Al-Fe (Table 2, A) or stainless steel regions (Table 2, B). The front side of HEX7-1 is similar to the front side of S-1, containing stainless steel/Co regions (Table 2, C), Al-Fe regions (Table 2, D), and Co-Cr-W regions. Again, a cobalt x-ray area scan reveals areas of high cobalt concentration, Figure 5. This cobalt-rich region viewed at high magnification, Figure 6, contains two structures E and F which are cobalt-chromium-tungsten particles. One of these particles (Table 2, E) contains low nickel (1.9%) while the other (Table 2, F) contains significantly more nickel (11%). The matrix between these particles is principally aluminum (Table 2, G), probably in an oxidized state, as indicated by the low total composition (oxygen is not directly measured in the x-ray spectrum).

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Class 2: Sample D10A, Figures 7(a), (b), was determined to be a particle which was markedly different from the particles of Class 1. D10A was found to be a stainless steel enriched with cobalt, (Table 3). The particle had a similar composition on the front and rear surfaces, as well as on the sides. Locally, the cobalt composition was similar point to point, Figures 7(c), (d), with one exception. A small (ca. 5 μm) cobalt-chromium-tungsten inclusion (Table 3, A) was found in the matrix of cobalt-enriched stainless steel.

The mass of the particle was estimated as follows:

Calculation of the Mass of Particle D10A

(1) On the front side, the image of the particle is bracketed by a rectangle 29 μm x 58 μm (scale calibrated from a Leitz stage micrometer).

(2) On the back side, the image of the particle is bracketed by a rectangle 32.5 μm x 58 μm.

(3) The thickness is 6 μm.

(4) The volume of the particle is 10,700 μm^3 or 1.07×10^{-8} cm^3.

(5) Considering the density of the particle as 7.8 g/cm^3 (the density of iron), the mass of the particle is 8.3×10^{-8} g.

(6) Considering the average cobalt concentration in the particle is 13 weight percent, the cobalt mass is 1.1×10^{-8} g.

The particle was analyzed at beam energies as high as 25 keV, which provided a sampling depth of 1.5 μm. Since both surfaces were analyzed, approximately one-half of the mass of the particle was sampled. If we consider the center of the particle to be composed of the 65% Co - 20% Cr - 4% W alloy, an unlikely hypothesis in view of the analyses of the side of the particle, then the possible cobalt content of the particle increases to 3.3×10^{-8} g.

Analysis of Pump Components

(1) Corings: Corings were provided from the base metal of the pump housing, impeller, and wear ring. The analyses of the coring flakes (Table 4) show the base metal to be Fe-Cr-Ni-Mo with virtually no cobalt or tungsten.

(2) Worn Wear Ring: The abraded surface of the wear ring, Figure 8, was found to be heterogeneous. A series of eight analyses (Table 4, A, B, C, D, E, F, G, H) in a local region, Figure 9, reveals substantial differences in the content of the principal elements (Cr, Fe, Co, Ni) from point to point.

The wear ring was also examined in a polished cross-section, Figure 10. At higher magnification, Figure 11, at least three distinct phases are

3

observed: a) a continuous fine phase, b) a discontinuous fine phase and c) a coarse spherical phase. The analyses for these phases (Table 4) show that the two-phase matrix surrounding the spherical phase consists of a cobalt-nickel-chromium alloy while the spherical phase is a cobalt-chromium-tungsten alloy.

(3) New Wear Ring: A new, unabraded wear ring was examined in cross section. The front surface of the wear ring, Figure 12, which corresponds to the abraded surface of the worn wear ring, is found to consist of four distinct layers. The outer most layer, (Tables 5, A, B), consists of a high cobalt-nickel alloy, with about 10% chromium and 3-5% tungsten. The next layer, (Table 5, C), is multiphase, Figure 13, with the individual phases yielding the analyses listed. The third layer, (Table 5, D), is the 65 Co - 20 Cr - 4 W layer. Beneath this high cobalt layer is another cobalt-nickel-chromium layer, (Table 5, E). A multiphase layer, Figure 14, (Table 5, F), is found at the interface with the stainless steel.
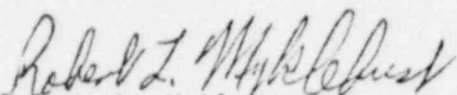
The back surface of the wear ring, Figure 15, consists of three distinct regions. The outermost layer, A, consists of an iron-chromium alloy. The innermost layer, B, consists principally of nickel. In addition inclusions, "C", of high molybdenum content are observed.

Discussion: With the exception of aluminum, the compositions of the two classes of particles can be derived from the components of the hard facing alloy of the wear ring and the stainless steel of the pump housing. For the class 1 particles (S-1, HEX7-1, etc.) the cobalt-chromium-tungsten inclusions correspond closely to the composition of the coarse spherical phase found in the wear ring hard facing alloy (65% Co - 20% Cr - 4% W). The stainless steel matrix of these particles is quite similar to the stainless steel base of the wear ring (60% Fe - 18% Cr - 11% Ni - 3% Mo), but enriched with cobalt (10%) and additional nickel (4 to 6% extra), which could be derived from the cobalt-nickel-chromium content of the wear ring. The aluminum which is frequently found as a coating on the class 1 particles must originate from a source other than the wear ring or pump components.
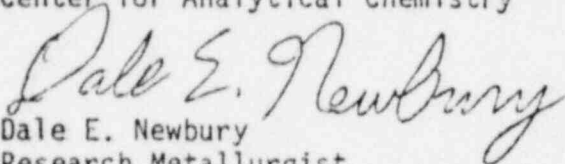
The class 2 particle (D10A), which has the composition of a cobalt-enriched stainless steel and which is similar to certain regions of the class 1 particles, could also be created from the wear ring and the base metal. Region A from the surface of the wear ring (Table 4) has a 3:1 Fe to Cr ratio similar to the base stainless steel, with an enrichment in cobalt and nickel which could be obtained from the Ni-Co hard facing alloy. D10A, in fact, appears to be a subclass of the class 1 particles rather than a member of a separate class. That is, D10A is similar in composition to the cobalt and nickel enriched stainless steel regions of the front surface of the class 1 particles.

The formation of these particles during the severe abrasion of the wear ring against the stainless steel surface of the pump housing is an attractive

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hypothesis. This mechanism explains the curious structure of the class 1 particles, e.g. S-1 and HEX7-1, which have a cobalt-free stainless steel rear face and a cobalt-enriched stainless steel front face, with occasional inclusions of the 65% Co - 20% Cr - 4% W alloy. The analysis of the surface of the wear ring itself reveals patches of stainless enriched regions on the cobalt-nickel-chromium hard facing alloy whereas iron is missing in the analyses of the cobalt-nickel-chromium facing examined in cross section. The stainless steel on the surface must have been deposited during the severe abrasion between the wear ring and the stainless steel pump housing. The further observation of the 65% Co - 20% Cr - 4% W alloy inclusions in the hard facing alloy cross section explains the occasional appearance of these inclusions in the reactor particles. The aluminum which is observed on the reactor particles may have been picked up during abrasion of the aluminum heat exchanger by the stainless steel - hard facing composite particles as they passed through the system in high velocity water flow.

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September 28, 1979

Table 1

X-ray Microanalaysis of Particle S1

	Al	Si	Cr	Mn	Fe	Co	Ni	Mo	W
Rear, scratched									
A	23	1.4	13	0.5	45	0.4	7.0	3.8	-
B	5.8	1.3	16	0.4	61	0.2	10	4.3	-
Front, rough									
C	30	1.1	12	0.1	21	12	7	1.8	-
D	9.3	1.3	17	0.3	38	9.3	13	3.3	-
E1	1.0	2.7	20	0	2.5	66	0.7	0.8	2.8
E2	0.9	2.2	21	0	2.2	65	0.6	0.7	2.9
E3	0.4	1.9	21	0	0.7	67	0.7	0.2	3.9

Table 2

X-ray Microanalysis of Particle HEX7-1

	Al	Si	Cr	Mn	Fe	Co	Ni	Mo	W
Rear									
A	36	0.4	4.3	*	14	1.0	2.3	1.1	0
B	2.4	1.3	14	*	51	2.1	9.0	2.0	0.6
Front									
C	4	0.8	15	*	30	11	10	1.5	0.9
D	23	0.6	9.7	*	20	8.5	7.0	1.1	0.6
E	9	1.7	15	*	2.4	44	1.9	0.3	2.9
F	0.6	2.4	16	*	1.5	46	11	0.1	2.8
G	42	0.6	1.7	*	7.5	2.0	0.4	0.7	0.1

*Not determined

Table 3

X-ray Microanalaysis of Particle D1OA

	Al	Si	Cr	Mn	Fe	Co	Ni	Mo	W
Front*	1.1	1.3	18	**	41	14	16	1.6	1.4
Rear*	1.4	0.7	18	**	42	13	15	1.8	1.7
Side	1.3	0.9	19	**	39	14	16	1.6	1.5

*Average of 8 analyses

	Al	Si	Cr	Mn	Fe	Co	Ni	Mo	W
A	1.2	1.2	21	**	15	37	9	0.9	2.7

**Not determined

Table 4

X-ray Microanalysis of Pump Components

	Al	Si	Cr	Mn	Fe	Co	Ni	Mo	W
Base metal (turnings)									
Wear Ring	0.5	1	18	1.7	65	0	11	6	0
Impeller	0.3	1.6	20	0.3	69	0	11	5	0
Housing	0.2	1.7	19	0.4	70	0	10	4	0
Surface of wear ring									
A	2.5	1.3	9.6	*	30	9.2	17	1.6	1.2
B	0.3	1.4	18	*	28	17	33	1.0	1.3
C	0.3	2.2	16	*	11	26	46	0.4	1.7
D	0.2	1.0	19	*	5.6	38	32	0.3	2.0
E	4.7	1.5	22	*	21	12	29	1.0	2.0
F	0.3	1.6	19	*	22	14	40	0.8	1.2
G	3.4	1.1	7.5	*	29	7.7	15	1.6	0.6
H	0.9	2.1	19	*	20	17	36	1.2	2.3
Wearing, polished cross section									
base metal	0.2	0.8	18	*	62	0.8	11	2.8	0
continuous free phase	0.3	0.9	27	*	1.3	47	11	0.3	1.3
discontinuous free phase	0.3	4.8	10	*	1.3	50	30	0.2	1.6
spherical phase	0.1	2.3	20	*	0.5	64	0.6	0	3.4

*Not determined

Table 5

X-ray Microanalysis of New Wear Ring

	Al	Si	Cr	Mn	Fe	Co	Ni	Mo	W
Front surface									
A	0.2	3.3	9.9	*	2.0	43	35	0.2	3.0
B	0.1	2.8	13	*	1.4	46	25	0.2	3.3
C1	0	3.3	9.0	*	1.2	43	29	0.2	5.2
C2	0	0.4	25	*	1.0	43	10	0.8	1.3
C3	0.1	1.4	13	*	0.8	51	12	0.3	8.3
D	0.1	2.0	18	*	0.5	65	0	0.3	5.0
E	0	3.6	8.3	*	1.8	39	36	0.1	3.6
F1	0.2	0.5	41	*	0.8	16	9	0.4	14
F2	0.1	4.2	8.3	*	4.5	11	63	0.2	3.7
F3	0	0.2	70	*	3.3	0.6	8.7	0.2	0.1
Rear surface									
A	0.1	0.3	11	*	80	0	0	0.3	0.3
B	0.2	0.2	0	*	0.1	0.2	91	0.2	0.4
C	0.5	0.5	0	*	0.1	0.1	0.2	96	0.9

*Not determined

Figure Captions

1(a) Particle S1, rear surface
 (b) Particle S-1, front surface

2 Cobalt x-ray area scan corresponding to Figure 1(b)

3(a) Particle S-1, inclusion SP-5B
 (b) Cobalt x-ray scan of inclusion SP-5B

4(a) Particle HEX7-1, rear surface
 (b) Particle HEX7-1, front surface

5 Cobalt x-ray area scan corresponding to Figure 4(b)

6 Structure containing high cobalt regions in Figure 5

7(a) Particle D10A, rear
 (b) Particle D10A, front
 (c) Particle D10A, front; "A" marks cobalt-rich region
 (d) Cobalt x-ray area scan corresponding to (c)

8(a) Optical micrograph of wear ring showing transition from fresh
      to worn region
 (b) Wear ring showing high wear area

9 Scanning electron micrograph of wear ring showing high wear region

10 Abraded wear ring cross section, optical micrograph

11 Abraded wear ring cross section, showing fine scale continuous phase, fine
   scale discontinuous phase, and coarse spherical phase

12 Front surface of new wear ring in cross section

13 High magnification of area C, Figure 12

14 High magnification of area F, Figure 12
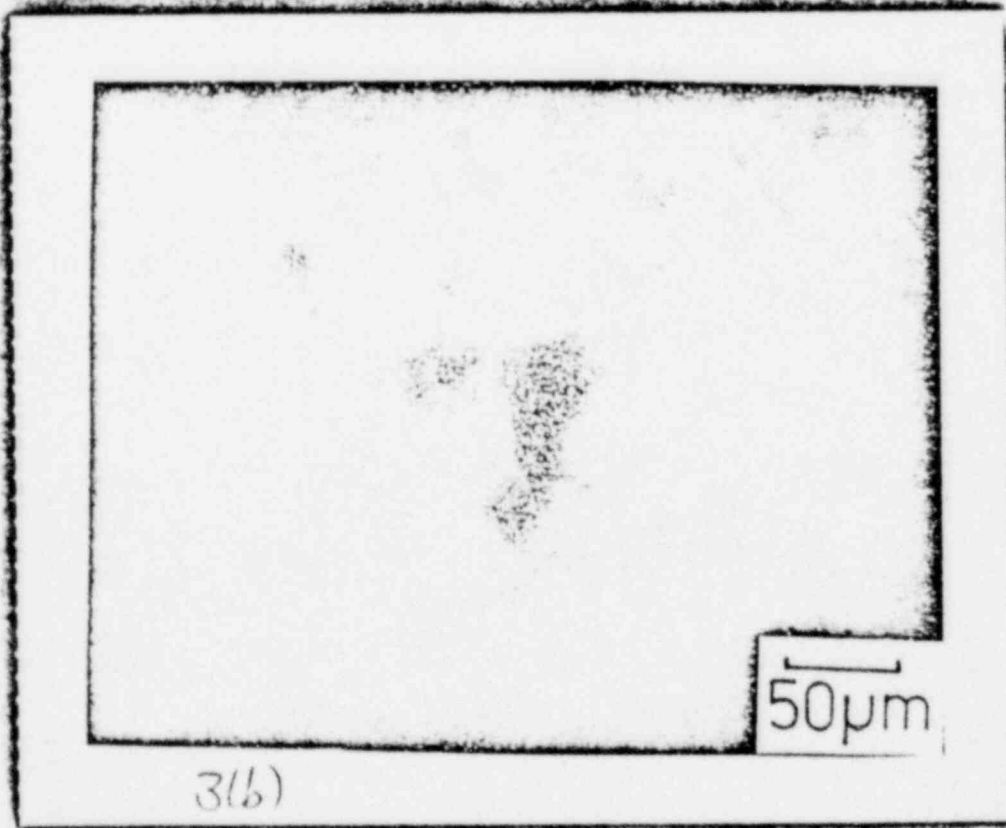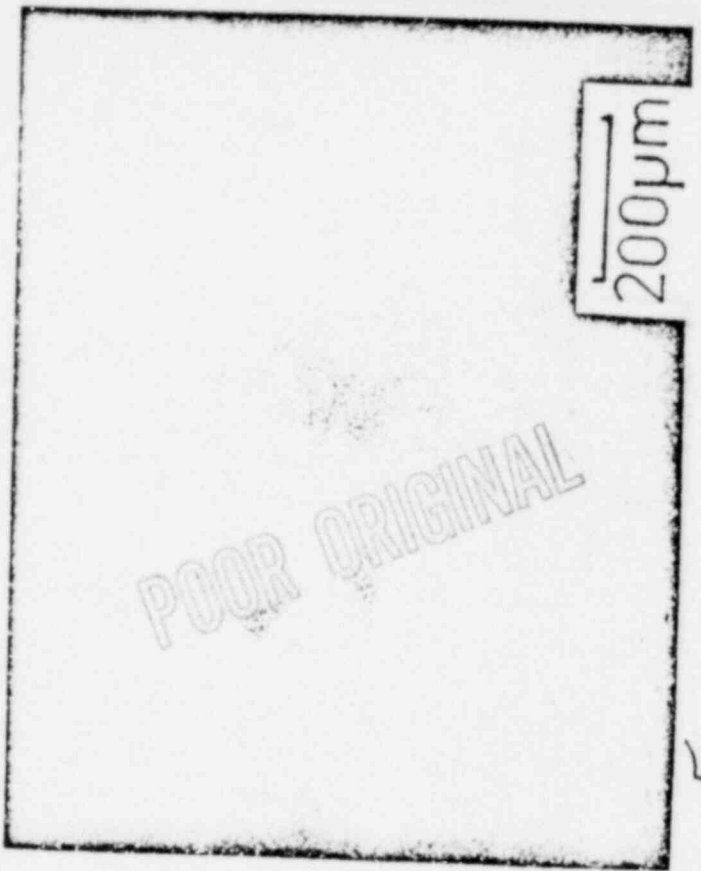
15 Rear surface of new wear ring
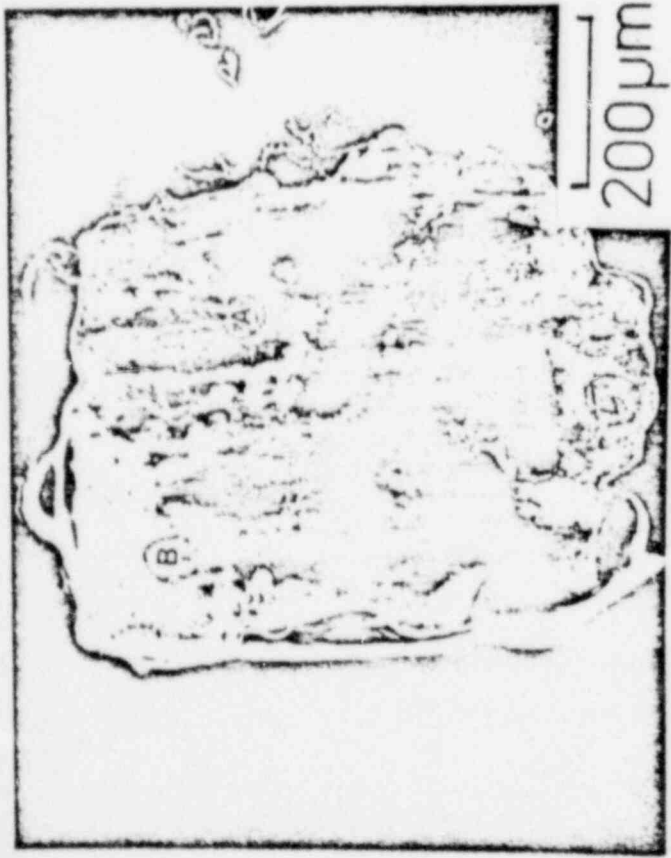
1 (a)
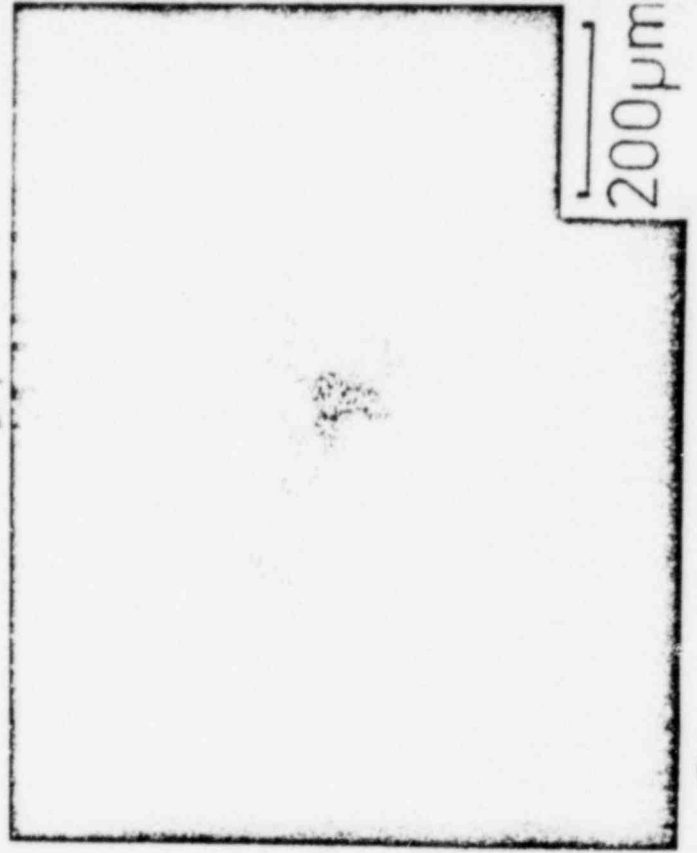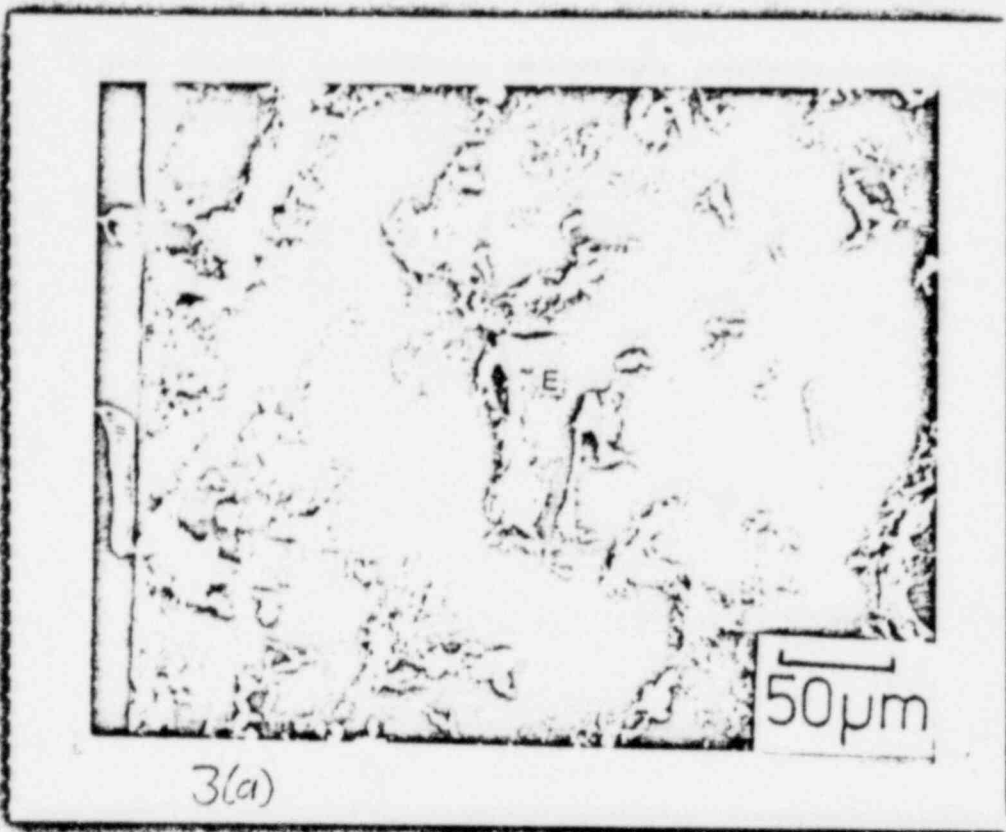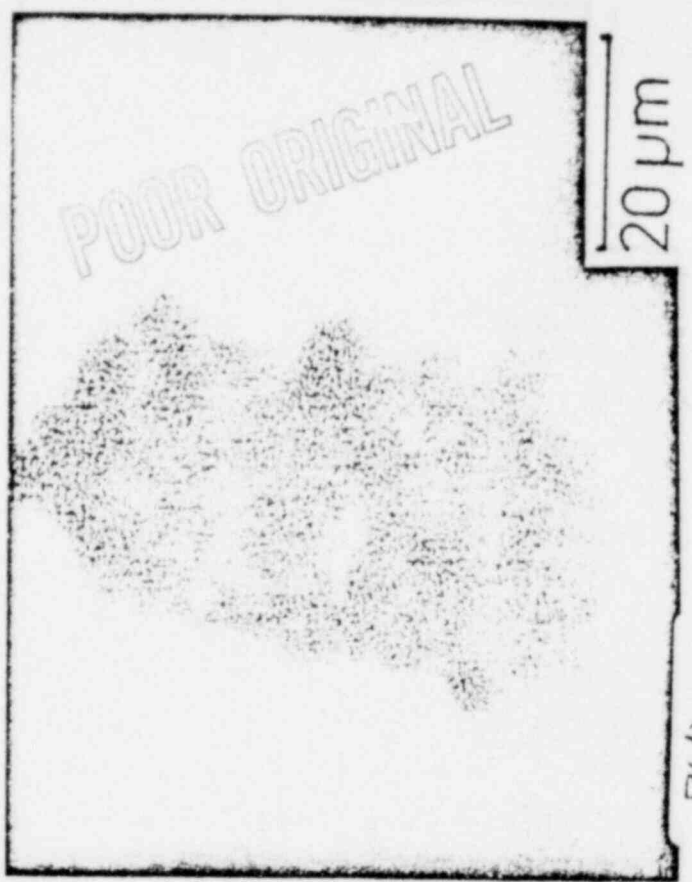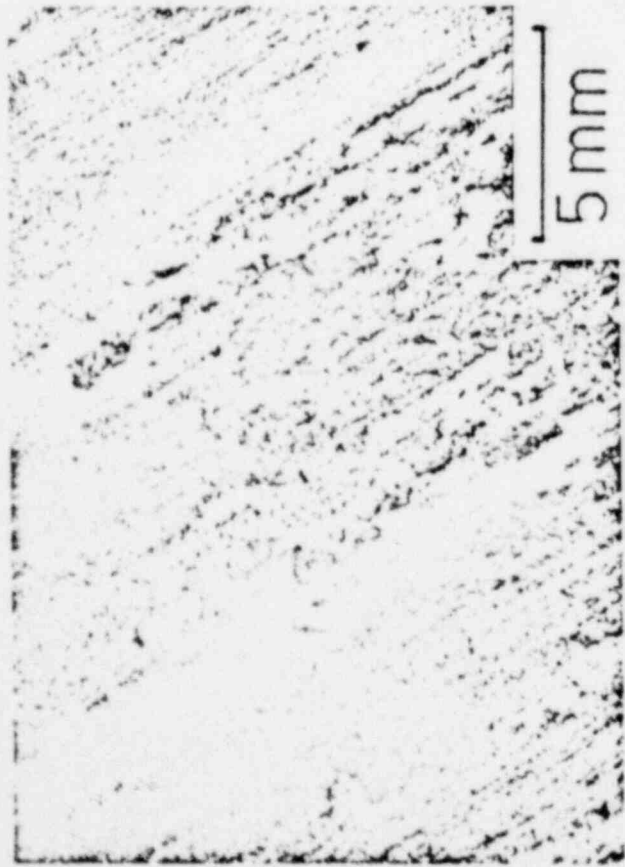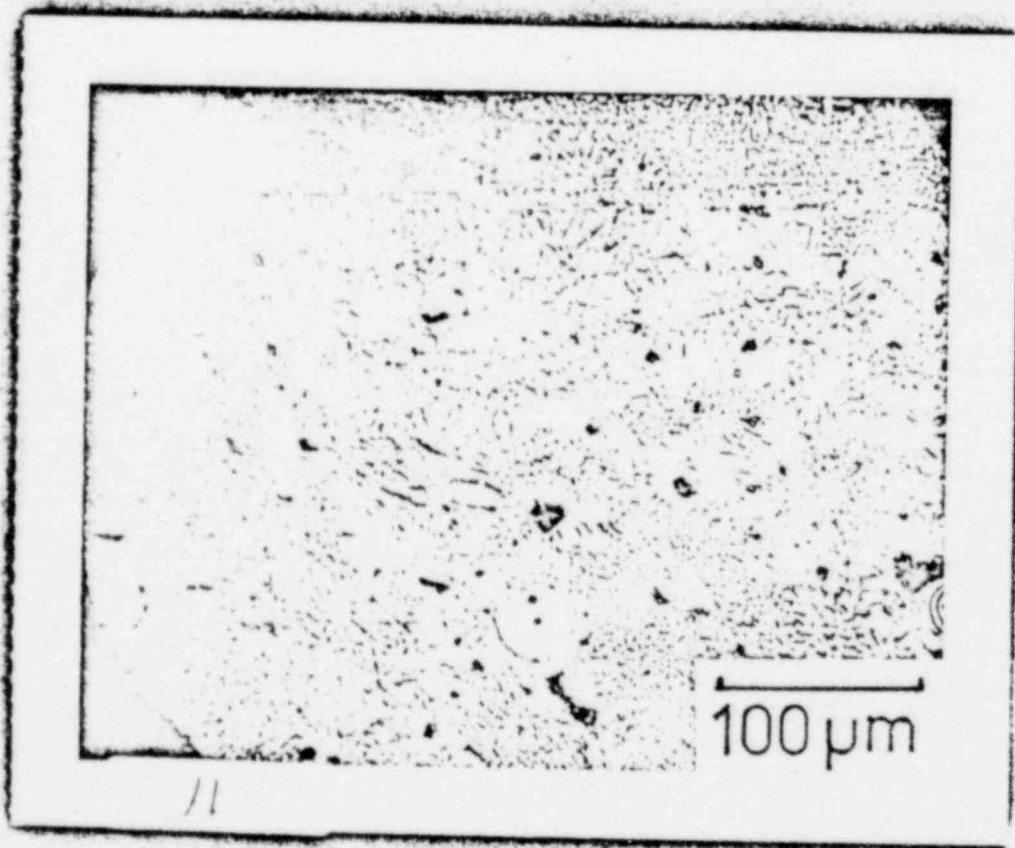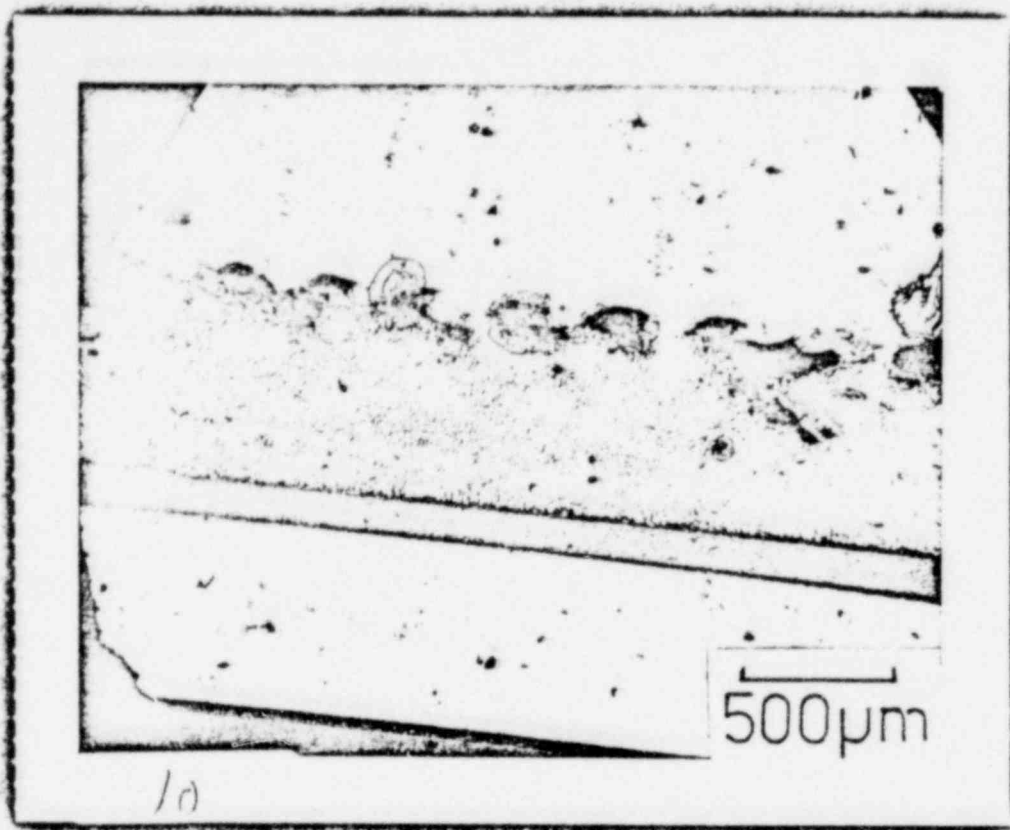
200μm

1 (b)

200μm

2.

200μm

3(a)

50μm

3(b)

50μm

4(b)

200μm

5

200μm

4(a)

200μm

6

20μm

7(c)   7(d)

7(a)   7(b)

20 μm

9

5 mm

8(a)

5 mm

8(b)

10

11

12

A B C D E F

100μm

14

F3 F2 F1

2μm

13

C3 C2 C1

POOR ORIGINAL

1μm

15

B C A

50μm

Appendix B

FORM NBS-148
(12-44)
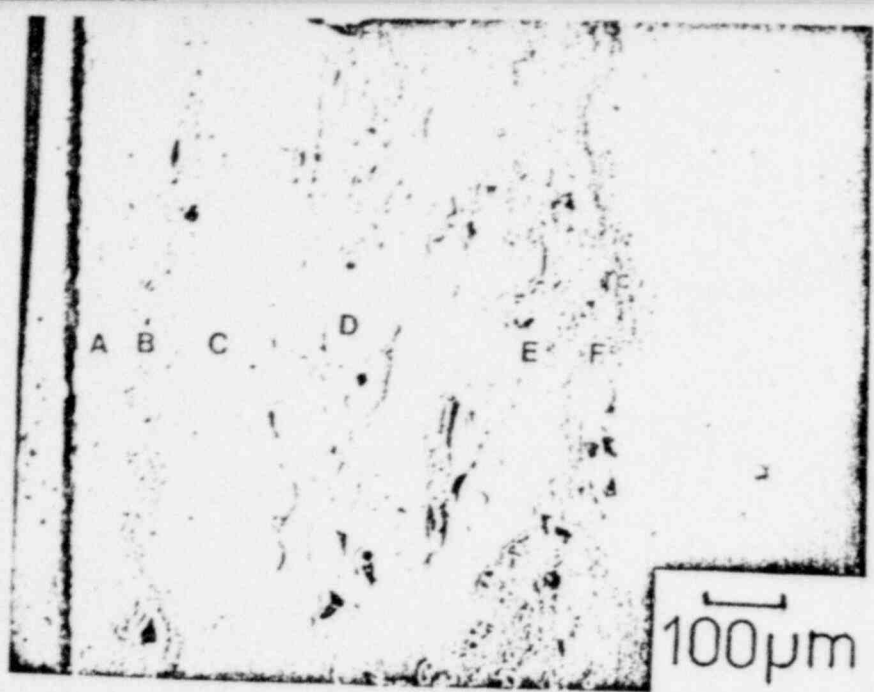
U.S. DEPARTMENT OF COMMERCE
NATIONAL BUREAU OF STANDARDS
WASHINGTON, D.C. 20234

REPORT OF ANALYSIS

Pump Wear Ring from NBS Reactor

Submitted by:

Elio Passaglia, Center for Materials Research

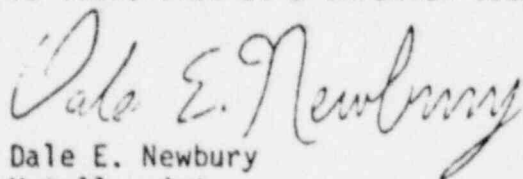Lab. No. 553-39054
Date Submitted: 10-3-79
Analyst: D. E. Newbury

Request: Determine the composition of the surface layer on the outer edge of the wear ring, as marked by the hardness indentations.
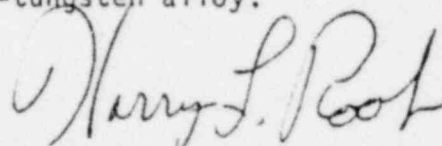
Description of Methods and Results: The Cameca electron microprobe equipped with an energy dispersive x-ray spectrometer was employed for the analysis. The NBS theoretical matrix correction procedure FRAME C was employed for the analysis. The results are estimated to be accurate within ±5% relative.

The surface layer on the wear ring was examined in two locations. In the first location, Figure 1(a), the surface layer was separated by a crack from the substrate. This area was analyzed at the points indicated in Figure 1(b). The analyses, (points 1, 2, and 3) Table 1, reveal that the surface layer consists principally of iron, nickel, chromium, and cobalt. This layer is apparently a cobalt and nickel enriched stainless steel. Just on the inside of the crack (points 4 and 5), the composition is radically different, consist of a nickel-cobalt alloy, with minor amounts of chromium and iron.

A similar result is obtained from the second surface layer, Figure 2. In this region, the surface layer again has the composition of a cobalt and nickel enriched stainless steel (points 6 and 7), Table 1. Locations examined at a depth at 10 um below the surface (points 8 and 9) show a radically different composition. Location 8 is a nickel-cobalt alloy, with minor amount of chromium and iron. Location 9, which corresponds in the image to a distinct phase which appears in relief due to polishing is identified as a chromium-cobalt-nickel-tungsten alloy.

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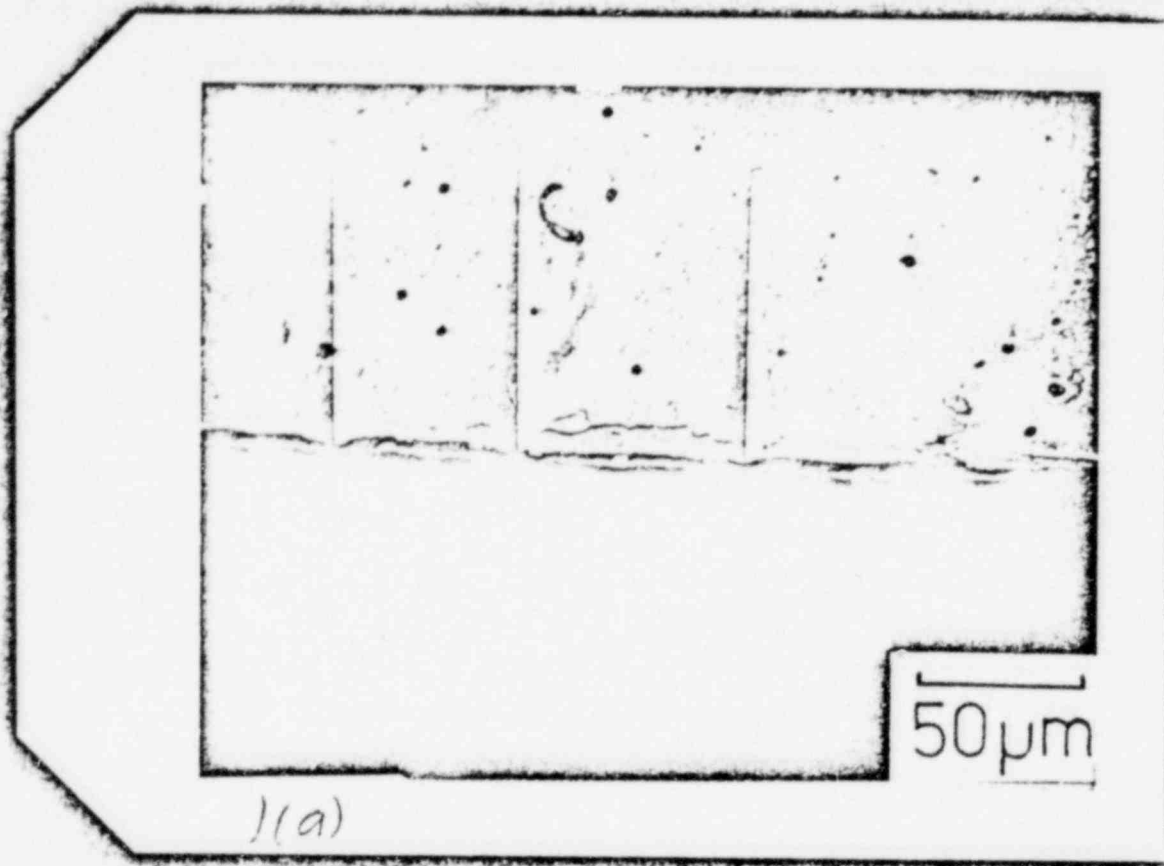Center for Analytical Chemistry
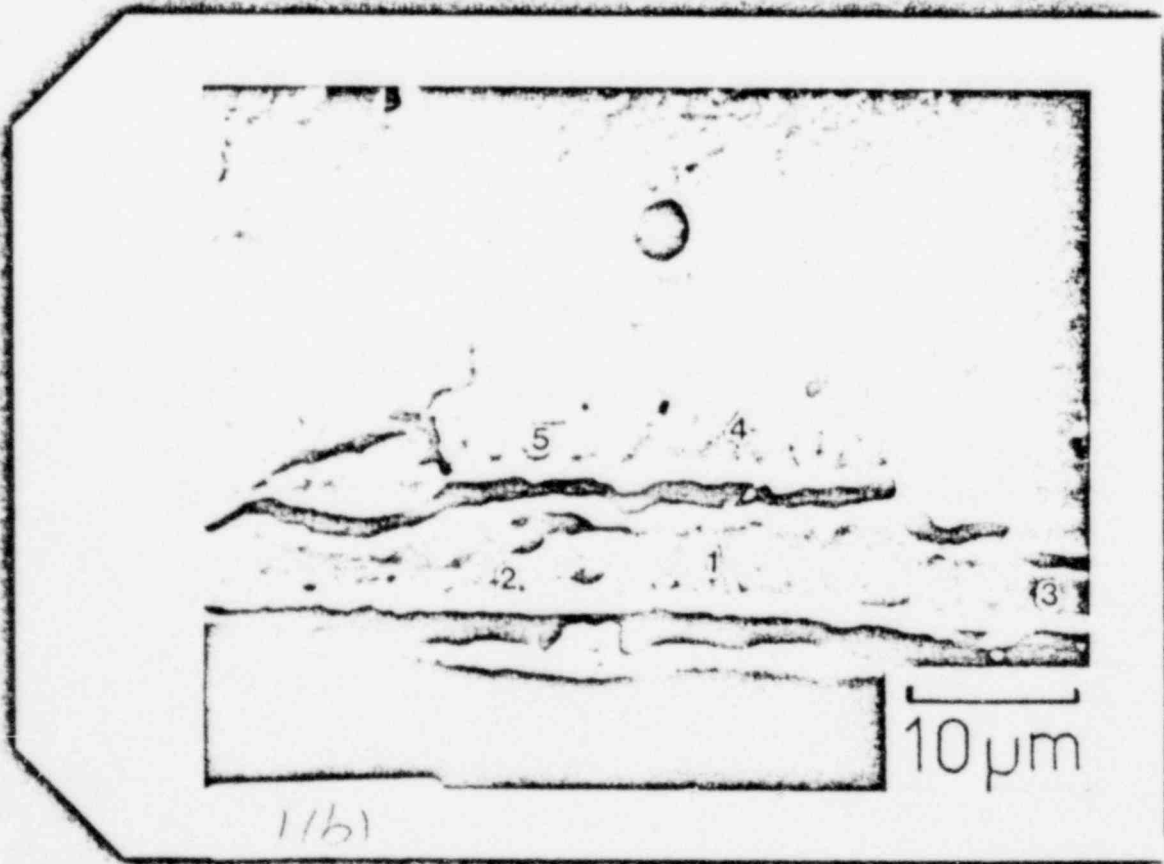
Enclosure

October 5, 1979

Table 1

Analyses of Surface Layer on Wear Ring

Location	Al	Si	W	Mo	Cr	Fe	Co	Ni
1	0.4	1.2	0.8	1.8	18.5	39.2	13.1	25.7
2	0.5	1.1	1.1	1.7	18.4	38.1	13.3	25.3
3	0.4	1.0	1.0	1.7	18.1	37.5	13.5	25.9
4	0.4	5.3	0.3	0.3	8.5	4.1	26.0	56.6
5	0.6	6.0	0.2	0.3	8.8	4.4	14.4	66.2
6	0.3	1.0	0.8	1.9	18.1	44.3	9.8	21.1
7	0.2	1.7	0.7	1.6	18.1	35.6	12.2	26.0
8	0.4	5.3	0	0.3	8.4	3.8	23.7	57.0
9	0.5	2.8	11.1	0.3	42.3	1.1	18.4	9.6

1(a)
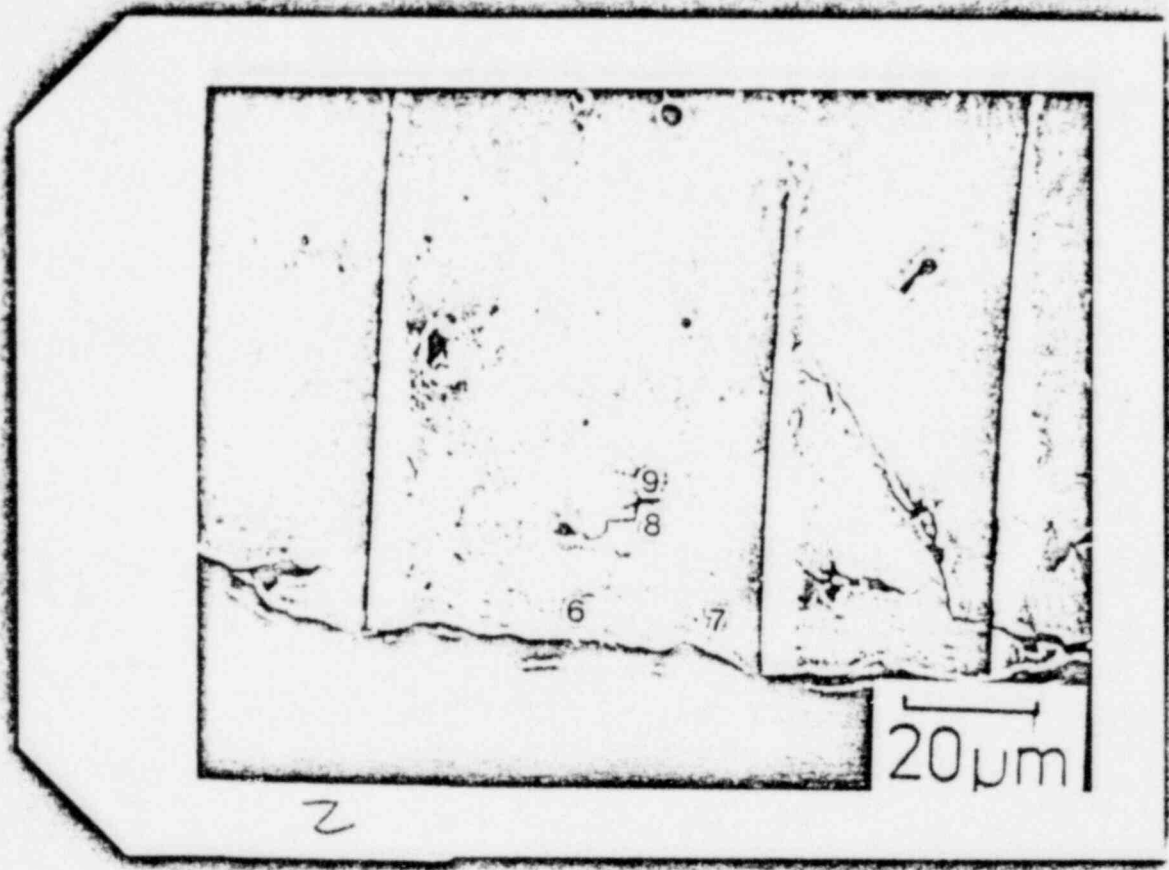
50 µm

1(b)

10 µm

20μm

Appendix C

October 23, 1979

REPORT OF ANALYSIS

To: Elio Passaglia, Deputy Director
Center for Materials Science

Subject: Cobalt and Cobalt-60 in Particles from NBSR Site

Objectives: To determine cobalt contents and cobalt-60 specific activities (in Ci Co-60/g Co-59) in particles found in and near the NBS Reactor.

Methods and Procedures: Six particles were assayed by neutron activation analysis at Oak Ridge National Laboratory (ORNL), using the pneumatic tube facility of the Oak Ridge Research Reactor (ORR). The nuclear reactions whose products were observed are given in Table 1. It is convenient that the experimental parameters are such as to permit measurement of both stable Co-59 and radioactive Co-60 simultaneously with the same detector, and thus the relative specific activity of the particles can be derived with little manipulation of the primary data. Two approaches are available for determining the Co-60 specific activity: the direct method from the Co-61/Co-60m ratio in one spectrum, and the conventional method from the ratio of separate measurements of the Co-60 gamma activity and of the Co-60m produced by neutron activation of stable Co-59.

The samples were assayed for Co-60 activity before and after irradiation with Ge(Li) detectors and with a precision ionization chamber. The results from all measurements agreed (Table 2). Each sample was irradiated in the ORR for 1 or 10 minutes at a flux of 5×10^{13} n/cm²sec, accompanied by a standard of known Co content. A thin Ge photon detector was used to measure the low-energy gamma- and x-rays of Co-60m and Co-61 in the presence of large quantities of high-energy Co-60 radiation. Each sample and its standard were counted at least twice; some were also counted with a conventional Ge(Li) detector. The detector used was connected to a Nuclear Data 50/50 analyzer system, based on a PDP-15 computer. Spectra were recorded on DEC tape, and peaks detected and integrated with the MONSTR program. Corrections were made for random coincidence, dead time, and radioactive decay, including decay during this dead-time-extended counting interval. All other parameters (cross sections, counting efficiency, reactor power variations) were arranged to cancel in the calculation of results and were separately monitored.

The specific activity of the particles can be measured directly by counting the activity of both the Co-60m and Co-61 in the same particle. Then

$$\frac{N_{60}}{N_{59}} = \frac{A_0^{61}}{A_0^{60m}} \; \frac{\sigma_{59} \, \Gamma_{60m} \, \epsilon_{60m}}{\sigma_{60} \, \Gamma_{61} \, \epsilon_{61}}$$

where

$$A_0 \equiv \frac{\lambda \, C \, e^{\lambda t_1}}{(1-e^{-\lambda \gamma})(1-e^{-\lambda \Delta})} \; \frac{e^{\lambda \delta} - 1}{\lambda \delta}$$

with λ = decay constant
  C = net counts in peak
  γ = irradiation time
  Δ = live time of count
  δ = dead time of count
  t_1 = time from end of irradiation to start of count
  σ = activation cross section
  Γ = gammas per decay
  ϵ = detector efficiency

In a separate measurement at ORNL the factor $K = \dfrac{\sigma_{59} \, \Gamma_{60m} \, \epsilon_{60m}}{\sigma_{60} \, \Gamma_{61} \, \epsilon_{61}}$ was determined as 0.2292 ($\pm 10\%$). The specific activity is then given as

$$\text{SA (Ci Co-60/g Co-59)} = (1151.6)(0.2292) \; \frac{A_0^{61}}{A_0^{60m}}$$

$$= 264 \; A_0^{61}/A_0^{60m}$$

Results, Observations, and Conclusions

1. The results are given in Table 3.

2. The same specific activities are given by the conventional and the direct procedures, with the exception of D10A. It was necessary to repackage this particle at ORNL to remove Cl-containing mounting medium. Since the particle was invisibly small, it could not be placed with assurance in the same position as the standard for irradiation and counting; for this reason the direct measurement is preferred. The two particles of highest mass and Co-60 activity, G2 and HEX 7-1, were irradiated for 1/10 as long as the others, with the result that the Co-61 peak was not observable.

3. Two different Co-59 standards gave the same counting rate per microgram of Co to within 1%, and a blank standard showed no Co-60m peak. This assures that the relation between count rate and Co content is properly calibrated.

4.   While six points do not define a distribution well, there is a cluster (G2 and D1) around 1 Ci/g Co, another (RF, G1, and HEX 7-1) between 10 and 20 Ci/g and particle D10A by itself at 335 Ci/g.

5.   Tungsten is present in particles D1 and G2, and not visible in D10A or G1.   Manganese is present in D1 and D10A.

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Attachments

cc: C. W. Reimann
      R. W. Burke

Table 1. Reactions Employed

| Determined | Reaction | $t_{\frac{1}{2}}$ | - - - - Product - - - - |
			Radiations
Stable Co-59	$^{59}Co(n,)^{60m}Co$	10.47 m	58.60 keV
Co-60	$^{60}Co(n,)^{61}Co$	99 m	67.4 keV
Manganese	$^{55}Mn(n,)^{56}Mn$	2.576 h	846.6, 1811.2 keV
Tungsten	$^{186}W(n,)^{187}W$	24.0 h	61.1 keV

Table 2. Cobalt-60 Activities

Sample ID	Ge(Li) NBS 1 Oct	Ge(Li) ORNL 3 Oct	Ion Ch. ORNL 4 Oct	Ge(Li) NBS 16 Oct	Mean	r.s.d.
	- - - - - - Co-60 Activity (μCi) - - - - - -					
G2	---	13.10	13.4	12.85	13.12	2.1%
D1	---	9.27	9.18	9.30	9.25	0.7%
RF	9.00	9.02	8.92	8.97	8.98	0.5%
G1	5.57	5.50	5.53	5.58	5.55	0.7%
HEX 7-1	---	---	238	257	---	---
D 10A	2.63	2.54	2.60	2.59	2.59	1.4%
Co-60 std. 1	3.43	3.35	---	3.40	3.39	1.2%
Co-60 std. 2	3.65	3.55	3.58	3.62	3.60	1.2%
Co-60 std. 3	2.93	2.82	2.88	2.86	2.87	1.6%

Table 3. Results of Measurements

Particle	Weight (μg)*	Wt. Co^{59} (μg)	Co^{59} conc, (percent)	Co^{60} activity (μCi)	Co^{60} Specific Activity (Ci Co^{60}/g Co^{59}) conventional	direct
G2	300.3	14.0	4.7	12.8	0.91	---
D1	66.2	6.95	10.5	9.30	1.34	1.36
RF	28.0	0.88	3.1	8.97	10.2	10.4
G1	15.7	0.48	3.0	5.58	11.6	11.6
HEX 7-1	134.3	13.1	9.7	257	19.6	---
D 10A	0.083*	0.0092	11	2.59	282	335

*Weights by R. L. Zeisler. Sample D 10A is not directly weighable; estimate is from micro-analysis groups' report of 8/30/79.

Appendix D

UNITED STATES DEPARTMENT OF COMMERCE
National Bureau of Standards
Washington, D.C. 20234

October 23, 1979

MEMORANDUM FOR Dr. Elio Passaglio

From: F.J. Schima

Subject: NBS Reactor speck labeled D10A

The photon emissions from the chip labeled D10A have been examined with Ge(Li) and Si(Li) spectrometer systems. The Ge(Li) detector measurement was straightforward as D10A made an excellent point source at the 25cm source position. A very well-characterized efficiency curve is available for this source position. Only the Co-60 radiations at 1173- and 1332-keV were observed. The corresponding activity value was determined to be 0.1044 MBq, at day 264 of 1979. The 1 σ Poissonian error of this one measurement was 0.20 percent and the best estimate of the systematic error limit was 1.25 percent.

On the other hand, the Si(Li) measurement was not as simple. The Mn K-alpha x rays, believed to be from the decay of Fe-55, were observed, but in the presence of the x rays of Cr, Fe, Co and Ni. These latter four x rays are most likely fluoresced by the Co-60 radiations. The measurement was at a non-conventional source distance of 1.8 cm. Also, 0.5 mm thick Be foil absorber was introduced to stop the Co-60 beta particles from entering the Si(Li) detector. After suitable decomposition of the spectrum, the detection rate of the Mn K-alpha x ray was determined to be 0.4807 cps at day 264 of 1979, with a 1 σ Poissonian error of 1.5 percent and an estimated systematic error limit of 7.0 percent, mostly due to the decomposition procedure. This Si(Li) detector setup was calibrated with the Fe-55 x-ray emission standard 4260-B #13, which consists of carrier free Fe-55 (chloride form) evaporated on stainless steel foil. Using $P_{k\omega k} = 0.2796$, the Fe-55 activity for D10A, assuming negligible source self absorbtion, was found to be 1194 Bq, as of day 264 of 1979. The 1 σ Poissonian error is 1.5 percent and the estimate systematic error limit is 10.6 percent. A second Fe-55 reference source was used to calibrate this Si(Li) detector setup and the result agreed to within 2 percent.

These results are the summary of single measurements of D10A on both a Ge(Li) and a Si(Li) detector system. Reproducibility of these values has not yet been established.