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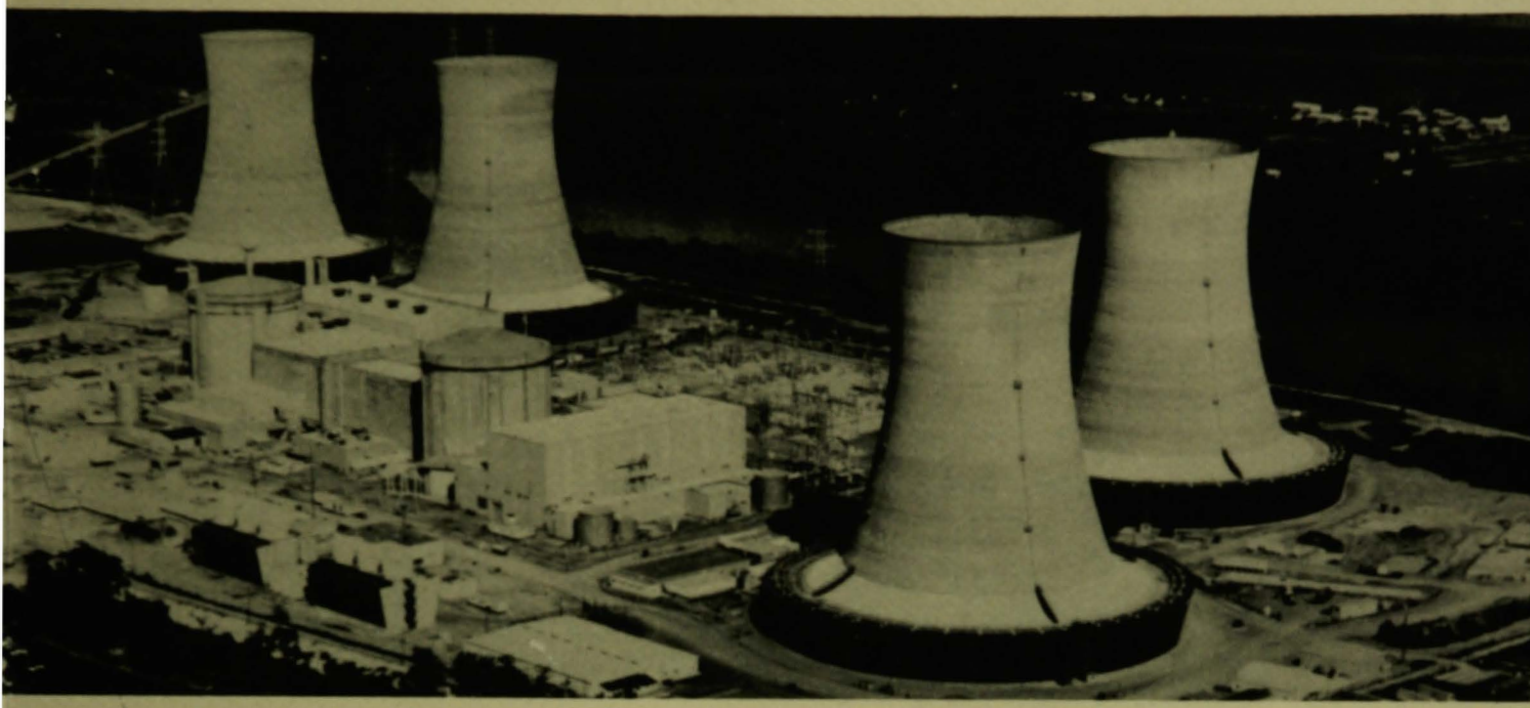

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TMI-2 REACTOR COOLANT SYSTEM SURFACE DEPOSIT EXAMINATION (DRAFT)

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Prepared for the
U.S. Department of Energy
Three Mile Island Operations Office
Under Contract No. DE-AC07-76ID01570

DISCLAIMER

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ABSTRACT

Samples from the TMI-2 reactor coolant system were examined to determine the nature of deposited materials on the surfaces of the samples. Manway cover backing plates were examined from both steam generators and from the pressurizer. A handhole cover liner was examined from the A-loop steam generator and a resistance thermal detector was examined from the A-loop hot leg. Analyses were performed to determine the elemental composition of deposits and the concentrations of deposited radionuclides. The results of surfaces analyses from the coolant loops external to the vessel are compared with the results of earlier studies of samples from the upper plenum region of the vessel. The decontamination studies, which were performed to identify preferred methods for cleaning contaminated surfaces, are also described.

EXECUTIVE SUMMARY

This report summarizes the results obtained by Battelle Columbus Division and EG&G Idaho, Inc. from the examination of samples collected from surfaces in the loops of the TMI-2 reactor coolant system. The samples examined were obtained from two steam generator manway cover backing plates (one each from steam generators A and B) a pressurizer manway backing plate, a handhole cover liner from the A-loop steam generator, and a resistance thermal detector (RTD) from the A-loop hot leg.

The examinations had two objectives. The first objective was to collect data regarding the release and transport of radioactive material in the core uncovery portion of the accident to provide a benchmark for the testing of methods for the analysis of severe accident source terms. It was recognized at the outset, however, that much of the information related to the core uncovery phase of the accident would be masked by conditions occurring after the vessel was refilled with water. The second objective was to develop and test methods for decontaminating surfaces.

Different types of analyses were performed on the various reactor coolant system surface samples. All of the samples were photographed and examined visually. The manway cover baking plates were subjected to the most complete examination, including autoradiography, optical metallography, scanning electron microscopy, gamma spectroscopy, and inductively coupled argon plasma, atomic absorption, and spark source mass spectroscopy. The examination of the handhole cover liner was more limited and was primarily used to verify the identification of the two steam generator manway backing plates with their respective steam generators. The RTD thermowell was subjected to a variety of decontamination steps to determine the relative effectiveness of different decontamination solutions. The elemental and radionuclide compositions of the removed material were also determined.

The physical appearances of the samples varied. The manway backing plate from the A-loop steam generator and the handhole cover liner were similar in appearance. They were essentially gray with patchy white areas of surface deposition. The pressurizer manway backing plate sample was uniformly gray, and the manway backing plate liner from the B-loop steam generator was smooth and shiny. The RTD thermowell was remarkably different in appearance. The tip of the thermowell that extended into the coolant flow is orange.

Metallurgical examination of the manway backing plate samples indicated surface layers ranging from 0 to 12 μm. Although the A steam generator sample had the highest gamma scan activity, it had the thinnest surface layer. The surface layers found on the in-vessel leadscrew samples were much thicker, ranging to ~100 μm. The metallurgical examinations of the backing plate samples showed evidence of indium and tin in addition to the major constituents of the steel.

The elemental analyses of the reactor coolant system (RCS) samples indicate significant transport of silver, aluminum, and tin from the core. Results obtained for tellurium are ambiguous. Although significant quantities of tellurium were found on some of the backing plate samples, none was found on the RTD thermowell, which is closer to the core.

Radioactive inventory analyses indicate that only a small fraction of the original inventory of fission products is currently deposited on reactor coolant system loop surfaces. The amount deposited in the pressurizer is extremely small, which is not surprising since the pressurizer surfaces were never subjected to the extreme accident environment. Based on the analysis of the loop samples, ~0.15% of the core inventory of cesium is currently associated with the surfaces of the coolant loops external to the vessel. This is comparable to the amount estimated to be deposited on upper plenum surfaces.

The analyses of surface samples described in this report provide some insights into the progression of the accident. The implications for source term methodology development and validation are quite limited, however. None of the results of the surface deposition analyses provide unambiguous evidence to confirm or refute current methodology. There are indications of cesium surface reactions, iodine-cesium association, aerosol deposition, transport of control rod materials, and transport of structural materials within the vessel and reactor coolant system. Under more controlled conditions these could have provided a data base for model validation, but a lack of understanding of boundary conditions or the influence of the extended period of submersion have confounded this possibility.

ACKNOWLEDGMENTS

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1. INTRODUCTION

The U.S. Department of Energy (DOE) has supported the examination of surface deposits in the reactor coolant system (RCS) of the Three Mile Island Unit 2 Reactor with two principal objectives. The first was to obtain data that could be used to check or validate the models used in the analysis of severe accidents in light water reactors. Because the TMI-2 accident progressed well into the regime of severe fuel damage in which substantial release and transport of radionuclides would have been expected within the reactor coolant system, examination of surface deposits offered a unique opportunity to benchmark severe accident codes against a full-scale accident. It was recognized at the outset, however, that much of the information relating to the core uncovery phase of the accident would be masked by conditions that had occurred over the long period following the accident in which these surfaces were submerged in water. The second objective was to support the development of methods for accident recovery by investigating different techniques for RCS decontamination. Studies to satisfy the two objectives are closely related, since the type of reaction that occurs between the radioactive species and the coolant system surface determines how difficult it is to decontaminate the surface.

The purpose of this report is to summarize results obtained by Battelle Columbus Division and EG&G Idaho, Inc. (EG&G) from their examination of samples collected from surfaces in the loops of the TMI-2 reactor coolant system. Manway cover backing plates from each of the two steam generators, a manway cover backing plate from the pressurizer, and a handhole cover liner from the A-loop steam generator were examined at Battelle, with some support analyses provided by EG&G and Westinghouse Electric Company. A resistance temperature detector (RTD) from the A-loop hot leg was examined by EG&G. The results of these examinations are compared with results obtained earlier for samples of control rod leadscrews and a leadscrew support tube, which are representative of the upper plenum region above the reactor core. From the combined data base, conclusions are drawn about the characteristics of the transport and deposition of radionuclides that occurred during the period of core uncovery.

1.1 Background

The examination of surface deposits from the TMI-2 Reactor Coolant System is an element of the "TMI-2 Accident Evaluation Program Sample Acquisition and Examination Plan."[1]

Although the March 28, 1979, accident at TMI-2 involved severe damage to the core of the reactor, it had no observable effects on the health and safety of the public in the area. That such a severe core-disruption accident would have no consequent health or safety effects has resulted in the questioning of earlier light water reactor (LWR) safety studies and estimates. In an effort to resolve these questions, several major research programs have been initiated by a variety of organizations concerned with nuclear power plant safety. The U.S. Nuclear Regulatory Commission (NRC) has embarked on a thorough review of reactor safety issues, particularly the causes and effects of core-damage accidents. Industrial organizations have conducted the Industry Degraded Core Rulemaking (IDCOR) Program. The U.S. Department of Energy (DOE) has established the TMI-2 Program to develop technology for recovery from a serious reactor accident and to conduct relevant research and development that will substantially enhance nuclear power plant safety.

Immediately after the TMI-2 accident, four organizations with interests in both plant recovery and accident data acquisition formally agreed to cooperate in these areas. These organizations, commonly referred to as the GEND Group -- GPU Nuclear Corporation, Electric Power Research Institute, Nuclear Regulatory Commission, and Department of Energy, are actively involved in reactor recovery and accident research. At present, DOE is providing a portion of the funds for reactor recovery (in those areas where accident recovery knowledge will be of generic benefit to the U.S. LWR industry) as well as the preponderance of funds for severe accident technical data acquisition (such as the examination of the damaged core).

The work described in this report, which was performed by Battelle Columbus Division, is part of the DOE program to involve private laboratories in the United States in the TMI-2 Accident Evaluation Program. The overall program is coordinated by EG&G for this report. The "TMI-2 Accident Evaluation Program (AEP) Report"[2] defines the program required to implement the DOE assignments and contains the guidelines and requirements for TMI-sample acquisition and examinations.

The already-completed portion of the Sample Acquisition and Evaluation (SA&E) Plan includes in situ measurements and sample acquisition and examinations involving private organizations and state and federal agencies. It has provided the postaccident core and fission product end-state data that indicate the following:

- The current estimate of damage and reconfiguration of the core that occurred during the first day of the accident is as follows:

Core Region	Percent of Core Material
Still standing rod bundle geometry	42
Loose debris (unmelted and previously molten core material mixture) below the cavity in the upper core region the cavity was 26% of the original core volume).	23
Previously molten core material:	35
Retained in core boundary	19
Escaped from core boundary	16

- Some uranium dioxide fuel melting occurred with temperatures up to at least 3,100 K.

- Between 10 and 20 metric tons of core and structural materials relocated into the space between the reactor vessel bottom head and the elliptical flow distributor.

- Fission product retention by the core materials was significant. Fission products released from the core were primarily retained by the reactor coolant system water and the reactor building basement water and concrete.

The examinations described in this report are part of the Reactor Coolant System Fission Product Inventory (RCS FPI) portion of the TMI-2 Accident Evaluation Program (AEP) SA&E Plan. This part of the plan also includes examination of loose deposits from reactor coolant system horizontal and low-point surfaces, in situ video and boroscope surveys of the reactor coolant system vessels and piping for locating core material deposits, and in situ gamma spectrometer surveys for locating uranium in the reactor coolant system. The AEP TMI-2 Program requirements and/or objective for the RCS FPI Sample Acquisition and Examination Program are:

> "All present experience in characterizing the plant indicates relatively small fission product deposition on the reactor system surfaces external to the reactor vessel. However, the primary cooling system surface deposition may provide the only benchmark for the fission product transport during the core degradation phase of the accident. In particular, the retained fission product chemical forms may be related to the fission product chemistry during the transient and the reactor vessel chemical environment during the accident. Analysis of the core material debris deposited in the RCS will be conducted to enhance our understanding of the plant hydraulic conditions during or shortly after the accident.
>
> "For each sample, the following characteristics should be determined:
>
> - Physical appearance,
> - Elemental and chemical composition,
> - Particle size distributions, density, and surface texture, and
> - Radiochemical measurements of retained fission product concentrations and chemical forms."

1.2 Description of System

1.2.1 RCS Description

The TMI-2 reactor coolant system piping and components are shown in Figure 1 and include the following:

- A reactor vessel containing the uranium-fueled core.
- Dual reactor cooling loops (A and B) consisting of candy-cane-shaped hot legs from the reactor vessel upper plenum to the steam generator tops, two single-pass type steam generators (Figure 2), dual (four total) cold legs from the steam generator bottom back to the reactor vessel via the four reactor coolant pumps.
- A pressurizer (Figure 3) connected to the cooling loops by a surge line from the A-loop hot leg to the pressurizer bottom and a spray line from the A-loop cold leg (downstream of pump RC-P-2A) to the pressurizer top.
- Dual core flood tanks connected to the reactor vessel.

1.2.2 Manway Cover Backing Plate Description

The manway cover backing plates are 3/4-in. thick, Type 304 stainless steel discs used to protect the carbon steel manway covers from the reactor coolant in the upper head regions of the pressurizer and steam generators (see Figures 2 and 3). The backing plate inner surfaces that were exposed to the reactor coolant are ~40 cm diameter.

1.2.3 Steam Generator Handhole Cover Liner

The handhole cover liner is also a Type 304 stainless steel plate that was exposed to the reactor coolant system environment in the upper head region of the A-loop steam generator. The diameter of the plate is ~17 cm.

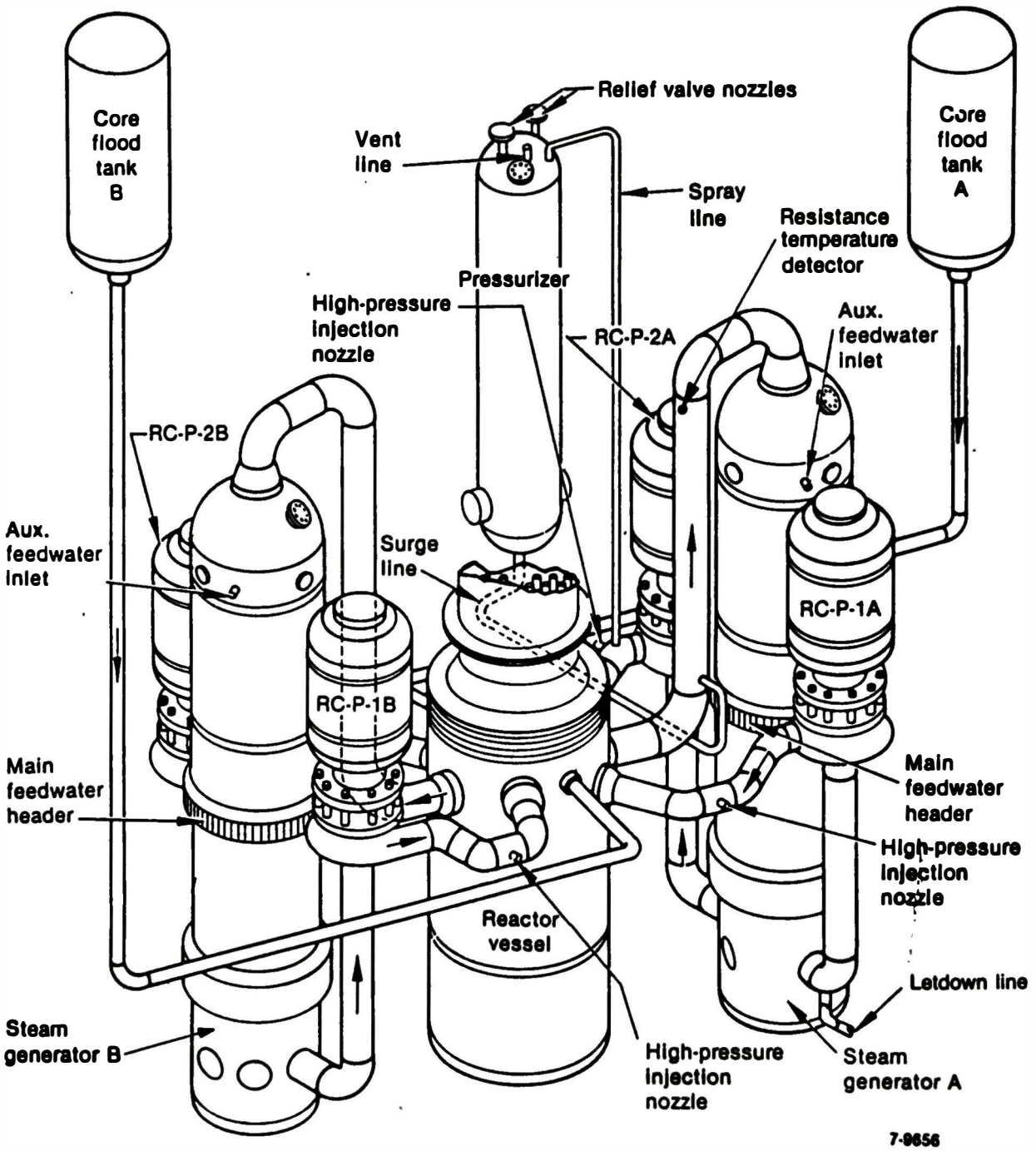


Figure 1. TMI-2 reactor coolant system piping and components.

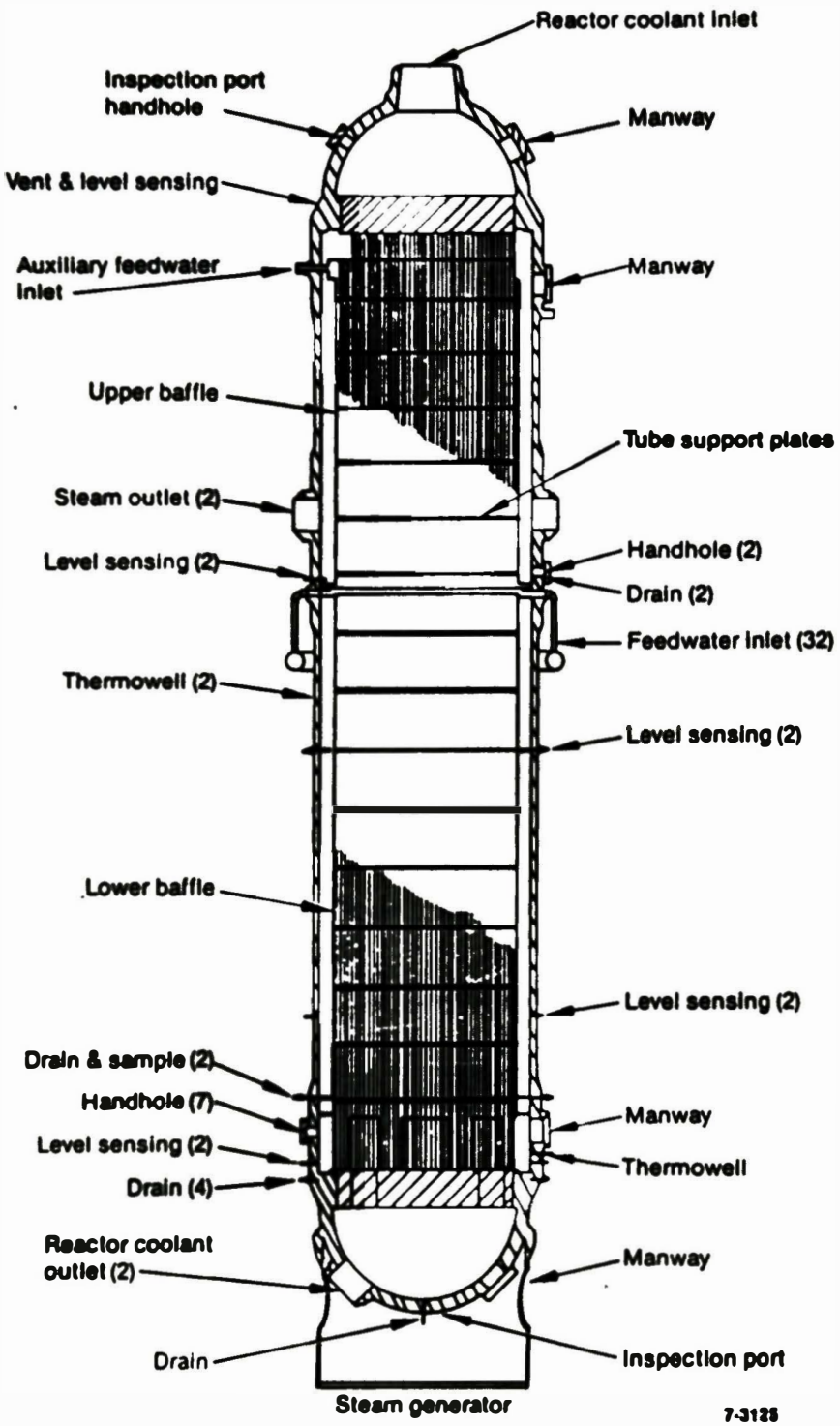


Figure 2. TMI-2 steam generator diagram.

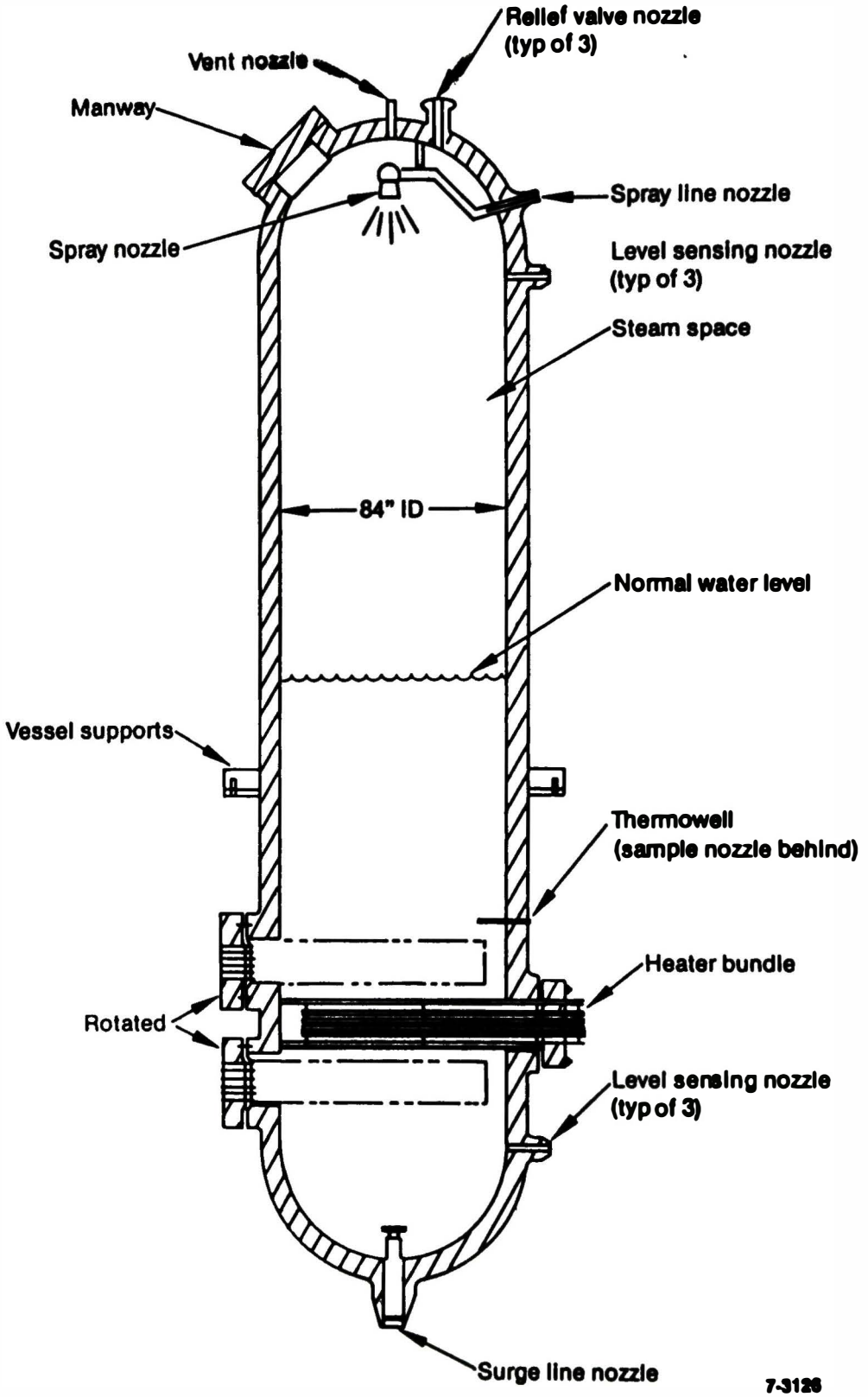


Figure 3. TMI-2 pressurizer layout.

1.2.4 RTD Thermowell

The resistance temperature detector examined had been used to measure primary fluid temperature in the candy cane of the A-loop hot leg. As shown in Figure 4, the tip of the RTD type 304 stainless steel extends into the primary coolant flow stream. The overall length of the thermowell is 19.9 cm from the base nut to the tip, which is ~4.3 cm in length.

1.2.5 Control Rod Leadscrews

Leadscrews are part of the control rod drive system used to raise and lower the control rods in the reactor core region. A diagram of the reactor vessel and internals is shown in Figure 5, which illustrates the location of the leadscrews. The control rods were fully inserted throughout the accident. A schematic of a leadscrew is shown in Figure 6. It comprises four major components: a male coupling which attaches to the control rod assembly, Type 17-4 PH stainless steel, 18 cm long; a lower extension, 304 stainless steel, 193 cm long; a threaded section, 17-4 PH stainless steel, 384 cm long; and an upper extension, 304L stainless steel, 137 cm long. Three leadscrews were removed from the vessel associated with core positions H8 (center), B8 (periphery of core at A-loop hot leg) and E9 (midradius) (Figure 7). The leadscrew support tube from the H8 position was also removed for examination. The leadscrew support tube, composed of Type 304 stainless steel, is located in the dome region of the vessel above the upper plenum.

1.3 TMI-2 Accident and Recovery Period Environment

The first 16 h of the TMI-2 accident is the period of main interest to accident analysis researchers. In particular, from 1 h 40 min, when reactor coolant pumps 1A and 2A were turned off, until 3 h 20 min, when high pressure injection was initiated, the core was uncovered for an extended period and underwent heatup, degradation, and fission product release. The principal purpose of the examination of the RCS samples has been to search for signatures of this period that could help to better

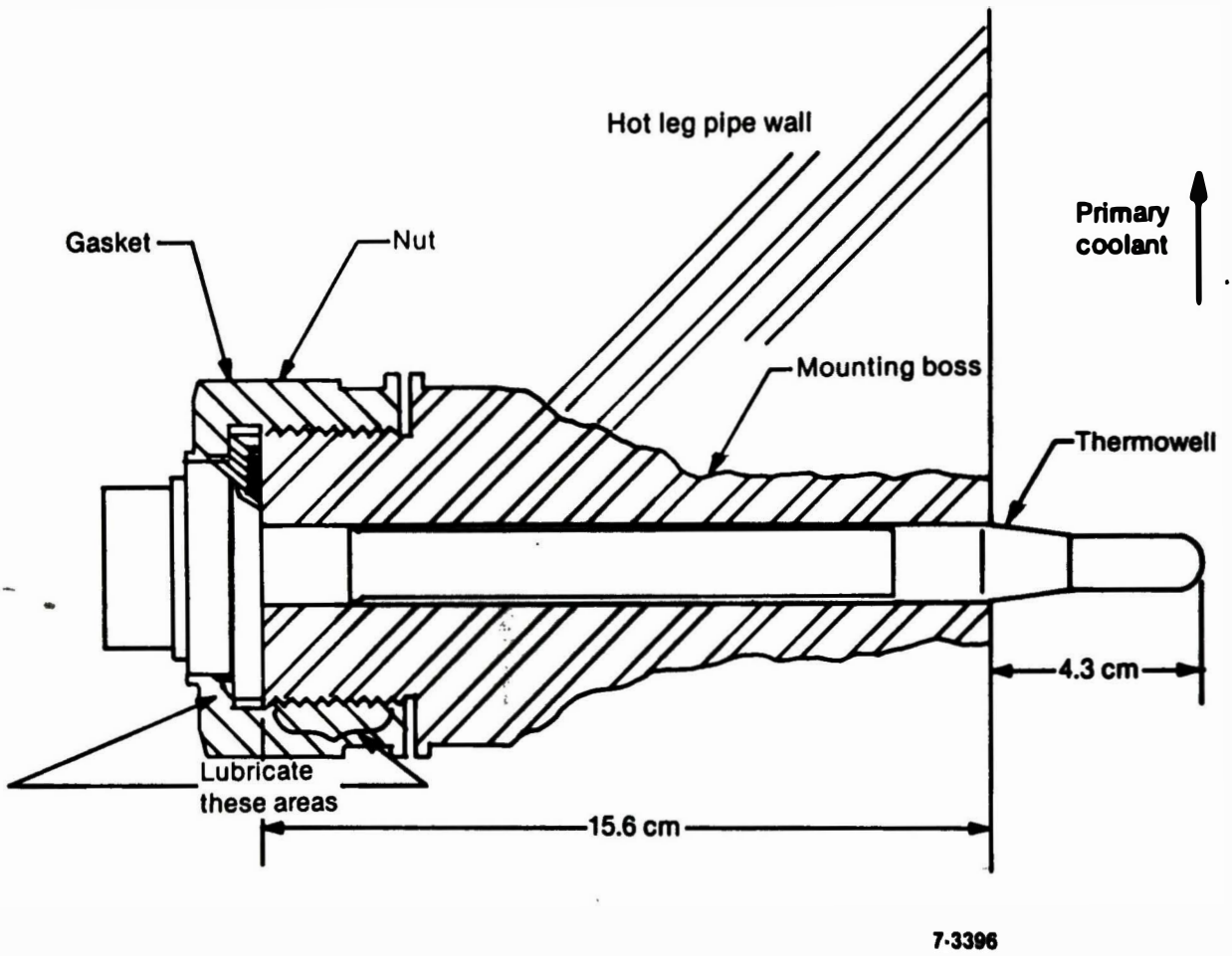


Figure 4. Thermowell installation into the plant piping.

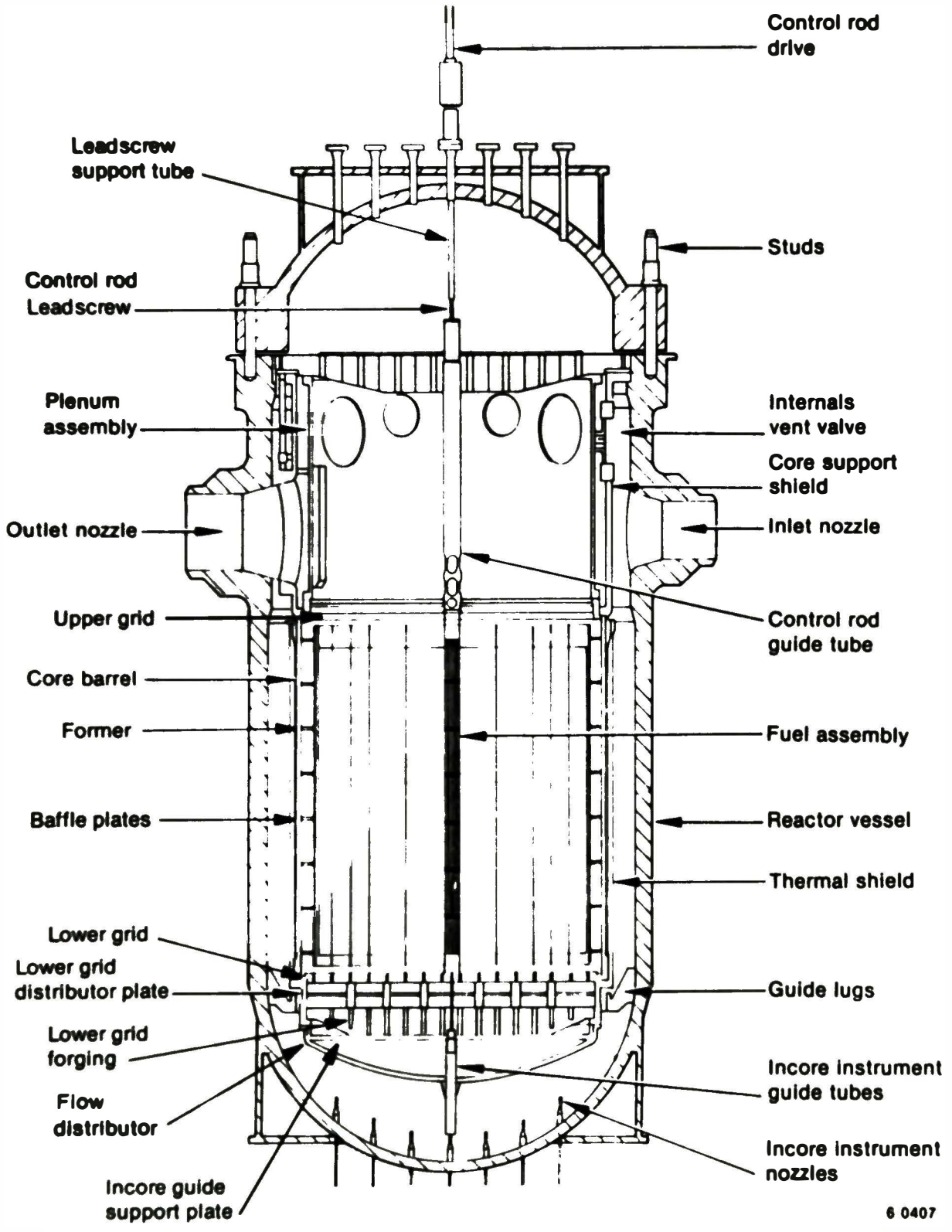


Figure 5 General arrangement of MI-2 reactor vessel and internals.

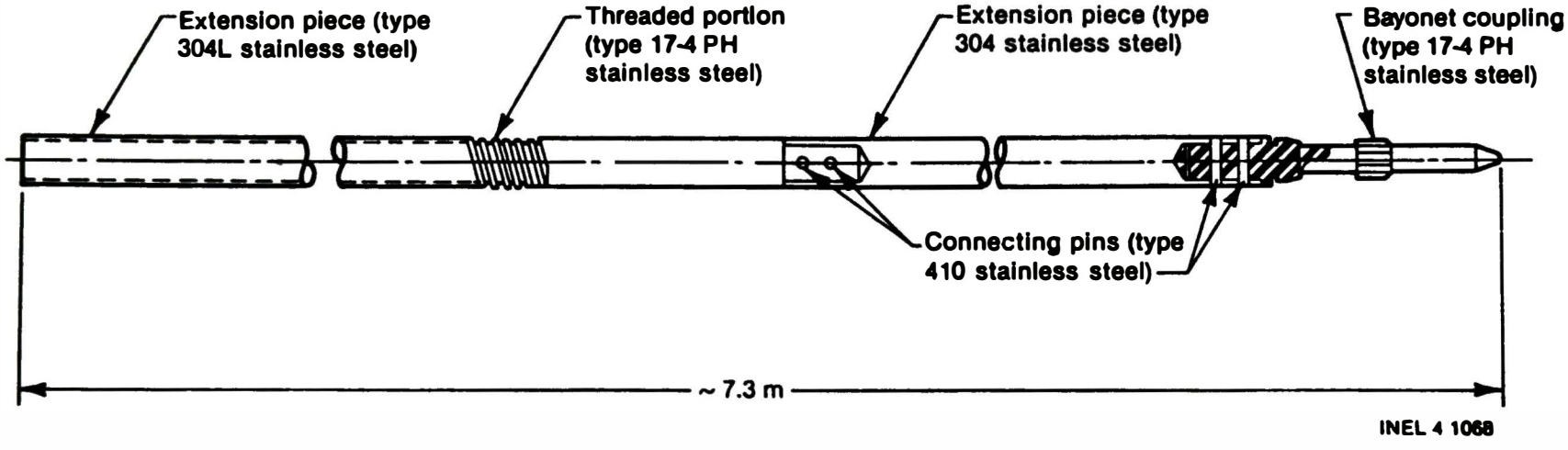


Figure 6. A schematic of a control rod drive leadscrew.

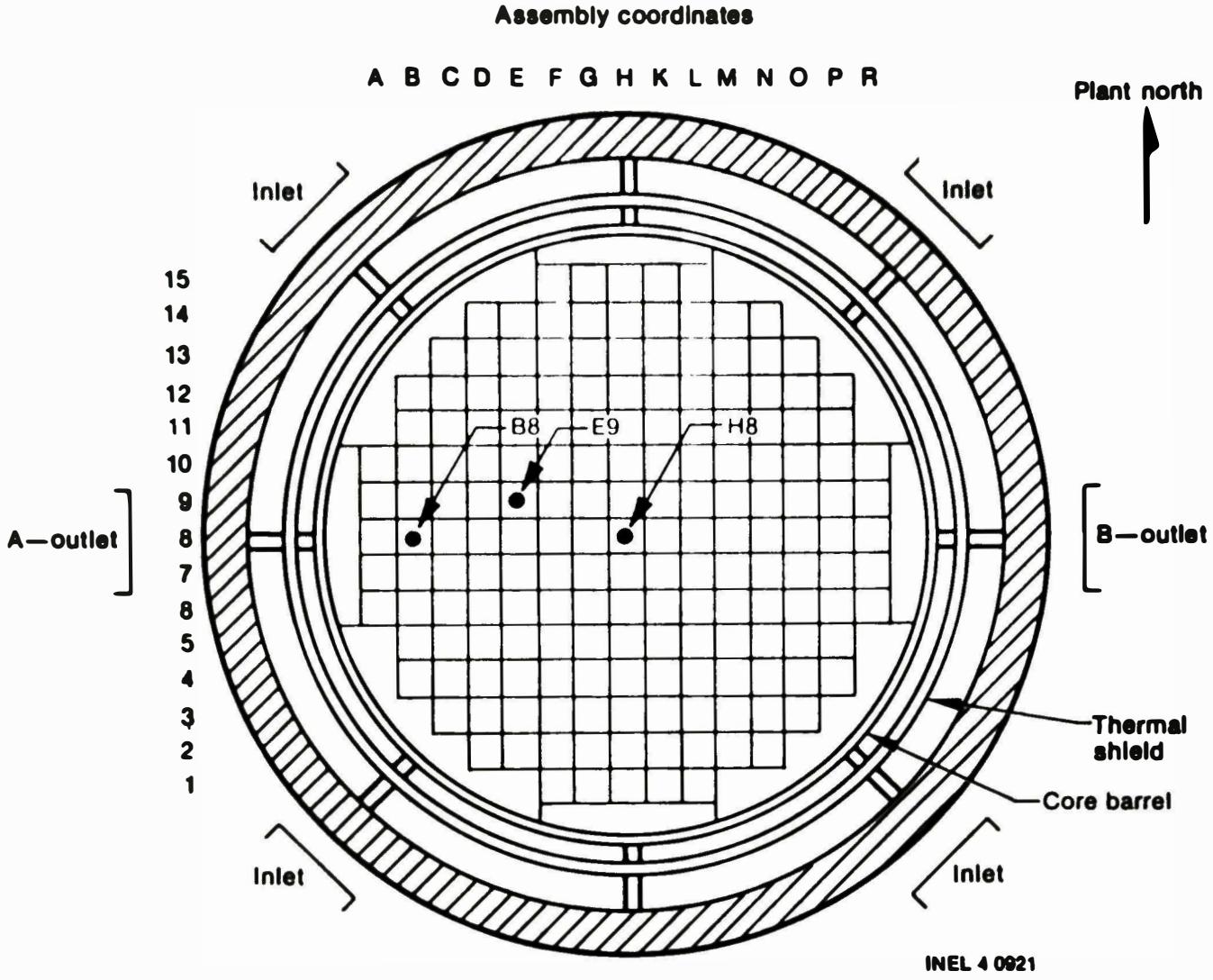


Figure 7. H8, B8, and E9 leadscrew locations in the TMI-2 core.

understand the phenomena occurring during the process of severe core degradation. It is recognized that much of the evidence that once existed on these plates has been altered by subsequent events, in particular an extended period of submersion in highly contaminated water.

The steam generator backing plates and handhole cover were taken from the tops of the A and B steam generators. Similarly, the RTD thermowell was removed from the candy cane of the A-loop hot leg. These regions were probably uncovered throughout the entire time period from 1 h 40 min until reactor coolant pump A was successfully restarted at 15 h 50 min. During the initial phase of core uncovery, the block valve to the stuck-open pilot operated relief valve was in the open position and the primary system was depressurizing through the relief line. It is unlikely that there was significant release of fission products from the fuel at this time, however. At 2 h 20 min the block valve was closed and remained closed until ~3 h 12 min, shortly before initiation of high pressure injection. During this period, when most of the release of fission products from the fuel is believed to have occurred, there was no loss from the primary coolant system. Some steam was flowing to each of the steam generators where it was condensed, probably carrying fission product vapors and aerosols with it.

By 3 h the temperatures at the top of the hot legs had risen to ~700 K where they remained until 10 h. Over much of this time period the hot legs and steam generators were apparently isolated from the core region. After high pressure injection, the lower portions of the hot legs were apparently filled with water, which was subcooled until 11 h. At this time, the A-leg temperature dropped to saturation indicating the partial reestablishment of steam flow through the hydrogen trapped in the region. The A-leg temperature subsequently increased by ~310 K, indicating that flow had again stopped. At 15 h 50 min water flow was reestablished in both loops.

The pressurizer manway cover is at the very top of the pressurizer. As discussed earlier, the block valve was closed during much of the period of fission product release from the fuel. In addition, the pressurizer contained a substantial quantity of water throughout the accident. Any fission products deposited on the cover plate either passed through the water pool in bubbles, were volatilized from the pool, or were deposited directly from the water while the pressurizer was completely filled.

Before and after water flow was reestablished (15 h 50 min), additional events occurred that affected the character and distribution of core materials and fission products which escaped from the reactor vessel to the reactor coolant system. The most significant events include the following:

- Reactor coolant pump RC-P-2B was energized from 174 to 192 min after accident initiation. This event is believed to have reflooded the overheated core region, fragmenting most of the standing fuel in the upper core region and creating the upper core region cavity, and causing circulation of core material particles and fission products throughout the B-loop components and also into the pressurizer.

- A reactor coolant sample taken at 283 min contained >500 Ci/mL gross activity.

- Forced circulation cooling of the reactor at 949 min (15 h 49 min) through the A-loop with reactor coolant pump RC-P-1A. This action flushed the noncondensable gases from the A-loop steam generator and hot leg upper regions.

- A reactor coolant sample taken at 36 h 15 min measured 1000 R/h on contact.

- At 6 days after accident initiation, the noncondensable gas bubble in the B-loop upper region appeared to be gone.

- Natural circulation cooling of the reactor commenced 30 days and 10 h after accident initiation using both coolant loops. Steam generator B was later isolated.

- Reactor coolant water cleanup using the SES/EPICOR-II system commenced 2 years and 106 days (7/12/81) after accident initiation and included cleanup of an equivalent of four reactor coolant system volumes of reactor coolant water.

- From the time of the accident until the manway cover was removed, the reactor coolant had been a water solution with the following target specifications.

  - pH 7.5 to 7.7
  - boron <4,350 ppm
  - buffer NaOH

- The RCS liquid volume was drawn down for initial reactor disassembly in July 1982 uncovering the pressurizer and steam generator upper regions.

- The reactor coolant system surface samples described in this report were collected prior to defueling the reactor.

2. EXAMINATION AND RESULTS

2.1 Examination Program

Five specimens were collected to characterize the depositions of radioactive materials on the surfaces of the reactor coolant system. Three manway cover backing plates that interface directly with the reactor coolant system fluid were retrieved, one from each of the steam generators and one from the pressurizer. From these specimens, four samples, each ~2 cm^2 in area, were taken from the center of each quadrant. The sample from Quadrant IV of the A steam generator was offset from the center of the quadrant, however, to include a region of high surface deposition. These samples were subjected to a variety of tests, as indicated in Table 1, to determine the elemental deposition, radionuclide deposition, and physical characteristics of the surface deposits.

The fourth specimen, the steam generator handhole cover liner, was analyzed primarily to confirm that the A-loop and B-loop manway cover backing plates had not been mislabeled in handling. The liner was subjected only to visual examination and gamma scanning. The physical appearance and pattern of deposited radionuclides on the liner were quite similar to those of the backing plate from the A-loop steam generator, indicating that the backing plates had been labeled properly.

The fifth specimen, the resistance temperature detector from the top of the A-loop candy cane, was decontaminated and the concentrations of deposited radionuclides were determined using gamma spectroscopy. Elemental analyses were also performed.

TABLE 1. EXAMINATIONS PERFORMED

	Cover Backing Plate A				Cover Backing Plate B				Manway Cover Backing Plate				Generator Hand-hole Liner	A-loop RTD Thermowell
	A1	A2	A3	A4	B1	B2	B3	B4	P1	P2	P3	P4		
Photography	x	x	x	x	x	x	x	x	x	x	x	x	x	x
Gamma Scan	x	x	x	x	x	x	x	x	x	x	x	x	x	x
Auto-radiography	x	x	x	x	x	x	x	x	x	x	x	x		
Optical Metallography	x	x	x	x	x	x	x	x	x	x	x	x		
SEM	x	x	x	x	x	x	x	x	x	x	x	x		
Gamma Spectroscopy	x	x	x	x	x	x	x	x	x	x	x	x		x
Sr-90/I-129	x	x	x	x	x	x	x	x	x	x	x	x		
Fissile/ Fertile[a]	x	x	x	x	x	x	x	x	x	x	x	x	x	
ICAP	x	x	x	x	x	x	x	x	x	x	x	x		
AAS	x	x	x	x	x	x	x	x	x	x	x	x		
Atomic Emission Spectroscopy														
Spark Source Mass Spectrometry	x	x	x	x	x	x	x	x	x	x	x	x		
Surface Deposit Removal	x	x	x	x	x	x	x	x	x	x	x	x		

a. Measurements performed by GPU.[4]

2.2 Summary Descriptions of Sample Preparation and Examination Techniques

2.2.1 Photography

The surface features of all of the specimens were documented by means of photographs in black and white. The RTD thermowell, which exhibited a bright orange-yellow appearance, was photographed in color.

2.2.2 Gamma Spectrometer Scanning

Gamma scans were performed for each of the RCS samples. Each of the manway backing plates was gamma-ray scanned along one complete diameter using the Battelle Waste Drum Counting System (WDCS).[5]

Two gamma-ray scans were performed across the diameter of the active surface of the steam generator handhole cover liner. The samples were positioned in contact with a 2-in. thick lead collimation shield. Through the center of the shield was a 1.3-cm diameter hole which permitted a small area of the liner (1.3 cm^2) to be gamma-ray counted. The detector, positioned on the other side of the shield, consisted of a 15% efficient [relative to a 3 x 3 in. NaI (Tl) detector] intrinsic germanium detector with a FWHM resolution of 2.4 keV at an energy of 1,332 keV. The scans were started at the edge of the liner and data were collected at 1-cm intervals. The relative gamma-ray activities of the most abundant isotopes (Cs-137 and Co-60) were determined as a function of distance along the axis of translation of the sample.

The gamma scan analysis was performed along the length of the RTD thermowell from the nut to the tip for a distance of 20 cm at 1-cm intervals. Two scans were performed with the thermowell rotated 180 degrees.

2.2.3 Autoradiography

To determine the distribution of gamma energy-emitting isotopes on the surface of the manway cover backing plates, the plates were autoradiographed. Each plate was covered on its active surface with a thin (0.1 mm) polyethylene film to prevent contamination of the radiographic film holder. A 35 x 43 cm X-ray film was contained in the film holder, which was then placed in intimate contact with each plate and exposed for 45 s (Plate P) or 60 s (Plates A and B).

2.2.4 Metallurgical Sample Preparation

Metallurgical examinations were performed on the manway cover backing plate samples.

Samples were obtained from the center of each quadrant of the plates, except for quadrant IV of Plate A. The surface deposit in quadrant IV, Plate A, occurred tangential to the center, so sampling for this quadrant shifted accordingly to include these surface deposits. Following completion of the nondestructive examinations, the plates were sectioned to obtain samples for chemical, radiochemical, and microscopic examination. A 1.59-cm wide strip was cut through the plates along the center of the quadrants of each plate. After the two strips were cut from each plate, a 0.63-cm wide section was obtained for optical and electron microscopy and a 1.27-cm wide section for chemical and radiochemical analyses. The samples were assigned letter/number designations corresponding to the origin, as shown in Table 1.

2.2.5 Optical Metallography

The manway cover backing plate metallography samples were mounted, ground, and polished using SiC and Al_2O_3 abrasives. The samples were examined in cross-section with an Olympus microscope along the edge containing the surface deposit. Black and white photographs were obtained from each sample.

2.2.6 Scanning Electron Microscopy

To identify the surface layers on the manway cover backing plate metallographic samples, the samples were examined with an ISI Model 100 scanning electron microscope equipped with a Tracor Northern energy dispersive system (EDS) analyzer.

2.2.7 Radiochemical Sample Preparation

After a review of potential dissolution methods, a destructive leach with radioactive tracers was chosen as the best method for the analysis of the surface deposits of the manway cover backing plate samples. In this method, the sample was supported such that the surface to be leached was immersed in a solution containing both carrier and radioactive tracers. Stable strontium and iodine were added as carrier material and Sr-85 and I-131 as tracers. Following equilibration of the carrier and tracer with the surface deposited species (this was determined to require 5-10 min), an unheated leach was begun. Multiple leaches were performed on samples A3 and P3 to determine the leach time required for the samples. It was determined that a cold leach period of ~4 h was required to place ≤97% of the measurable activity in solution. The samples were then slowly heated and the I-129 volatilized and trapped in dilute NaOH. Visual examination of the metal coupon after leaching indicated that all surface material had been removed from the leached surface. Although some redeposition was expected to occur, gross and isotopic radiation measurements indicated that the bulk of the activity had been removed. Upon completion of the leach process, the leach solution was diluted to 60 mL and stored in marked vials for subsequent chemical and radiochemical analyses.

Decontamination of the RTD thermowell surfaces is described in Section 2.2.13. The decontamination solutions were collected and used to perform radiochemistry for the RTD thermowell.

2.2.8 Sr-90 and I-129 Analyses

For the manway cover backing plates the bulk diluted solutions were analyzed in a known geometry via gamma spectroscopy. Subsequently, aliquots were removed from the bulk solution for Sr-90 and I-129 analyses. A 5-mL sample was removed and subjected to a standard $SrCO_3$ precipitation separation[6] followed by gamma spectroscopy for tracer measurements and liquid scintillation analysis (Packard TRICARB model). The radioiodine trapped in the NaOH was separated, analyzed by gamma spectroscopy for a yield determination, and subsequently activated using the Advanced Test Reactor (ATR) facility at the Idaho National Engineering Laboratory (INEL).

2.2.9 Fissile/Fertile Analysis

After removing the manway cover backing plates and handhole cover liner from the reactor coolant system, GPU collected scrape samples from the surfaces. Radiochemical analysis was used to determine the isotopic content of the transuranic isotopes in the samples. By comparing the ratios of plutonium isotopes to uranium, the burnup of the fuel deposited on the reactor coolant system surfaces can be inferred. A crude assessment can then be made of the point of origin of the fuel from within the core.

2.2.10 Inductively Coupled Argon Plasma Spectroscopy (ICAP) and Atomic Absorption Spectroscopy (AAS) Analysis

Approximately 35 mL of the bulk diluted solutions were analyzed by ICAP for the elements B, Na, Mg, Al, Si, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, Mo, Ag, Cd, Sn, Sb, Te, and Pb using a computer-controlled Instrumentation Laboratories Model 200 Inductively Coupled Plasma Emission Spectrograph. In addition, cesium was determined by AAS, which is more sensitive than ICAP for this element. The IL951 Atomic Absorption Spectrophotometer features double beam, dual-wavelength measurement with selection of integration time plus peak area or height analysis.

2.2.11 Spark Source Mass Spectrometric (SSMS) Analysis

Aliquots (5 mL) from the original diluted solutions from the manway cover plates were analyzed with an AEI Model MS702R SSMS instrument.

2.2.12 Atomic Emission Spectroscopy

The RTD Thermowell surface deposit samples were analyzed for elemental composition by atomic emission spectroscopy with arc-spark excitation. The spectrographic analyses were done on a 3.4 m Javel-Ash, Ebert mount spectrograph with a Custom Varisource Excitation Unit.

2.2.13 RTD Thermowell Surface Deposit Removal

The surface deposits on the RTD thermowell were removed by means of decontamination solutions.

2.2.13.1 Decontamination of the RTD Thermowell Tip (~4.3 cm). The decontamination of the thermowell tip was performed as defined in Reference 7. The solutions used, temperature, and time of each immersion are listed in Table 2. Also given are the gross radiation measurement readings performed on the RTD thermowell after each decontamination solution was removed. The gross radiation readings are semiquantitative and are probably not more accurate than a factor of 2. Originally, six decontaminations were planned. The thermowell tip measured ~3 Rads/h at near contact upon completion of decontamination 6, so decontaminations 7-12 were added. Upon completion of the final decontamination, No. 12, the RTD well was returned for a measurement of retained surface activity by gamma analysis. About 0.1% of the Cs-137 remained on the thermowell; no other radionuclides were detected.

2.2.13.2 Decontamination of the Thermowell Body. As a result of the difficulty in decontaminating the RTD thermowell tip, it was decided to serially decontaminate the top 15 cm body of the RTD in two sections. During decontamination of the body of the RTD thermowell, the tip was

TABLE 2. RTD THERMOWELL TIP SERIAL DECONTAMINATION SOLUTIONS

Decontamination No.	Solution Used	Time (h)	Temp (K)	Radiation Reading Decontamination (Rads/h)
1	Demineralized Water	24	294	4.5
2	Borated water-3500 ppm boron as boric acid, pH=7.5 with 1% Triton x-100 surfactant	24	294	3.8
3	Sodium carbonate/hydrogen peroxide 5.0 wt% Na_2Co_3 and 1 wt% H_2O_2	1	294	3.8
4	Two step APC a. 10% NaOH - 3% $KMNO_4$	1	294	--
	b. Oxalic acid (25 g/L and dibasic ammonium citrate 50 g/L	1	294	3.0
5	Nitric/hydrofluoric acid 10 wt% HNO_3-0.1 M HF	1	294	3.0
6	Nitric/hydrofluoric acid	2	294	3.0
7	Nitric/hydrofluoric acid	3	366	2.5
8	Nitric/hydrofluoric acid 40 wt% HNO_3	3	366	2.8
9	Hydrochloric acid (IN)	1	366	0.5
10	Hydrochloric acid (IN)	1	366	0.2
11	Hydrochloric acid (IN)	1	366	<0.2
12	Hydrochloric acid (IN)	1	366	<0.15

protected with acid-resistant tape to prevent further etching. Also, when one portion of the body was decontaminated, the other portion was protected. The decontaminations performed on the body of the RTD are listed in Table 3. Upon completion of the RTD thermowell body decontamination, the thermowell was again gamma scanned for retained activity.

2.3 Physical Appearance

2.3.1 Manway Cover Backing Plates, Handhole Cover Liner, and Thermowell

Each of the manway cover backing plates was visually examined through the hot cell window. The surface features were documented by means of black and white photographs (shown in Figures 8-10). In general, the plates exhibited no unusual features on their surfaces. Plate P appeared to be covered by a uniform gray surface layer. Plate A, on the other hand, appeared to have only patchy white areas of surface deposits. The surface of Plate B was smooth and shiny. These observations are consistent with the subsequent Cs-137 gamma scans of the plates.

The handhole cover liner was visually examined at the hot cell. The surface features were documented by means of black and white photographs, as illustrated in Figure 11. In general, the liner exhibited no unusual features on its active surface. The surface appeared to be covered by a uniform gray surface layer with patchy dark and light areas. These features are similar to those observed on Manway Cover Backing Plate A.

The RTD was received in the laboratory as a complete assembly. The thermal elements were still in the thermowell; and the attachment wires, conduit, and cable head were in place. A plastic sleeve was attached to the thermowell flange nut to protect the thermowell surface. The tip of the thermowell (end 4.3 cm) was an orange color; the remaining surfaces

TABLE 3. RTD THERMOWELL BODY ANALYSIS - (4.3 - 19.0cm)

Location	Decontamination Number	Solution Used	Time (h)	Temperature (K)
Bottom 4.3 to 11.8 cm	1	Hydrochloric acid (IN)	2	294
	2	Nitric/ hydrofluoric 10 wt% 0.1 MHF	2	
Top 11.8 to 19.0 cm	1	Hydrochloric acid (IN)	2	294

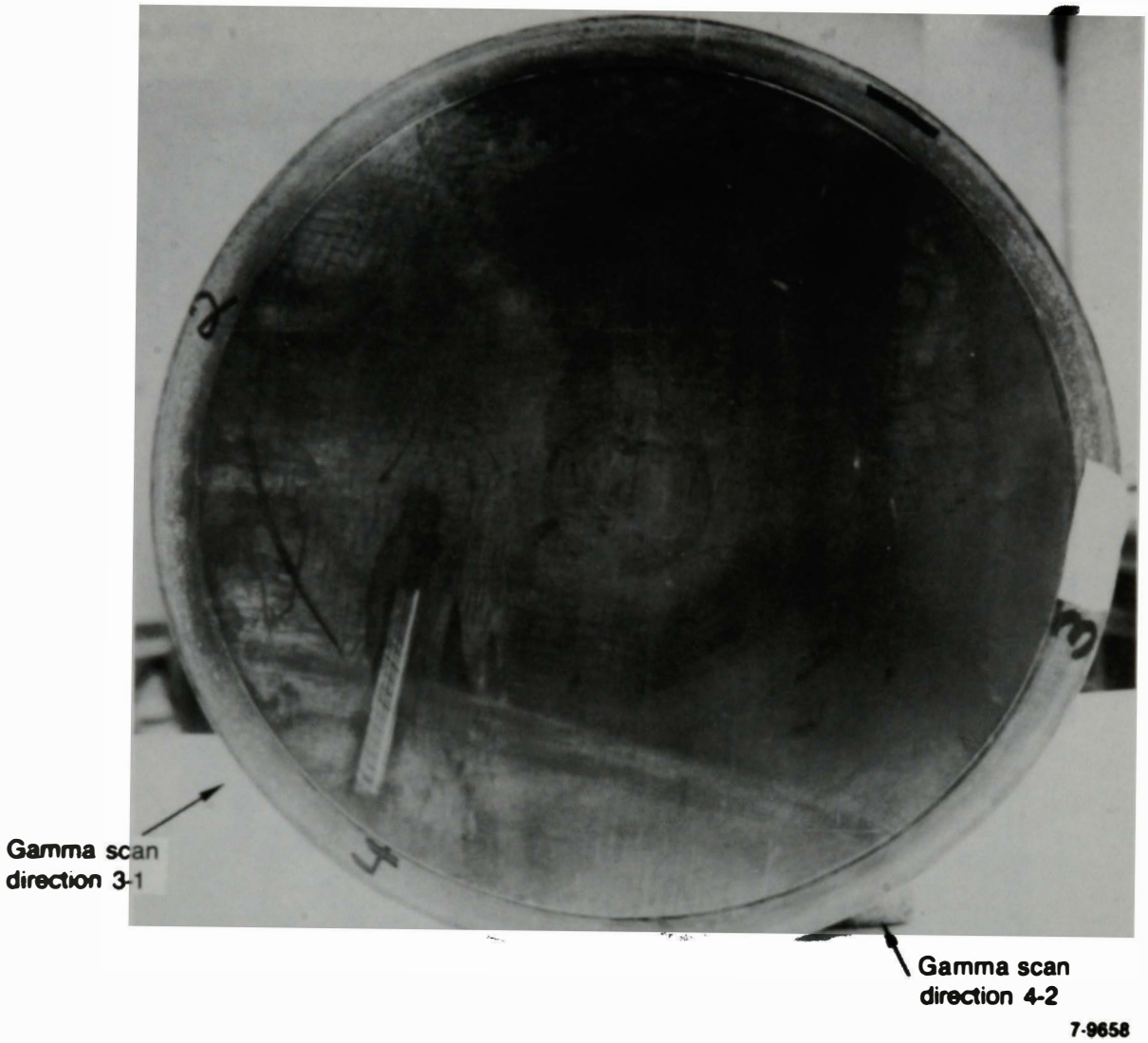
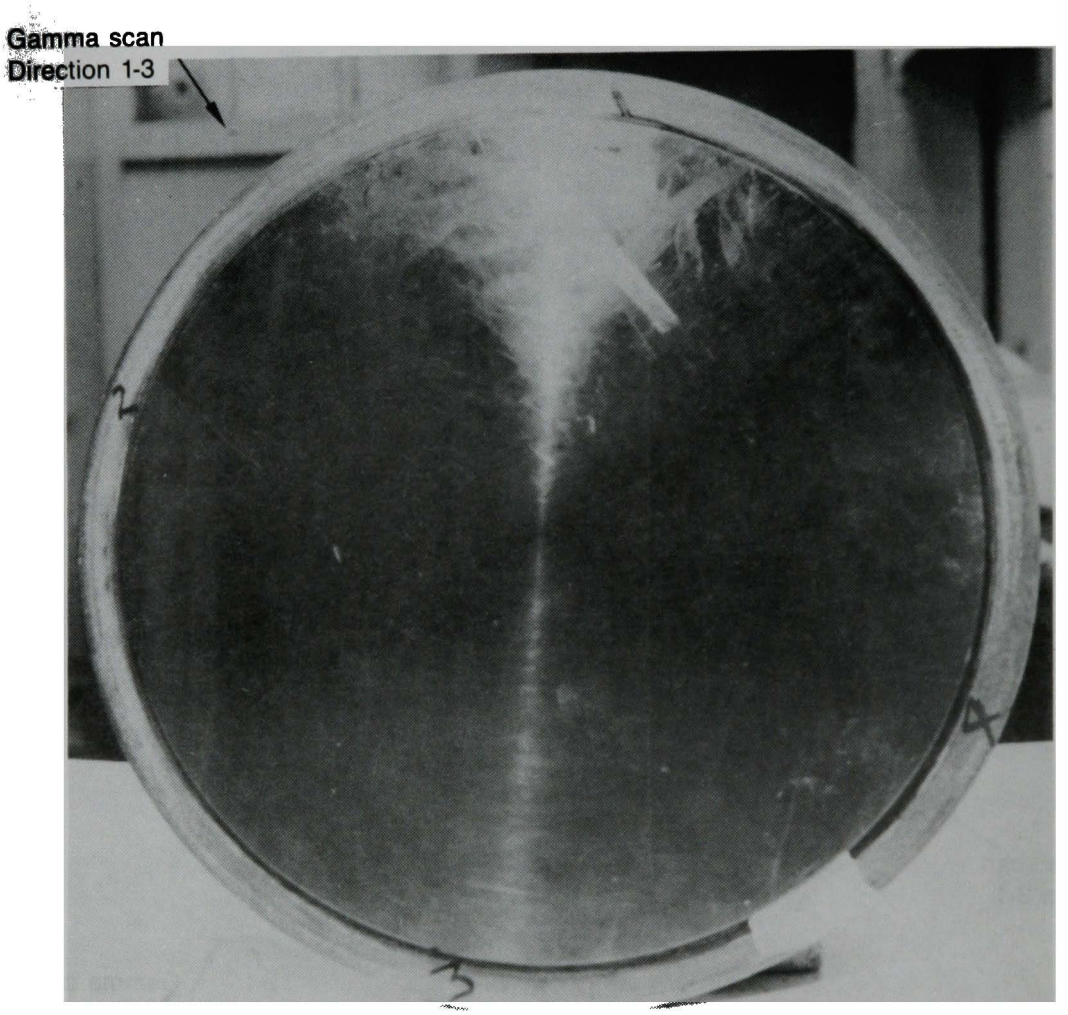
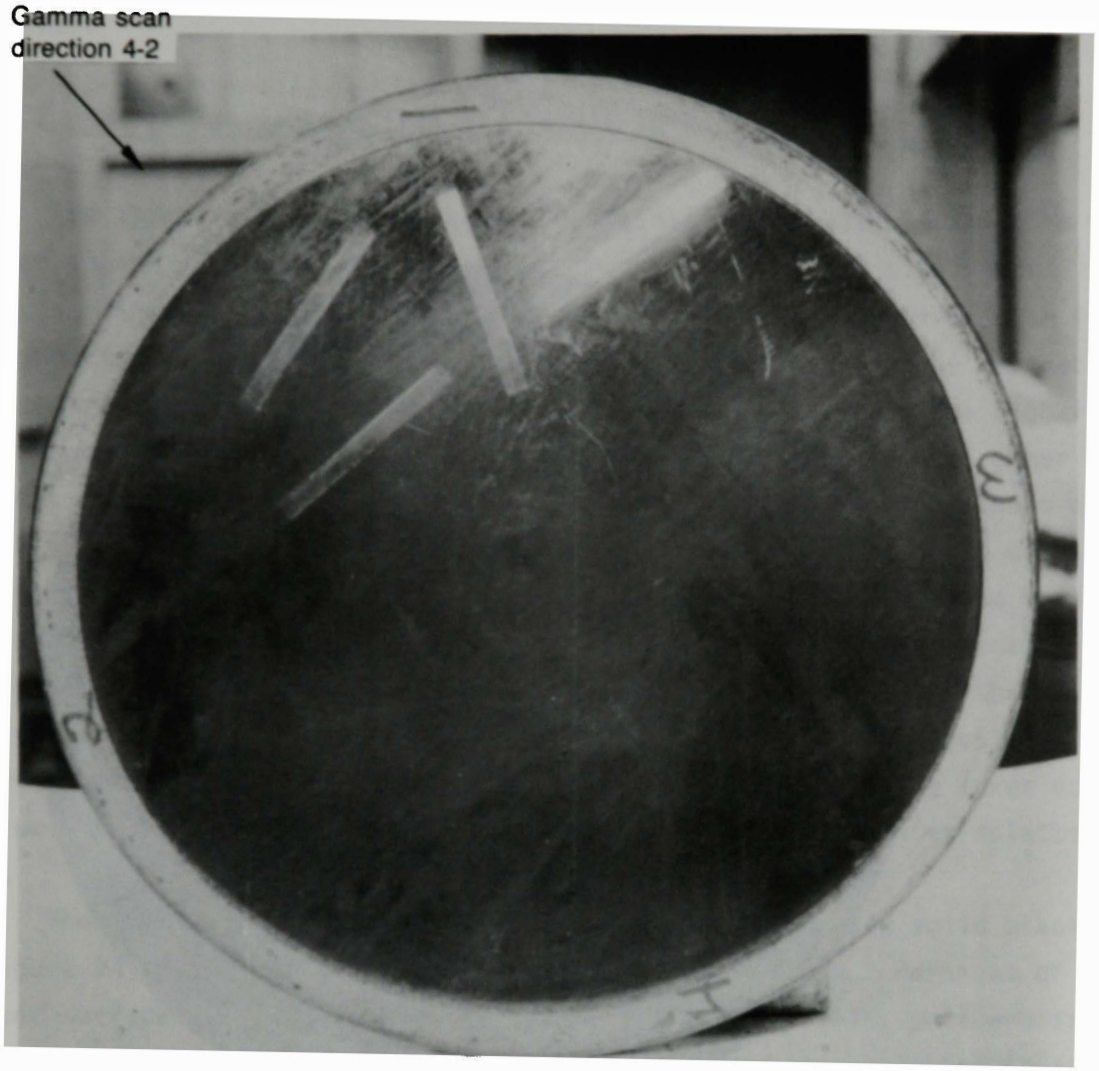


Figure 8. View of active surface of Plate A.



7-9657

Figure 9. View of active surface of Plate B.



7-9859

Figure 10. View of active surface of Plate P.

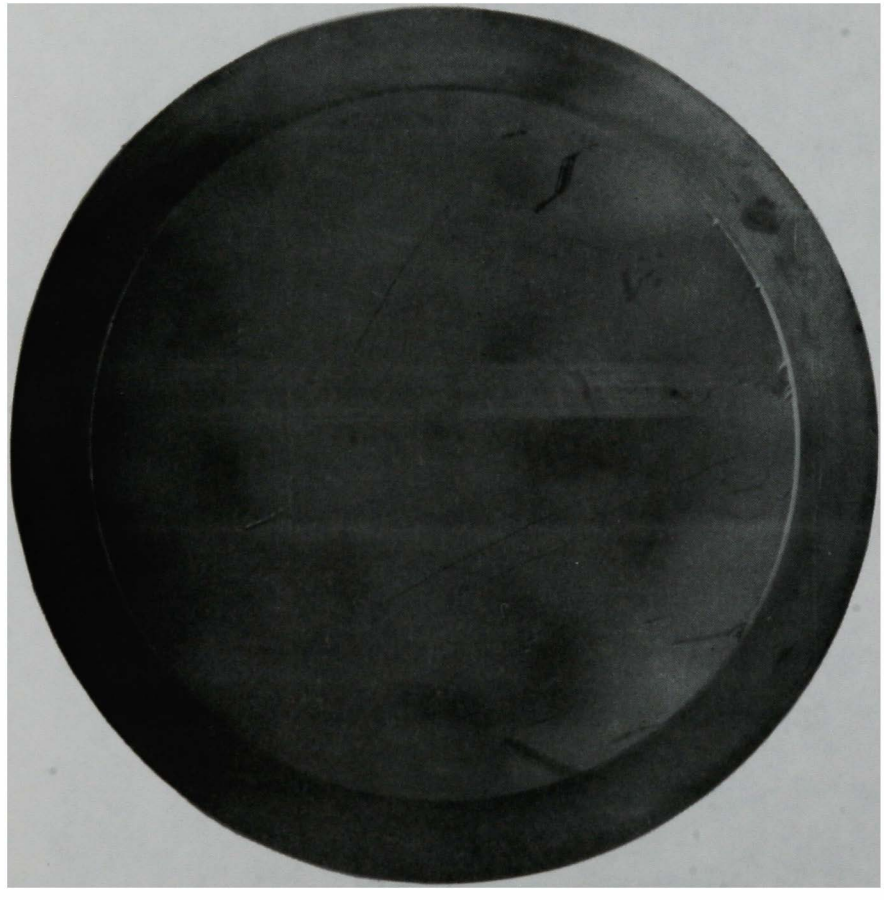


Figure 11. View of active surface of handhole cover liner.

were brown or brown/black (Figure 12). As received, the RTD thermowell measured 19.9 cm from the thermowell base nut to the tip. There was no visible or measured loose radionuclide material contained in the plastic sleeves.

2.3.2 Comparison to Leadscrew and Leadscrew Support Tube (LST)

The appearance of the leadscrew sections varied with location in the upper plenum. The section closest to the core was coated with black debris. Several flakes of silver-colored material were observed both in the debris and on the surface of the leadscrew section near the bayonet. In contrast, the loose material deposited on the surfaces of the higher leadscrew sections and the collected debris were gray in color, rather than black. The relative quantity of surface debris increased from almost none at the lower end of the leadscrew to a heavy coating near the top of the plenum assembly region. A distinct stepped decrease in debris was observed just above the top of the plenum assembly. The surface of the section near the upper end of the leadscrew was visually clean and shiny and there was no observable debris. The visual characteristics of debris deposition and corrosion were similar on leadscrews B8 and H8 except that the lower end of B8 was coated with white material that looked similar to solder splatter.

The entire outside surface of the LST section appeared solid black (Figure 13). The deposits were thin and largely uniform. Portions of the outer surface on one side were partially covered by a thin, yellow-orange deposit. The flat bottom of the section, which had been oriented toward the reactor core, showed adhesion of metallic particles ranging in size from barely visible to about 1 mm in diameter. The largest particles appeared spherical. The inside surface also appeared black with regions of yellow-orange surface material which covered less of an area.

Based on the physical appearance of the surface samples during the accident, conditions in the upper plenum of the reactor were more severe than in the reactor coolant loops.

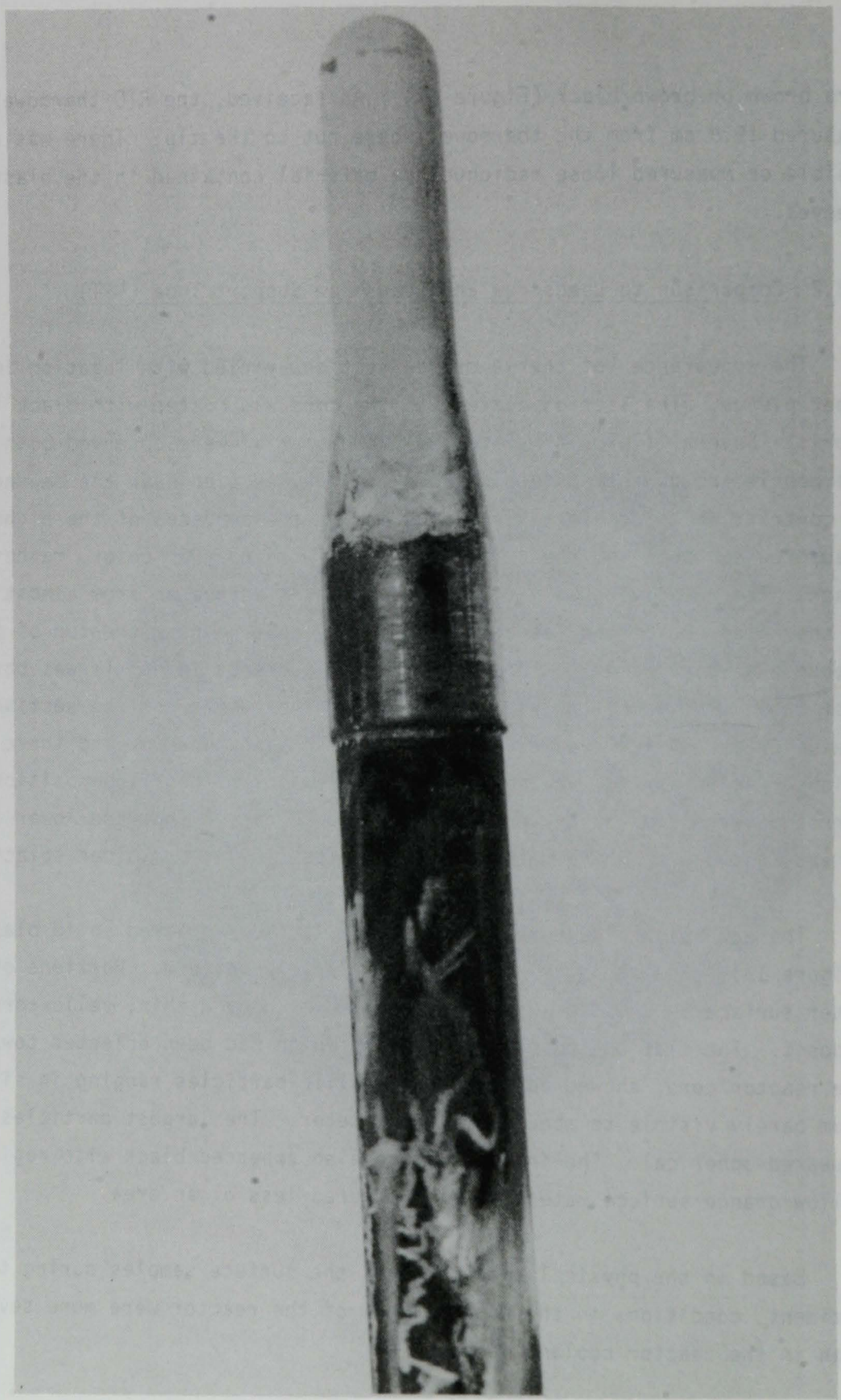


Figure 12. Overall view of the RTD thermowell.

Figure 13. Photograph showing the LST section.

2.4 Metallurgical Examination Results

2.4.1 Manway Cover Backing Plates

Figures 14-16 show the optical micrographs for the three specimens. A thin surface layer (4-12 μm) is visible on the specimens from Plates B and P, but this layer is less than 2 μm thick on the specimens from quadrants I-III of Plate A (samples A1, A2, and A3). The surface layer on sample A4 was thicker (3-4 μm) than on samples A1-3, in agreement with the visual examinations. On each sample, however, there were no measurable differences in the thickness within the surface layers.

Detailed examination of the samples showed no major differences in the structure and morphology of the surface layer within each sample. SEM micrographs of the samples indicate that the surface layers are relatively thin. Not surprisingly, EDS analysis of the surface layer showed it to consist mainly of iron, chromium, and nickel. The proximity of these matrix elements tend to overwhelm the electron energy peaks from any other element. However, the presence of indium and/or tin was also revealed in specimens A1 and A4. Since EDS cannot resolve the electron energy peaks of indium and tin, electron microprobe (wave length dispersive) analysis was also performed on specimen A4 to resolve any uncertainties. Both tin and indium were shown to be unambiguously present in the surface layer of the specimen.

2.4.2 Comparison to Leadscrew and Leadscrew Support Tube

Surface samples from the H8 leadscrew were metallographically examined to determine the thickness and nature of the surface layer. In general, three layers were observed: an inner layer next to the base metal, a thin middle layer, and an outer layer. Near the bottom of the leadscrew these layers were 28 μm, 1 μm, and 20 μm, respectively. The total layer thickness increased with elevation and was 114 μm at the top of the leadscrew. The top edges of the threads contained the largest layers, probably as the result of gravitational settling of aerosols.[8]

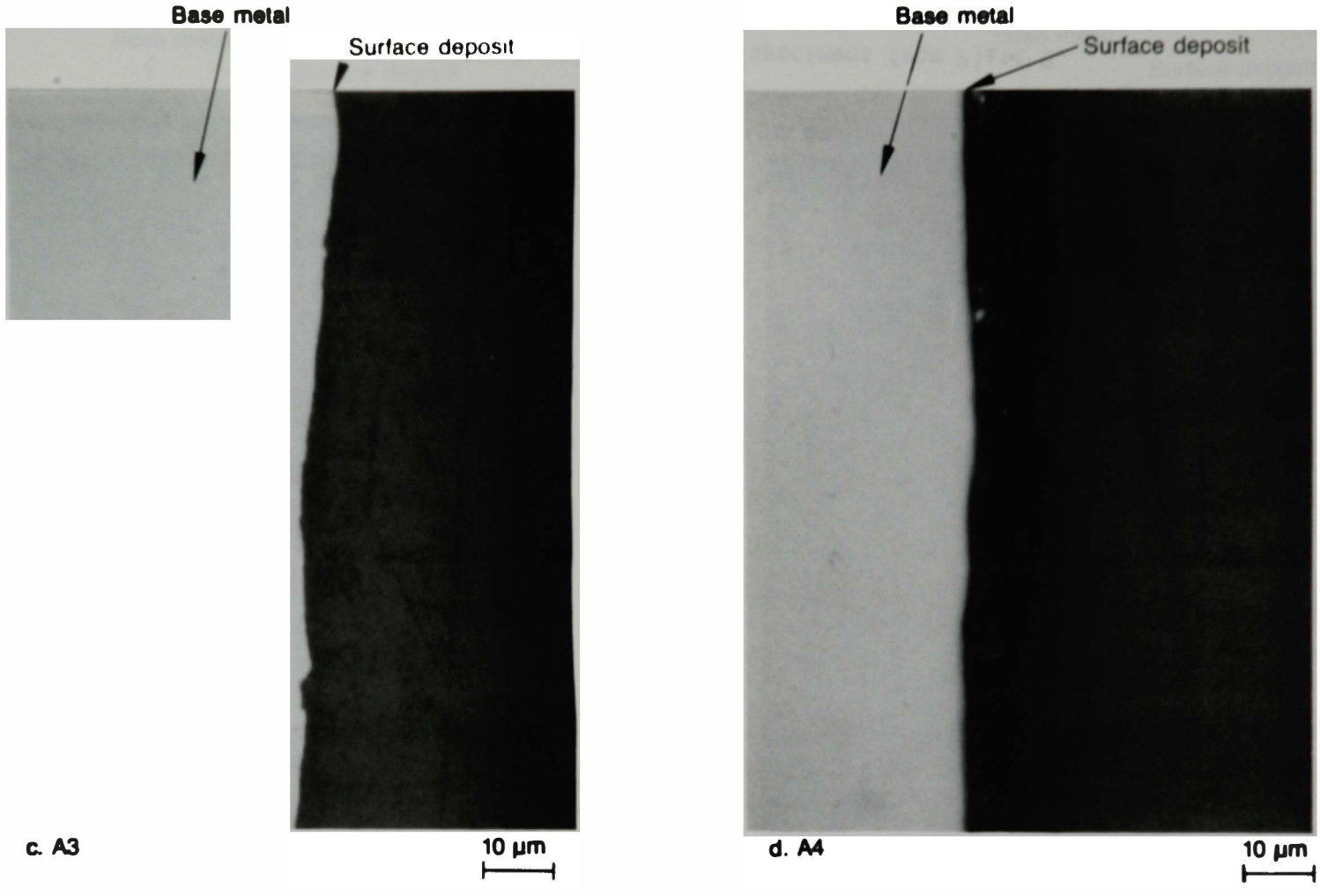


Figure 14. Optical micrographs of specimens from Plate A.

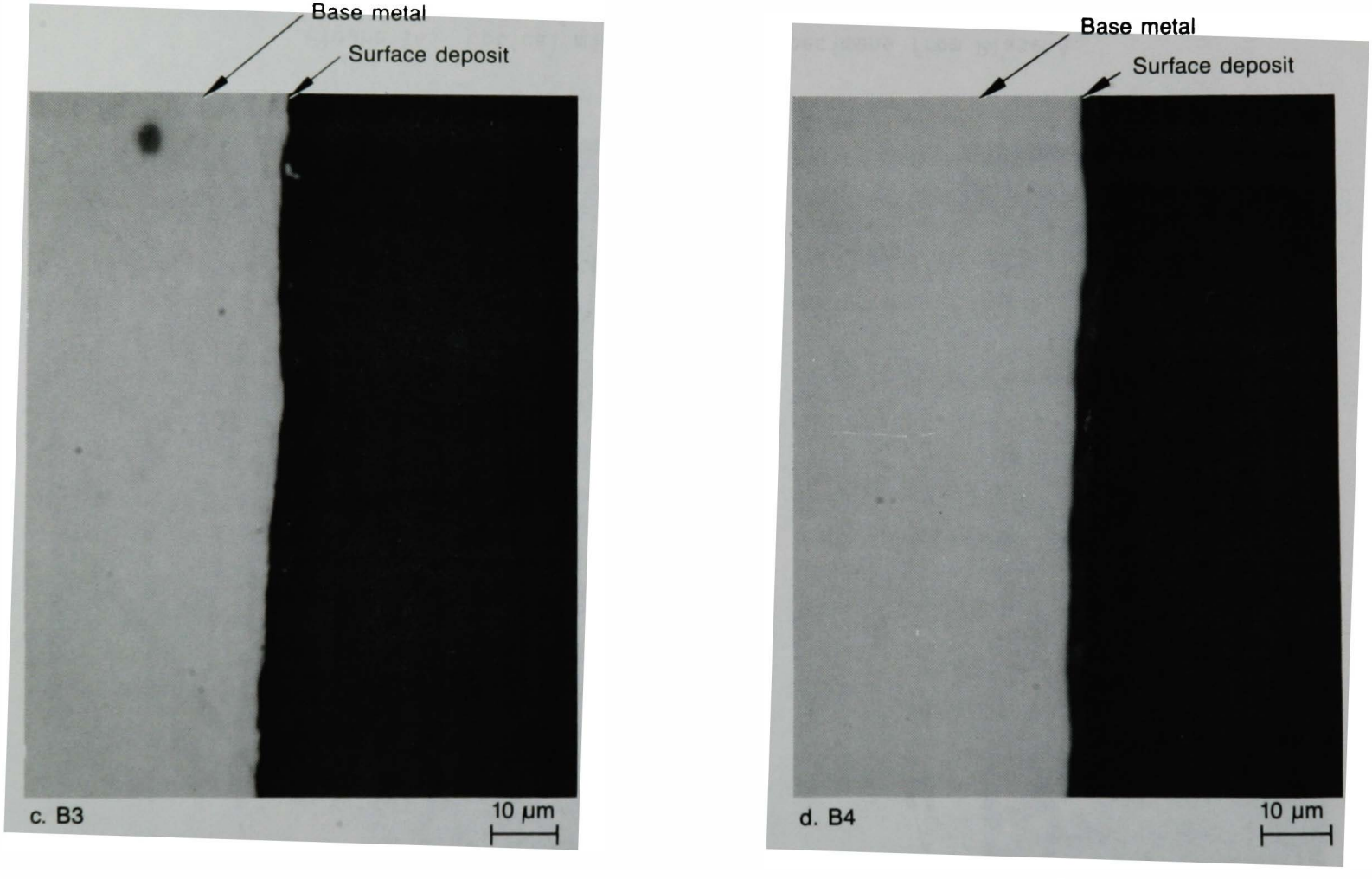


Figure 15. Optical micrographs of specimens from Plate B.

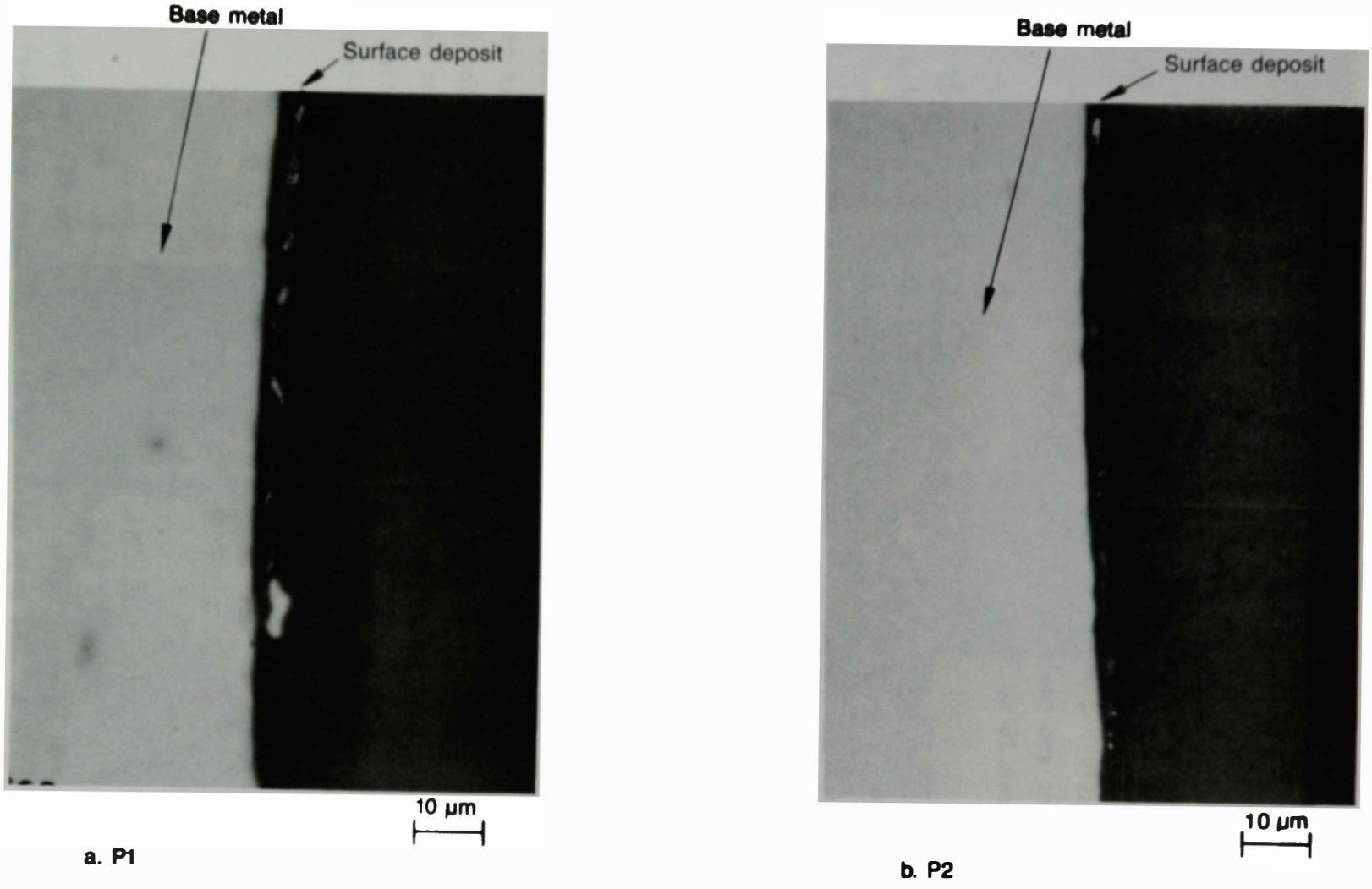


Figure 16. Optical micrographs of specimens from Plate P.

In their examination of a section of the H8 leadscrew located 14 ft above the core, B&W also observed three surface layers. They attributed the innermost layer, which was 3 μm thick, to an in-service oxide layer. The middle layer was porous in appearance but tightly adherent. The thickness of this layer varied from 10 to 90 μm. Silver-rich metal globules of ~15-300 μm were bonded to the outside surface of this adherent deposit. Electron microprobe results indicated the presence of indium in the silver globules but no cadmium. The outside layer was loosely adherent and could be removed by brushing. The thickness of this layer varied from 25 to 75 μm.

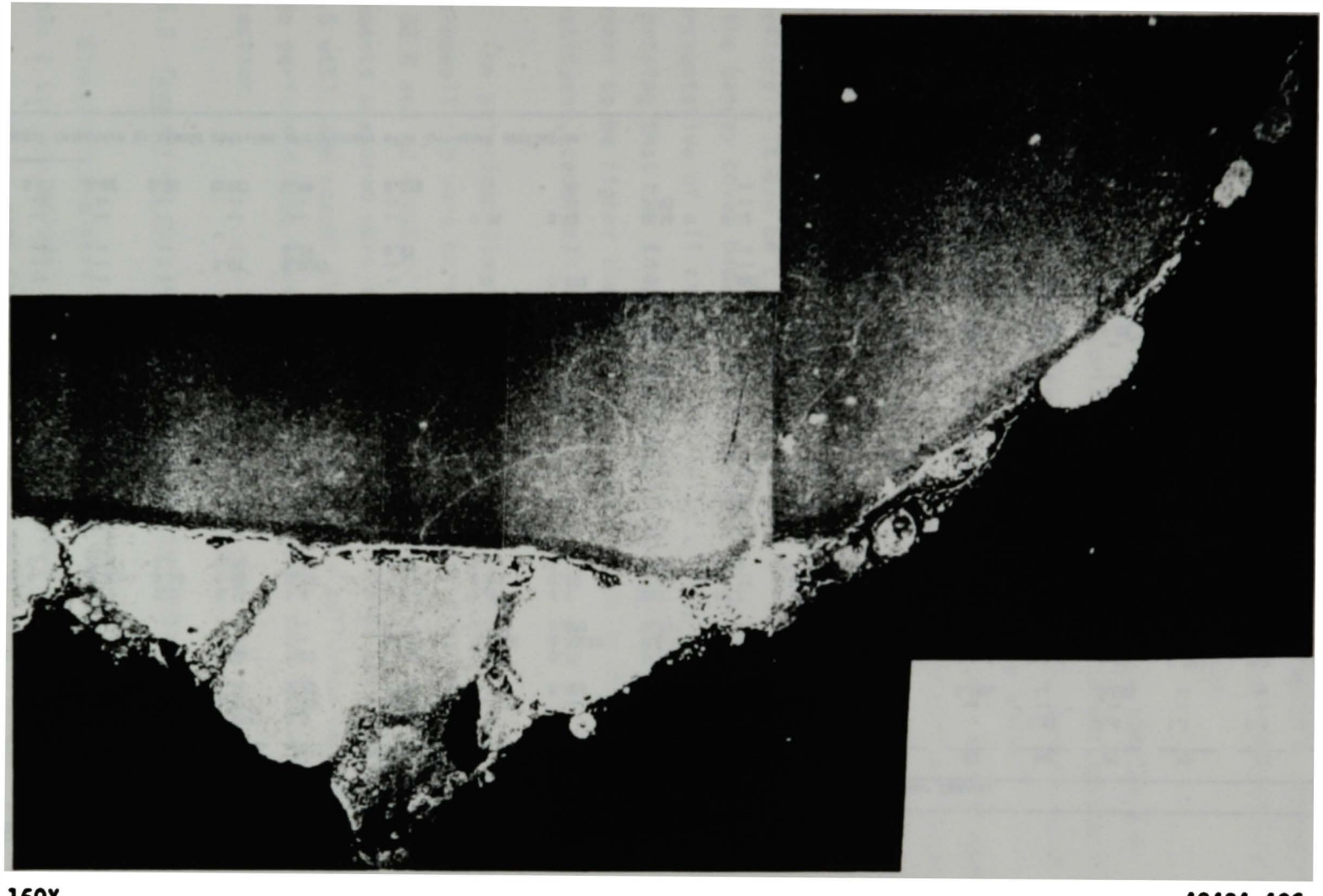
Samples from the surface of the B8 leadscrew were also examined. The results were qualitatively similar except that the surface layers on these specimens tended to be thinner than on the H8 leadscrew.[8]

Only two surface layers were identified on the leadscrew support tube: an inner adherent layer and an outer loosely adherent layer. Unlike the leadscrews, which are made of Type 17-4 pH stainless steel, the support tube is made of 304 stainless steel. The loosely adherent layer varied widely in thickness. The adherent layers on the inside and outside surfaces of the annular tube were quite similar (ranging in thickness from 15 to 30 m and from 15 to 40 m, respectively) but the loosely adherent layer on the outside of the tube was better defined and thicker than on the inside surface (4 to 35 μm compared to 0 to 20 m on the inside surface). Metallic globules were also found on the outside of the tube but not on the inside surface. As indicated in Figure 17, the end of the tube facing the core had a large number of metallic globules. The globules were primarily composed of silver, indium, and some cadmium.[6]

2.5 Elemental Analysis Results

2.5.1 Manway Cover Backing Plates and RTD Thermowell

The results of the elemental analyses for all of the RCS samples are tabulated in Table 4. The high values for iron. chromium, and nickel in



160X

48434-436

Figure 17. Appearance of the bright metallic globules at bottom end of the LST section.

TABLE 4. ELEMENTAL SURFACE DEPOSITION BY ICAP SPECTROSCOPY (mg/cm^2)

	Leadscrew Samples						Manway Cover Samples											
Element	MB-2	MB-11	MB-15	BB-2	BB-7	Thermowell[a]	A1	A2	A3	A4	B1	B2	B3	B4	P1	P2	P3	P4
Ag	--	0.002	--	0.004	2.9	0.15	1.4	--	1.4	0.72	0.63	4.4	--	0.51	1.4	1.1	0.9	0.39
Al	0.37	0.29	0.37	0.07	0.23	--	0.093	--	0.07	0.083	--	--	--	--	--	--	--	0.052
B	2.8	2.0	2.2	0.10	0.75	8.3	0.017	0.003	0.02	--	--	--	0.004	--	0.009	--	--	--
Ca	0.36	0.48	1.2	0.10	0.01	--	0.02	0.10	0.34	0.064	0.019	0.034	0.046	0.023	0.057	0.045	2.2	0.031
Cd	--	--	--	--	--	--	0.0006	0.00075	0.0048	0.026	0.00029	--	--	--	--	0.0004	--	0.00048
Co	--	--	--	--	--	--	0.051	0.033	0.092	0.004	0.021	0.19	0.028	0.003	0.005	0.068	0.080	0.048
Cr	1.6	1.5	3.3	0.15	1.4	--	12.0	8.1	22.7	10.7	8.9	49.1	8.4	9.4	13.8	15.8	17.3	9.8
Cu	0.028	0.27	0.34	0.66	0.28	0.11	0.23	0.15	0.36	0.18	0.17	0.97	0.12	0.14	0.095	0.13	0.06	0.016
Fe	5.7	7.4	6.0	0.79	3.3	15.0	53.0	32.1	102.0	44.9	36.5	188.0	35.4	40.4	57.7	65.6	71.6	43.8
K	--	--	--	0.01	0.02	--	9.3	4.1	13.1	6.0	2.7	2.1	3.0	7.3	4.0	4.2	8.1	4.0
Mg	0.01	0.36	0.13	0.011	0.04	--	--	--	--	--	--	--	--	--	--	--	--	--
Mn	0.04	0.05	0.07	0.02	0.43	--	0.69	0.51	1.3	0.60	0.40	2.6	0.40	0.48	0.76	0.94	0.93	0.52
Mo	--	--	--	--	--	--	0.12	0.069	0.21	0.12	0.074	0.40	0.077	0.078	0.088	0.083	0.10	0.042
Na	1.0	0.61	1.2	0.15	0.26	--	1.1	1.2	1.5	2.0	1.4	0.54	1.1	0.90	0.51	0.60	2.4	1.3
Nb	--	--	--	0.0096	0.031	--	--	--	--	--	--	--	--	--	--	--	--	--
Ni	1.1	0.32	0.43	0.11	0.50	--	6.4	4.3	11.8	5.4	4.7	25.8	4.6	5.2	7.3	8.2	9.3	5.3
P	--	--	--	--	--	--	8.7	1.9	3.5	4.1	2.1	3.1	1.4	12.2	4.0	2.5	9.8	2.5
Pb	--	--	--	0.19	0.14	--	--	--	2.8	3.0	--	--	--	2.8	--	1.4	1.1	--
Si	0.76	0.66	1.2	0.29	8.7	--	--	--	0.8	--	0.83	0.97	--	--	--	--	--	--
Sn	--	--	--	0.26	1.1	--	0.015	0.006	0.013	0.044	0.011	0.054	0.03	0.006	0.02	0.008	0.025	0.006
Sr	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
Te	--	--	--			--	--	0.033	--	0.028	--	0.13	--	0.045	0.035	0.045	--	0.045
Ta	--	--	--	0.02	0.0077	--	--	--	--	--	--	--	--	--	--	--	--	--
U	--	--	--	--	0.11	0.61	--	--	--	--	--	--	--	--	--	--	--	--
V	--	--	--	--	--	--	0.28	0.18	0.36	0.22	0.9	1.0	0.18	0.14	0.23	0.33	0.24	0.21
Zn	--	--	--	--	--	--	--	--	0.03	--	--	--	--	--	--	--	0.03	--
Zr	0.03	0.02	0.03	0.021	0.49	0.078	--	--	--	--	--	--	--	--	--	--	--	--

a. Elemental detection by atomic emission spectroscopy with arc-spark excitation.

the manway cover backing plate samples indicate that a significant amount of base metal was probably removed during the dissolution operation. On the other hand, there does appear to have been considerable transport of core materials and fission products, as indicated by the relatively high values for silver, cerium, and tellurium, respectively.

In Table 5, the results obtained from the surface samples are extrapolated to the entire surfaces of the reactor coolant system loop piping (6×10^5 cm^2) steam generators (2.4×10^8 cm^2), upper plenum (4.25×10^5 cm^2) and pressurizer (9×10^5 cm^2). Only those elements that could be related to the transport of core material were included in the tabulation. Considering the wide variability in measured values and the extent of the extrapolation, these values should be used for illustrative purposes only. The implied quantity of tellurium deposited on the surfaces of the RCS loop, for example, exceeds the available inventory. It can be concluded that the localized deposition of tellurium on the manway cover backing plates is high. Clearly, these samples are not representative of all the steam generator surfaces. It is somewhat surprising that the fraction of silver deposited on the RCS surfaces appears to be higher than that of the more volatile control rod constituent, cadmium.

The principal elements detected in the elemental analysis of the RTD thermowell tip were boron and iron, which were present in relative weights of 33.8 and 62.4 wt% of the elements detected in the sample. Other elements measured were silver (0.6 wt%), zirconium (0.3 wt%), uranium (2.5 wt%), and copper (0.4 wt%). Cadmium and tellurium measurements were also performed, but these elements, if present, were below limits of detection.

2.5.2 Comparison to Leadscrew and Leadscrew Support Tube

Elemental analyses for five leadscrew samples are also shown in Table 4 for comparison. In general, the quantities of deposited materials are small and in many cases near or below detectable limits. The amount of

TABLE 5. IMPLIED SURFACE DEPOSITION[a]

Element	Core Inventory (kg)	Origin	Implied Mass Deposited (kg)			Fraction of Core Inventory		
			Upper Plenum	RCS Loop	Pressurizer	Upper Plenum	RCS Loop	Pressurizer
Ag	2,200	Control rod	6.2	2.7(2)	0.85	2.8(-3)	0.12	3.9(-4)
Al	215	Burnable poison	1.0	7.4	1.2(-2)	4.7(-3)	3.4(-3)	5.6(-5)
Cd	138	Control rod	--	0.84	2.0(-4)	--	6.1(-3)	1.4(-6)
Sn	369	Zircaloy	0.34	5.4	1.3(-2)	9.2(-4)	1.5(-2)	3.5(-5)
Te	3.65	Fission product	--	7.1	2.8(-2)	--	1.9	7.7(-3)

a. The values in this table involve the extrapolation of data with high variability to much larger surface areas. It is quite possible that this extrapolation is not valid, as evidenced by the implied deposition of tellurium on RCS surfaces which exceeds the available inventory. These results are provided for illustration only.

silver on the leadscrews (see Table 5) indicates that less than 1% of the original control rod silver deposited in the upper plenum. There is a significant amount of aluminum on the leadscrew surfaces that apparently originated from burnable poisons.

Absolute values for inner layer surface deposition of elements was not determined for the leadscrew support tube. Relative amounts of the principal elements were determined for pieces of the loosely adherent and adherent layers. Typically, only the steel materials were measurable on the surface. A loosely adherent sample did, however, incorporate one or more metallic globules and indicated a large fraction of silver, indium, cadmium, and tin in the sample.

2.6 Radioactive Inventory Analysis Results

2.6.1 Gamma Scans

The relative Cs-137 activity is plotted in Figure 18 as a function of distance from the edge of the manway cover Plate A (initial gamma scans had shown the surface activity to contain mainly Cs-137 with very minor amounts of Co-60 and Cs-134 present). Results for Plates B and P are also shown in Figure 18. The Cs-137 activity profiles show very low count rate for all scans except near the edges of the plates. In fact, Plate P shows significant Cs-137 activity only within 1 cm of the edge; the activity profile goes to near zero across the remaining surface. Plate B, on the other hand, exhibits a Cs-137 profile very similar to that obtained on Plate A, although the overall count rate was approximately five times lower than on Plate A.

Gamma scans were performed on the handhole cover liner. The results are shown in Figure 19. The Cs-137 activity profile shows an uneven distribution across the liner surface, although there does appear to be a slight increase in activity near the edges of the liner. This Cs-137 activity distribution appears to be somewhat similar to that obtained previously on the manway cover backing Plate A.

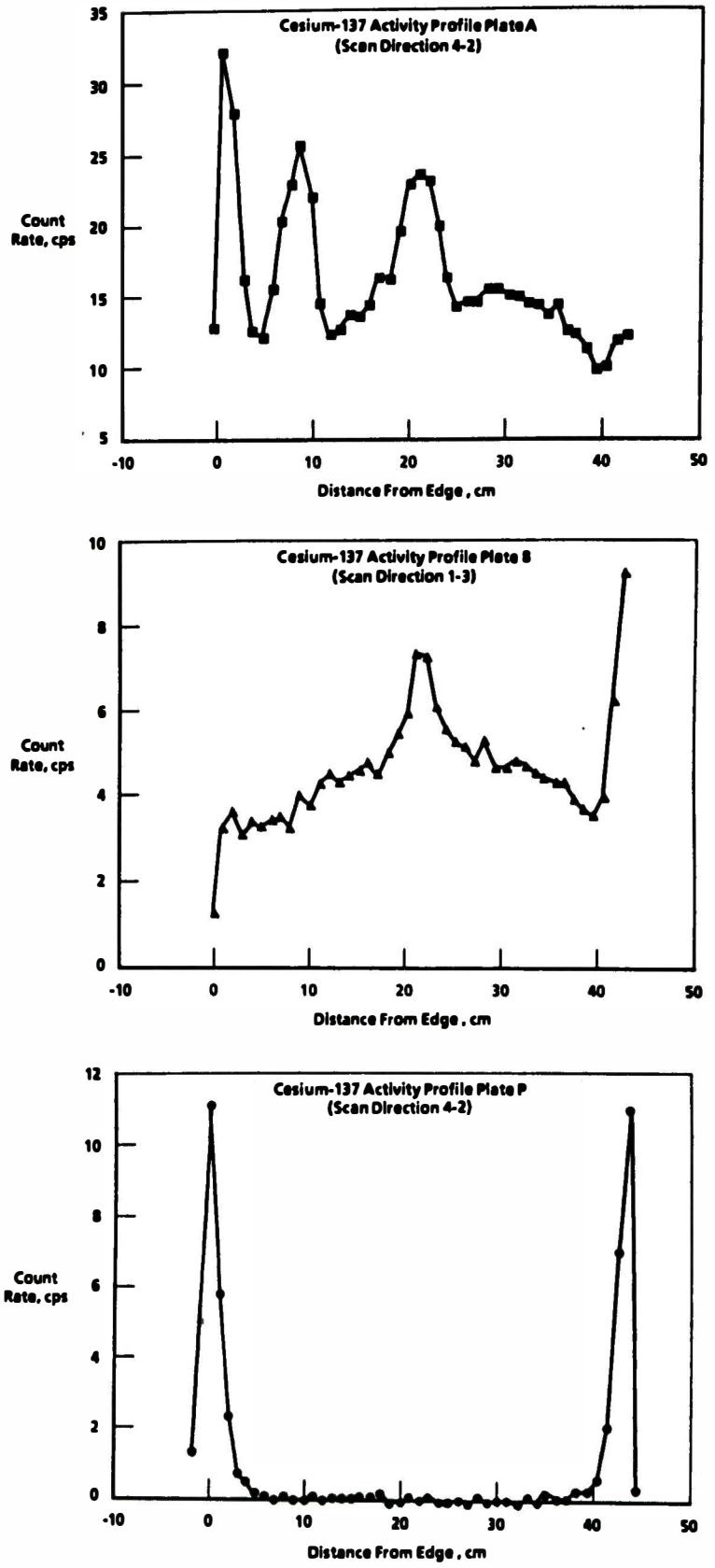


Figure 18. CS-137 activity profile of manway cover Plates A, B, and P.

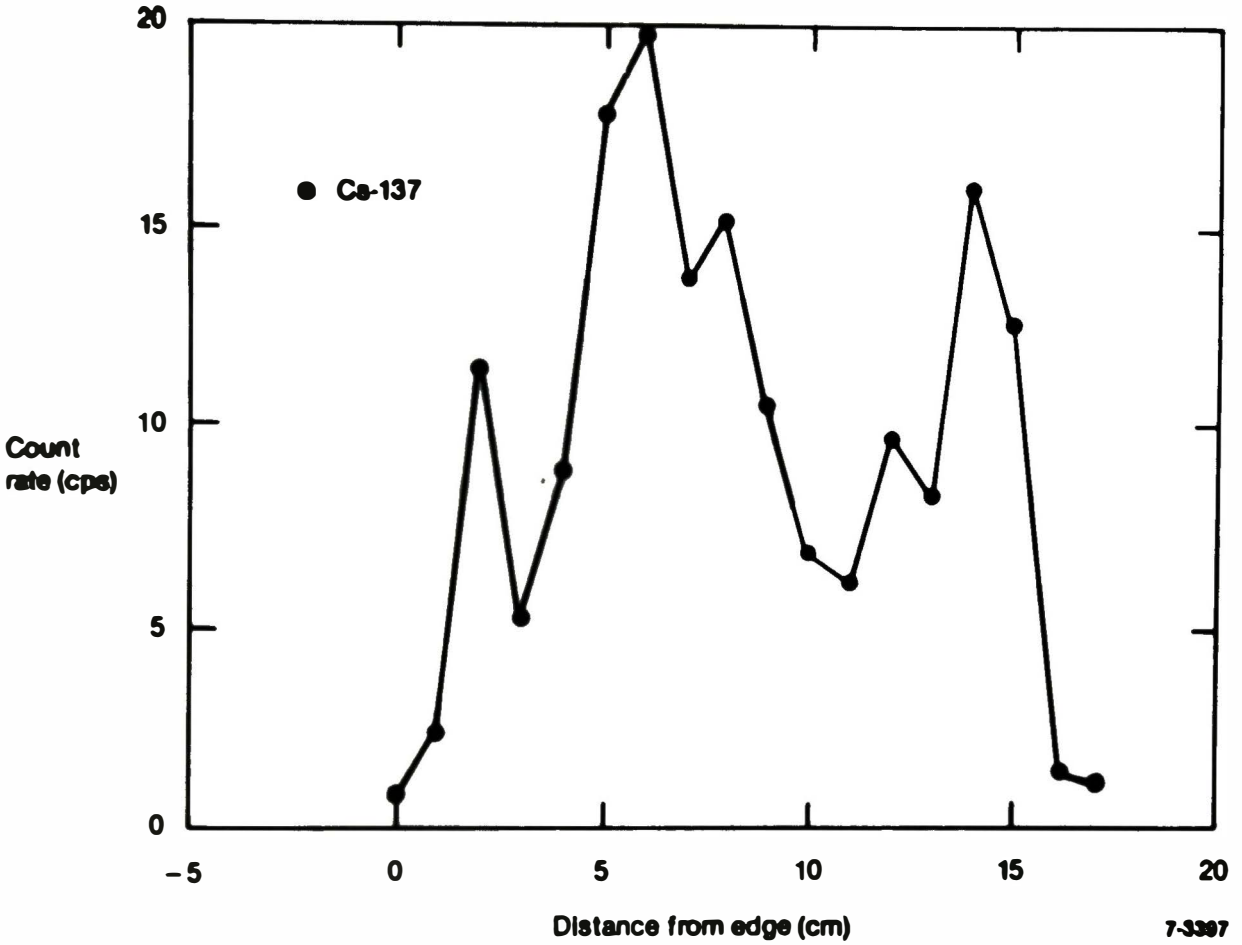


Figure 19. Activity profile of A-loop steam generator handhole cover liner.

The RTD thermowell was gamma scanned on two sides at 0 and 180 degrees at 10-cm intervals from the thermowell nut (flange) to the tip. The Cs-137 analysis results for the radionuclides detected are plotted in Figure 20. The amount of Cs-137 deposited near the end of the body of the thermowell closest to the flow stream is essentially the same as on the thermowell tip.

2.6.2 Autoradiographs

The autoradiographs of the manway cover backing plates are shown in Figures 21, 22, and 23. Very little gamma activity is evident in the autoradiographs, which is not surprising since the gamma scans had previously shown very little activity on the surface of the plates. Closer inspection of the radiographs reveals that the gamma activity is uniformly distributed over the surface, in agreement with the results of the gamma scans. The Plate P radiograph shows significant gamma activity only at the very edges of the plate.

2.6.3 Radiochemical Analysis

Table 6 lists the total radionuclide content leached from the inner surface of each coupon from the manway cover backing plates corrected to April 1, 1986, to facilitate comparison with other data.[5]

The A and B samples from the steam generators tend to have significantly higher concentrations (10-100 times) than the P samples from the pressurizer, which appear to have come from an area of lower radionuclide surface deposition. Also the Cs-137/Cs-134 ratio is less, which suggests that the deposited cesium in the pressurizer came from a different region of the core than that deposited in the A and B sample locations.

The surface activity of the RTD thermowell is also shown in Table 6. The measured values for most of the radionuclides (I-129 is the exception) are higher than for the steam generator coupons, which is to be expected because the RTD thermowell was closer to the core.

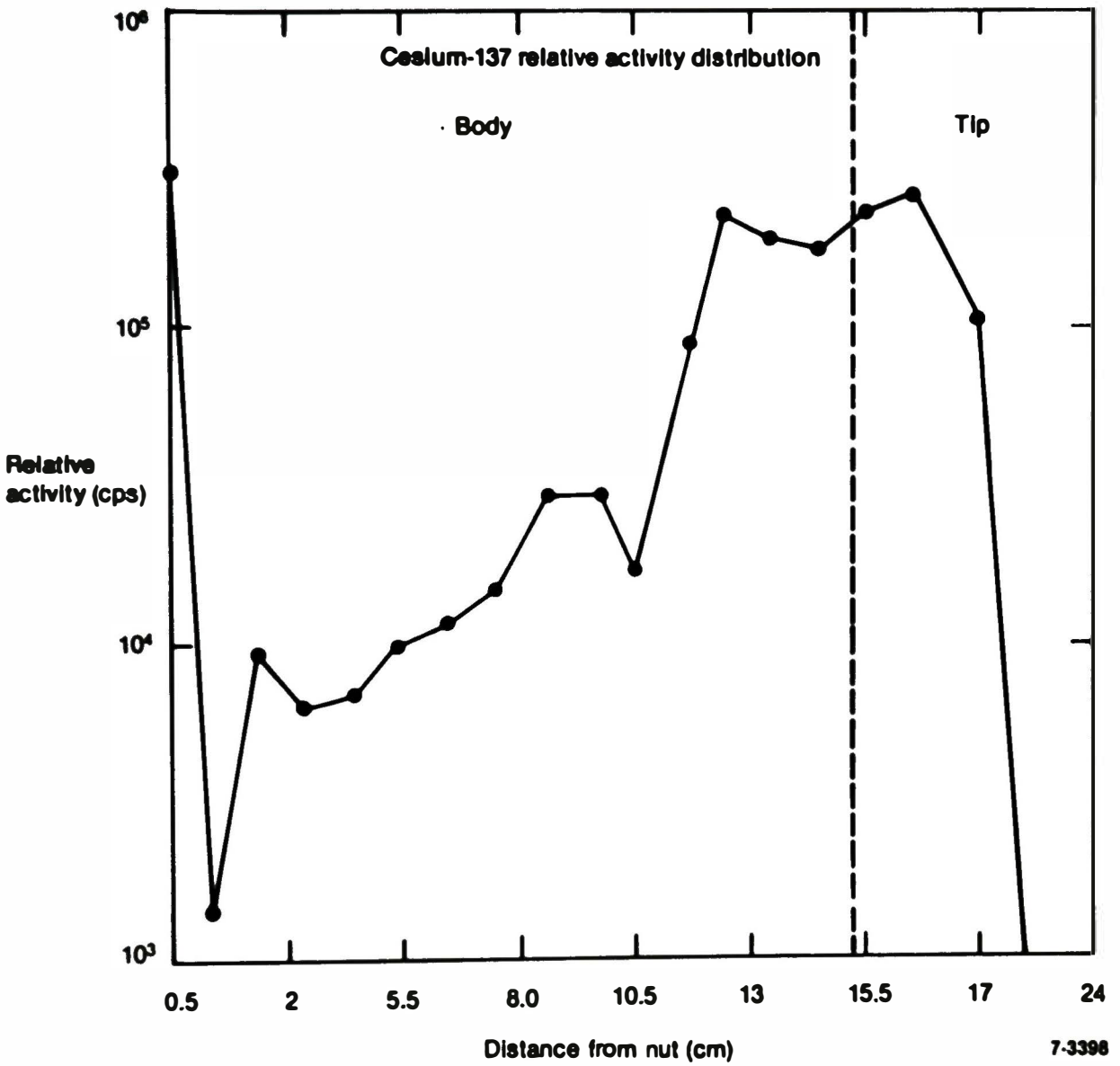


Figure 20. CS-137 activity profile RTD thermowell.

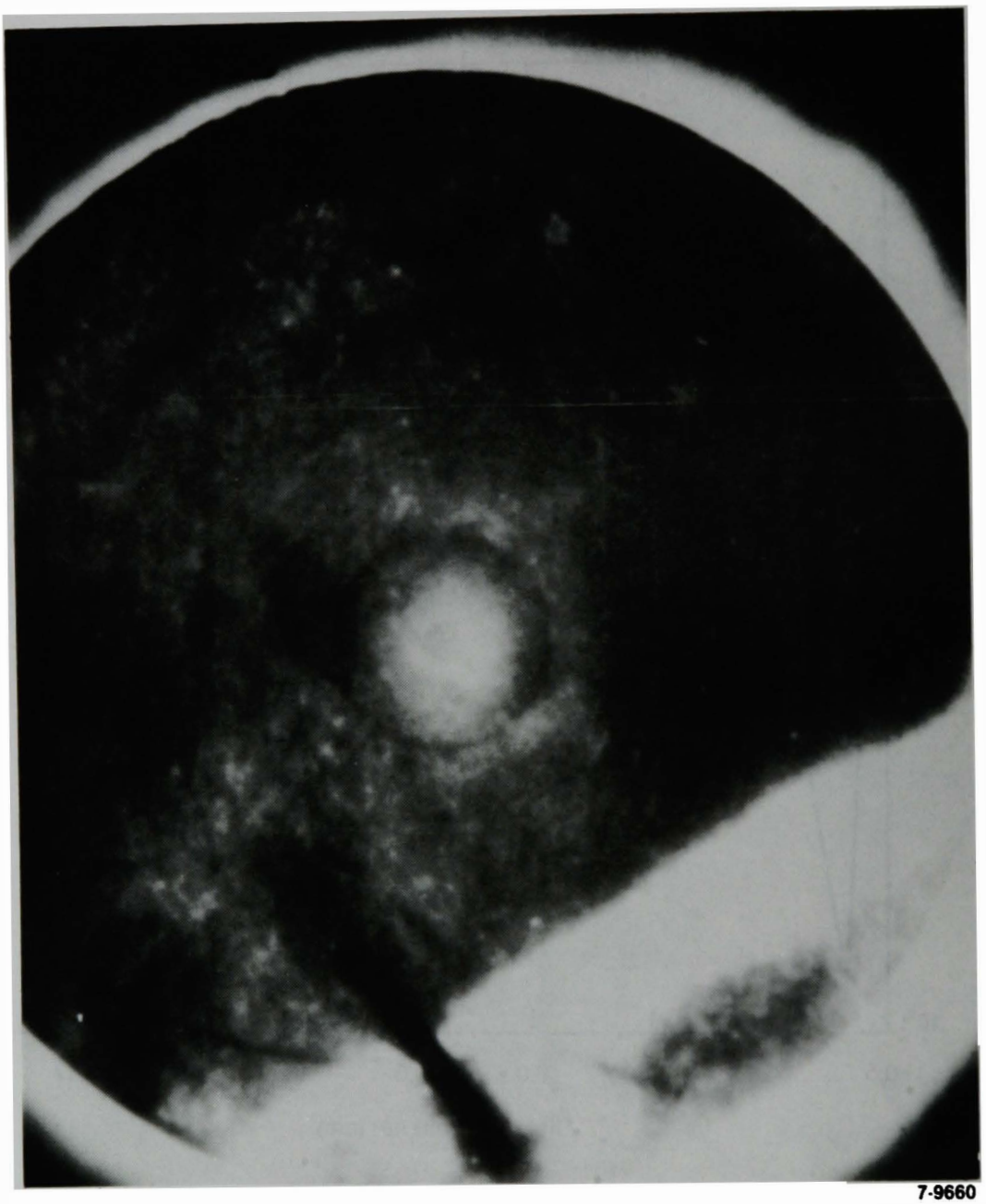


Figure 21. Autoradiograph of Plate A.

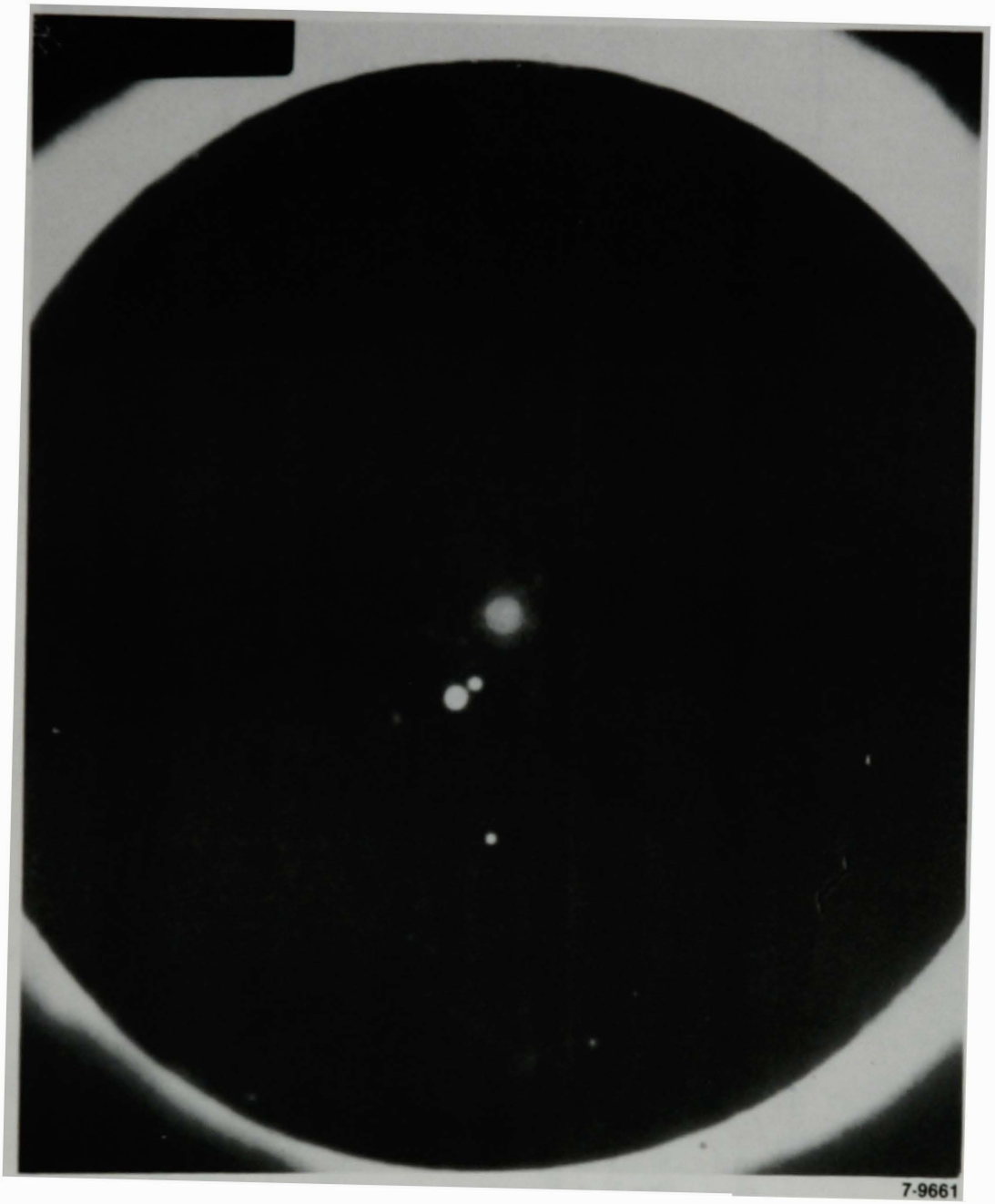


Figure 22. Autoradiograph of Plate B.

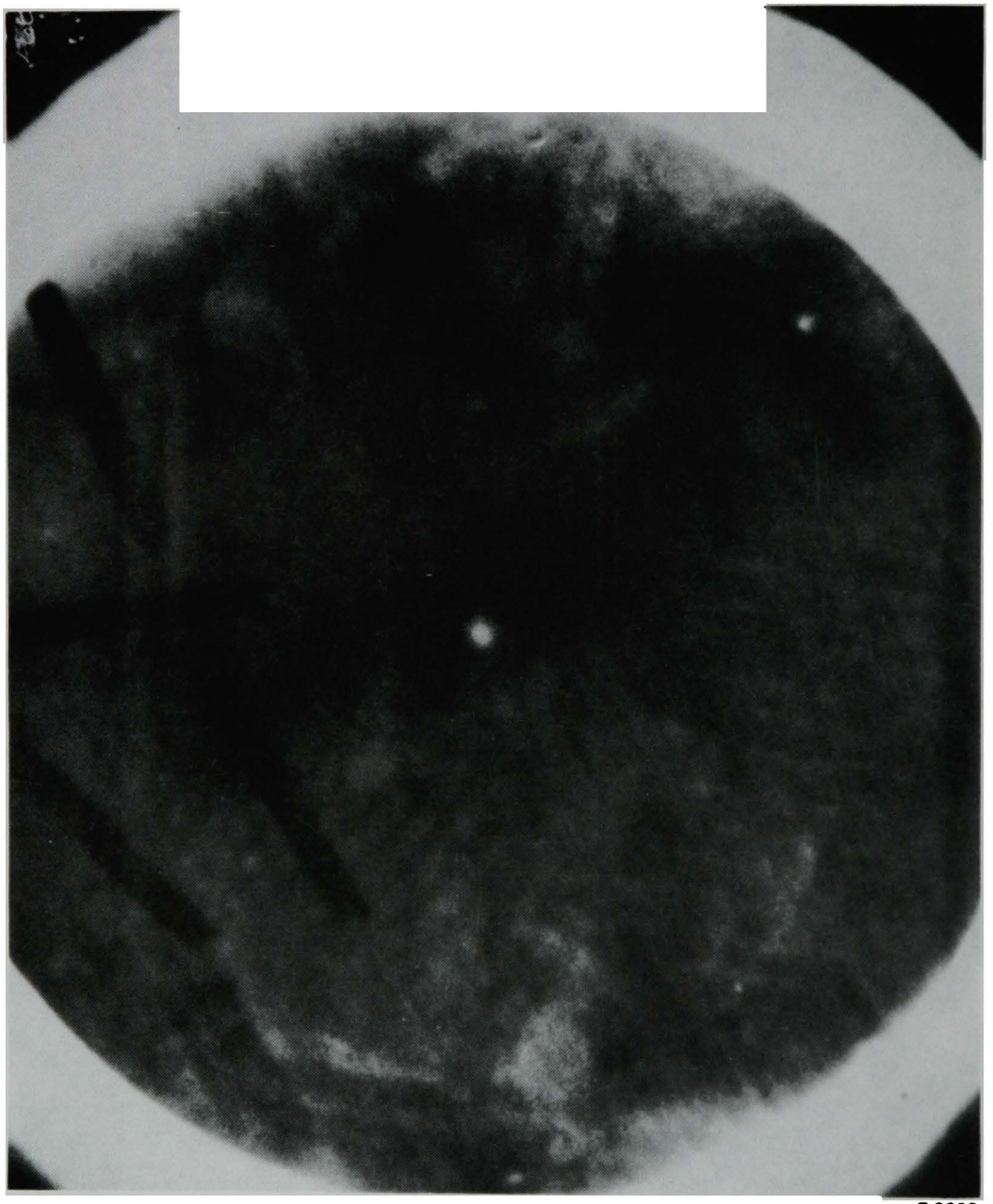


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Figure 23. Autoradiograph of Plate P.

TABLE 6. RADIONUCLIDE SURFACE ACTIVITY

Sample	Surface Activity ($\mu Ci/cm^2$)[a]						
	Co-60	Sr-90	Sb-125	Cs-134	Cs-137	Ru-106	I-129
Leadscrew - H8	5.3(-1)	2.7(0)	1.0(1)	1.2(1)	4.6(2)	--	1.4(-4)
Leadscrew - B8	5.8(-1)	7.5(0)	5.6(0)	8.2(0)	2.8(2)	2.2(-1)	6.8(-5)
RTD Thermowell	1.1(-1)	8.9(0)	8.3(-1)	4.7(-1)	2.0(1)	--	8.0(-6)
Manway Backing Plate - SGA	7.4(-2)	1.8(-1)	5.0(-1)	1.6(-1)	5.8(0)	5.3(-1)	7.2(-6)
Manway Backing Plate - SGB	4.4(-2)	9.3(-2)	2.8(-2)	5.5(-2)	1.9(0)	--	--
Manway Backing Plate - Pressurizer	2.4(-2)	4.0(-1)	4.3(-3)	2.1(-3)	4.8(-2)	--	--

a. Corrected to April 1, 1986.

Table 7 shows the data with the surface areas of the upper plenum (4.25×10^6 cm^2), loop piping (6×10^6 cm^2), steam generators (2.4×10^8 cm^2), and pressurizer (9×10^5 cm^2)to obtain an estimate of the surface deposition in the entire reactor coolant system. The fractions of the fission product species deposited on the primary system surfaces are all small. This does not imply, however, that the fractions deposited during the accident sequence were small, since deposit of aerosols and soluble species could have been washed away from the surface. The fission product inventory fractions deposited in the pressurizer are extremely small, both because of the small surface available and the low concentrations of materials.

2.6.4 Surface Deposit Removal

The objectives for removing the surface deposition from the manway cover backing plates were directed more at the measurement of the amount of deposited radioactive material than toward the determination of the effectiveness of different decontamination techniques. As noted in Section 2.2.7, a cold leach period of ~4 h was required to place 97% of the measurable activity in solution. Visual examination of the metal coupon after leaching indicated that all surface material had been removed from the leached surface.

The RTD thermowell tip was serially decontaminated as shown in Table 2. The concentration of individual radionuclides removed from the tip during the 12 decontamination steps was measured for each solution. The fraction of each radionuclide removed for each step is shown in Table 8. In general, aggressive acid leaching was required to remove most of the surface activity from the thermowell tip, indicating tightly adherent, insoluble chemical species for most radionuclides.

TABLE 7. TOTAL SURFACE ACTIVITIES

	Total Activity (Ci)			Fraction of Core Inventory		
Radionuclide	Upper Plenum	RCS Loop	Pressurizer	Upper Plenum	RCS Loop	Pressurizer
Co-60	2.3(0)	1.5(1)	2.1(-2)	--	--	--
Sr-90	2.2(1)	8.7(1)	3.6(-1)	3.5(-5)	1.4(-4)	5.8(-7)
Sb-125	3.3(1)	6.5(1)	3.9(-3)	1.6(-3)	3.0(-3)	1.8(-7)
I-129	4.3(-4)	1.7(-3)	--	1.7(-3)	7.4(-3)	--
Cs-134	4.3(1)	2.8(1)	1.9(-3)	2.9(-3)	1.5(-3)	1.0(-7)
Cs-137	1.6(1)	1.0(1)	4.3(-2)	2.2(-3)	1.4(-3)	5.9(-8)
Ru-106	4.7(-1)	1.3(2)	--	1.7(-5)	4.6(-3)	--

TABLE 8. FRACTION OF TOTAL ACTIVITY REMOVED FROM RTO THERMOWELL TIP BY EACH DECONTAMINATION SOLUTION

Radionuclide	Total Radionuclide Concentration Removed (μCi)	Fraction Removed (%)											
		Decon Solutions				Acid Leaches							
		1	2	3	4	5	6	7	8	9	10	11	12
Mn-54	3.4(-3)	11.8	0	0	0	0	0	0	0	0	0	88.2	0
Co-60	2.17(0)	1.0	0.2	3.3	1.4	24.4	4.7	1.2	13.7	5.9	42.2	2.4	1.8
Sr-90	1.47(+2)	8.6	4.9	6.5	9.9	5.6	3.7	21.8	6.6	3.9	21.8	3.4	3.4
Sb-125	2.07(0)	1.0	0.5	8.9	13.5	38.2	16.9	1.4	12.1	7.2	0	0	0
Cs-134	1.46(+1)	0.1	<0.1	0.1	5.5	0.8	9.6	2.8	25.2	5.5	50.0	<0.1	0.6
Cs-137	3.12(+2)	0.1	<0.1	0.1	5.3	0.8	9.3	3.0	25.4	5.3	47.4	2.4	0.6
Ce-144	4.08(0)	0.4	0.1	3.8	2.2	26.5	11.8	0.7	40.4	12.3	0	1.4	0.5

Neither [two-step outer (4.3 to 11.8 cm) surface decontamination and the single-step inner (11.8 to 19.5 cm) surface decontamination] procedure removed large fractions of the radionuclides. Comparison of before and after gamma scans indicates the decontamination steps each removed about 70% of the surface contamination.

The combined acid leaching and the use of HCl only as a leaching agent yield about the same overall decontamination effectiveness. The experience with the decontamination of the tip indicates that a combination of strong acid leaching and elevated temperatures is required to totally solubilize and remove the adhered activity.

3. INTERPRETATION

There were two objectives for the examination of specimens from the TMI-2 reactor coolant system surfaces. The principal objective was to learn more about the processes of radionuclide release, transport, and deposition that occurred in the accident. The second objective was to examine different techniques for surface decontamination to determine which were the most effective. In this section of the report, results obtained from the examinations will be reviewed to identify insights that can be drawn about the course of the accident.

3.1 Origin of Surface Layers

The surface layers observed on the reactor coolant system surface samples are substantially different than those of the leadscrew samples from the upper plenum. The surface layers on the B-steam generator and pressurizer manway backing plates are in the range of 4 to 12 μm thick. The A-steam generator backing plates had an even thinner surface layer. Since the pressurizer always contained water, it is likely that the pressurizer sample was never exposed to a severe accident environment and that the surface layer resulted from the operational history preceding the accident. Because of the similarity between the surface layers on the steam generator samples and pressurizer sample, it further appears likely that they are of similar preaccident origin. Radionuclides and structural materials released from the core apparently interacted with this preexisting layer.

In contrast, the surface layers in the upper plenum appear to have been largely determined by the accident environment. Most of the cesium is associated with the adherent intermediate layer. Since the cesium is tightly bound, it is likely that it chemically reacted with the layer while in a vapor state, as observed in high temperature vapor deposition experiments with cesium hydroxide. Globules of molten control rod material were apparently carried into the upper plenum by high steam velocity at some time after substantial reaction had occurred with the leadscrew and

support tube surfaces. The globules appear to be at the interface between the adherent layer and loosely adherent layer. The adherent layer is apparently composed of aerosolized material which deposited and settled either from the gas phase during the core uncovery period of the accident or from liquid after core recovery.

3.2 Transport of Control Rod Materials

Control rod materials were apparently transported within the reactor coolant system as vapors and as particles. As discussed previously, droplets of control rod material were carried by rapidly flowing steam into the upper plenum at some period while the core was uncovered. The composition of the droplets indicates that some preferential vaporization of the more volatile constituents cadmium and indium had occurred prior to deposition of the droplets. Nevertheless, less than 1% of the core inventory of silver is estimated to be deposited on upper plenum surfaces. In contrast, if the reactor coolant system samples are representative of deposition in the reactor coolant system, including the large steam generator surface area, 12% of the silver inventory is on these surfaces. In comparison, only 0.6% of the cadmium inventory is represented on the reactor coolant system surfaces. It is not clear whether the silver was transported throughout the reactor coolant system as a vapor, an aerosol, or by liquid transport after the accident.

3.3 Transport of Structural Materials

The mass of zirconium on reactor coolant surface samples is below detectable limits. Tin, a constituent of Zircaloy, is unambiguously present in these samples and was observed in the EDS spectrum as well. These results are consistent with the leadscrew analyses in which comparatively little zirconium was found except in brushoff debris near the bottom of the leadscrews.

3.4 Transport of Cesium

Because of its half life and volatility, cesium was found to be the principal source of radioactivity on all the reactor coolant system surfaces. Nevertheless, the total inventory of cesium integrated over all surfaces is small. As a result, it is not possible to draw conclusions about how the bulk of the cesium was transported during the core uncovery phase of the accident. The surface deposition of cesium on the leadscrews is substantially higher than on the RTD thermowell, which is in turn higher than on the steam generator samples. Not surprisingly, the amount deposited decreased with distance from the core.

The isotopic ratios of Cs-134 and Cs-137 on the backing plates may provide some clue as to the progression of the accident. The ratios are 0.052 and 0.055 for the A and B steam generator samples and 0.084 for the pressurizer sample indicating that the pressurizer cesium originated from a region of higher burnup.

3.5 Relationship between Cesium and Iodine

It was previously concluded from the deposition of Cs-137 and I-129 determined in the analysis of leadscrew samples[8] that cesium and iodine transported with similar modes. It was not possible to draw any conclusion from the reactor coolant system samples, largely because of the small quantities of iodine-129 present.

3.6 Transport of Tellurium

The amount of tellurium present on the reactor coolant system surfaces is difficult to estimate because of the variability of the results for different samples. In many cases the quantity was below detectable limits; on some samples, however, there was considerable tellurium present. Based on the rate of reaction of tellurium with steel, it would be surprising to observe a substantial quantity of tellurium transported into the reactor coolant system unless it was transported as an aerosol or by liquid after the accident.

3.7 Transport of Fuel

Before shipping the manway cover backing plates, GPU collected scrape samples which were subsequently analyzed to determine the abundance of uranium and plutonium isotopes. The plutonium concentration in fuel on the pressurizer surface was lower than on the steam generator surfaces, indicating that the fuel in the pressurizer came from a region of lower burnup. A possible explanation is that the deposition in the pressurizer resulted from material transported at the time of reactor coolant pump operation at 2 h 54 min. The cesium (probably in the form of CsOH) transported with the steam into the pressurizer was from the hottest, high burnup region of the core, whereas fuel carried into the pressurizer came from the fragmentation of lower burnup fuel at the top of the core. In general, the plutonium concentrations in fuel on the reactor coolant system surfaces are lower than would be expected from the core averaged ORIGEN concentration. From examination of the core it is apparent that a substantial portion of the higher burnup region of the core melted and resolidified within the vessel.

Substantial quantities of fuel debris were apparently transported around the reactor coolant system. Fuel debris that was deposited on the reactor coolant system surfaces was associated with the loosely adherent layer that could be brushed from the surface. Although a small amount of uranium was measured on the RTD thermowell, none was measured on the manway cover plates. The uranium found on the leadscrews was primarily loosely adhered and near the bottom.

3.8 Effects of Extended Period of Submersion

It is not possible to quantify the effect of water submersion on the deposits of materials that occurred during the period of core uncovery. It is likely that at some point in the accident, the mass of fission product material deposited on reactor coolant system surfaces on reactor coolant system surfaces was substantially higher than the amounts measured on reactor coolant system surfaces as reflected in terms of fraction of core

inventory in Table 3. The analyses performed on surface specimens were not able to reveal how much material that may have been deposited on the samples during the accident sequence was subsequently removed by water immersion or how much of the material currently on the specimens may have been deposited during submersion, rather than during the period of core uncovery.

3.9 Decontaminatability of RCS Surfaces

The general conclusions from the leaching studies are that an acid solution will decontaminate the RCS surfaces. By immersing the components for 3 to 4 h in the acid solution at 280 to 300 K, over 95% of the surface activity can be removed. Other neutral or basic solutions are not very effective.

3.10 Implications of Deposition Data to Source Term Understanding

The analyses of surface samples described in this report provide some insights into the progression of the accident. The implications to source term methodology development and validation are quite limited, however. None of the results of the surface deposition analyses provide unambiguous evidence to confirm or refute current methodology. There are indications of cesium surface reactions, iodine-cesium association, aerosol deposition, transport of control rod materials, and transport of structural materials within the vessel and reactor coolant system, that under more controlled conditions could have provided a data base for model validation but which are confounded by lack of understanding of boundary conditions or the influence of the subsequent environment. The greatest contribution to the ambiguity in the interpretation of the sample results is the years of submersion in water.

The review of the applicability of surface disposition data to code development and validation that has been performed by the authors has been quite limited. There is considerable information regarding source term

behavior contained in the surface deposition data that have been collected. More careful examination of these data by source term model developers could identify uses that have not been identified in this report.

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