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THI-2 LEADSCREW DEBRIS PYROPHORICITY STUDY

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ii

# ABSTRACT

Debris removed from the surface of a leadscrew from the TMI-2 Reactor Building was examined to assess the potential for the debris to become pyrophoric. Elemental analyses were performed to identify ca didate phases that could be pyrophoric, and X-ray diffraction was used to determine if any of these phases was actually present. However, none of the candidate phases were found. Based on differential scanning calorimetry, no exothermic reactions were observed upon heating the debris to  $500^\circ$ . In air. Particle size distributions for the debris were obtained from analyses of micrographs of the debris. A light blockage instrument was also used to determine the particle size distribution. These analyses indicated that particles larger than 10 µm accounted for most of the debris volume, although the majority of the particles were actually smaller than 10 µm. Gamma spectroscopy indicated that most of the radioactivity in the debris, and on the leadscrew after debris removal, was due to mixed fission products such as 137Cs and 134Cs.

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# CONTENTS

ACK NOWL EDGEMENTS	ii
AB STRACT	iii
INTRODUCTION	1
SAMPLE COLLECTION	2
CHARACTERIZATION OF LEADSCREW DEBRIS	7
ICP Analysis	9
SEM Analyses	11
Particle Size Analysis	16
X-Ray Diffraction Analyses	19
Thermal Analysis	22
Radioactivity Measurements	22
DISCUSSION AND CONCLUSIONS	28
REFERENCES	30

iv

# TMI-2 LEADSCREW DEBRIS PYROPHORICITY STUDY

# INTRODUCTION

The head lift, plenum removal, and other operations associated with fuel removal and cleanup of Three Mile Island Unit 2 (TMI-2) will potentially expose some of the core debris to air. If the debris contains a sufficient concentration of finely divided, oxidizable particles, a pyrophoric reaction possibly could occur. Pyrophoric materials potentially present in the core debris include zirconium metal and zirconium-rich alloys, partially or completely hydrided zirconium, partially oxidized zirconium metal, and certain combinations of silver and zirconium.

Based on this concern, Pacific Northwest Laboratory (PNL) conducted a comprehensive study for the U.S. Department of Energy of the pyrophoricity characteristics of a sample of core debris from a section of TMI-2 leadscrew. The test specimen was a 9-in.-long section cut from the lower half of the center core (8-H) leadscrew. An adjacent specimen was used by the Babcock & Wilcox Company to characterize the contamination in the surface deposits under the loose debris.<sup>1</sup>

The specific elements of the sample preparation, characterization, and pyrophoricity test program included:

1

Removal of the loosely adherent debris from the leadscrew specimen by brushing under Freon to collect a complete and representative sample

Radiological characterization of the leadscrew specimen before and after removal of the debris using thermoluminescent dosimeters (TLDs), and analysis of the removed debris by gamma spectroscopy Determination of the size distribution of the particles in the debris by direct measurement with a light-blockage instrument and by quantitative image analysis of scanning electron micrographs of the debris

Evaluation and correlation of particle size, morphology, and composition using scanning electron microscopy (SEM) with energy-dispersive X-ray fluorescence (EDX)

Identification of the crystalline phases present using powder X-ray diffraction (XRD)

Direct determination of the pyrophoric potential of the debris by differential scanning calorimetry (DSC)

Measurement of the major constituents in the debris by inductively coupled plasma (ICP) spectrometry.

The following sections of the report describe the details of the analyses and present the results for each of the evaluation areas. Although the data and conclusions are felt to be valid for the particular debris sample investigated, it should be recognized that this sample may not be representative of the debris associated with other components and at  $\iota$  ther locations within the reactor vessel or balance of the primary system.

#### SAMPLE COLLECTION

Sample collection from the 9-in. section of the 8-H leadscrew (Figure 1) was accomplished by brushing the leadscrew with a soft brush while the leadscrew was submerged under electronics grade Freon TF in a glass tank. The entire operation took place in the shielded containment shown in Figure 2. To ensure that all loose debris associated with the leadscrew was collected, the inner bag and plastic sleeve in which the



3

Figure 1. Nine-inch section of 8-H leadscrew from TMI-2.



Figure 2. Shielded containment used for the collection of loose debris from the 8-H leadscrew section.

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leadscrew was packaged were opened while under the Freon. This reduced the possibility of losing debris by electrostatic forces and minimized loss due to airborne dispersion.<sup>2</sup> After opening the bag and plastic sleeve, the leadscrew was removed and both the bag and plastic sleeve were brushed with a pure bristle brush and rinsed with the Freon in the tank. Next, the bag and plastic sleeve were removed from the Freon and packaged to allow future examination if desired. The leadscrew was then carefully brushed with a pure bristle brush, giving special attention to the sides and bottom of the threads.

The debris removed from the leadscrew was clearly visible during the brushing (Figure 3). The larger, heavy particles immediately settled to the bottom of the glass tank. In addition, clouds of fine particles were produced by each stroke of the brush. Eventually, the Freon became generally cloudy from the fine debris. Approximately two hours were required for the Freon to clear.

The clear Freen was then decanted from the tank in an effort to remove any adhesive that might have dissolved from the tape used to seal the inner bag and plastic sleeve in which the leadscrew was initially packaged. The sample debris, still in the glass tank, was then washed with clean Freen, the Freen was allowed to stand until clear, and then it also was decanted and packaged separately from the Freen used during the leadscrew brushing. Finally, the sample was removed from the glass tank by again adding clean Freen and then pumping both the Freen and the sample debris from the tank into a glass flask for transport to the analytical laboratory. A peristaltic pump w th Tygon tubing was used to remove the sample and decant the Freen from the glass tank.

To check for residual debris, stick-tape samples were collected throughout this procedure. Approximately 1/2-in. square pieces of tape were pressed against the surface to be sampled to collect any loose debris that might have remained. This type of sample was collected from the inner bag, plastic sleeve, and the leadscrew after they were removed from the Freon, and from the bottom of the glass tank after the sample debris was removed. The sample locations on the bottom of the glass



Figure 3. Glass tank used to contain the 8-H leadscrew section and Freon during sample collection. Debris removed from the leadscrew section is clearly visible on the bottom of the tank.

tank are shown in Figure 4, and the radiation level of each sample is listed in Table 1. The relatively low radiation levels indicate that most of the debris was transferred from the tank.

Sample No.	Radiation Level at Contact (mrad/h)
1	0.5
2	0.5
3	6
4	5
5	8
6	10
7	10
8	100
9	28
10	25
11	100
12	100

TABLE 1. RADIATION LEVELS OF STICK-TAPE SAMPLES FROM BOTTOM OF GLASS TANK

#### CHARACTERIZATION OF LEADSCREW DEBRIS

Debris collected from the leadscrew surface was characterized several ways to determine if any pyrophoricity hazard existed. The elemental composition of the debris was determined using ICP and SEM/EDX. X-ray diffraction (XRD) patterns were obtained to determine the major chemical constituents of the debris. Quantitative image analysis of selected SEM micrographs and light-blockage particle counting were used to determine particle size distributions in the debris. Gamma spectroscopy was used to identify gamma emitting isotopes in the debris, and TLDs were used to determine beta and gamma dose rates from the debris. DSC was used to identify any exothermic reactions caused by heating the debris in air. In addition, XRF was attempted on

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Figure 4. Location of stick-tape samples for residual contamination measurements on the bottom of the glass tank.

a sample (approximately 0.3 g) of the debris. However the bulk analysis could not be done since the specimen radioactivity saturated the [silicon (lithium)] detector in the instrument.

As discussed earlier, the loose debris was collected by scrubbing the leadscrew section under Freon TF in order to (a) collect most of the particulate, (b) reduce the spread of smearable contamination, and (c) minimize any pyrophoricity hazard associated with the debris. Vacuum filtration was used to recover the debris for analysis.

The debris was collected on Teflon filters having a 0.25  $\mu$ m pore size. Because fines quickly plugged the filter, approximately 3/4 of the Freon was evaporated (at ~50°C) to reduce the total number of filtrations. Just prior to filtration, the beaker containing the debris/Freon mixture was vigorously swirled to suspend the debris. This was done so that the debris collected on each filter would be representative of the original mixture.

The debris was fairly dense and settled quickly from the Freon after agitation. Most of the radioactivity appeared to be concentrated in the heavier particulates. The debris appeared as a fine, dull-gray powder after drying. A total of 4 g of loose debris were collected from the Freon. Control of radioactive contamination during filtration and specimen preparation operations was difficult. All of the analyses were based on debris from the filters.

#### ICP Analysis

Debris (0.3 g) was scraped from one of the filters, fused with potassium hydroxide in a nickel crucible, and discolved in deionized water with diluted hydrochloric acid for elemental analysis by ICP. Table 2 lists the elements that were analyzed by ICP and their respective weight percents in the debris. Only about 32% of the specimen composition could be determined by ICP analysis. Analysis for potassium and nickel was not possible due to the specimen preparation method. A significant fraction of the 68% weight difference is thought to be

silver based on SEM/EDX analyses and oxygen from XRD analysis. ICP analysis indicated that the primary constituents were: iron (9%), zirconium (7%), silicon (7%), and aluminum (4%).

Elemental Analysis - Performed for	Weight Percent of Debris	Normalized to 100%
<b>A</b> I	4.20	13.1
8	0.67	2.1
Ba	0.06	0.2
Ca	0.44	1.4
Ce		
Cr	0.79	2.5
Dy		<b>~~</b>
Fe	9.42	29.3
K	fusion	fusion
La	-	
Li	**	·
Ma	0.07	0.2
Mn	0.09	0.3
Mo	0.04	0.1
Na	1.24	3.9
Nd		
Ni	fusion	fusion
Rh		
Ru	-	
Si	7.18	22.3
Sr	0.01	
Te	• • • •	
Ti	0.66	2.1
Zn	0.13	0.4
<b>Zr</b>	<u>7.17</u>	<u>_22.1</u>
	32.17%	100%

TABLE 2. ANALYSIS OF LEADSCREW DEBRIS BY ICP

#### SEM Analyses

Two SEM specimens were prepared from the leadscrew debris collected on the filter. The specimens were prepared by pressing double-stick carbon tape against the filter and then shaking off any loosely bound material. Since the debris layer on the filter was relatively thick, two specimens were taken from the same location on the filter (M-71 was the first specimen and M-72 was the second specimen). Specimen M-71 had a higher debris concentration and an apparent higher percentage of larger particles than specimen M-72. Both specimens were carbon coated prior to SEM examination, and an SEM electron beam voltage of 25 keV was used. After preparation, the SEM specimens remained relatively nonsmearable and presented few radioactivity contamination control problems.

SEM micrographs of a typical area on specimen M-71 are shown in Figure 5. The debris consists of two types of particles: large solid particles measuring 5 to 25  $\mu$ m in diameter and agglomerates, made of 1  $\mu$ m particulates. Table 3 tabulates the major elements, based on EDX analyses for particles shown in Micrograph No. 0529 (Figure 5). The major particle compositions exclusive of elements with atomic numbers below aluminum were uranium, aluminum/silicon, zirconium/silver, and iron.

Region	Major	Minor	Trace
1	U	Fe, Zr, Al, Si	
2	Al, Si	Zr, U, Ti, Fe	Ni
3	Zr	Al, Si	Fe
4	U	Al, Si, Ti, Fe	
5	U	Al, Si, Fe	•
6	Zr, Ag, U	Fe, Al, Si	
7	Al, Si, Fe	U, Ti	
8	Zr	Fe, Al, Si, U, Ni, Cr, Ag	
9	Fe		Al, Si

TABLE 3. EDX ELEMENTAL ANALYSIS OF M-71



The particle density was lower for specimen M-72 than for specimen M-71. SEM micrographs of typical areas on specimen M-72 are shown in Figures 6 and 7, and the corresponding major elements for selected particles in these regions are provided in Table 4. Figure 6 suggests that the aluminum/silicon, iron, zirconium, and zirconium/silver particles were all agglomerates of smaller particulate. The uranium particles appeared to be faceted crystals (see Micrograph No. 0541 in Figure 6). The uranium/zirconium particles may be uranium particles coated with zirconium agglomerate (see Micrograph No. 0540 in Figure 6).

Region	Major	Minor	Trace
1	U	Zr, Ag	Al, Si, Ti, Fe
2	Ag, U, Fe, Ni, Al, Si	Ti, Zr	
3	Ag, U, In	Al, Si, Ti, Fe, Ni	Mo
4	Cr, Fe	Al, Si, Ti	Ni, Cu, Zr
5	U, In	Fe	Al, Si, Zr
6.	U, In, Fe, Ni, Cu	Al, Si	Ag
7	Ag, U, In, Fe, Ni	Cu, Cr, Al, Si	
8	Cu, S, A1, Si	Ti, Fe	In
9	Fe, Cr, Ni	U, Zr, Mo, In	Al, Si, Ti

TABLE 4. EDX ELEMENTAL ANALYSIS OF M-72

Micrographs in Figures 5 and 6 are based on secondary electrons emitted from the specimen surface due to the electron beam excitation. Images that are based on backscattered electrons from the beam cause particles composed of high atomic number elements, uranium, for example, to appear brighter than particles composed of lower atomic number elements, such as iron, chromium, and aluminum. Comparison of backscattered and secondary electron images of the same area on M-72 (Figure 7) suggests that the higher atomic number particles, probably e.g. uranium, are larger than 5  $\mu$ m.









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Figure 6. SEM examination of specimen M-72.



Figure 7. Backscattered and secondary electron SEM examination of specimen M-72.

# Particle Size Analysis

Particle sizes were analyzed using quantitative image analysis of the SEM micrographs and an HIAC light-blocking instrument for particles suspended in water. The HIAC analyzed for particles in the 1.5 to 100 µm range; particles above 0.15 µm could be detected in the analyses of the SEM micrographs. The HIAC measured 822,606 particles suspended in water with Triton dispersant, and the Quantamet results are based on an analysis of 629 particles examined by SEM.

Figure 8 shows the cumulative population distributions based on particle size and equivalent spherical volume for both the HIAC and the Quantamet analyses. Based on particle diameters, 50% of the population is between 0.7 and 2.5  $\mu$ m for the Quantamet analysis and between 3 and 6.8  $\mu$ m for the HIAC analysis. However, the population distribution curve based on volume percent for the Quantamet analysis does not have a tail at the upper end. This suggests that the SEM examination missed most of the very large particles in the debris. The population distribution curves based on volume fraction indicate that 50% of the volume is made up of particles between 5.5 and 10  $\mu$ m according to the Quantamet analysis and between 8 and 25  $\mu$ m according to the HIAC analysis.

Figure 9 shows a histogram that summarizes the size distribution of 142 particles of different compositions. Particles centaining zircenium including zirconium/silver and zirconium/uranium, accounted for 20% of the total number. If aluminum/silicon particles are excluded (see Discussions and Conclusions), then the zirconium-containing particles represent 32% of the total number. Table 5 summarizes the mean diameter and volume fractions of the particles tabulated for the histogram (Figure 9).







Figure 9. Particle counts by composition.

# TABLE 5. APPROXIMATE PARTICLE DISTRIBUTION ON SPECIMEN M-72

Excluding Al-Si & Other Particles	~**	Particle Count
Zr = 6%		87
Zr-Ag = 10%	32%	
Zr-U = 16%		
U = 17%		
$Fe^{+} = 38\%$		•
Other - 13%		
Including Al-Si & Other Particles		
Zr = 4%		142
Zr-Ag = 6%	20%	
Zr-U = 10%		
U = 11%		•
Fe <sup>+</sup> = 23%		· ·
Al-Si = 39%		
Other = 7%		

# X-Ray Diffraction Analyses

All diffraction patterns were taken with a Philips diffractometer that was fitted with a diffracted-beam monochromator. XRD patterns were obtained for three specimens, each prepared slightly differently. The first specimen was prepared by collecting debris on double-stick carbon tape that was mounted on a glass slide. Approximately 1 sq cm of the tape was used as a diffractometer sample. The second sample consisted of debris sandwiched between two layers of 0.25 mil Mylar foil. The third sample was prepared by coating a layer of debri. on an aluminum slide with Krylon spray lacquer. This specimen preparation method generally contributes little scattering background.

A usable X-ray diffraction pattern was obtained for each of the specimens; however, in every case the background was high. Mormal background levels were obtained with the specimens in place with the

X-ray beam off, indicating the background was not due to specimen radioactivity. The background rose only after the X-ray beam was turned on, signifying incoherent scattering. Since incoherent scattering is not a problem for well-crystallized specimens mounted on aluminum with a Krylon spray, and since the high background was not due to specimen radioactivity, much of the debris was either poorly crystallized or amorphous.

The diffraction patterns contained many lines that often overlapped. Due to the complexity of the patterns and the high background, not all lines could be identified. Table 6 summarizes the diffraction lines that were obtained. Only three phases were positively identified:

- 1. A face-centered cubic (fcc) spinel with a lattice parameter of 8.39 Å (probably  $Fe_3O_4$ )
- 2. An fcc phase with a lattice parameter of 5.467 A
- 3. An fcc phase with a lattice parameter of 5.33 A.

The latter two phases were respectively oxygen-deficient and oxygen-rich UO<sub>2</sub>. Three additional phases were tentatively identified:

- 1. U0<sub>3</sub>•2H<sub>2</sub>O
- 2. FeO·OH
- 3. Zr0<sub>2</sub>.

The lines for these three phases were weak, and too many of the lines overlapped with other lines for positive identification. The high aluminum/silicon content of the debris suggested that a significant quantity of zeolite might be present. Since many zeolite patterns have spacings similar to the schoepite  $(UO_3 \cdot 2H_2O)$  strong lines, this possibility cannot be discounted.

Metallic uranium cannot be excluded with certainty because the strongest lines would be obscured by lines from other phases. The

Line (Å)	Intensity
7_27	very strong
4.88	moderately weak
4.12	weak
3.59	very strong
3.34	sharp, moderately weak
3.24	weak
3.15	sharp, strong
3.08	moderate
2.96	weak
2.83	broad, moderately weak
2.73	moderate
2.70	weak
2.66	broad, moderately weak
2.53	sharp, moderately strong
2.49	mcderately weak
2.10	weak
1.93	sharp, moderate
1.88	broad, moderate
1.79	broad, weak
1.65	moderately weak
1.63	broad, very weak
1.61	broad, weak
1.54	broad, weak
1.49	moderately weak
1.45	very weak

TABLE 6. X-RA	( DIFFRACTION	PATTERN	SUMMARY
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absence of any of the weaker lines for metallic uranium suggests that it is not present in significant quantity. The same applies to uranium hydride. Although its two strong lines coincide with those of magnetite and uranium dioxide, its third strong line was absent from the diffraction pattern. The strong lines for metallic zirconium, zirconium-silver alloys, and zirconium hydride were also absent from the diffraction pattern, indicating that these phases were not present in significant amounts.

#### Thermal Analysis

DSC was used to determine if the debris removed from the leadscrew would undergo an exothermic reaction upon heating in air. The measurements were made using a Perkin Elmer DSC. A 1.15 mg sample of the debris in an aluminum pan covered with a crimped aluminum lid was analyzed. Heating the specimen from room temperature to 500°C produced only a small, spread out, endothermic peak in the region from 310°C to 450°C (see Figure 10). This peak was probably due to a slight phase change rather than a chemical reaction, since the specimen only absorbed 46 cal/g over the 140°C interval. The absence of any detectable exotherm strongly suggests that a pyrophoric reaction due to exposure of the debris to air is unlikely.

#### Radioactivity Measurements

Gamma spectroscopy and TLD measurements were used to characterize the radioactivity in the debris and on the leadscrew before and after debris removal. Activation products, such as  $^{60}$ Co, contributed only a minor amount of the radioactivity on the leadscrew or in the debris.

Gamma spectral data for a 0.235 g debris sample on filter caper contained inside two plastic vials was obtained using a germaniumlithium drifted coaxial detector. The <sup>22</sup>Na and <sup>60</sup>Co standards were measured in the same counting geometry as the debris sample. The results are summarized in Figure 11 and Table 7. Most of the gamma activity was due to  $13^7$ Cs,  $13^4$ Cs, 125Sb, 144Ce, and 60Co. The 137Cs



Figure 10. DSC analysis of leadscrew debris.





contributed 58%, <sup>134</sup>Cs contributed 13%, <sup>60</sup>Co contributed 12%, and <sup>125</sup>Sb and <sup>144</sup>Ce each contributed approximately 8% of the total gamma activity. The specific activity of the debris for <sup>137</sup>Cs was calculated to be approximately 1275  $\mu$ Ci/g. The specific activity for <sup>134</sup>Cs was 165  $\mu$ Ci/g, and for <sup>60</sup>Co was 75  $\mu$ Ci/g. TLD measurements of the 0.235 g debris sample gave a dose rate of 4 mr/hr through 125 mils of aluminum at 8 in.

Energy (ke¥)	Integral	Area	Relative Area	% Gamma Contribution to Total Activity	Radionuclide
75-86	115082	2624			Pb X-rays + Ce-144
133.5	105899	23369	4.67E7	7.4	Ce-144
176.4	64913	<b>4286</b>	8.93E6	1.4	Sb-125
427.8	65614	10516	3.00E7	4.8	Sb-125
463.4	34918	1847	5.60E6	<1.0	Sb-125
511.0	27443	3011	9.71E6	1.5	Annihilation
569.3	15332	1052	3.63E6	<1.0	Cs-134
604-606	46962	14426	5.15E7	8.2	Cs-134 + Sb-125
635.9	17886	813	3.01E6	<1.0	Sb-125
661.6	136326	95126	3.66E8	57.9	Cs-137
795.8	29863	6438	2.93E7	4.6	Cs-134
1173.2	11286	4559	3.51E7	5.6	Co-60
1332.5	8498	4651	4.23E7	6.7	Co-60

TABLE 7. GAMMA SPECTRUM OF TMI-2 LEADSCREW DEBRIS ~ SAMPLE 14B

Using TLDs, gamma and beta dose rate measurements at the leadscrew surface were obtained before and after cleaning. Figure 12 shows the TLD configurations that were used, with the results tabulated in Table 8. Various filters were used for the TLDs, ranging from thin Mylar to 125-mil aluminum, to allow separation of beta and gamma radiation. The Mylar is transparent to most of the beta energies, while the 125-mil aluminum foil absorbs all of the beta radiation and photons with energies of up to 50 keV. Table 8 shows that the radioactivity was nonuniformly distributed on the leadscrew both before and after cleaning. While the apparent decontamination factor due to cleaning was 40 for beta radiation



Figure 12. Position of TLDs on leadscrew before and after cleaning.

	Bet	a	Gan	Gamma	
Position	Before cleaning c	After Cleaning <sup>d</sup>	Before <u>Cleaning</u> e	After <u>Cleaning</u> f	
1	14.0	5.5	5.1	2.8	
2	152.0	18.7	15.9	5.2	
3	1080.0	46.1	54.0	10.7	
4	1170.0	55.4	55.0	17.9	
5	1170.0	40.0	53.0	17.4	
6	1190.0	37.1	60.2	19.7	
7	1240.0	28.3	58.8	21.0	
8	1280.0	19.6	66.5	23.5	
9	1200.0	16.7	64.8	26.7	
10	1220.0	34.6	75.5	30.3	
11	1500.0	25.7	90.2	30.2	
12	1750.0	37.9	103.0	30.7	
13	2170.0	44.5	110.0	28.1	
14	1810.0	20.3	59.9	22.7	
15	54.5	14.3	13.5	16.3	
16	14.5	16.3	6.0	8.9	

TABLE 8. THI LEADSCREW BEFORE AND AFTER CLEANING-READINGS R/HR AT CONTACT

a. Average beta decontamination factor is 41.3.

b. Average gamma decontamination factor is 3.1.

c. Average beta dose rate for positions 3 through 14 before cleaning was 1400 R/hr.

d. Average beta dose rate for positions 3 through 14 after cleaning was 33.9 R/hr.

e. Average gamma dose rate for positions 3 through 14 before cleaning was 70.9 R/hr.

f. Average gamma dose rate for positions 3 through 14 after cleaning was 23.2 R/hr.

and 3 for gamma radiation, the relative spatial distribution of radioactivity on the leadscrew was unchanged. Gamma spectra of the leadscrew suggested that most of the radioactivity was due to 137Cs followed by 134Cs, and that the 60Co intensity was small.

### DISCUSSION AND CONCLUSIONS

In summary, the surface contamination on the leadscrew is due to debris from failed fuel elements and primary side corrosion products (oxides). SEM examination of the debris found particles that were primarily uranium, uranium/zirconium, zirconium, zirconium/silver, iron, and silicon/aluminum. All particles except the uranium and uranium/ zirconium appeared to be agglomerates of smaller particulates. By population, most of the particulates were smaller than 10 µm, but particles larger than 10 µm were responsible for most of the volume. Backscatter SEM micrographs suggested that the particles containing uranium were generally larger than other particles. The ICP elemental analysis supports the SEM observations that uranium, zirconium, iron, silicon, and aluminum account for most of the composition. The high silicon and aluminum levels were a surprise, and may have been due to zeolite contamination, cross contamination with dirt, or filler material from an RTV sealant applied to the tank used to clean the leadscrew.

Based on the elemental composition of the debris, possible pyrophoric species in the debris are metallic zirconium, zirconium/silver alloy, or uranium, as well as zirconium and uranium hydrides. However, XRD analysis of the debris found no evidence that these potentially pyrophoric species are present in significant quantities. Furthermore, based on a DSC analysis of the debris, no exothermic reaction occurred when the debris was heated in air between room temperature and 500°C. Chemical and thermal characterizations of the debris thus indicate that the possibility of a pyrophoric process involving the debris from the leadscrew is quite small.

Radiochemical characterization of the debris and leadscrew found that most of the radioactivity was due to mixed fission products. The total specific activity for the debris was about 1500  $\mu$ Ci/g. The dried debris was easily airborne and difficult to work with, although it did settle quickly from an agitated Freon mixture. Since dose rates on the leadscrew were nonuniform along the leadscrew length, the composition of the debris may vary significantly between different specific locations.

# REFERENCES

1. G. M. Bain and G. O. Hayner, <u>Initial Examination of the Surface</u> Layer of a 9-Inch Leadscrew Section Removed From TMI-2, Final Report, EPRI RP2056-2, Task 1, June 1983.

30

2. A. D. Zimon, <u>Adhesion of Dust and Powder</u>, New York: Plenum Press, 1969.