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INTERIM REPORT ON THE TMI-2 PURIFICATION FILTER EXAMINATION

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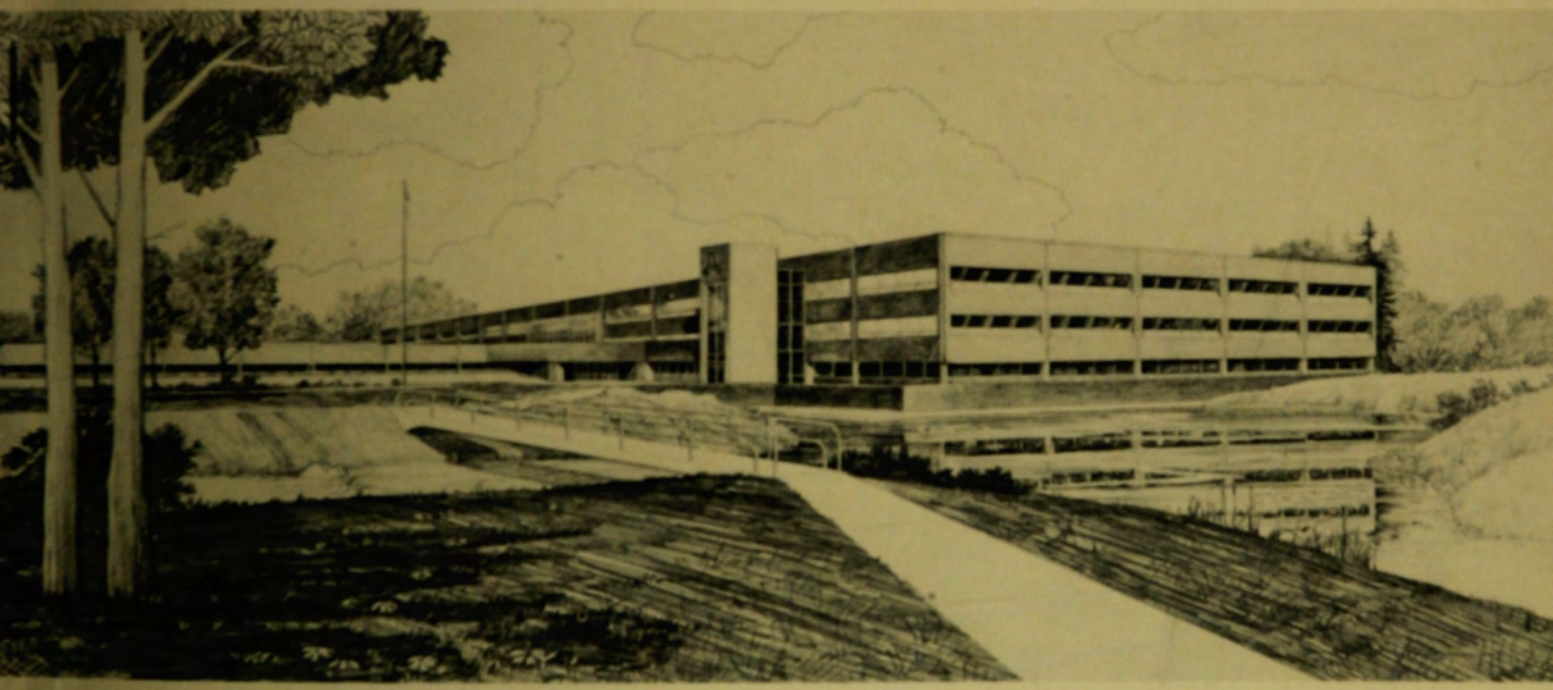
Idaho National Engineering Laboratory
Operated by the U.S. Department of Energy



This is an informal report intended for use as a preliminary or working document

Prepared for the
U.S. DEPARTMENT OF ENERGY
Idaho Operations Office
Under DOE Contract No. DE-AC07-76ID01570

EG&G Idaho

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ABSTRACT

Filters from the purification/makeup system of the Three Mile Island Unit 2 Reactor were examined after the March 28, 1979, accident to determine the character of the debris transported to the filters. The general condition of the filters is presented. Material was removed from the filters and examined. The elemental and radionuclide makeup of the debris is discussed. Distribution of particle size and shape is presented for some of the material examined. This is an interim report. When the investigation is completed, another report summarizing all of the data will be issued.

SUMMARY

Part of the cold leg coolant flow from the TMI-2 reactor is diverted to the purification/makeup system during normal reactor operation. The system was online during the TMI-2 accident. The filters in the purification/makeup system were removed after the accident and the material on the filters was examined.

The purification/makeup system did not receive coolant containing core debris until about 16 h into the accident, at which time the first filter in line (MUF-5A) became blocked and was by-passed. The second MUF-5 filter was by-passed shortly thereafter. The remaining filters were online at various times during the accident and for an extended period after the accident. Five filters were examined: three large filters upstream of the makeup tank, and two smaller filters downstream of the makeup tank. In addition to the purification/makeup system filters, the eight vacuum cartridge filters used to vacuum loose debris out of the system filter housings were examined.

Most of the filter paper and debris was missing from the three large filters. Small Filter MUF-4A was intact, loaded with dark debris, and its frame collapsed. Small Filter MUF-5B was intact, with only a small amount of debris on it, and its filter paper swollen.

The filter debris was analyzed to determine elemental and radionuclide makeup of the debris. Less than 100 g of UO_2 was received at EG&G Idaho with the purification/makeup system and vacuum filters. Non-fuel-rod components generated the largest amount of debris. Stainless steel/Inconel alloying elements were found in 67% of the particles examined; control rod materials were found in 60% of the particles. Most particles contained many different elements. The activity of ^{137}Cs, ^{134}Cs, ^{125}Sb, $^{106}Ru/Rh$, and ^{90}Sr in the debris was greater than can be accounted for by the fuel content of the debris. It is most probable that these fission products were released from the fuel, deposited on aerosolized control rod and spacer grid material, and transported to the filters with these particles, or were washed from the TMI-2 debris bed.

Distribution of particle size and shape was determined on particles from Filters MUF-5B and MUF-2B and homogenized Sample 0104 from Filter MUF-5B. The mean particle size was <6 μm, and size range was from <1 to 50 μm. It is noted, however, that the coolant containing the reactor debris had to travel through a steam generator, letdown coolers, and a block orifice before it reached the filters. These structural components may have removed relatively large particles from the coolant before it reached the filters. Also, the components provided surfaces for deposition of smaller particles.

Particle shape varied from very irregular to nearly spherical. The mean sphericity was 0.61. The spherical particles were probably control rod materials, the irregular particles generally stainless steel/Inconel or zirconium components.

ACKNOWLEDGMENTS

We thank the personnel at Argonne National Laboratory-East, Los Alamos National Laboratory, Exxon Nuclear Idaho Company, and EG&G Idaho Hot Cell, Physic, and Fuel Behavior Branches who have worked with, examined, or analyzed the data presented in this report. We specially thank the principal investigators: C. V. McIsaac (EG&G Idaho), R. L. Nitschke (EG&G Idaho), N. D. Stalnaker (Los Alamos National Laboratory), and R. V. Strain (Argonne National Laboratory). We also specially thank P. B. Hembree for assistance in preparing the figures.

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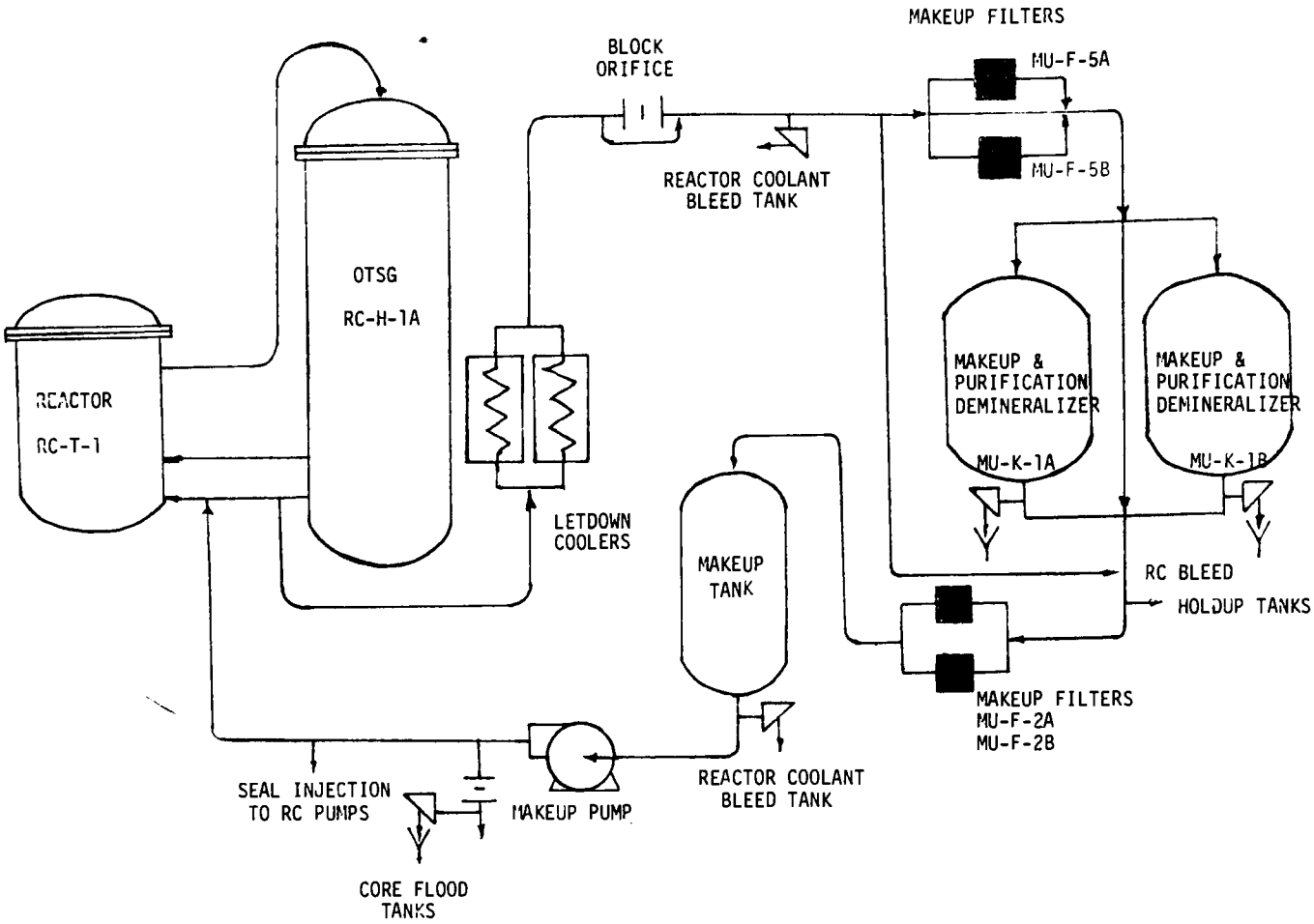
INTERIM REPORT ON THE TMI-2 PURIFICATION FILTER EXAMINATION

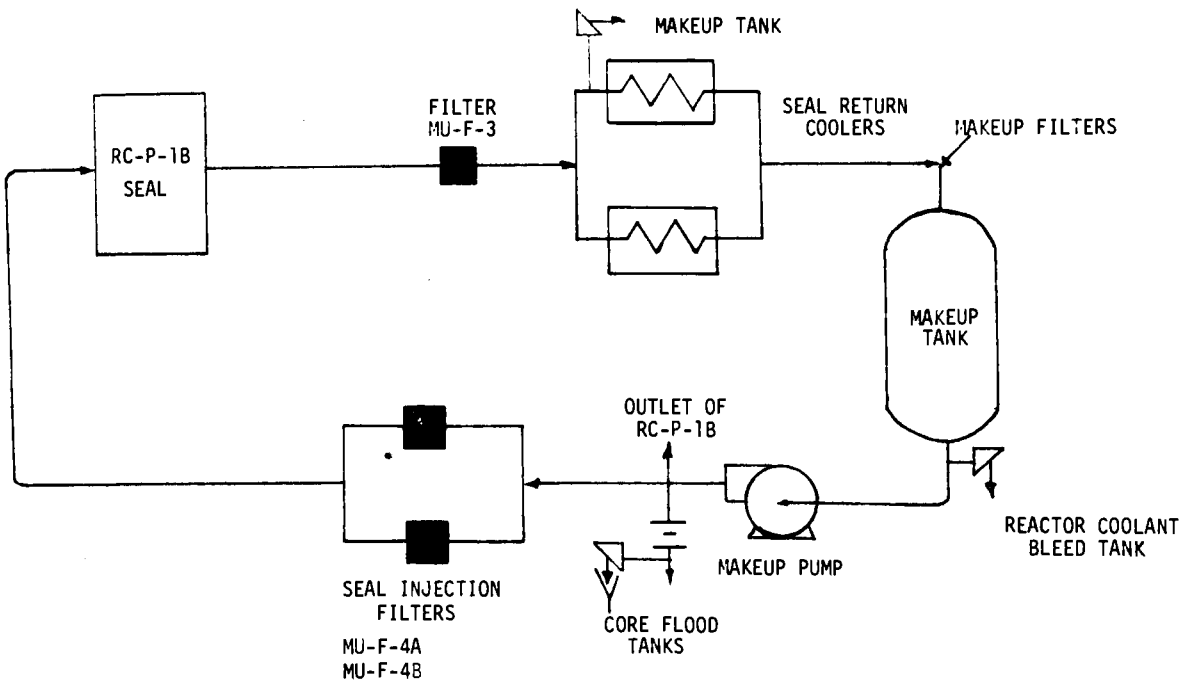
INTRODUCTION

The accident at Three Mile Island Unit-2 (TMI-2) on March 28, 1979, provides a unique opportunity to investigate severe accident damage and advance the knowledge of light water reactor safety. Recognizing this, four organizations joined together after the accident to acquire technical data during reactor recovery. These organizations, commonly referred to as the GEND group (General Public Utilities, Electric Power Research Institute, Nuclear Regulatory Commission, and the Department of Energy) are conducting a variety of data acquisition and reactor recovery tasks. One of these tasks is an examination of the purification/makeup filters and vacuum cartridge filters from the TMI-2 reactor. The examinations performed at EG&G Idaho, Inc., and the Los Alamos National Laboratory (LANL) were funded by the Department of Energy. The work performed at Argonne National Laboratory-East (ANL-E) was funded by the Nuclear Regulatory Commission.

The TMI-2 purification/makeup system purifies and recirculates reactor coolant and supplies water to the reactor coolant primary pump seals. A fraction of the primary coolant water is diverted from the primary coolant cold leg through the purification/makeup system and back into the cold leg. A simplified schematic of the system is shown in Figure 1. The flow leaves the cold leg through letdown coolers and a block orifice. At this point, some of the flow may be diverted to reactor coolant bleed tanks. The flow then travels through Filter MUF-5A and/or -5B, one of two purification/makeup demineralizer tanks, Filter MUF-2A and/or -2B, and into a makeup tank. Either one or both of the MUF-5 and -2 filters, and the demineralizer tanks, can be valved into the line or by-passed completely. Some of the flow from the makeup tank is diverted through two seal injection filters (MUF-4A and -4B) and then to cool the reactor coolant primary pump seals. The flow through the pump seals then proceeds through another filter (MUF-3), through seal return coolers, back to the makeup tank, and then back to the primary coolant cold leg.



(A)



(B)

Figure 1. Schematic of the purification/makeup filter system in the TMI-2 reactor.

All the "A" filters were on-line when the accident began. However, the TMI-2 primary coolant flow was stopped from 1 h 40 min to 15 h 50 min after the accident began, except for two brief periods (recorded as spikes) at about 2:54 and 15:33 h into the accident. The purification/makeup system was not valved off during this time, but there was very little coolant available in the cold leg for the system to divert. At about 4 h 26 min into the accident, the first major relocation of core material occurred. Therefore, the filters in the purification/makeup system first received coolant containing core debris about 11 or 12 h after the first major core material relocation occurred. Shortly after flow was reestablished, Filter MUF-5A became blocked and MUF-5B was added to the system. Shortly after that, both MUF-5 filters were by-passed and the inlet valve to MUF-2B was opened.

The flow rate into the purification/makeup system and the primary coolant pump seals was 40 gpm prior to the accident (i.e., all purification/makeup system flow was being diverted to the primary coolant pump seal loop). The seal injection flow was reduced from 31.5 to 9.5 gpm between March 30 and 31, and the purification/makeup system flow rate over this period was estimated to be ~25 gpm.

Coolant from reactor coolant bleed Tank B was batch fed into the makeup tank for approximately one week after the accident. After that, the coolant in reactor coolant bleed Tank A was fed into the makeup tank. Approximately 47,000 gal of coolant were fed through the system before the filters were by-passed. However, the coolant from the reactor coolant bleed tanks was probably clean coolant from the TMI-1 reactor system, and should not have affected the material on the filters.

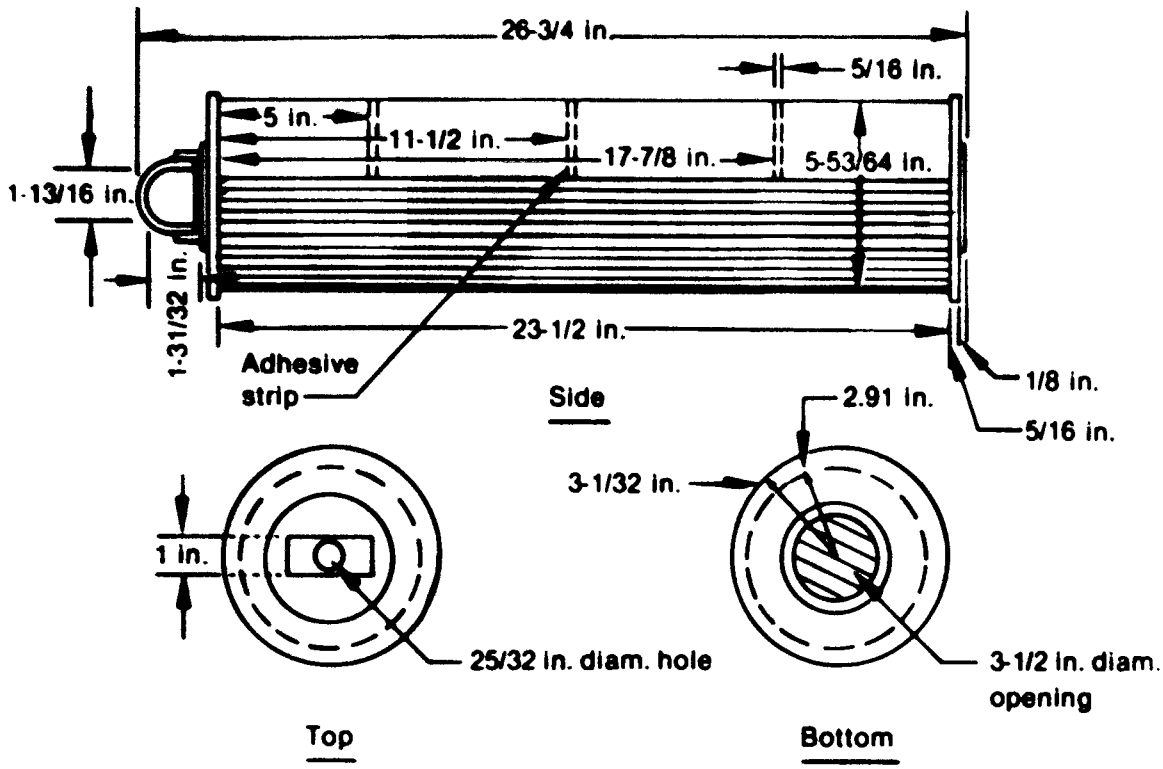
Three large (~0.6 m long) and two short (~0.3 m long) purification/makeup filters were received at the EG&G Idaho hot cells in April of 1982. The long filters (MUF-5B, -2A, and -2B) were located in the system upstream of the makeup tank. The short filters (MUF-4A and -4B) preceded the injection to the primary pump seal. Each filter had a cylindrical perforated stainless steel backing covered by pleated paper. The pleated paper on the large filters was composed of a minimum of four layers of

paper. The outer paper layer was a treated composite of cellulose and glass fiber cardboard type paper, and the multiple inner layers were thin and fine grained. The smaller filters were composed of a perforated stainless steel backing covered with only one layer of treated cellulose cardboard type paper. The filters are illustrated in Figure 2.

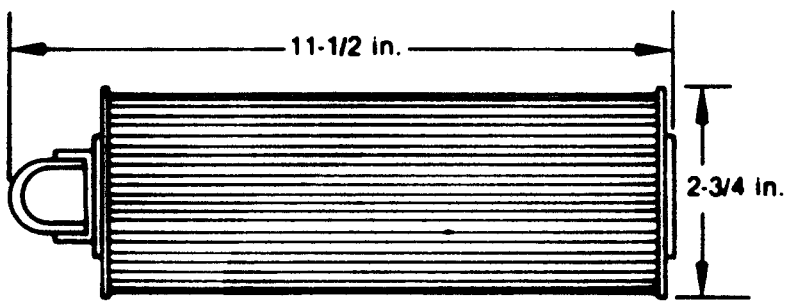
In addition to the purification/makeup system filters, eight vacuum cartridge filters were used to vacuum loose debris out of the filter housings after the system filters were removed. These filters were also sent for examination. The vacuum filters were enclosed in metal canisters and sealed on both ends by spring closed caps, as illustrated in Figure 3.

The primary goals of the filter examinations are to provide data about debris character to the defueling group for consideration in the defueling plans, and provide information to the Off-Site Core Examination Program on light water reactor behavior during accident conditions. Specifically, the filter examination will assist the reconstruction of the TMI-2 accident events, indicate which core components are prone to form fine particulate that redistributes through the primary coolant system of a light water reactor during a severe accident, and provide data on debris chemistry and radionuclide concentrations. To accomplish these objectives, the chemistry of the bulk material and individual particles must be measured along with the particle shape and size distributions.

The information described in this preliminary report is an accumulation of data from EG&G Idaho, Exxon Nuclear Idaho Company (ENICO), ANL-E, and LANL. Details of the examination plan as specified in July 1982 are provided in Appendix A.



Filters MUF 5B, 2A and 2B



Filters MUF 4A and 4B

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Figure 2. Schematic of the TMI-2 purification/makeup filters.

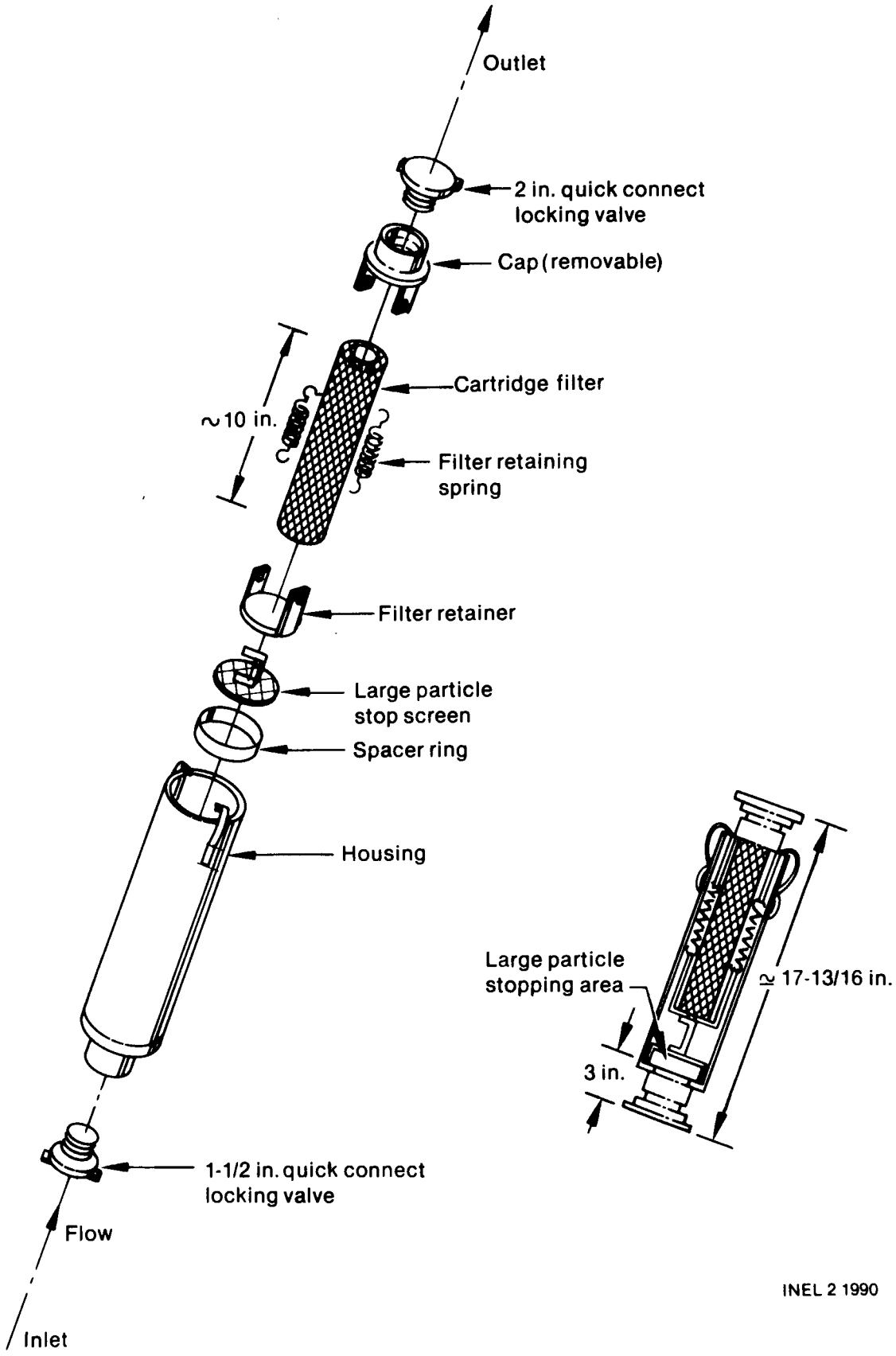


Figure 3. Schematic of the TMI-2 vacuum filter.

The condition of the filters upon receipt at EG&G Idaho and the results of recent particle size, shape, and chemistry measurements performed at EG&G Idaho are discussed in Appendix B. The results of the examination of Sample TMI 0104 at LANL are presented in Appendix C. And the results of the preliminary ANL-E examinations of five samples from Filters MUF-5B and -2A are presented in Appendix D. Additional analysis work is currently in progress at ANL-E and LANL and is described in Tables 1 and 2. No additional examinations are scheduled at EG&G Idaho. All of the analyses will be combined in a final GEND report on the TMI-2 Purification Filter Examination when the ongoing work is completed.

All data have been consolidated, analyzed, and compared in this report. This report discusses the general condition of the as-received filters. It addresses the chemistry of both the bulk debris material and the individual particles, presents particle morphology and size distributions, and finally presents the interim conclusions.

TABLE 1. PLAN FOR ADDITIONAL ANALYSES OF TMI-2 FILTER SAMPLES AT LOS ALAMOS NATIONAL LABORATORY

Experimental Objectives	Method	Filter Samples to be Analyzed	Relevance to TMI Information Base	Remarks
Determine if debris is stratified on filters and if so, how	Electron microprobe elemental mapping of metallographically mounted filter sections	MUF-5B and -2B	Obtain temporal information on events of the accident	Some of these examinations are in progress. After examining filter debris, gamma scanning was deemed inapplicable.
Determine elemental composition of individual particles	Electron microprobe on metallographically mounted samples	MUF-5B, -2A, -2B	Obtain data on what components of the core failed and how	--
Determine H_2, He, Kr, and Xe in debris	Dynamic gas release (mass spectrometry)	MUF-5B, -2A, -2B	Obtain data on gas retention capacity of debris	The high carbon content of these samples may complicate these analyses.
Determine particle size of debris	Image analysis of metallographically mounted filter sections	MUF-5B, -2A, -2B	Obtain data that could be useful in determining the mode of core failure	Difficulties could be encountered in distinguishing filter material from debris.
Determine gross elemental composition	Emission spectroscopy	MUF-5B, -2A, -2B	Obtain data on what components of the core failed and were transported outside the core	These data should help resolve the discrepancies on the elemental composition of the debris.
Determine U and Pu isotopic composition	Mass spectroscopy	MUF-5B, -2A	Obtain data on how the core failed and how to process debris outside the core	--
Determine radioactive element composition	Gamma spectroscopy, gross alpha and gross beta counting, etc.	MUF-5B, -2A, -2B	Obtain data on the release of radioisotopes from the core	Several samples will be fully or partially analyzed in order to discern any differences between samples.
Determine gross gamma ray activity	Gamma scanning spectroscopy	MUF-5B	Obtain information on the uniformity of samples	Those samples not analyzed by other methods will be scanned for unusual gamma ray activity.

TABLE 2. PLAN FOR ADDITIONAL ANALYSES OF TMI-2 FILTER SAMPLES AT ARGONNE NATIONAL LABORATORY-EAST

Experimental Objectives	Method	Filter Samples to be Analyzed	Relevance to TMI Information Base
Determine if debris is stratified and determine particulate character	Image analysis of metallographically mounted filter sections	MUF-5B, -4A, -2A	Obtain data that may be useful in determining core failure modes and determining debris particulate sizes in the primary coolant system
Determine elemental composition, shape, and size of individual particles	SEM/EDS	MUF-4A, -2A, -5B	Obtain data on what components of the core failed and how
Determine elemental composition of individual particles	Electron microprobe on metallographically mounted samples	MUF-4A, -2A, -5B	Obtain data on what components of the core failed and how and determine the particulate composition
Determine gross elemental composition	Emission spectroscopy	MUF-4A, -2A	Obtain data on the release of radioisotopes from the core
Determine filter composition	Emission spectroscopy or X-ray fluorescence	MUF-4B	Obtain data of the filter composition and determine if Si is filter fiber material
Determine radioactive element composition	Gamma spectroscopy	MUF-4B, -2A	Obtain data on the release of radioisotopes from the core

GENERAL CONDITION OF THE FILTERS

Three large and two short purification filters, and eight vacuum cartridge filters were shipped in their original steel canisters from TMI-2. Each filter was in an inner plastic bag just large enough to hold the filter, the mouth of the bag always open. These inner bags with filters were contained in a larger plastic bag partially filled with vermiculite (an adsorbent material). The outer bag was generally sealed with tape.

The physical condition of Filters MUF-5B, -2A, -2B, -4A, and -4B when they were removed from their containment bags is illustrated in Figures 4 through 8, respectively. As Figures 4, 5, and 6 illustrate, there was little filter paper or debris left on the steel frames of the three large filters. However, Filter MUF-4A (Figure 7) was received with almost all the filter paper and debris still on the metal filter frame. This filter was scanned to determine the concentrations of gamma-emitting isotopes on the filter. Figure 9 shows the results of the gross gamma scan of Filter MUF-4A. Much of the gamma-emitting debris was on one end. Figure 10 shows a gamma spectrum of the filter at about mid-height. The primary radioactive elements were Cs, Sb, Ru, and Ce/Pr; the activation elements were Ag, Mn, and Co. The filter paper was very friable and debris and paper fell off as the filter was handled. Also, the top half of the metal filter frame was collapsed, as shown in Figure 7. There was no indication on the filter paper that the filter had been crushed during shipping. Thus, a pressure differential during use at TMI probably caused the filter to collapse. This may have occurred after the filter had been loaded with debris. Nearly all of the filter paper and debris was scraped from the filter. The MUF-4A filter paper and debris weight was 82.7 g.

Short purification Filter MUF-4B was also received intact. However, it was not loaded with the dark debris found on all the other filters, and the filter paper was swollen, as shown in Figure 8. The filter contained cesium and cobalt, as indicated by the gamma spectrum shown in Figure 11. Cesium was the major fission product on the filter. The gross gamma scan of Filter MUF-4B is shown in Figure 9, along with the MUF-4A scan. Although the gamma intensity of Filter MUF-4B was much lower than the gamma radiation

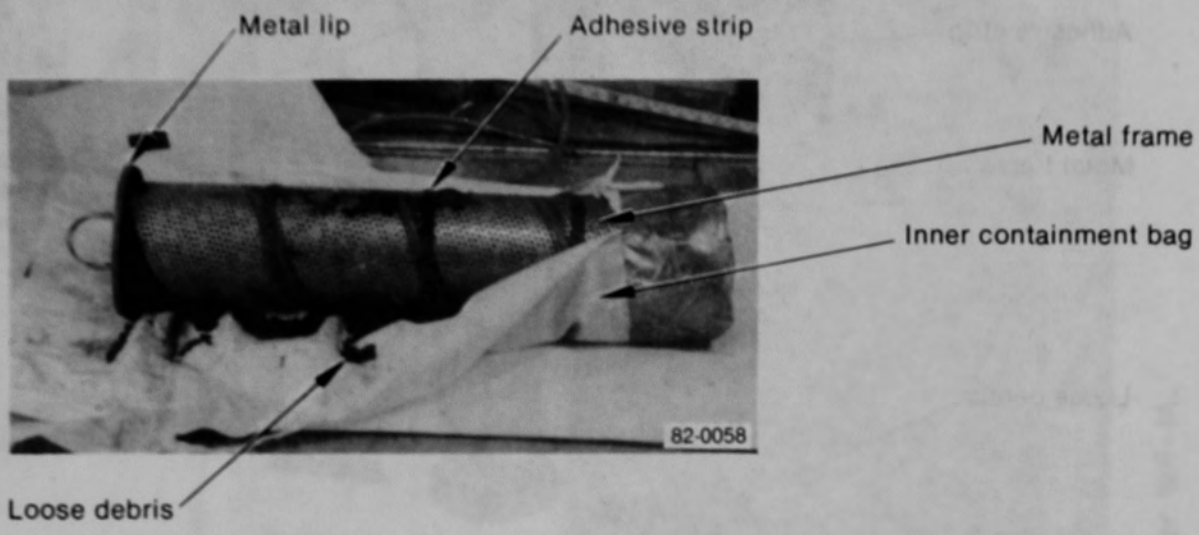


Figure 4. Purification/makeup Filter MUF-5B.

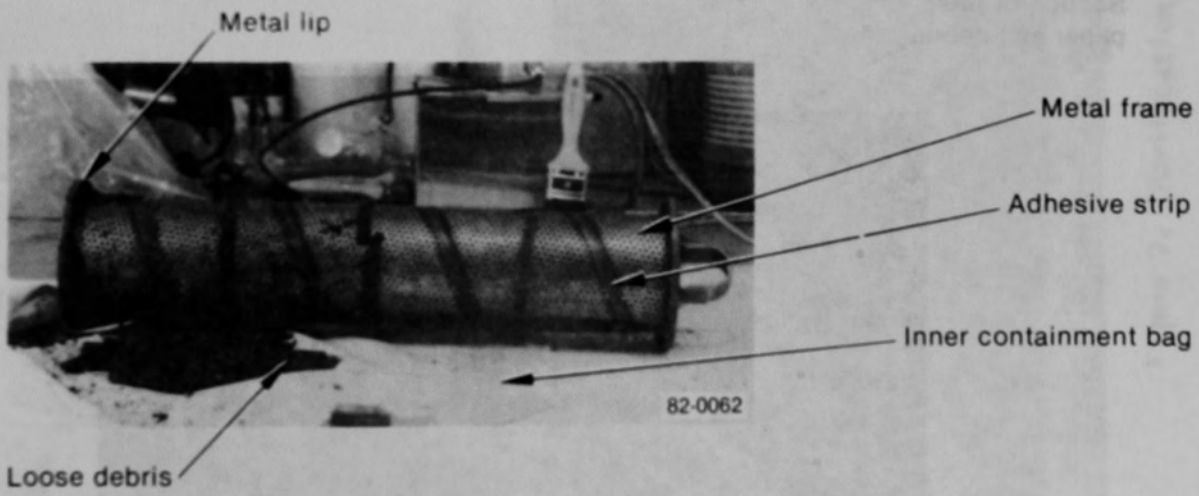
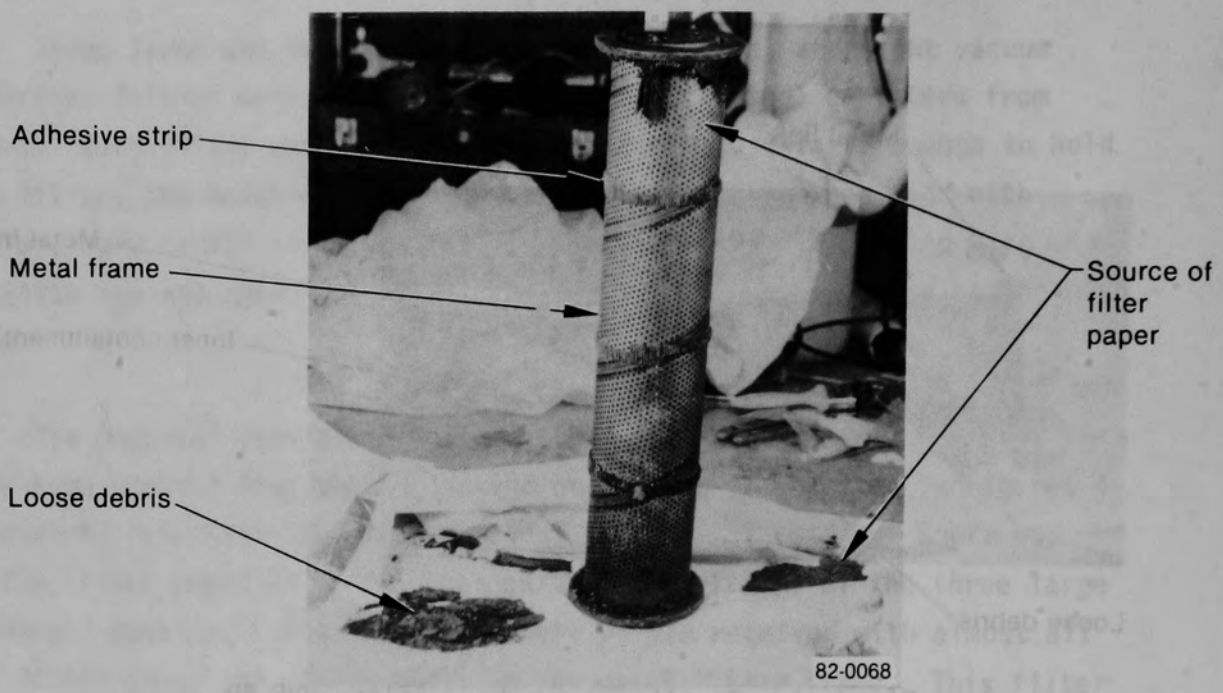


Figure 5. Purification/makeup Filter MUF-2A.



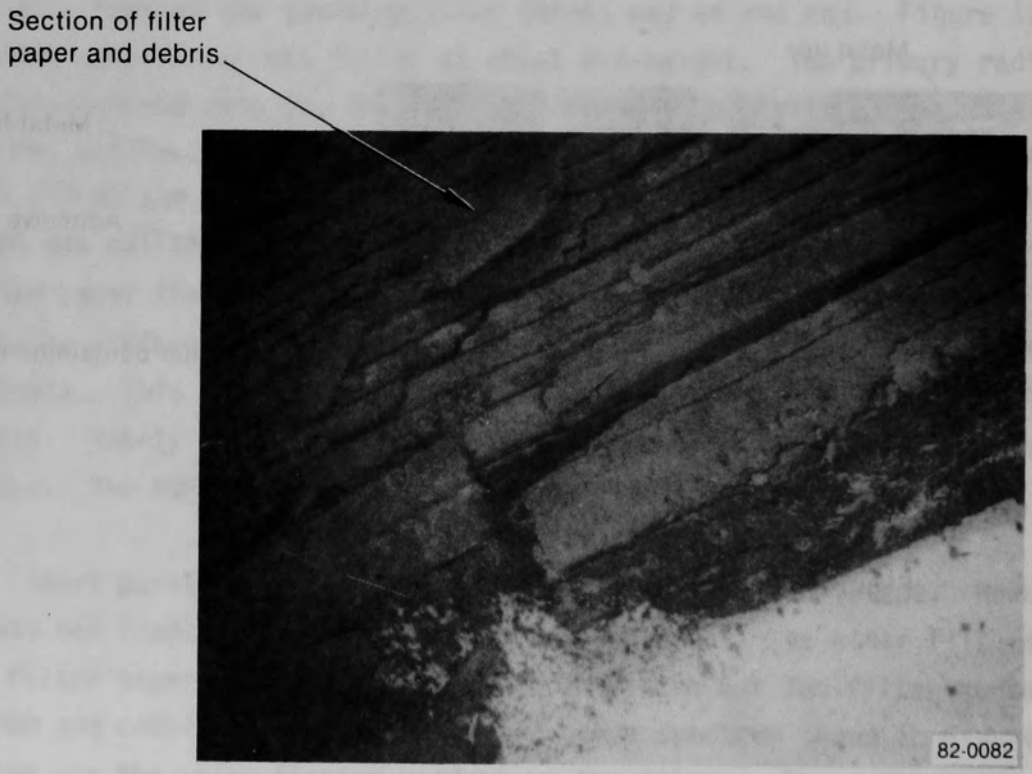


Figure 6. Purification/makeup Filter MUF-2B.

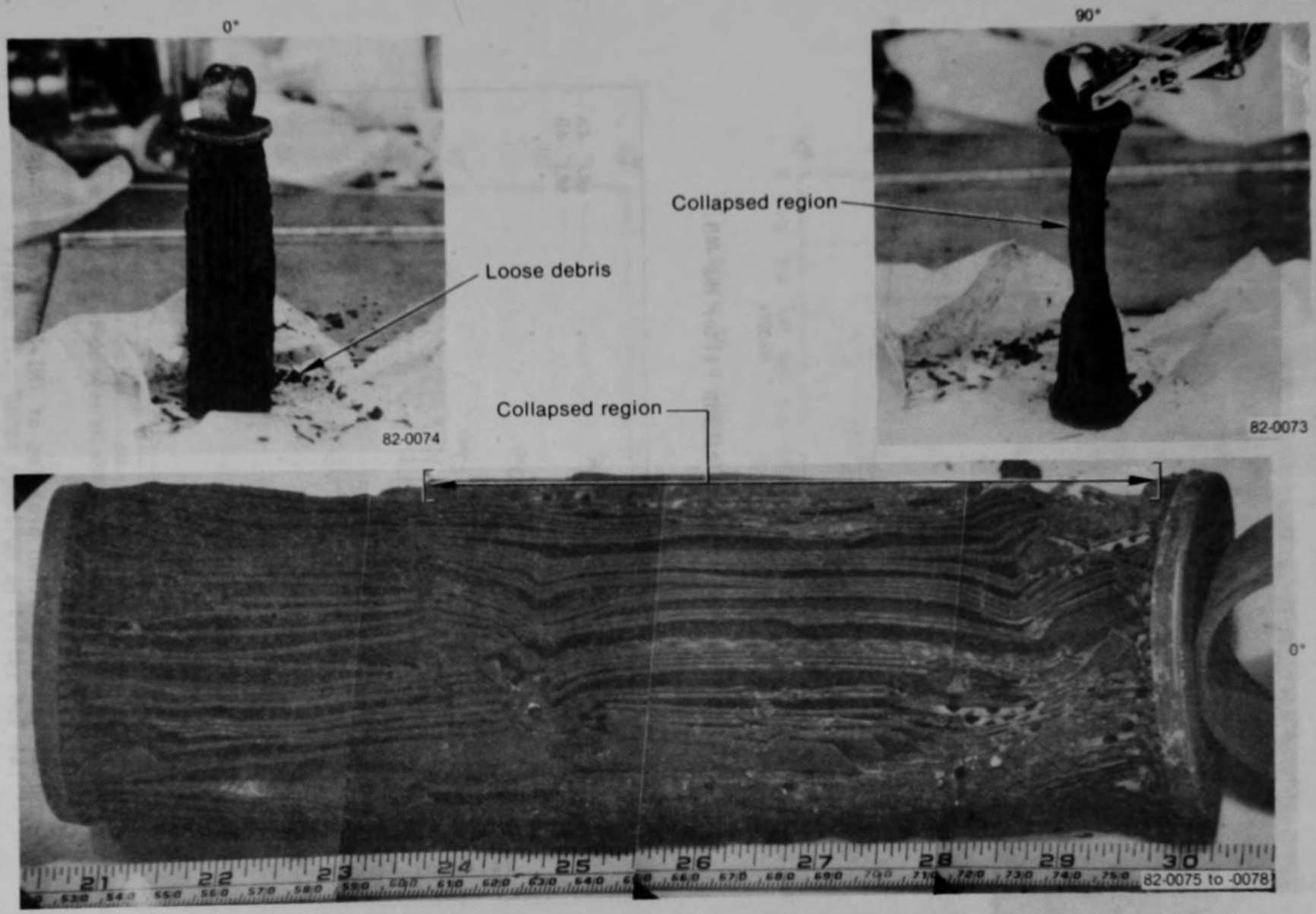


Figure 7. Purification/makeup Filter MUF-4A.

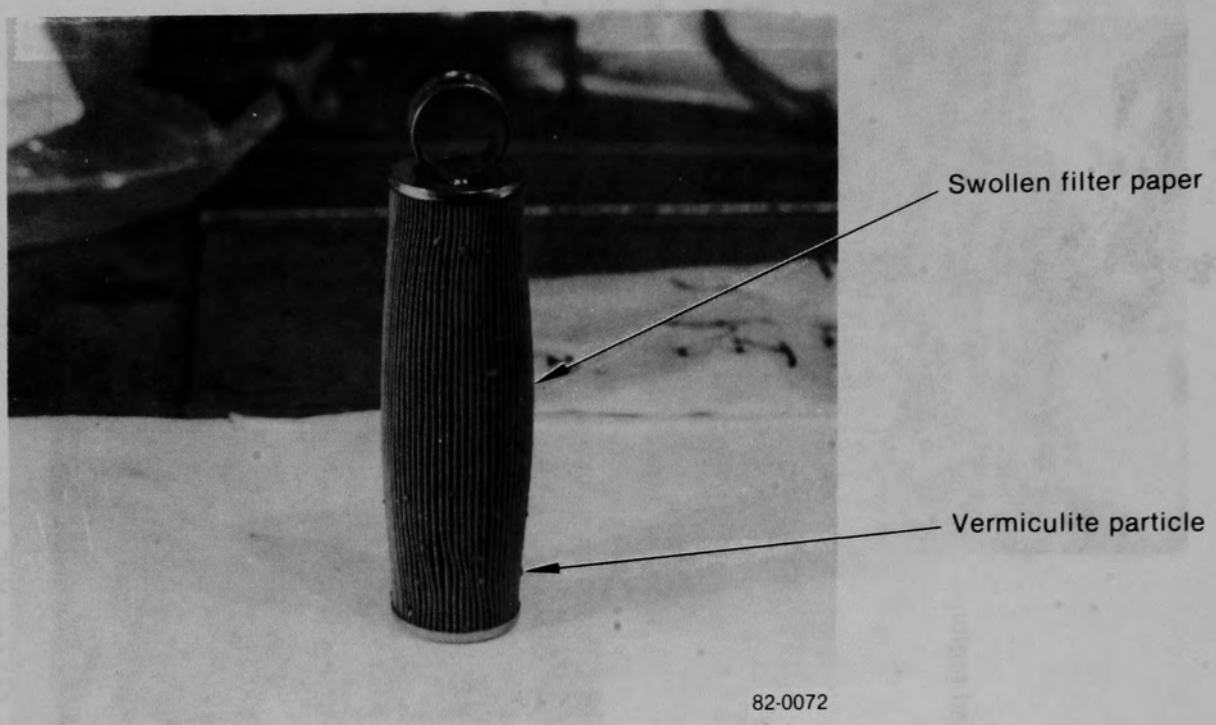


Figure 8. Purification/makeup Filter MUF-4B.

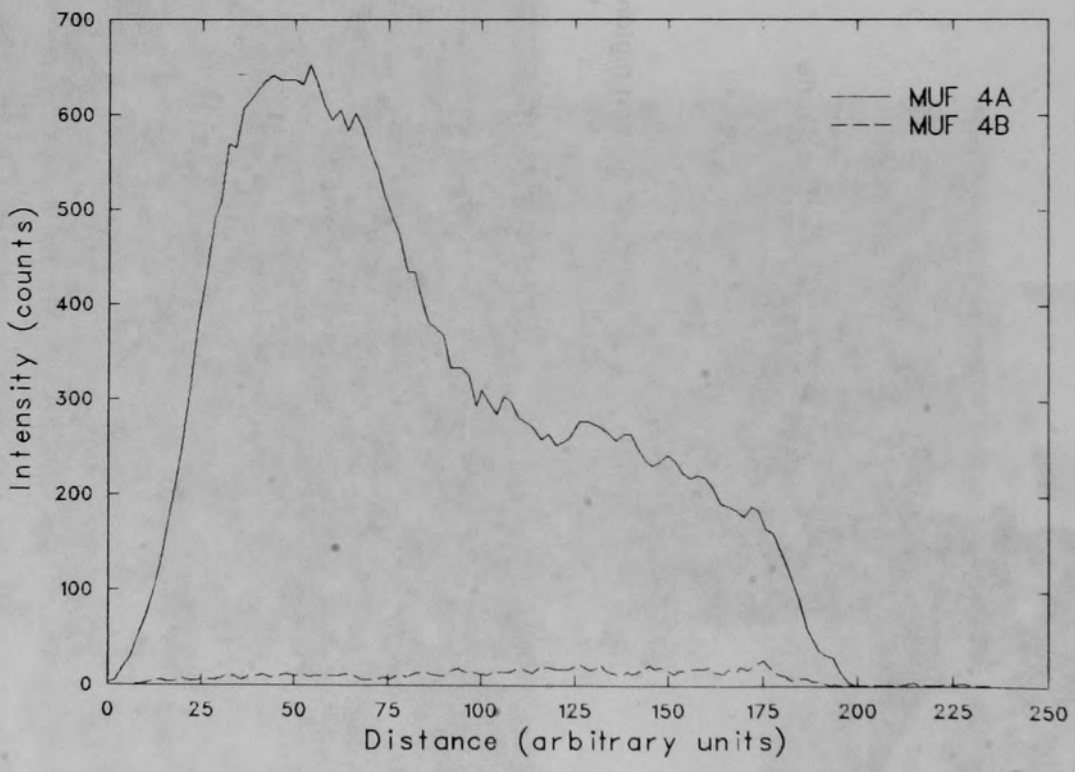


Figure 9. Gross gamma radiation profiles of TMI-2 Filters MUF-4F and -4B.

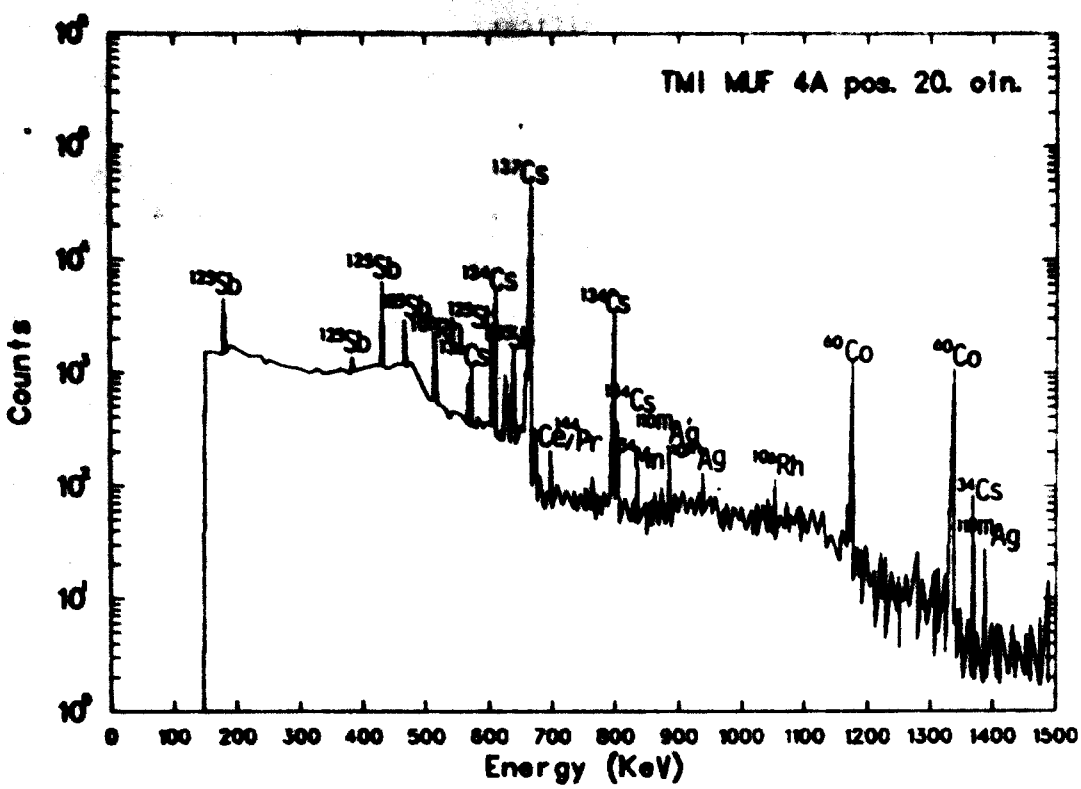


Figure 10. Gamma spectrum of Filter MUF-4A.

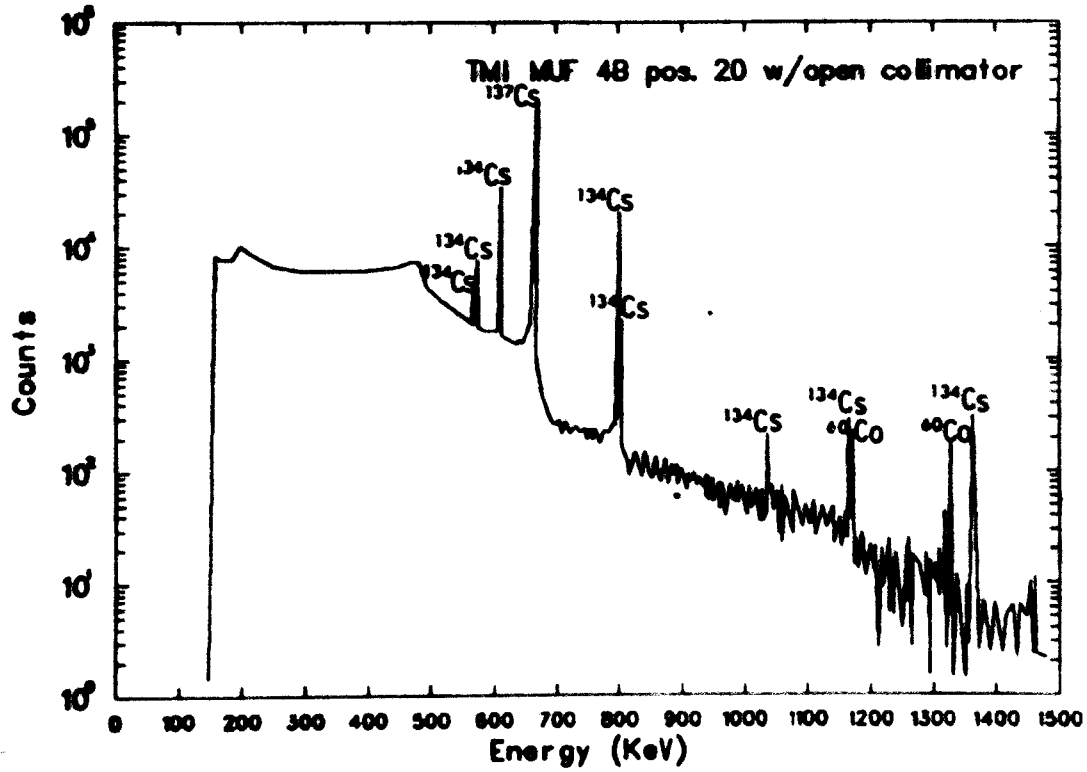


Figure 11. Gamma spectrum of Filter MUF-4B.

of Filter MUF-4A, it was still significant. The absence of significant quantities of other fission products, such as Ce, suggests that there was very little fuel on Filter MUF-4B. The uranium concentration based on the spectral gamma scan was <0.007 g. Apparently, there was no flow through Filter MUF-4B during or immediately after the accident, and the flow through Filter MUF-4B occurred only during a period when water soluble elements were present.

The eight vacuum filters received at the EG&G Idaho hot cells were opened, gamma scanned, and repackaged for archive storage. A typical vacuum filter after removal from the metal canister is shown in Figure 12. The radioisotope loading on these filters and on the MUF-4A and -4B purification filters is described in Table 3.

The gross gamma scans of vacuum filters VAC-MUF-4A and -4B are shown in Figure 13. Although the radioactive loading was low on VAC-MUF-4B, as expected, it did pick up cesium and cobalt from the filter housing of MUF-4B.

The total weight of the debris (filter paper and material collected from the filter) from each filter is given in Table 4. The filters, although severely deteriorated, had much of their original debris loading with them in their containment bags. Filters MUF-5B and -2B probably had about 50% of their original loading, and Filters MUF-2A, -4A, and -4B had ~100% of their original loading.[a] Most of the vacuum filters were also loaded with debris. The exact amount of uranium contained in the debris is not known, but an uranium concentration of 5 to 7% was estimated, based on analyses of debris from the first MUF-5B filter Samples 0104 and 0105. If 5 to 7% of the debris is uranium, then between 70 and 100 g of uranium were received with the purification and vacuum filters. This estimate matches the GPU estimate of ~11 g for Filter MUF-5B.

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a. It is assumed that the original loading on Filters MUF-5B, -2A, and -2B was at least as much per unit of surface area as the loading on Filter MUF-4A (which was received with almost all the filter paper and debris in place on the stainless steel frame).

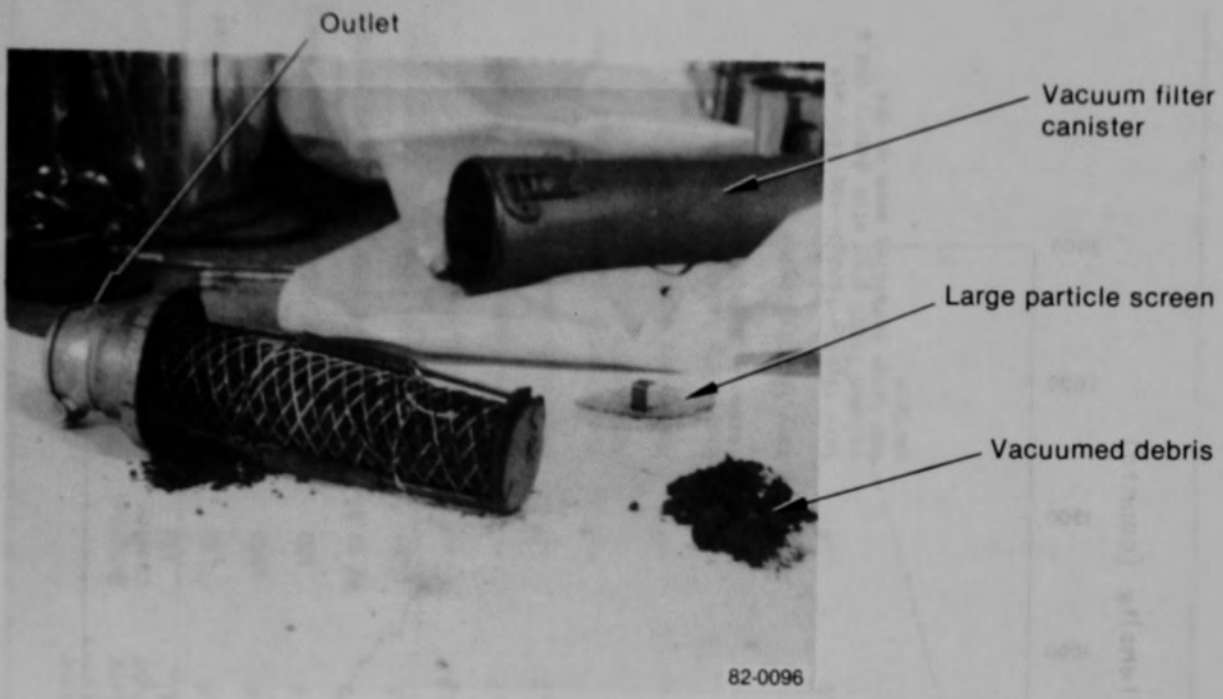

Figure 12. Vacuum Filter VAC-MUF-2A-1.

TABLE 3. PURIFICATION AND VACUUM FILTER RADIONUCLIDE INVENTORY NUCLIDE ACTIVITY (μCi)[a]

Filter	^{137}Cs	^{125}Sb	$^{106}Ru/Rh$	^{134}Cs	^{60}Co	^{110m}Ag	^{54}Mn	$^{144}Ce/Pr$	Total μCi
MUF-4A	295 000	59 500	36 700	24 100	12 500	1 350	587	52 900	486 000
VAC-MUF-4A	4 190	376	478	345	223	10	8	847	6 480
MUF-4B	17 770	<17	<20	1 480	13	<1	<5	<74	19 200
VAC-MUF-4B	23 700	<19	<22	1 960	25	<1	<1	<81	25 700
VAC-MUF-2A-1	437 000	130 000	47 100	36 800	15 100	1 110	484	54 900	72 200
VAC-MUF-2A-2	7 880	1 940	748	650	266	22	9	973	12 500
VAC-MUF-2A-3	124 000	23 200	9 970	9 900	2 810	224	121	12 200	18 200
VAC-MUF-2B	12 000	<14	<16	992	5	<1	<1	<57	13 000
VAC-MUF-5A	2 660	479	216	221	194	5	5	252	4 030
VAC-MUF-5B	4 900	139	46	404	33	<1	<1	88	5 610
Total	932 000	216 000	95 300	76 900	31 200	2 720	1 220	122 000	1 480 000

a. Decay corrected to 9/1/82.

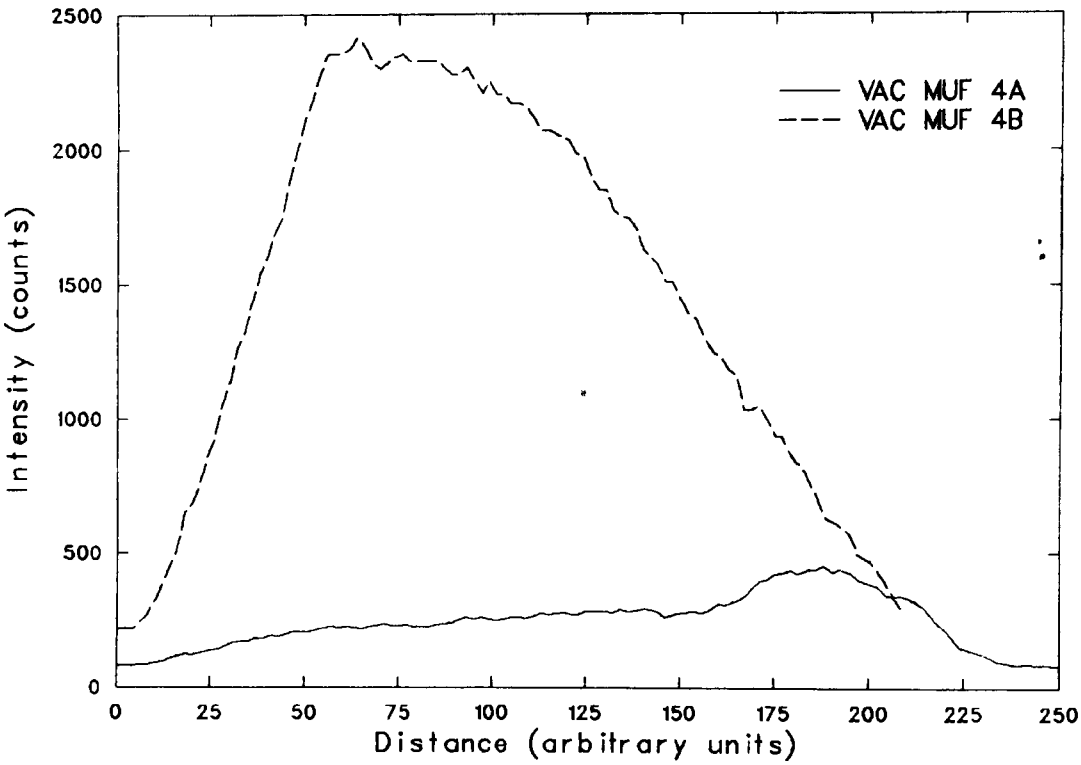

Figure 13. Gross gamma scans of vacuum Filters VAC-MUF-4A and -4B.

TABLE 4. NET WEIGHT OF DEBRIS COLLECTED FROM TMI-2 PURIFICATION FILTERS

Filter	Debris Weight[a] (g)	Estimated Content (g)		Storage Drum Identification	Filter and Debris Weight (g)	Original Loading (%)	Description
		Weight[b]	Gamma Scan				
MUF-5B	203.9	14.3	--[d]	--[c]	--[d]	~50	Filter paper completely deteriorated
MUF-4A	82.7	5.8	5.1	03	--[d]	~100	Filter heavily loaded and intact
MUF-4B	--[d]	0	0.007	03	--[d]	100	Filter very lightly loaded and intact
MUF-2A	405.7	28.4	--[d]	--[c]	--[d]	80 to 90	Filter paper completely deteriorated
MUF-2B	206.2	14.4	--[d]	--[c]	--[d]	~50	Filter paper completely deteriorated
VAC-MUF-2A-1	340[e]	23.8	5.3	09	577.6	--[d]	Heavily loaded
VAC-MUF-2A-2	~6[e]	0.4	0.09	09	245.7	--[d]	Lightly loaded
VAC-MUF-2A-3	~90[e]	6.3	1.18	03	330.5	--[d]	Moderately loaded
VAC-MUF-2B	~0	0	0.006	RCBTA	236	--[d]	Very lightly loaded--side had a few dark spots
VAC-MUF-4A	~80[e]	5.6	0.08	04	327	--[d]	Moderately loaded
VAC-MUF-4B	~0	0	0.008	04	242	--[d]	Moderately loaded--one side was loaded with dark debris
VAC-MUF-5B	~0	0	0.009	04	269	--[d]	Very lightly loaded
VAC-MUF-5A	~0	0	0.02	14	242.5	--[d]	Very lightly loaded--one side was colored light gray with debris and a few large particles were on the surface

a. Damp weights.

b. Estimate based on the assumption that 7% of the debris was UO_2.

c. Metal filter frames were discarded as hot waste.

d. Not applicable.

e. Estimation based on the assumption that the cartridge filter weighed 240 g.

DEBRIS CHEMISTRY

The results of analyses to determine the elemental and radionuclide makeup of debris from Filter MUF-5B are presented in this section. Most of the data are from a sample of material that fell off the filter at TMI and sent to Babcock and Wilcox (B&W) where it was homogenized by milling with glass rods. Part of the sample was then sent to EG&G Idaho where it was further subdivided into two samples, 0104 and 0105. Results are reported from the original B&W sample, from ENICO on Samples 0104 and 0105, and from LANL on Sample 0104. Results are also presented on ENICO Sample 0111, which were obtained directly from Filter MUF-5B in the hot cells at EG&G Idaho, and from a nonhomogenized sample also obtained from this same filter. Analyses of additional samples taken directly from Filter MUF-5B and others are still on-going.

Elemental Makeup

Emission spectroscopy, electron microprobe, and scanning electron microscope/energy dispersive spectroscopy (SEM/EDS) measurements of the elemental makeup of the debris from Filter MUF-5B are reported in Tables 5, 6, and 7. Emission spectroscopy measures elemental composition in a bulk sample (a few milligrams), consisting of many particles, whereas the electron microprobe and SEM/EDS techniques analyze individual debris particles. As an aid to interpreting the measurements of debris composition, Table 8 presents the composition by weight and the melting and boiling temperatures of various materials in the core that were possible sources of the debris.

The emission spectroscopic data are presented as a percentage of the total weight of the sample. As shown in Table 5, the elements measured do not total 100% of the sample weight. The principal reason for this is that most of the elements in the debris are probably in the form of oxides, and oxygen is not measurable by this technique. The debris was found to be made up primarily of control materials, Ag (~12%), Cd (~11%) and In (~5%); fuel rod cladding materials, Zr (5 to 25%) and Sn (2 to 3%);

TABLE 5. EMISSION SPECTROSCOPIC ANALYSIS OF THE MUF-5B FILTER DEBRIS

	Elemental Composition of Debris wt%			
		ENICO		
Element	B&W[a]	Sample 0104[a]	Sample 0105[a]	Sample T 0111[b]
C	--[c]	17.5	18	--[c]
Ag	6 to 12	11.1	13	12.2
Al	0.6	0.55	0.43	~0.5
B	2	0.62	0.64	~0.1
Ca	0.2	--	--	--
Cd	11	11.4	11.2	--
Co	0.08	--	--	--
Cr	0.4	1.0	0.8	2.1
Cu	0.5	0.22	0.23	~0.5
Fe	7	5.7	5.2	3.9
Gd	<0.1	--	--	--
In	--	5.7	5.5	4.5
K	--	--	--	~4.5
Mg	<0.05	0.02	0.02	~0.01
Mn	0.1	0.10	0.10	~0.06
Mo	0.8	0.82	0.86	~0.08
Na	0.06	--	--	--
Nb	--	0.05	0.03	--
Ni	6	4.9	4.5	~0.9
Pb	<0.03	--	--	--
Si	1.8	<0.3	<0.3	~0.16
Sn	3	2.3	2.2	--
Ti	0.1	0.07	0.07	--
U	6	~5	~5	7.27
V	<0.05	--	--	--
Zn	<0.2	--	--	--
Zr	>25	5.4	5.7	12.6
Total	77.27	72.75	73.78	47.78

a. Homogenized sample that fell from the filter during a 1981 filter removal attempt.

b. Specimen taken from the MUF-5B filter. This specimen was homogenized in a plastic jar with Al_2O_3 milling pellets.

c. Not measured.

TABLE 6. ELECTRON MICROPROBE ANALYSIS BY LOS ALAMOS NATIONAL LABORATORY OF SAMPLE 0104 FROM FILTER MUF-5B[a]

Particle	Normalized Intensity Ratios (%)												Diameter (μm)
	Mg	Al	Si	Zr	Mo	Ag	Cd	In	Cr	Fe	Ni	U	
1	--	2.9	3.8	--	18.1	--	34.3	--	--	23.5	17.5	--	12.3
2	--	--	--	--	--	--	--	--	0.8	1.5	--	97.6	3.1
3	37.8	--	27.4	--	34.7	--	--	--	--	--	--	--	7.7
4	0.4	0.2	0.2	--	4.7	59.1	--	32.2	--	0.6	2.6	--	9.2
5	38.0	--	18.4	--	43.5	--	--	--	--	--	--	--	12.6[b]
6	--	1.7	2.0	30.6	1.2	--	26.3	--	1.4	20.7	16.1	--	14.8
7	--	1.2	0.5	4.8	9.4	--	31.8	--	1.0	23.1	28.2	--	16.7
8	--	2.6	86.3	--	--	--	-	--	1.0	2.6	7.4	--	5.1
9	--	--	0.3	--	--	--	--	97.6	--	1.2	0.9	--	4.3
10	0.1	0.2	0.2	0.5	5.4	52.8	--	--	1.6	11.6	27.6	--	18.0
11	--	0.3	0.6	2.0	2.8	--	44.2	--	0.7	7.9	41.4	--	16.0
12	--	0.4	0.3	17.6	--	--	76.1	--	--	1.6	4.0	--	6.6
13	--	2.2	2.6	--	44.1	--	43.1	--	--	4.6	3.1	--	6.2
14	--	--	7.0	--	18.8	--	--	43.2	6.1	14.1	--	--	19.2[b]
15	--	11.3	14.4	--	23.4	39.0	--	--	--	--	11.9	--	20.3[b]
16	--	--	99.9	--	--	--	--	--	--	--	--	--	1.7[c]
17	--	--	99.9	--	--	--	--	--	--	--	--	--	10.2[c]
18	--	0.4	0.2	47.0	--	43.1	2.2	--	--	5.4	1.7	--	5.6
19	--	7.0	6.4	--	22.4	--	28.8	--	--	17.7	17.7	--	4.2[b]
20	31.2	--	34.3	--	34.4	--	--	--	--	--	--	--	12.0[b]
21	--	0.5	0.4	--	11.2	19.4	--	18.3	6.2	11.4	32.5	--	3.1
22	--	--	0.1	--	0.7	20.8	78.4	--	--	--	--	--	6.7
23	41.5	12.4	--	--	46.1	--	--	--	--	--	--	--	10.7[b]
24	--	--	99.9	--	--	--	--	--	--	--	--	--	1.7[c]
25	--	0.3	0.2	0.9	2.5	13.9	67.8	--	1.0	4.1	9.4	--	24.8
26	--	--	--	0.4	0.7	15.6	81.7	--	--	0.4	1.1	--	6.4
27	--	0.1	--	--	1.7	--	2.7	--	0.6	84.0	10.9	--	6.7
28	--	1.6	0.6	0.8	19.7	--	65.5	--	2.3	8.1	1.4	--	9.4
29	--	0.2	--	60.4	--	16.8	--	6.5	0.7	4.1	9.4	--	7.0
30	--	--	--	--	--	--	--	--	--	0.2	--	99.8	4.9

TABLE 6. (continued)

Particle	Normalized Intensity Ratios (%)												Diameter (μm)
	Mg	Al	Si	Zr	Mo	Ag	Cd	In	Cr	Fe	Ni	U	
31	--	0.3	0.1	6.0	1.3	14.1	73.8	--	0.8	2.1	1.5	--	6.4
32	--	2.1	1.0	8.8	18.6	--	37.4	--	1.0	20.6	10.4	--	2.0
33	--	1.2	0.7	--	18.5	--	51.8	--	1.8	7.6	12.5	--	4.0
34	--	5.1	4.0	7.4	15.3	--	30.7	--	3.3	15.6	18.6	--	5.6
35	--	1.2	1.4	1.5	14.2	--	27.3	--	1.5	28.1	24.8	--	9.0
36	33.0	--	60.9	--	--	--	--	--	--	6.1	--	--	9.4
37	--	1.3	0.7	3.1	8.3	--	--	27.0	4.8	35.4	7.8	11.5	4.4
38	26.6	--	73.4	--	--	--	--	--	--	--	--	--	10.0[b]
39	35.2	--	59.8	--	--	--	--	--	--	4.9	--	--	10.0
40	--	--	--	--	--	--	--	--	--	--	--	99.9	4.0
41	--	0.4	0.3	14.6	0.7	--	54.2	--	2.1	6.6	5.9	15.1	8.5
42	32.2	--	17.0	--	50.9	--	--	--	--	--	--	--	10.0[b]
43	--	--	3.4	13.1	--	--	--	--	--	5.2	3.8	74.5	9.0[b]
44	--	0.7	1.9	--	7.9	--	46.4	--	0.7	10.7	31.8	--	12.9
45	--	--	--	--	2.8	--	90.7	--	--	1.6	4.9	--	7.1
46	54.7	--	45.3	--	--	--	--	--	--	--	--	--	15.0[b]
47	--	3.6	2.3	--	15.5	--	36.2	--	3.4	16.2	22.8	--	10.0

a. Particles <1 μm were ignored in this analysis.

b. Low yield, ion-exchange media.

c. Particle moved during chemical analysis. Particles 3, 5, 8, 23, and 42 contained low concentrations of Ca. Particles 8 and 13 contained low concentrations of Na.

TABLE 7. SEM/EDS RESULTS FROM EG&G IDAHO, PARTICLES REMOVED DIRECTLY FROM FILTER MUF-5B

Particle Elemental Composition	Frequency (Number of Particles)	Total Particles Examined %
Zirconium	2	7
Uranium[a]	4	14
Zirconium--Uranium[b]	2	7
Zirconium--Uranium--Stainless Steel/Inconel	2	7
Zirconium--Control Materials--Stainless Steel/Inconel	1	4
Zirconium--Uranium and/or Control Materials--Stainless Steel/Inconel[c]	12	43
Control Material[d]	2	7
Aluminum	3	11

a. Particle X-ray spectra generally included small zirconium peaks.

b. Particle X-ray spectra generally contained small peaks from elements associated with stainless steel or Inconel (chromium, iron and nickel).

c. Uranium L-series X-ray peaks were not observed. However, the counting statistics were not sufficient to rule out uranium.

d. X-ray peaks are too broad to be the result of X-ray activation of one element. These particles probably contain Cd, In, and Ag elements from control rods.

TABLE 8. SOURCE OF REACTOR DEBRIS

Material Source	Composition	Composition w%	Melting Temperature (K)	Boiling Point at 0.1 MPa (K)
Fuel cladding	UO_2	100	3136	--
	Zr	98	2200	3851
	Sn	<2	505	2533
	Cr	<0.25	2163	2755
	Fe	<0.25	1808	3273
Reacted cladding and Ceramic Spacers	ZrO_2	100	2973	~5273
Spacer grid			1533 to 1559	--
(Inconel 718)	Ni	52.5	1726	3005
	Cr	19.0	2163	2755
	Fe	18.5	1808	3273
	Ta	5.1	3269	5698
	Mo	3.5	2883	5833
	Al	0.5	936	2740
	Si	0.2	1693	2873
	Cu	0.15	1356	2868
Control rod			1060	--
neutron	Ag	80	1233	2485
adsorber	In	15	429	2273
	Cd	5	394	1038
Stainless steel			1698	--
(Grade CF-3M)	Fe	65 to 76	1808	3273
	Cr	18 to 20	2163	2755
	Ni	8 to 12	1726	3005
	Mn	trace	1244	2097
	Si	trace	1693	2873
Burnable poison			2303	--
shaping rods	Al_2O_3	98.6 to 99	2318	3253
	B_4C	1 to 1.4	2743	>3773
Test rods		--	2303	--
	UO_2	--	3136	--
	Gd_2O_3	--	--	--
Instrumentation				
Sheaths	Inconel	--	--	--
Insulation	MgO	--	3073	3873

fuel, U (5 to 7%); and stainless steel and Inconel, Fe (~6%), Ni (~5%), Cr (~1%), Mo (~0.8%) and Si (~0.3%). Also, large amounts of carbon (18%), probably from the filter paper and possibly the demineralizer resin beads, were measured in two of the four samples analyzed by emission spectroscopy.

Forty-seven particles >1 μm in diameter from the homogenized Sample 0104 were analyzed by LANL using the election microprobe. These results, having an estimated accuracy of ±20%, are presented in Table 6. Twenty-eight particles removed directly from Filter MUF-5B and not homogenized were also examined by SEM/EDS at EG&G Idaho. These results are presented in Table 7. The single particle analyses show that occasionally the debris particles may contain essentially one metallic element, such as zirconium or uranium, but generally the particles have complex compositions containing a number of elements from the TMI-2 fuel, cladding, stainless steel and Inconel, and control materials. Seven of the 75 particles examined by LANL and EG&G Idaho essentially contain uranium as the only metallic element; three are only silicon; another three basically aluminum bearing; two contain essentially zirconium; one contains mostly indium; one contains more than 90% cadmium; and five contain only a combination of control materials (Ag, In, or Cd). The rest of the particles (71%) contained mixtures of zirconium and tin from the fuel rod cladding and control rod guide tubes; uranium from the fuel; iron, nickel, and chromium from the stainless steel or Inconel components; and/or silver, indium, and/or cadmium from the control materials. Fifty (67%) of the particles contain one or more of the principal components of stainless steel or Inconel (Fe, Ni, Cr). Forty-five (60%) contain one or more control materials (Ag, In, Cd). Thirty-six (48%) contain zirconium. Twenty-six (35%) contain uranium. Fourteen (19%) of the particles contain elements of all four major components: fuel, fuel rod cladding and control rod guide tube, stainless steel/Inconel, and control materials. Eighteen (24%) contain fuel rod cladding, stainless steel/Inconel, and control materials. And 41 (55%) contain stainless steel/Inconel and control materials.

More particles (67%) contain one or more of the major elements of stainless steel or Inconel (Fe, Ni, Cr) than any other group of metallic elements. The most likely source of stainless steel is the control rod

cladding, though the Quick Look Examination of the TMI-2 reactor, has shown that the fuel assembly end boxes were also heavily damaged. The principal source of Inconel is the spacer grids. Elemental ratios for these materials, based on the information in Table 8, are presented in Table 9, along with elemental ratios determined from the measurements reported in Tables 6 and 7. Based on the measured Ni/Fe and Mo/Fe ratios, it would appear that the debris likely contains material from both stainless steel and Inconel components, though a somewhat higher Cr/Fe ratio would be expected (from the Inconel source) than was measured. The very high Mo/Fe ratio measured on individual particles at LANL by electron microprobe analysis is not understood, nor is the high Cu/Fe ratio measured in the bulk emission spectroscopic analysis. The foregoing evaluation suggests that both stainless steel control rod cladding and Inconel spacer grids are substantial contributors to the debris collected on the purification/makeup system filters.

TABLE 9. STRUCTURAL MATERIAL MASS RATIOS WITH RESPECT TO IRON COMPARED TO SIMILAR RATIOS OF TMI-2 FILTER DEBRIS

Material	Ni/Fe	Cr/Fe	Cu/Fe	Mo/Fe	Si/Fe	Al/Fe	Fe/ Total Weight
Stainless steel	0.14	0.27	0	0	0	0	0.65 to 0.76
Inconel	2.84	1.03	0.01	0.19	0.01	0.03	0.19
Bulk filter debris	0.8	0.2	0.06	0.1	0.1	0.09	0.06
LANL microanalysis	1.7	0.2	--	2.7	1.1	0.08	--

It is also apparent from the bulk elemental data and the individual particle data that Ag, In, and Cd were released from the control rods and were major contributors to the debris transported to the purification/makeup filters. The substantial content of zirconium and uranium bearing particles in the debris, in conjunction with control materials and the stainless steel and Inconel components, is consistent with the picture of severe core damage revealed by the Quick Look Examinations.

More than 70% of the particles containing cadmium examined at the LANL with the electron microprobe had essentially no silver or indium in them, but instead had steel, Inconel, and/or zirconium components. In fact, cadmium and indium do not occur together in any of the particles analyzed by LANL. The cadmium probably vaporized to a greater extent than did the silver or indium, because of its larger vapor pressure, and separated from those control rod components. Vaporization of cadmium from failed control rods has been observed experimentally at ORNL[1] in simulated core-melt tests using light water reactor (LWR) type fuel and control rods. In these atmospheric pressure tests, the control rods burst at about 1678 K because of high cadmium vapor pressure. These tests indicate that cadmium volatized, left the control rods, and formed CdO when oxygen was available. In the absence of oxygen, vaporized cadmium formed small spherical particles that agglomerated, agglomerates being $\leq$8.6 μm in diameter. The spherical particles and agglomerate structures were very similar in morphology to those found in the TMI filter debris.

The LANL results indicate that iron is associated with cadmium in 60% of the particles containing cadmium. This would not be expected based on the thermodynamics of the cadmium iron phase system. Cadmium is virtually insoluble in solid iron.[2] In fact, the solubility of cadmium in iron is only 2 K 10^{-4} to 3 x 10^{-4} w% at 973 K.[3] However, cadmium vapor may have condensed on iron-bearing particles to produce the particle chemistry measured.

Aluminum was present in 21 of the 47 particles analyzed by LANL, and aluminum was the principle constituent in 3 of the 28 particles analyzed by EG&G Idaho. However, the intensity of aluminum in the electron mircroprobe analyses is <5%, except on ion-exchange media particles. Pieces of the burnable poison (pelletized Al_2O_3 - B_4C) in the sample should produce particles with nearly 100% intensity for aluminum. Three such particles were found by the SEM/EDS analysis. The Al/Fe ratio measured higher than is available from Inconel (see Table 9), which may be due to reactions between burnable poison pellets, stainless steel, and the Inconel components (Fe, Ni, Cr) frequently found with Al in the debris.

The silicon measured in the debris is higher than would be expected from the silicon content of Inconel (see Table 9). Sources of silicon could be the glass rods used to mill the debris sample at B&W (three particles in the LANL sample showed only Si), or possibly, the filter paper itself.

Resin beads that may have entered the purification lines from the demineralizer tank were looked for in the filter debris. The resin beads were 380 μm diameter ion-exchange resins that cleaned the primary coolant of tramp radioactive elements. No intact resin beads were observed by SEM in the MUF-5B or -2B filter examination. Detection of the chemical elements making up the resin beads by SEM/EDS or microprobe analysis is difficult, however, since the resin bead composition is H, C, O, Li, N, S, and the characteristic X-rays have too low an energy for efficient detection. Several particles analyzed by electron microprobe and reported in Table 6 were found to give unusually low yields of X-rays for the size of the particles and are suspected to contain ion-exchange resin material.

Three particles were examined on the molecular optical laser examiner (MOLE) for chemical form. Two of the three particles examined contained ZrO_2, and one of the two was almost entirely ZrO_2. However, the crystallographic state of this particle was that of high-temperature cubic ZrO_2, which was stabilized by some means, perhaps rapid cooling and/or trace element additions. The Raman spectra indicated that its structure changed to the thermodynamically stable monoclinic structure because of laser heating during examination. The other zirconium particle contained silver and possibly indium. Although, the particles were mounted in indium foil so the X-rays may or may not have been from indium in the particle. The Raman spectrum indicated a zirconium oxide monoclinic crystalline structure. The third particle examined produced a Raman spectrum that did not closely fit available standard spectra, though the spectrum most closely resembled the standard monoclinic ZrO_2 spectrum. The MOLE technique provided unique information on chemical and crystallographic structures and could be beneficially applied to other particles if funding were available.

Radionuclide Makeup

The major isotopes found by spectral gamma scanning the filters were listed in Table 3. The predominant gamma-emitting nuclides are the fission products ^{137}Cs, ^{134}Cs, ^{125}Sb, $^{106}Ru/Rh$, and $^{144}Ce/Pr$, and the activation products ^{60}Co, ^{100m}Ag, and ^{54}Mn. The measurements on the specific activity of the fission product radionuclides are related to the fission product inventory per unit weight of uranium in this section. Results of isotopic analyses on the fissile nuclides found in the debris are also reported. All the data reported in this section are from Filter MUF-5B.

Gamma spectral analyses of debris from Samples 0104 (LANL) and 0105 (EG&G Idaho) are presented in Tables 10 and 11, respectively. The fission product isotopic activity per gram of sample is converted to activity per gram of uranium, assuming the debris contains 5% uranium by weight as indicated by the emission spectroscopic results in Table 5. Also in Tables 10 and 11, the measured activities of fission products are compared with activities in the fuel calculated for the TMI-2 power history with the ORIGEN-2 code. The measured and calculated ^{144}Ce activities are in good agreement. This result is both expected and gratifying: expected because ^{144}Ce is nonvolatile and should remain with the fuel, and gratifying because the result suggests that the comparison of the measured and calculated activities is valid. It is noteworthy that the measured activity of the cesium isotopes is about eight times that expected from the inventory in the fuel. The ratio is about 9 for ^{90}Sr, 20 to 70 for ^{125}Sb, and 2 for ^{106}Ru. The presence of these fission products in the debris in amounts greatly exceeding the inventory in the fuel strongly suggests that these isotopes were released from the fuel during the high temperature portion of the accident and deposited on debris particles (probably aerosols), which were subsequently transported to the purification/makeup filters. The presence of these isotopes in the filter debris suggests that they were well-fixed on, or within, the debris and in a form insoluble in water.

TABLE 10. GAMMA SPECTROSCOPY ANALYSIS BY LOS ALAMOS NATIONAL LABORATORY OF SAMPLE TMI 0104 FROM FILTER MUF-5B

Isotope	Sample Number[a]	Specific Activity (μCi/g)	Average Specific Activity (μCi/g)[b]	Average Experimental Activity (μCi/gU)	Calculated Activity (μCi/gU)	Ratio of Measured to Activity (μCi/gU)
^{144}Ce	2	527.6				
	6	573.6	550.6 ± 13.0	11,000	11,830	0.93
	9	550.5				
^{137}Cs	2	3540				
	6	3730	3606 ± 62	73,500	9,560	7.7
	9	3748				
^{134}Cs	2	259.5				
	6	266.7	266.7 ± 4.2	5,300	640	8.3
	9	273.9				
^{125}Sb	2	612.5				
	6	652.8	637.2 ± 12.5	12,800	620	21
	9	646.4				
^{106}Ru	2	289.2				
	6	307.5	313.6 ± 16.2	6,300	3,460	1.8
	9	344.1				
^{110m}Ag	2	6.77				
	6	8.10	7.4 ± 0.5	--	--	--
	9	--				
^{60}Co	2	222.7				
	6	240.6	228.7 ± 6.0	--	--	--
	9	222.7				
^{54}Mn	2	8.0				
	6	9.0	9.0 ± 0.6	--	--	--
	9	10.0				

a. The sample number refers to the cut of the sample taken for analysis, Sample 2 was 0.0175 g, Sample 6 was 0.0158 g, and Sample 9 was 0.0148 g.

b. The specific activities are for October 29, 1982.

c. Silver chloride crystals formed in the samples and resulted in low values for ^{110m}Ag.

TABLE 11. FISSION PRODUCT ANALYSIS BY EG&G IDAHO OF SAMPLE 0105 FROM FILTER MUF-5B[a]

Nuclide	Activity[a] (μCi/g)	Activity (μCi/gU)	Calculated Activity (μCi/gU)	Ratio of Measured to Calculated Activities
^{144}Ce	920[a] ± 30	18,400	24,360	0.76
^{137}Cs	3810[a] ± 30	76,200	9,740	7.8
^{134}Cs	416[a] ± 5	8,300	840	9.9
^{125}Sb	2650[a] ± 20	53,000	760	70
^{106}Ru	620[a] ± 30	12,400	6,040	2.1
^{90}Sr	4000[b]	80,000	8,500	9.4

a. The activity was measured January 6, 1982, and the sample weight was 0.0239 g. The uranium concentration was assumed to be 5% of the total weight.

b. The activity was measured February 2, 1982. The uranium concentration was assumed to be 5% of the total weight.

c. The calculated activity values are based on ORIGEN-2 code calculations.

The results of analyses of the fissile content of the debris from Filter MUF-5B are presented in Table 12. The ^{235}U values measured in the homogenized samples taken from material that had fallen off the filter at TMI (B&W Samples 0104 and 0105) are mainly in the range of the fuel enrichments found in the major portion of the core (1.98 and 2.64%), rather than the peripheral assemblies (2.96% enrichment). The enrichment (2.81%) measured in Sample 0111 (debris removed from the filter in the hot cell) is, however, closer to that of the peripheral assemblies. This result tends to suggest that fuel rods in the peripheral assemblies may have ballooned and ruptured. This is a tentative conclusion, at best, based on only one measurement of a small sample.

TABLE 12. FISSILE ISOTOPIC ANALYSES FROM FILTER MUF-5B

	Concentration w%				
		ENICO			
Isotope	B&W	Sample 0104	Sample 0105	Sample 0111	Core Average
^{234}U	0.016	0.02	0.02	0.16	--
^{235}U	2.299	2.56	2.43	2.81	2.16
^{236}U	0.063	0.09	0.08	0.16	0.08
^{238}U	97.622	97.33	97.51	96.88	97.76
^{238}Pu	0.027	--	--	--	--
^{239}Pu	90.88	--	--	--	89.81
^{240}Pu	7.504	--	--	--	8.30
^{241}Pu	1.499	--	--	--	1.82
^{242}Pu	0.082	--	--	--	--
^{241}Am	2.9×10^{-3}	--	--	--	--
^{244}Cm	2.1×10^{-5}	--	--	--	--
^{242}Cm	2.9×10^{-7}	--	--	--	--

DEBRIS MORPHOLOGY

Distribution of particle shape was obtained from a debris sample from Filter MUF-5B. Distribution of particle size was determined on material removed from Filters MUF-5B and -2B at EG&G Idaho and the homogenized Sample 0104 from Filter MUF-5B. The data from the homogenized sample is not relevant and is presented in Appendix D.

Particle Shape Distribution

The particles collected from the debris of Filter MUF-5B varied in shape from nearly spherical particles to very irregular particles and large agglomerated groups of particles. Agglomerates and individual particles on a MUF-5B sample examined at ANL-E are shown in Figures 14a and b. Debris particles examined at the EG&G Idaho hot cells are shown in Figures 15a,b,c. These particles tend to be chemically very complex, as discussed in the previous section and indicated in Figures 15a,b,c.

A polished cross section of a piece of filter paper is shown in Figure 16. The small spherical particles were generally sandwiched between the filter paper with the irregular shaped particles and, therefore, the irregular shaped particles were last to be deposited. The irregular particles were also found in aggregates with spherical particles intermixed.

The sphericity of the particles from the MUF-5B debris was determined from SEM photomicrographs. Sphericity is the ratio of a projected diameter to the diameter[a] of a circle that just circumscribes the particle, and the overall sphericity of the particles is a gross indicator of the particle shape. A particle sphericity histogram of the debris examined on the SEM is illustrated in Figure 17. The mean sphericity of the particles was about 0.61, indicating quite irregular particles.

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a. The projected particle diameter is the diameter of a circle whose area is equal to the area defined by the profile of the particle.

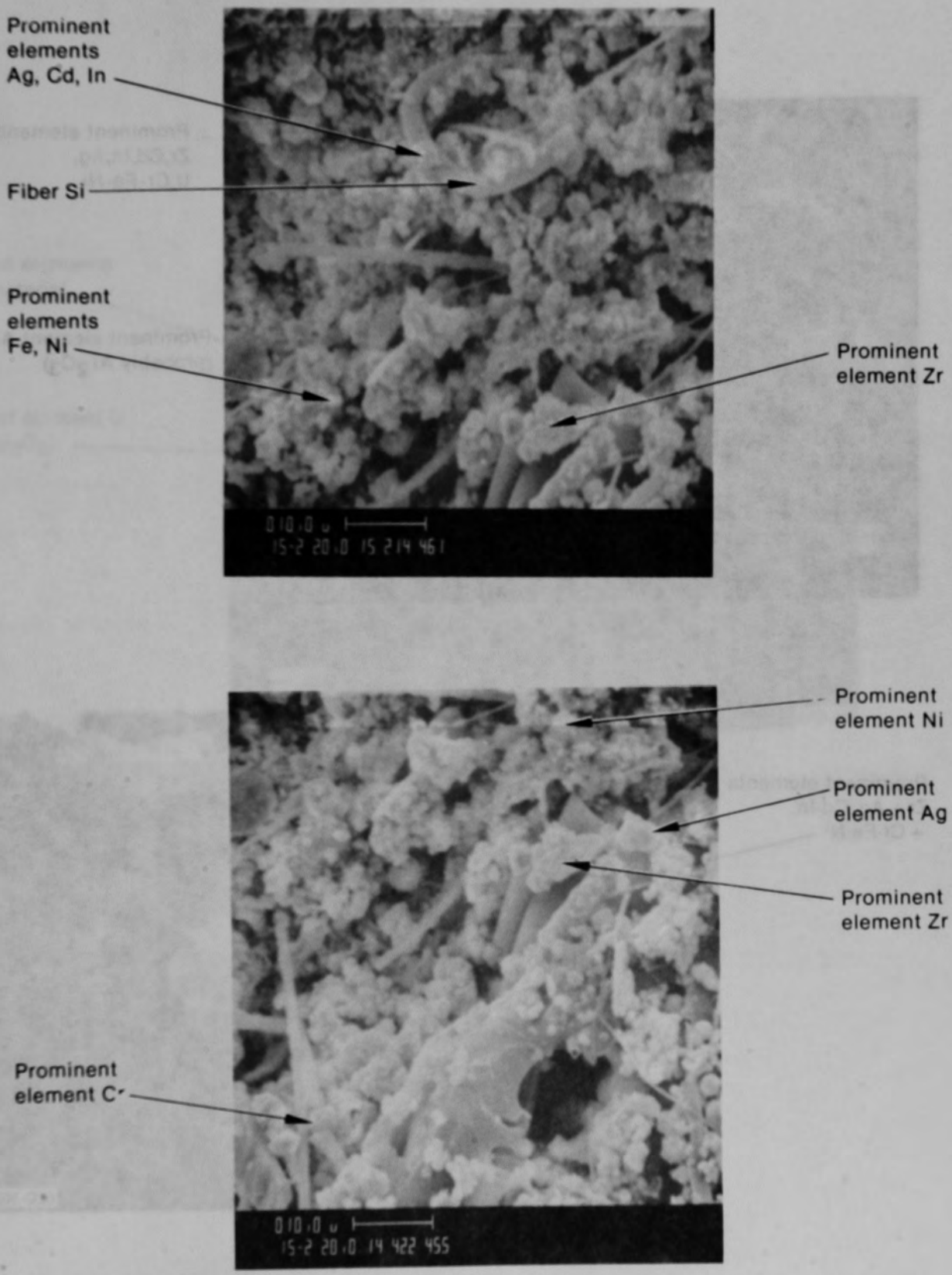

Figure 14a,b. Typical filter MUF-58 debris.

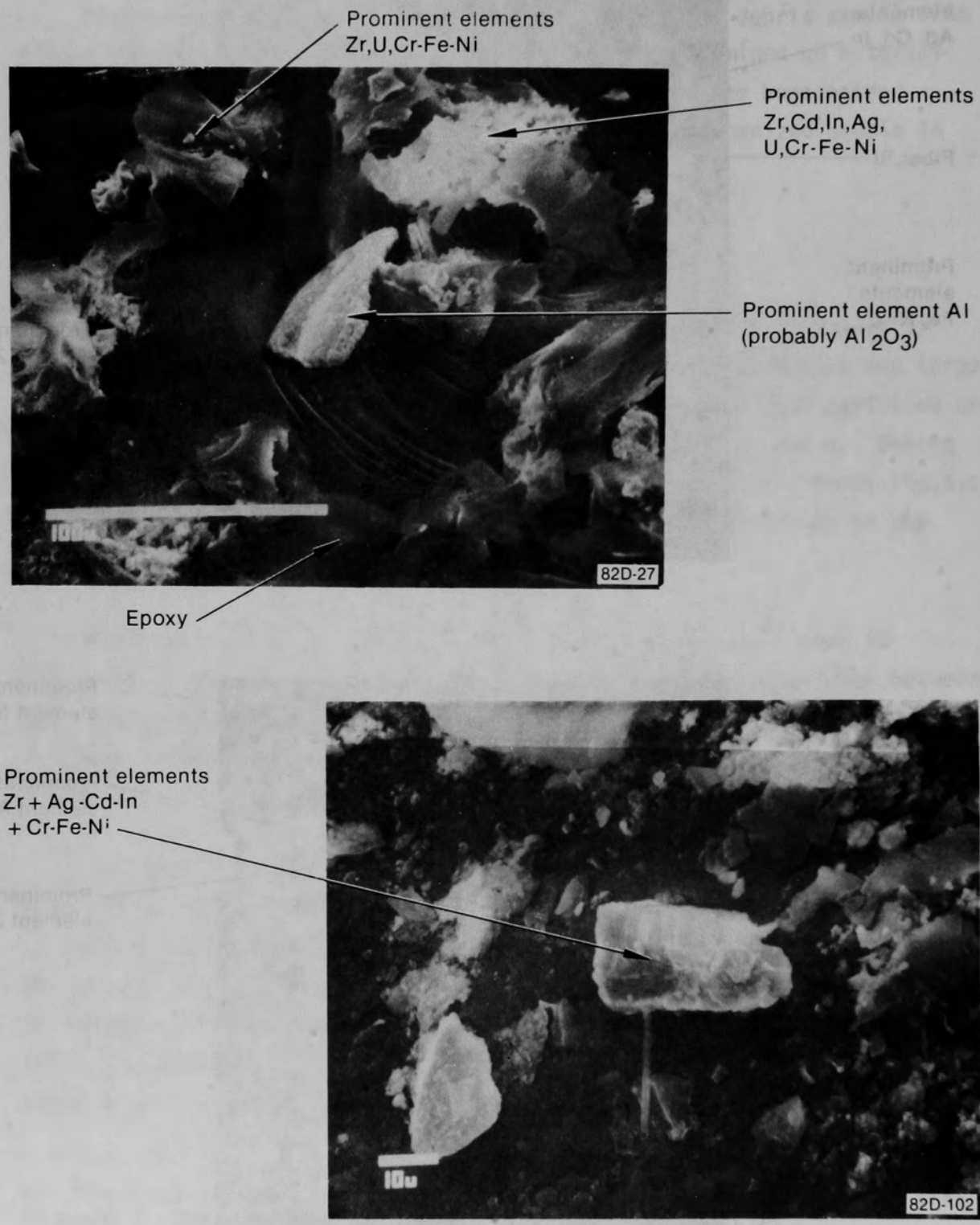

Figure 15a,b. Particles from Filter MUF-5B.

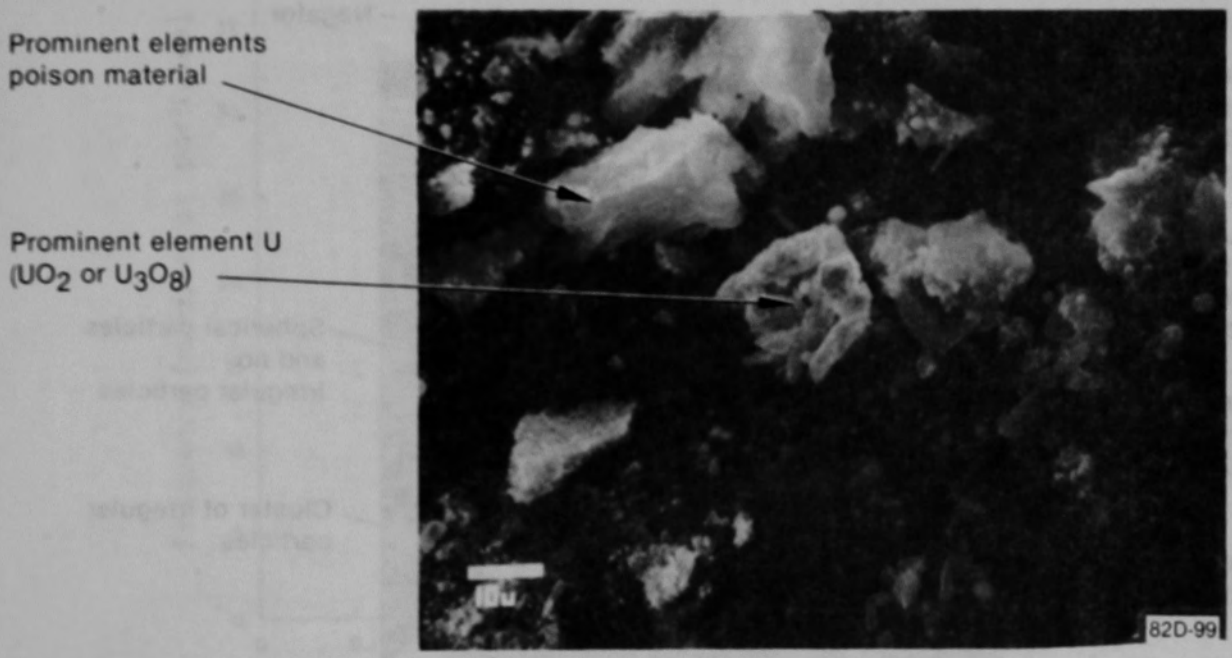

Figure 15c. Particles from Filter MUF-5B.

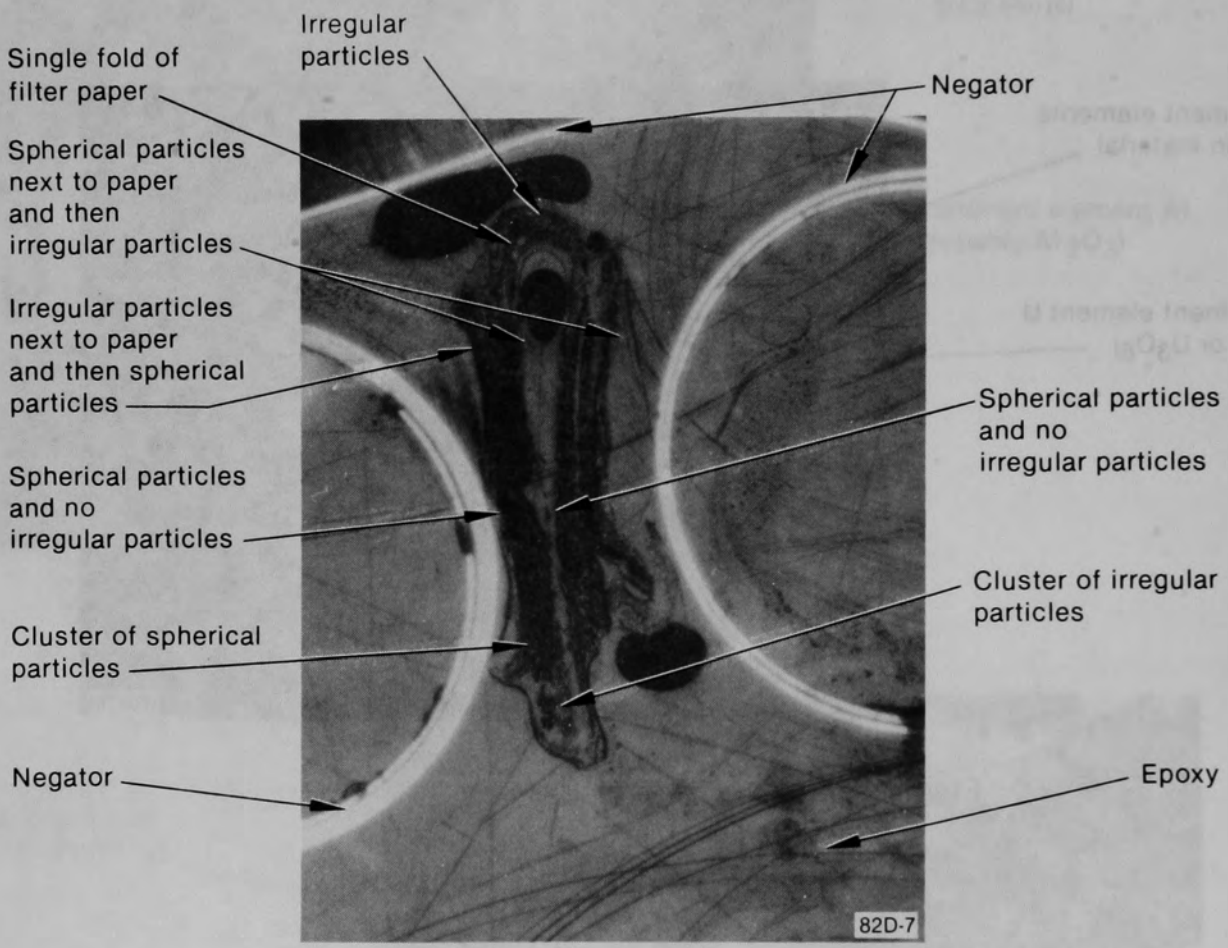

Figure 16. Cross section of a single fold of Filter MUF-5B paper.

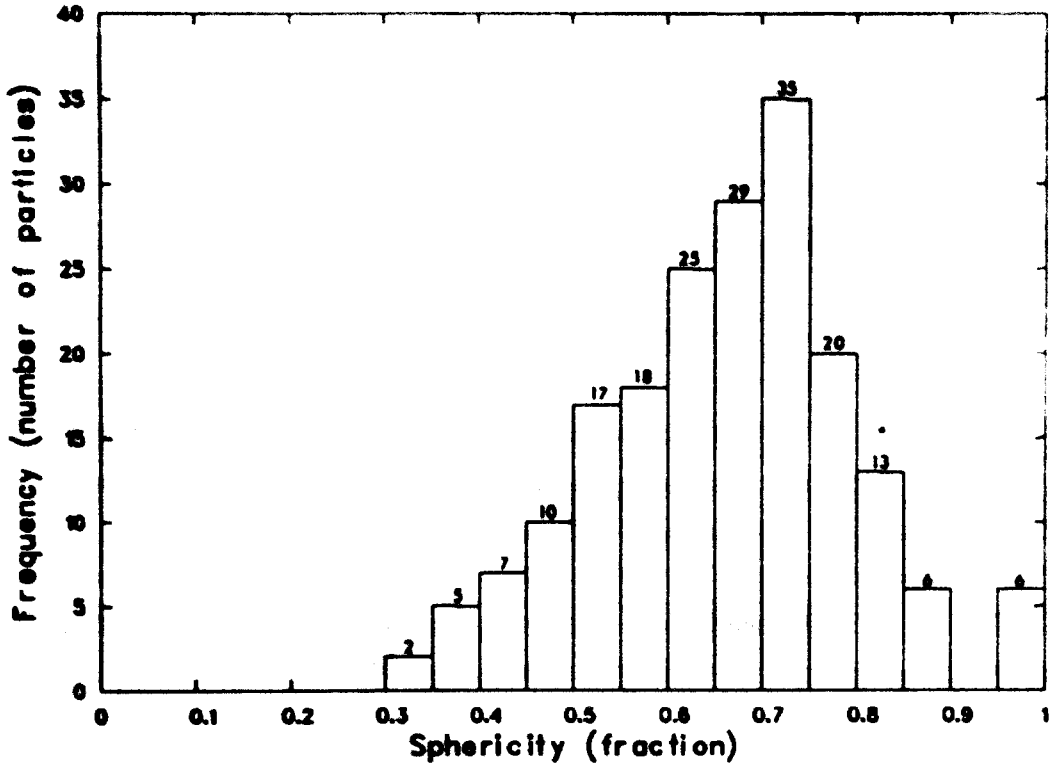

Figure 17. Sphericity distribution of particles from Filter MUF-5B.

The spherical or nearly spherical particles were probably control rod material (silver and/or cadmium). That is, the EG&G Idaho analyses indicated that the particles had X-ray peaks at energies characteristic of silver and cadmium. The spherical particles also had a mean diameter <2 μm instead of the ∿6 μm estimated for all particles. Thus, there may be a correlation between size, shape, and composition. A shape-size-composition relation was also suggested in the ANL-E analyses. The ANL-E analyses also indicated that the numerous spherical particles seem to be control rod materials, and in some cases the spherical particles appeared to be pure silver or cadmium. Zirconium particles were generally found as either large rectangular pieces or small flakes, and steel bearing particles were generally found as small rectangular particles.

Particle-Size Distribution

Particle-size distributions were determined from samples from Filters MUF-2B and -5B, and from the homogenized Sample 0104 from Filter MUF-5B. The particle diameters on Filters MUF-5B and -2B ranged from ∿50 μm[a] down to submicrometer size. A histogram of particle sizes on MUF-5B is shown in Figure 18. A cumulative distribution of particle sizes from Filters MUF-5B and -2A are shown in Figure 19. The apparent mean particle diameters are ∿6 μm. However, these particle distributions may be skewed to a higher mean particle diameter than actual because particles <1μ m were not accurately counted, and the SEM examinations of the debris indicated a large number of particles <1 μm. This is illustrated by the profile of projected diameters shown in Figure 19. The projected diameter measurements were obtained manually, and the <3-μm particles that could be resolved were classified in a single group; thus, the large number of <1-μm particles force the cumulative projected diameter plot in Figure 19 to begin at ∿90%.

Particles in the micron size range were expected on the filters because the coolant containing the reactor debris had to travel through the

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a. Measurements using the projected diameter technique indicate a maximum diameter of 50 μm; measurements using a Feret's diameter technique indicate that the maximum diameter is about 70 μm.

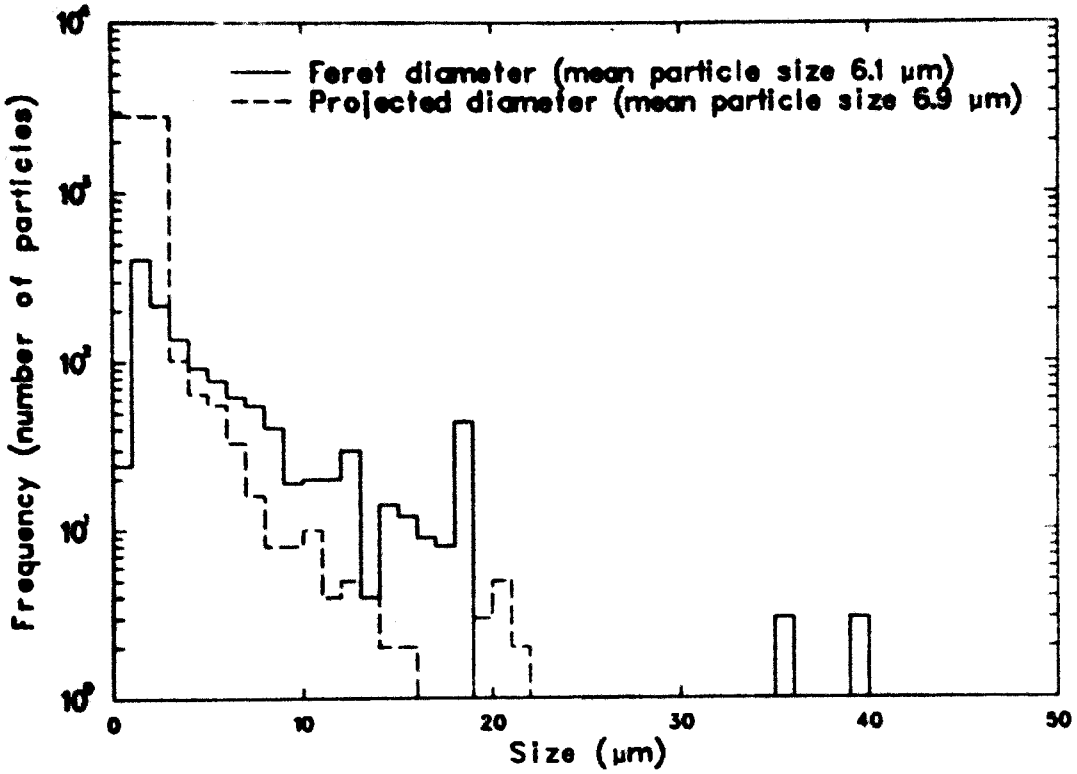

Figure 18. Size distribution of particles from Filter MUF-58.

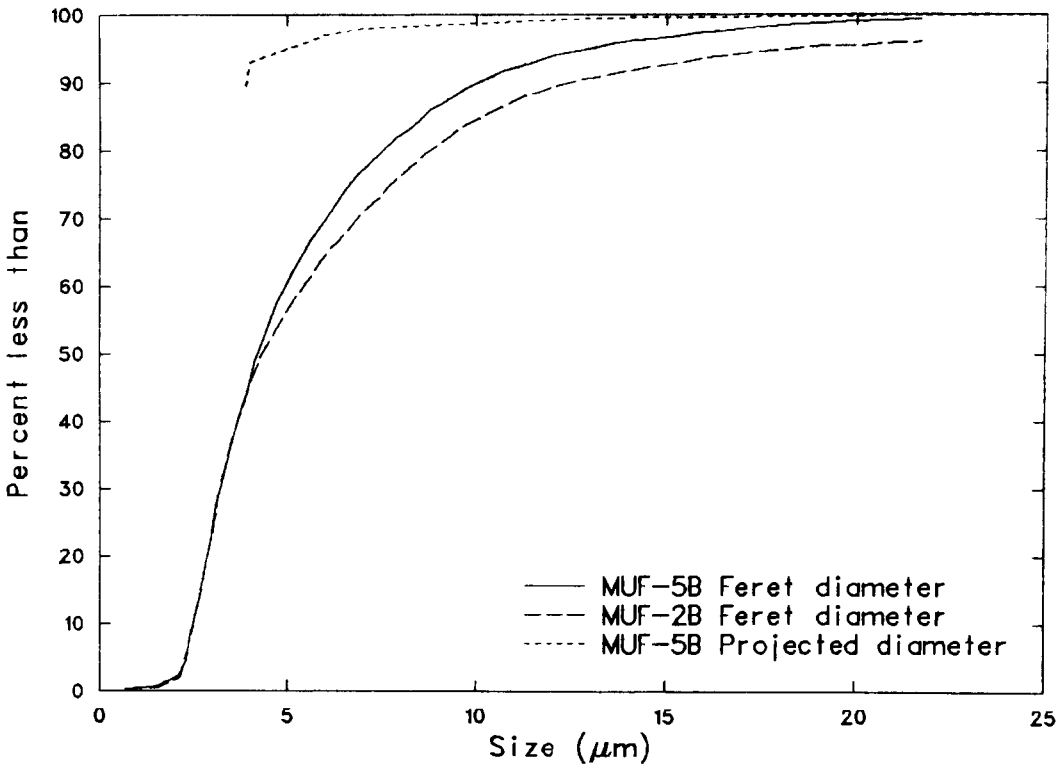

Figure 19. Cumulative particle size distribution from Filters MUF-5B and 2A

steam generator, the letdown coolers, and block orifice before it reached the filters. These structural components may have limited the maximum particle size that would reach the filters and also provide surfaces for deposition of small particles.

In summary, the majority of particles deposited on the filters were <10 μm (>80%), with a mean size of ~6 μm, and were irregular in shape. However, additional work will be needed to determine a size and shape distribution of the micron and smaller particles since these have higher biological implications should any debris escape to the atmosphere. Additionally, the MUF-2B, and -4B filter debris should be examined to determine the particle size and shape distributions on those filters.

INTERIM CONCLUSIONS

The objectives of the TMI-2 purification/makeup filter examination were to determine the character of the debris in the primary coolant system and collect and analyze data that will assist in the reconstruction of the accident events. Preliminary conclusions that can be drawn from the data and analyses areas follow:

1. The filter paper did not withstand the posttest environments they were exposed to. That is, they became severely deteriorated after ~2.5 years in water, covered with radioactive debris.

2. Less than 100 g of UO_2 was received with the purification and vacuum filters. Approximately 11 g of UO_2 were received with the MUF-5B filter, consistent with the GPU estimate.

3. Non-fuel-rod components generated the largest amount of debris. Stainless steel/Inconel alloying elements were found in 67% of the particles examined, and control materials were found in 60% of the particles.

4. Electron microprobe and SEM/EDS data show that some debris particles have simple compositions, being dominated by one metallic element such as uranium or zirconium, but most contain many elements.

5. The activities of ^{137}Cs, ^{134}Cs, ^{125}Sb, $^{106}Ru/12h$, and ^{90}Sr in the debris are greater than can be accounted for by the fuel content of the debris. It is postulated that these fission products were released from the fuel, deposited on particles (aerosols) generated from control rod cladding, spacer grids, and control materials, and transported to the filters with these particles.

6. The mean particle size is <6 μm, and the particle size range is from <1 to 50 μm.

7. The shape of the debris particles is extremely varied, ranging from nearly spherical to very irregular particles. The mean sphericity of the particles is 0.61.

8. The cadmium and/or silver from the control rods apparently formed most of the spherical particles. The irregular particles generally contained larger amounts of stainless steel/Inconel alloying elements, or zirconium.

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APPENDIX A

PROPOSED EXAMINATION OF FILTER DEBRIS FROM THE TMI-2 PURIFICATION/MAKEUP SYSTEM

APPENDIX A

PROPOSED EXAMINATION OF FILTER DEBRIS FROM THE TMI-2 PURIFICATION/MAKEUP SYSTEM

The filter examination as proposed in July 1982 is provided in this appendix.

Purification/Makeup Filter Examination

A limited examination will be performed on the purification/makeup Filters MUF-2A, -2B, -4A, and -4B. An extensive examination will be performed of the debris from Filter MUF-5B. The following tasks will be performed during the examination.

1. Each filter will be removed from the outer polyethylene bag. The outer bag contains a water adsorbing material, vermiculite. The bag and adsorbent material will be disposed. The filters were packaged in double polyethylene bags.

2. Sample handling will be documented and appropriate photodocumentation will be taken.

3. The weights of the filter and inner polyethylene bags will be determined.

4. Appropriate filter debris samples will be collected as archive specimens.

5. Uranium and plutonium isotope ratios and concentrations will be determined.

6. Radionuclide and elemental concentrations will be determined.

7. The carbon content will be determined.

8. The resin bead concentration and morphology in the debris will be determined (except Filters MUF-4A and -4B).

9. The filter and inner polyethylene bag will be stored in a suitable container.

In addition, the following tasks will be performed on the debris from Filter MUF-5B:

10. Weight fractions of nonfuel debris will be determined.

11. The particulate character of fuel and nonfuel debris will be determined. This characterization will consist of the following:

    a. Particle size distributions

    b. Particle shapes

    c. Particle surface area estimates

    d. Elemental compositions of particles

    e. Phases in the particles.

12. Elemental and/or particle size stratification in the material caked on the filter will be determined.

13. The oxidation state of uranium and the oxygen to metal ratio of the fuel will be determined.

14. Compounds and phases of the debris particles will be identified.

15. Retained fission gas release rates under controlled temperature-time heating cycles will be determined.

The information described above will be collected from the various participants and reported in a single comprehensive report. The final report will be both a compilation and interpretation of the data.

Vacuum Filter Examination

The vacuum filters are loaded with debris vacuumed from the purification/makeup filter housings. Only a limited examination of the vacuum filters will be performed, with the exception of vacuum Filter VAC-MUF-5A. Filter MUF-5A was the first filter to collect debris from the reactor core and became overloaded. It was, however, discarded at TMI. The VAC-MUF-5A was used to clean the filter housing of MUF-5A so it probably collected some of the debris that first reached the purification/ makeup filters. Filter VAC-MUF-5A will therefore be more thoroughly examined than the other vacuum filters.

The examination of the vacuum filters will consist of the following tasks:

1. Each filter canister will be removed from the storage drum and polyethylene containment bags.

2. Filter handling will be documented and appropriate photodocumentation will be taken.

3. Each filter canister will be weighed.

4. Each filter canister will be stored in a suitable storage container.

In addition, the following task will be performed on the VAC-MUF-5A vacuum filter:

5. Radionuclide concentrations and the particle size distribution will be determined.

The specific techniques to be used are provided in Tables A-1 through A-5.

TABLE A-1. ANALYSES TO BE PERFORMED BY EG&G PHYSICS

Analysis Technique	Isotope, Element, Molecule, Physical Parameter
Gamma spectrometry	^{134}Cs, ^{137}Cs, etc.
Beta spectrometry	^{90}Sr
Neutron activation on analysis and/or delayed fission neutrons	Total fissile (^{235}U equivalent)
Neutron activation analysis	^{129}I
MOLE	Molecular species
Neutron activation analysis	Elemental cations: Li, B, Na, Mg, Al, Si, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, Ag, Cd, In, Sn, Gd

TABLE A-2. ANALYSES TO BE PERFORMED BY ENICO

Analysis Technique	Isotope, Element, Molecule, Physical Parameter
Direct current emission spectroscopy (semiquantitative) or X-ray diffraction	Elemental cations: Li, B, Na, Mg, Al, Si, K, Ca, Cr, Mn, Fe, Co, Ni, Cu, Zn, Zr, Ag, Cd, In, Sn, Gd
Ion chromatography	Anions: Fluorides, Chlorides, Bromides, SO_4^{-2}, PO_4^{-3}, NO_3^-, NO_2^-
Combustion furnace	Total carbon, organic carbon
Beta spectrometry	Gross beta, ^{3}H, ^{14}C, ^{55}Fe, ^{63}Ni, ^{90}Sr
Alpha spectrometry	^{241}Am[a], 242,243, ^{244}Cm[a], Gross alpha
Isotope dilution mass spectroscopy	U isotopes[a]
Alpha spectrometry and isotope dilution mass spectroscopy	Pu isotopes[a]

a. Duplicate U, Pu, Am, and Cm analyses will be performed.

TABLE A-3. MATERIALS EXAMINATIONS TO BE PERFORMED BY EG&G LIGHT WATER REACTOR--FUEL AND FISSION PRODUCT RESEARCH BRANCH

Analyses Technique	Physical Parameter Determined
Photovisual	• General description of filters and filter debris • General character of the samples from which to base in-depth the examination
Gross weights	• Documents initial sample quantity • Provides bases for further examination
Microsieves	• Ranks fuel sizes between 1 and 37 μm
Metallography	• Particle shapes • Formation process[a] • Particle deposition sequence • Deposition sequence
SEM/EDS	• Particle shapes • Formation process[a] • Phase composition of particles • Elemental distribution in particles • Particle size distribution in the <37 μm range • Particle deposition sequence
Auger/ESCA	• Molecular structure • Depth profile of elements and compounds • Elemental analysis--especially low atomic number elements--oxygen in particular
X-ray powder diffraction[b]	• Crystallographic form • Molecular structure • Stoichiometric composition

a. Molten or liquified fuel will be indicated by spherical particles, dendrites, large nonsintered grains, and no inert gas bubbles in the particle matrix. Frothy-bubbly appearance in iron particles will also indicate melting.

b. This work will be done by ENICO and will be followed by LWR-FRD personnel.

TABLE A-4. EXAMINATIONS TO BE PERFORMED BY LOS ALAMOS NATIONAL LABORATORY

Examination Method	Information to be Obtained
Photovisual	• General description of sample as received • Documentation of sample handling throughout examination process
Metallographic examination	• Optical record of sample • Gross particle size distribution • Particle morphology • Phase differences
Electron microprobe	• Detailed particle size distribution • Elemental distribution in sample (elements with atomic numbers >10) • Particle morphology • Particle deposition sequence • Tentative phase identification
Ion microprobe	• Elemental (and isotopic) distribution in sample • Particle deposition sequence • Tentative phase identification
Auger/ESCA	• Elemental distribution in sample • Elemental oxidation states
X-ray diffraction	• Phase and compound identification • Microcrystalline size distribution
Infrared spectrometry[a]	• Chemical compound identification

a. Method to be tried to determine applicability.

TABLE A-5. CHEMICAL CHARACTERIZATIONS TO BE PERFORMED BY LOS ALAMOS NATIONAL LABORATORY

Analysis Technique	Information to be Obtained
Emission spectroscopy	• Elemental composition (semiquantitative for 46 metallic elements and the rare earths)
X-ray fluorescence	• Semiquantitative for major and minor elements with atomic numbers >10
Ion chromatography	• Quantitative for common anions

TABLE A-5. (continued)

Analysis Technique	Information to be Obtained
Mass spectrometry	• U and Pu isotopic distributions • U and Pu assay using isotope dilution • Burnup to a low limit of 0.001% using isotope-dilution determination of Nd, U, and Pu • Assay and isotopic distribution of Kr an Xe in plenum and retained in fuel
Radiochemistry (α, β, and γ spectrometry)	• Determination of radionuclides (fission and activation products)
Interstitial gas analysis (gas fusion)	• Total C, and H_2 and O_2 especially on zircaloy clad
Dynamic gas release measurements	• Determine release rate and composition of retained gases, including Kr and Xe isotopes, under controlled temperature-time heating cycles
Wet chemical analysis	• Determine specific elements of interest including components (Ag, B, Cd, Gd, In, U, and Zr) of fuel, burnable poison, and control rods.
Thermogravimetry	• Determine oxygen-to-metal ratio of uranium oxide fuel
Electrochemical determination of uranium	• Determine effective oxidation state of uranium in fuel

APPENDIX B
TMI-2 PURIFICATION/MAKEUP FILTERS AND FILTER DEBRIS EXAMINATION AND ANALYSES

APPENDIX B

TMI-2 PURIFICATION/MAKEUP FILTERS AND FILTER DEBRIS EXAMINATION AND ANALYSES

This appendix provides additional details concerning the condition of the filters upon receipt at EG&G Idaho and the results of recent particle size, shape, and chemistry measurements performed at EG&G Idaho.

FILTER CONDITION

Photodocumentation, visual observation, and gross weight data were collected on each of the filters received. After the containment bags were opened, the filters and debris were left uncovered and allowed to dry in the hot cell. The total weight of debris (deteriorated filter paper and reactor core debris) for each filter is provided in Table B-1. Samples were collected from the filters and distributed to the various participant laboratories. Specimen designation, primary storage container description, gross weight of specimens and containers, net specimen weight radiation fields, and a general description of each specimen are provided in Tables B-2 through B-6.

Purification/Makeup Filter MUF-5B

The first TMI-2 filter samples were collected approximately one year before the filters arrived at EG&G Idaho. TMI-2 operations personnel attempted to pull Filter MUF-5B out of its housing, but were unable to do so during their first attempt on February 28, 1981. However, during the removal attempt, some of the filter material was inadvertently scraped from the filter. This material was recovered and portions were sent to B&W. B&W[1] milled the debris and two samples were sent to EG&G Idaho.[2] The largest sample was given the Experimental Number 0104, weighing about 1.5 g, and had a radiation dose of 25 R/h beta-gamma at near contact and a radiation dose of 3 R/h gamma at near contact. The smaller sample was given an experimental number of 0105. It had a weight of about 0.5 g. The EG&G Idaho sample was redivided and sent to ENICO and LANL. Chemical analyses were performed by B&W, EG&G Idaho, and ENICO and the results were documented

TABLE B-1. NET WEIGHT OF DEBRIS COLLECTED FROM TMI-2 PURIFICATION FILTERS

Filter	Debris Weight[a] (g)	Estimated UO_2 Content (g)[b]	Storage Identification Filter and Debris Drum	Percent of Weight (g)	Original Loading	Description
MUF-5B	203.9	14.3	--[c]	--[d]	<50	Filter paper completely deteriorated
MUF-4A	82.7	5.8	03	--[d]	~100	Filter heavily loaded and intact
MUF-4B	--[d]	0	03	--[d]	100	Filter very lightly loaded and intact
MUF-2A	405.7	28.4	--[c]	--[d]	80 to 90	Filter paper completely deteriorated
MUF-2B	206.2	14.4	--[c]	--[d]	<50	Filter paper completely deteriorated
VAC-MUF-2A-1	340[e]	23.8	09	577.6	--[f]	Heavily loaded
VAC-MUF-2A-2	~6[e]	0.4	09	245.7	--[f]	Lightly loaded
VAC-MUF-2A-3	~90[e]	6.3	03	330.5	--[f]	Moderately loaded
VAC-MUF-2B	~0	0	RCBTA	236	--[f]	Very lightly loaded--one side had a few dark areas of debris
VAC-MUF-4A	~80[e]	5.6	04	327	--[f]	Moderately loaded
VAC-MUF-4B	~0	0	04	242	--[f]	Moderately loaded--one side was loaded with dark debris
VAC-MUF-5B	~0	0	04	269	--[f]	Very lightly loaded
VAC-MUF-5A	~0	0	14	242.5	--[f]	Very lightly loaded--one side was colored light gray with debris and a few large particles were on the surface

a. Damp weights.

b. Estimate based on the assumption that 7% of the debris was UO_2.

c. Metal filter frames were discarded as hot waste.

d. Measurement was not made.

e. Estimation based on the assumption that the cartridge filter weighed 240 g.

f. Not applicable.

TABLE B-2. DESCRIPTION OF SPECIMEN SENT TO LOS ALAMOS NATIONAL LABORATORY

Specimen Label	Container Type	Weight (g) Gross	Weight (g) Net	Radiation Field (R/h) Gamma	Radiation Field (R/h) Beta-Gamma	Description
MUF-5B Sample 0104	--	--	0.25	--	--	A portion of the B&W homogenized 0104 sample was sent July 8, 1982.
VAC-MUF-5A	Plastic bag in a steel pipe	--	242.5	2.5	15	Vacuum MUF-5A filter cartridge. This filter has a small quantity of debris on one side of the filter.
MUF-5B 111D GRAB, LANL	Aluminum can	73	43.2	30	>50	"As is" sample--sample taken from debris found in the bottom of the plastic containment bag.
MUF-5B 111F Homog., LANL	Plastic vial	24	13.5	30	>50	Homogenized sample--sample collected from debris found in the plastic container bag. Debris was homogenized in a rubber lined jar filled with Al_2O_3 pellets.
MUF-5B Top 3	Large plastic vial	11.9	1.2	10	>50	"As is" sample--paper section collected from the top of the MUF-5B filter frame.
MUF-5B Center 2	Large plastic vial	11.1	0.4	8	>40	"As is" sample--paper section collected from the center of the filter.
MUF-5B Center 2	Small plastic vial	5.5	0.7	1	10	"As is" sample--section of paper and glue seam left on the filter. The section is from the center of the filter.

TABLE B-2. (continued)

Specimen Label	Container Type	Weight (g)		Radiation Field (R/h)		Description
		Gross	Net	Gamma	Beta-Gamma	
MUF-5B Bottom 1	Small plastic vial	6.0	1.0	20	40	"As is" sample--debris scraped from the bottom ring of Filter MUF-5B.
MUF-5B Top 3	Small plastic vial	5.4	0.5	15	>50	"As is" sample--paper an debris section that fell from the top of the filter. This piece was picked up off blotting paper.
MUF-2A 111AA	Aluminum can	97.5	67.9	35	>50	"As is" sample--debris collected from debris found in the plastic containment bag. Debris from Filter MUF-2A.
MUF-2B 111BA	Large aluminum can	77.9	10.1	20	>50	"As is" sample--section of filter paper and debris that fell from the top of Filter MUF-2B. This section fell off as the filter frame was raised to an upright position.

TABLE B-3. DESCRIPTIONS OF SPECIMENS SENT TO ARGONNE EAST

Specimen Label	Container Type	Weight (g)		Radiation Field (R/h)		Description
		Gross	Net	Gamma	Beta-Gamma	
MUF-2A 113AJ ANL-E	Aluminum can	39.0	27.7	--	--	"As is" sample--sample collected from debris found in the plastic containment bag of Filter MUF-2A.
MUF-2A Homogenized .113AK, ANL-E	Aluminum can	12.0	1.5	--	--	Homogenized sample--ball milled sample collected from debris found in the plastic containment bag of Filter MUF-2A.
MUF-4A 112AJ ANL-E	Aluminum can	22.0	10.8	--	---	"As is" sample--sample collected from debris and filter paper removed from the filter surface.
MUF-5B, ANL-E 111EJ	Aluminum can	19.0	8.5	--	--	"As is" sample--sample collected from debris found in the plastic containment bag of Filter MUF-5B.
MUF-4B, Paper section	Aluminum can	7.7	3.9	--	--	Excised paper section from Filter MUF-4B.

TABLE B-4. DESCRIPTION OF ARCHIVE SPECIMENS

Specimen Label	Container Type	Weight (g)		Radiation Field (R/h)		Final Disposition (drum number)	Description
		Gross	Net	Gamma	Beta-Gamma		
MUF-5BH Archive 111G	Aluminum can	39.5	20.45	--	--	02	Homogenized sample--sample from Filter MUF-5B. Sample collected from debris found in the plastic bag. Debris was homogenized in a plastic jar filled with Al_2O_3 pellets.
MUF-5B Archive 111E	Aluminum can	--	45.8	--	--	02	"As is" sample--sample taken from debris found in the plastic containment bag of Filter MUF-5B.
MUF-2A Archive Number 1	Aluminum can	222	147.8	--	--	02	"As is" sample--sample collected from debris found in the plastic containment bag of MUF-2A.
MUF-2A Archive	Plastic bottle	--	--	--	--	02	Homogenized sample--sample from Filter MUF-2A. Consists of milling balls and milling container.
MUF-2B Archive 113BC	Large aluminum can	80	12	--	--	02	"As is" sample--sample collected from debris found in the plastic containment bag of Filter MUF-2B.
MUF-2B	Plastic bottle	--	--	--	--	02	Homogenized sample--sample from Filter MUF-2B. Sample consists of milling balls and plastic milling container.
MUF-4A Archive 0112AB	Aluminum can	60	30.5	--	--	02	"As is" sample--sample collected from Filter MUF-4A. This debris was found in the plastic containment bag.
MUF-4A Archive 0112AC	Aluminum can	57.5	28.5	--	--	02	"As is" sample--sample collected from Filter MUF-4A. This debris fell out of the containment bag onto the hot cell table.
MUF-4A Archive Homogenized	Aluminum can	40.0	11.5	--	--	02	"As is" sample--debris and paper scrapped of Filter MUF-4A.
MUF-4A	Plastic bottle	--	--	--	--	02	Homogenized sample--sample from Filter MUF-4A. Sample consists of milling balls and plastic milling container.
MUF-4B Number 3	Aluminum can	8.0	3.0	--	--	02	"As is" section of paper cut out of the center of the filter.

TABLE B-5. DESCRIPTION OF SPECIMENS COLLECTED FOR MICROANALYSIS

Specimen Label	Container Type	Weight (g)		Radiation Field (R/h)		Final Disposition (drum number)	Description
		Gross	Net	Gamma	Beta-Gamma		
MUF-5B Top	Small plastic vial	15	4.9	10	>50	TRA-Cell 2	"As is" sample--section of debris and filter paper that fell from the top of Filter MUF-5B during visual examination.
MUF-5B	Glass vial	16	0.4	0.005	0.1	TRA-Cell 2	"As is" sample--large particle found among the debris of Filter MUF-5B.
MUF-5B Met .111C	Large plastic vial	12.5	7.7	15	>50	TRA-Cell 2	"As is" sample--debris collected from the containment bag of Filter MUF-5B.
MUF-2A Met 113AB	Plastic vial	10.0	5.2	20	>50	TRA-Cell 2	"As is" sample--debris collected from the containment bag of Filter MUF-5B.
MUF-2A Number 3	Plastic vial	--	--	2	20	TRA-Cell 2	"As is" sample--debris was collected from Filter MUF-2A.
MUF-2B Met 113BB	Plastic vial	13	2.2	30	>50	TRA-Cell 2	Filter paper and debris section. Cut from the intact filter paper section from Filter MUF-2B.

TABLE B-6. DESCRIPTION OF SPECIMENS COLLECTED FOR CHEMICAL AND RADIOCHEMICAL ANALYSES

Specimen Label	Container Type	Weight (g)		Radiation Field (R/h)		Final Disposition (drum number)	Description
		Gross	Net	Gamma	Beta-Gamma		
MUF-5B Physics 111B	Plastic vial	5.1	<0.1	--	--	08	"As is" sample--sample collected from the containment bag of MUF-5B.
MUF-5BH Physics 111I	Plastic vial	6.2	1.4	--	--	08	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-5B.
MUF-5B CPP 111A	Plastic vial	7.5	2.5	--	--	05	"As is" sample--sample collected from the containment bag of MUF-5B.
MUF-5BH CPP 111H	Plastic vial	6.7	2.0	--	--	05	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-5B.
MUF-2A Physics 113AD	Plastic vial	4.8	<0.1	--	--	08	"As is" sample--sample collected from the containment bag of MUF-2A.
MUF-2A Physics 113AG	Plastic vial	5.0	0.45	--	--	08	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-2A.
MUF-2A CPP 113AB	Plastic vial	10.0	4.9	--	--	05	"As is" sample--sample collected from the containment bag of MUF-2A.
MUF-2A CPP 113AG	Plastic vial	33.0	3.0	--	--	05	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-2A.
MUF-2B Physics 113BE	Aluminum vial	5.0	~0.5	--	--	08	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-2B.
MUF-2B 113BF	Aluminum vial	30.0	2.3	--	--	05	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-2B.

TABLE B-6. (continued)

Specimen Label	Container Type	Weight (g)		Radiation Field (R/h)		Final Disposition (drum number)	Description
		Gross	Net	Gamma	Beta-Gamma		
MUF-4A Physics 113AF	Aluminum vial	5.0	0.4	--	--	08	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-4A.
MUF-4A Physics	Aluminum vial	6.0	1.6	--	--	08	Homogenized sample--ball milled sample. Sample collected from the containment bag of MUF-4A.
MMUF-4B	Aluminum vial	6.5	1.9	--	--	08	Paper section from filter. Paper was excised from the top of the filter.
VAC-MUF-5A	Plastic vial	--	0.176	--	--		"As is" sample--sample knocked from the canister VAC-MUF-5A.

in earlier interim reports. A 250-mg specimen from the 0104 sample was shipped to LANL on July 8, 1982.

The MUF-5B filter was received at the EG&G Idaho hot cells in 1982. The MUF-5B filter was double bagged in plastic bags. The bags were received intact. However, the inner containment bag (containment bag in contact with the filter) was open at the top. The inner containment bag was split lengthwise to open the bag for filter removal.

Except for a small section at the top of Filter MUF-5B (see Figure B-1), almost all of the filter paper had fallen off the metal filter frame. A glue-paper seam ran along the length of the filter that remained with the filter frame except for a small section removed from the filter at midheight. This piece will be examined by LANL. Debris also remained in the top and bottom metal retaining lips. The debris found in the containment bag with the filter had the appearance of wet dark mud.

The intact paper sections at the top of the filter were collected. One section was retained by EG&G Idaho for analysis, one sent to LANL, and the third archived. Samples were also collected from the filter at midheight and from the bottom rim of the filter. These samples are described in more detail in Table B-2. A rather large debris particle (see Figure B-2) was collected from the loose debris. SEM/EDS analysis showed it to be primarily silicon.

Approximately 204 g of debris were collected from the containment bag and filter surface. The filter paper could not be extracted from the top and bottom rims. It was estimated that only about 50% of the filter paper and filter debris was received with this filter.[a] The debris was not completely dried prior to weighing, so the debris weights are only approximate and are probably high.

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a. It is estimated that a fully loaded large filter would have held between 400 and 500 g of filter paper and debris. This estimate was based on the assumption that the weight ratio of the filter paper and debris was proportional to the surface area ratio of the filters and by the observation that Filter MUF-4A was essentially intact with no significant loss of filter paper or debris. Thus, Filter MUF-4A represented the expected filter paper and debris weight per surface area.

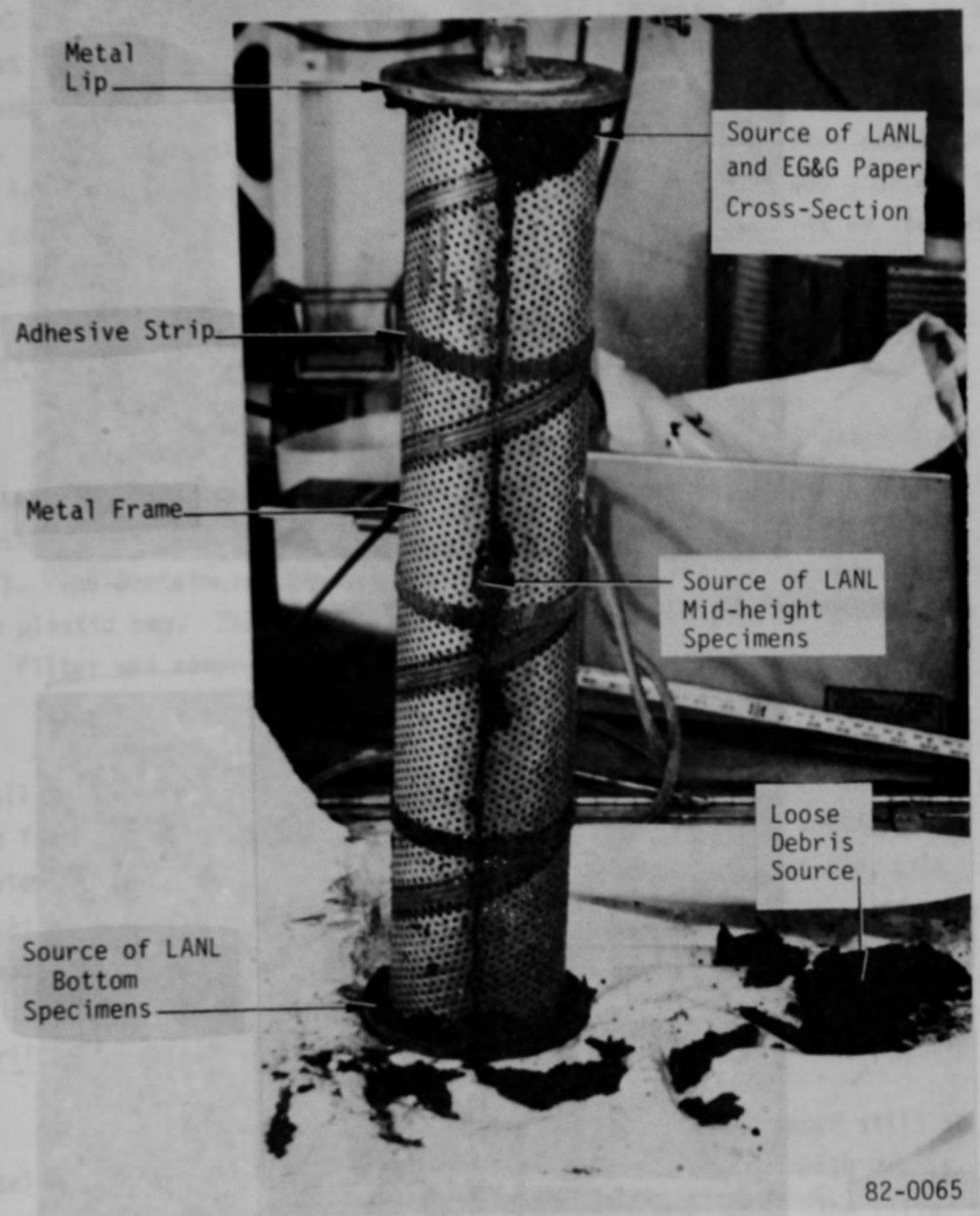
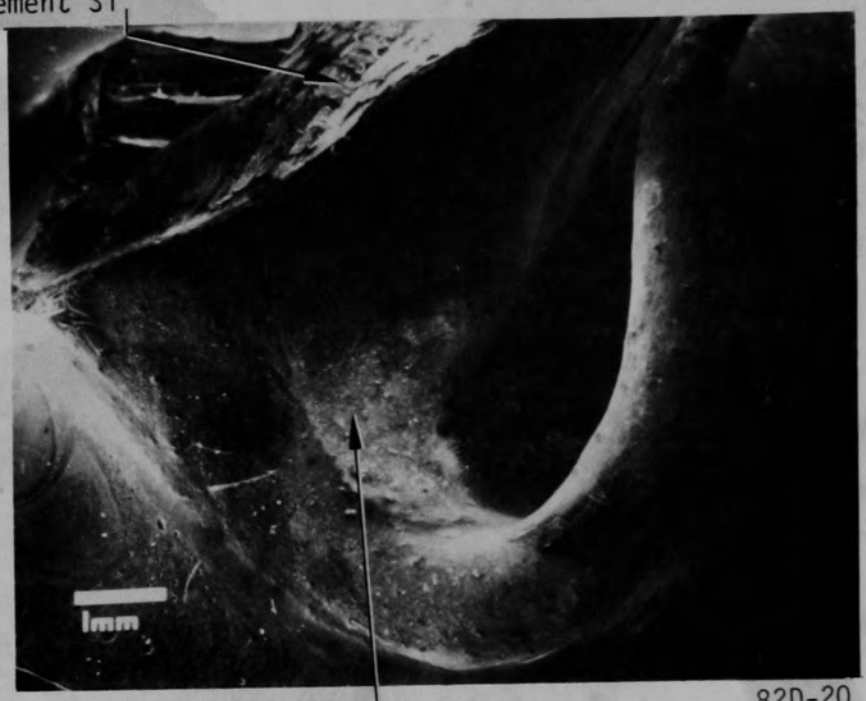
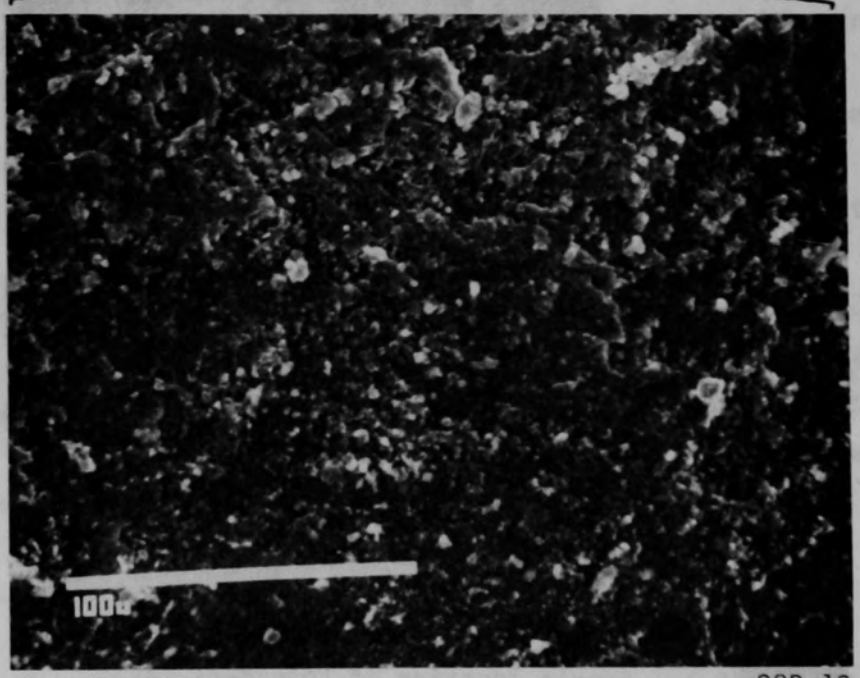


Figure B-1. Sample location of specimens taken from Filter MUF-5B.



82D-20

a. SEM Overall Structure



82D-19

b. Surface Covered with Control Material and Steel/Inconel Material.

Figure B-2. Large debris piece found in the MUF-5B debris.

Purification/Makeup Filter MUF-2A

Practically all of the filter paper had fallen off the Filter MUF-2A metal frame before it was received in the hot cell. The vermiculite in the first plastic bag was dark and wet, as illustrated in Figure B-3. Water probably leaked from the filter through small holes in the inner containment bag. A small amount of vermiculite was knocked into the filter debris while the inner containment bag was being opened. Approximately 406 g of debris was collected from Filter MUF-2A. Very little debris was left on the metal filter frame.

Purification/Makeup Filter MUF-2B

This filter was contained in only one bag. As with the other plastic filter containment bags, this one was open at the top and some vermiculite probably fell through the opening as the filter was unloaded in the hot cell. The containment bag was torn where the top filter rim rubbed against the plastic bag. The plastic bag in this region was brittle and tore as the filter was removed for examination.

All of the filter paper had fallen off the filter frame except for a small section at the top, as illustrated in Figure B-4, which broke off as the filter frame was stood upright. The originally intact section was divided into three pieces for analysis: EG&G Idaho received one, LANL received one, and one was archived. Debris on this filter was quite dry compared to the debris found on the other filters.

Purification/Makeup Filter MUF-4A

This filter was received with almost all the filter paper still on the metal filter frame. The filter paper was dark and covered with debris. It was very friable and fell apart as the filter was handled. The filter paper and debris were only slightly damp.

The metal filter frame was collapsed, but the filter paper showed no sign of being smashed. Nearly all of the filter paper was scraped off this

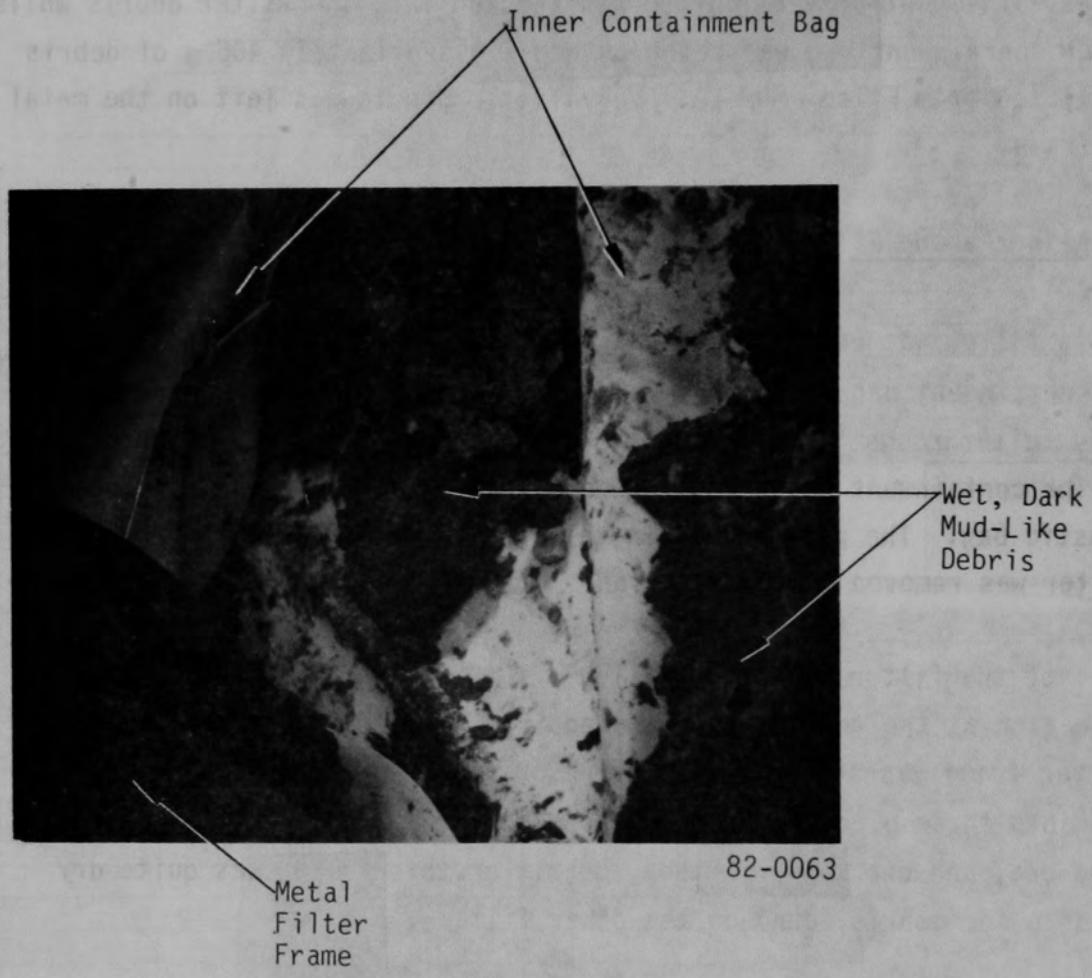
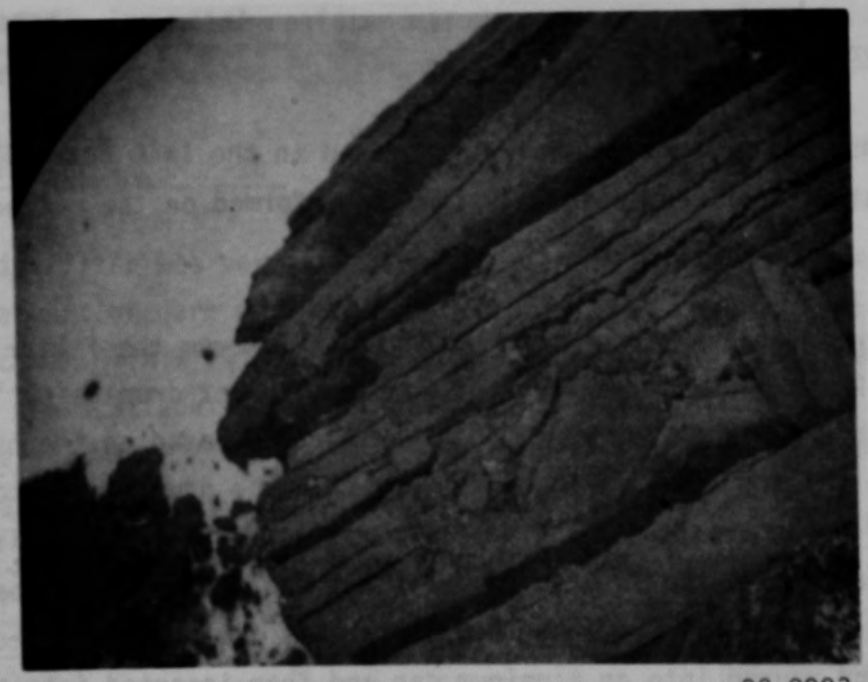


Figure B-3. Condition of filter and filter debris on Filter MUF-2A.

82-0070

Intact Filter Paper



82-0083

b. High Magnification of the Section of Filter MUF-2B Paper Sent to LANL

Figure B-4. Intact filter paper section found on Filter MUF-2B.

filter. The filter material weight is the dry weight. A total weight of 82.7 g of debris and filter paper was collected. Samples from this filter were collected and distributed as indicated in Tables B-2 through B-6. This filter was gross and spectral gamma scanned, and the results of these examinations were reported by Nitschke.[3] The filter housing was put in a steel storage container and stored in a 55-gal drum, which will be stored at the EG&G TMI-2 archive storage area.

Purification/Makeup Filter MUF-4B

This filter was received intact and was not loaded with the dark debris found on the other filters. The filter paper was, however, swollen at the center. The metal frame was not deformed, so the midheight bulge was due solely to swollen filter paper. There were small pieces of vermiculite on the filter surface, which probably got there during handling of the filter and filter bag. This filter was gross and spectral gamma scanned, and the results were reported by Nitschke.[3] This filter was put in a steel storage container and stored in a 55-gal drum, which will be stored at the EG&G Idaho TMI-2 archive storage area.

Vacuum Canister Filter VAC-MUF-2A-1

The VAC-MUF-2A-1 vacuum filter was examined in the INEL hot cell. However, no material or chemical analysis was performed on the collected debris.

This filter was supposed to have been used to vacuum the filter housing of Filter MUF-2A. However, the labeling on the shipment drum read, "In line VAC filter - MUF-2A, -2B, -5A, -5B, and Floor Area." Based on the heavy loading of the filter, it is believed the label on the drum is an accurate description of those areas the vacuum filter was used to clean. This filter was heavily loaded with debris. In fact, it was the most heavily loaded vacuum canister received. The loose debris of this filter was collected in the hot cell and poured into an aluminum can and then inserted into the center of the cartridge filter. The vacuum cartridge and aluminum can were then inserted into a steel storage container (a steel pipe welded on one

end and a cap on the other). The steel storage container was then put in a 55-gal drum, which will be stored at the EG&G Idaho TMI-2 archive storage area. All vacuum filter cartridges were stored in this manner.

Vacuum Filter VAC-MUF-2A-2

This vacuum cartridge filter was moderately loaded with debris, most of which stayed on the filters or on the metal cartridge walls, so no loose debris was collected. This filter was gross and spectral gamma scanned. However, no material or chemical analysis was performed, nor is any analysis of this filter or filter debris anticipated. The metal canister was discarded as hot waste, and the filter was stored for archiving.

Vacuum Filter VAC-MUF-2A-3

This vacuum filter was moderately loaded, holding approximately 90 g of debris. The tare weight of the filter cartridge was assumed to be 240 g. This filter was gross and spectral gamma scanned. No material or chemical analysis was performed, nor is any analysis of this filter or filter debris anticipated. The metal canister was discarded as hot waste and the filter was stored for archiving.

Vacuum Filter VAC-MUF-2B

This filter was very lightly loaded, with one side of the filter having small patches of dark material, the remainder of the filter surface relatively clean. This filter was gross and spectral gamma scanned. No material or chemical analysis was performed, nor is any analysis of this filter or filter debris anticipated. The metal canister was discarded as hot waste, and the filter was stored for archiving.

Vacuum Filter VAC-MUF-4A

This filter was moderately loaded, the entire surface covered with dark debris. A small amount of debris fell off the filter and was collected. This filter was gross and spectral gamma scanned. No material or chemical

analysis was performed, nor is any analysis of this filter or filter debris anticipated. The metal canister was discarded as hot waste, and the filter was stored for archiving.

Vacuum Filter VAC-MUF-4B

This filter was moderately loaded with debris and one side was particularly dark. This vacuum filter should not have had any dark debris on the filter surface, since the purification/makeup MUF-4B filter was devoid of dark debris. There was, nevertheless, some dark patches (light coating of debris) on the filter. This filter was gross and spectral gamma scanned. No material or chemical analysis was performed, nor is any analysis of this filter or filter debris anticipated. The metal canister was discarded as hot waste, and the filter was stored for archiving.

Vacuum Filter VAC-MUF-5B

This vacuum filter was very lightly loaded. However, the net weight of this filter was 269 g, compared to the 242 g and 233 g of the other lightly loaded filters. The reason for the higher weight was not obvious, unless it was still damp. This filter was gross and spectral gamma scanned. No material or chemical analysis was performed, nor is any analysis of this filter or filter debris anticipated. The metal canister was discarded as hot waste, and the filter was stored for archiving.

Vacuum Filter VAC-MUF-5A

This filter was very lightly loaded. However, it was highly radioactive, as it measured 15 R/h beta-gamma at near contact and 5 R/h gamma at near contact. This filter had a light gray coating of debris on one side, and there were also some large particles visible on the fiber surface, and a few particles were lodged in the wrapping twine. A 0.176-mL/g sample was removed from the metal canister; analyzed with neutron activation analysis, weighed, and gamma scanned. Gamma analysis of the VAC-MUF-5A sample is presented in Table B-7 along with those for the MUF-5B Samples 0104 and 0105, the latter data having been decay-corrected to 7/21/82.[4] The

TABLE B-7. GAMMA SPECTROMETRY ANALYSIS OF THE TMI-2 PURIFICATION FILTERS

Radionuclide	Activity (μCi/g) MUF-5A Vacuum[a]	MUF-5B Sample 104[b]	MUF-5B Sample 105[b]
^{54}Mn	4.7 ± 0.6	12 ± 1	12.3 ± 0.5
^{60}Co	132 ± 2	190 ± 7	193 ± 3
^{95}Zr	5.9 ± 0.9	--[c]	--[c]
^{95}Nb	7.8 ± 0.6	--[c]	--[c]
$^{106}Ru/Rh$	460 ± 10	410 ± 20	422 ± 7
^{110m}Ag	4.6 ± 0.8	28 ± 2	26.7 ± 0.6
^{113}Sn	--[c]	2 ± 1	1.5 ± 0.6
^{125}Sb	365 ± 5	2360 ± 30	2330 ± 10
^{134}Cs	161 ± 2	343 ± 6	348 ± 3
^{137}Cs	1790 ± 10	3850 ± 20	3810 ± 10
^{144}Ce	705 ± 7	620 ± 10	583 ± 6
^{154}Eu	4 ± 1	--[c]	--[c]
^{155}Eu	7 ± 2	--[c]	--[c]

a. Determined 7/21/82.

b. Decay corrected to 7/21/82.

c. Not detected.

fission and activation product concentrations in the VAC-MUF-5A sample are substantially lower than concentrations in the MUF-5B samples, with the exception of two nuclides, ^{106}RU and ^{144}Ce. The concentration of uranium (5.5 wt%) in Filter MUF-5B Samples 0104 and 0105 was applied to the VAC-MUF-5A sample to obtain an uranium concentration. The estimated uranium concentration was 0.025 g. The VAC-MUF-5A cartridge filter was sent to LANL, and they may examine this filter and the material on it in FY-83.

MICROANALYSIS RESULTS

Microanalysis of filter debris provided information on the particle size and shape distributions, on the phase structures and elemental composition of the particles, and on the radiation levels of the specimens.

Particle Size and Shape Distribution

Particle size and shape distributions were determined from polished cross sections of MUF-5B and -2B debris particles. A cross section of MUF-2B filter paper is shown in Figure B-5. High magnification photographs of portions of this cross section are shown in Figure B-6. The overall particle size distribution for Filter MUF-5B and -2B was obtained from these photographs and other similar photomicrographs. Particles from the purification/makeup filters debris were generally very small. The particle sizes ranged from submicron particles to <50 μm particles using the projected diameters and from submicron to 75 μm using Feret's diameters.

The particle size distribution of the spherical particles was also determined. This distribution is illustrated in Figure B-7. The mean size is $\leq$ μm.

The particle size and shape distributions were measured with an image analyzer and by manual techniques. The particle diameters from the image analyzing equipment are Feret's diameters and the manual measurements are projected diameters. The projected particle diameter is the diameter of a circle whose area is equal to the area defined by the profile of the particle.[5] The Feret's particle diameter is the mean length between two tangents on opposite sides of the apparent outline of the particles.[5]

Particle shape varied tremendously in the Filter MUF-5B and -2B debris. Particle shapes range from spherical particles to very irregular particles, as illustrated in Figures B-7, B-8, and B-9. Analyses were performed on paper sections from Filters MUF-5B and -2B. Figure B-6 shows particles from Filter MUF-2A, and Figures B-8 and B-9 show particles from Filter MUF-5B. Figure B-8 shows the deposition pattern found on Filter MUF-5B. Layers of

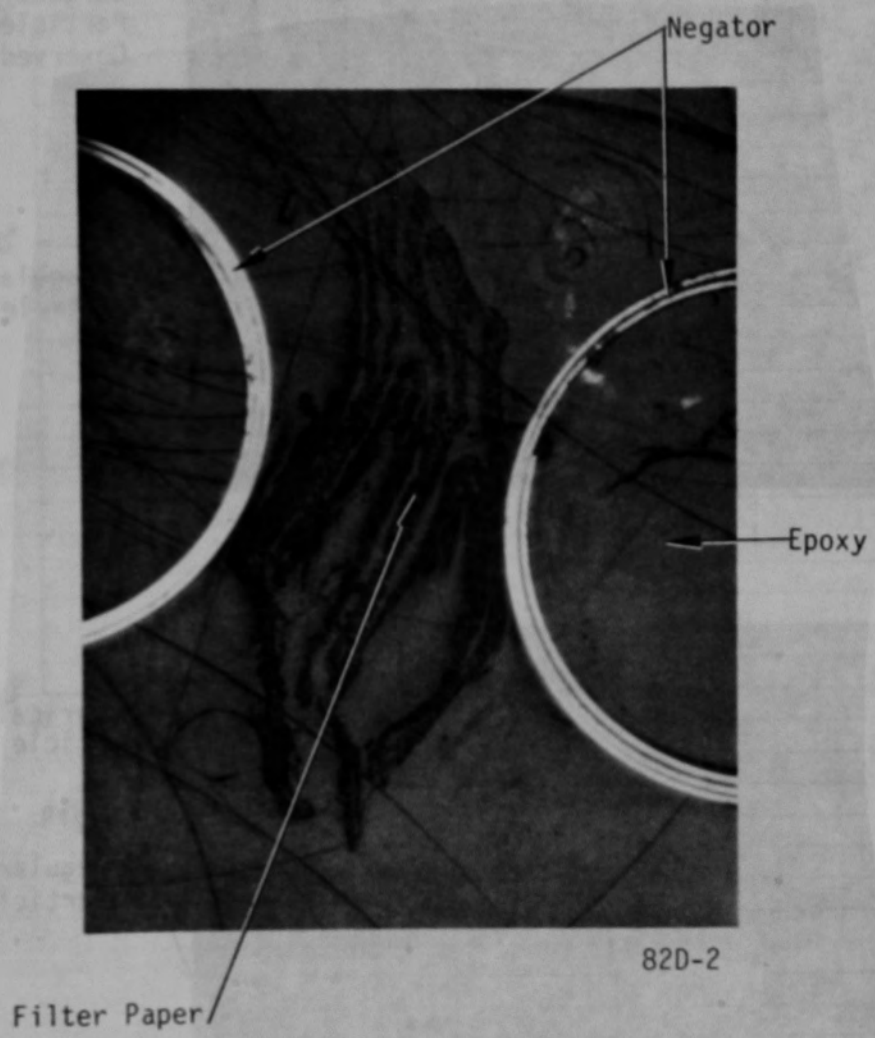
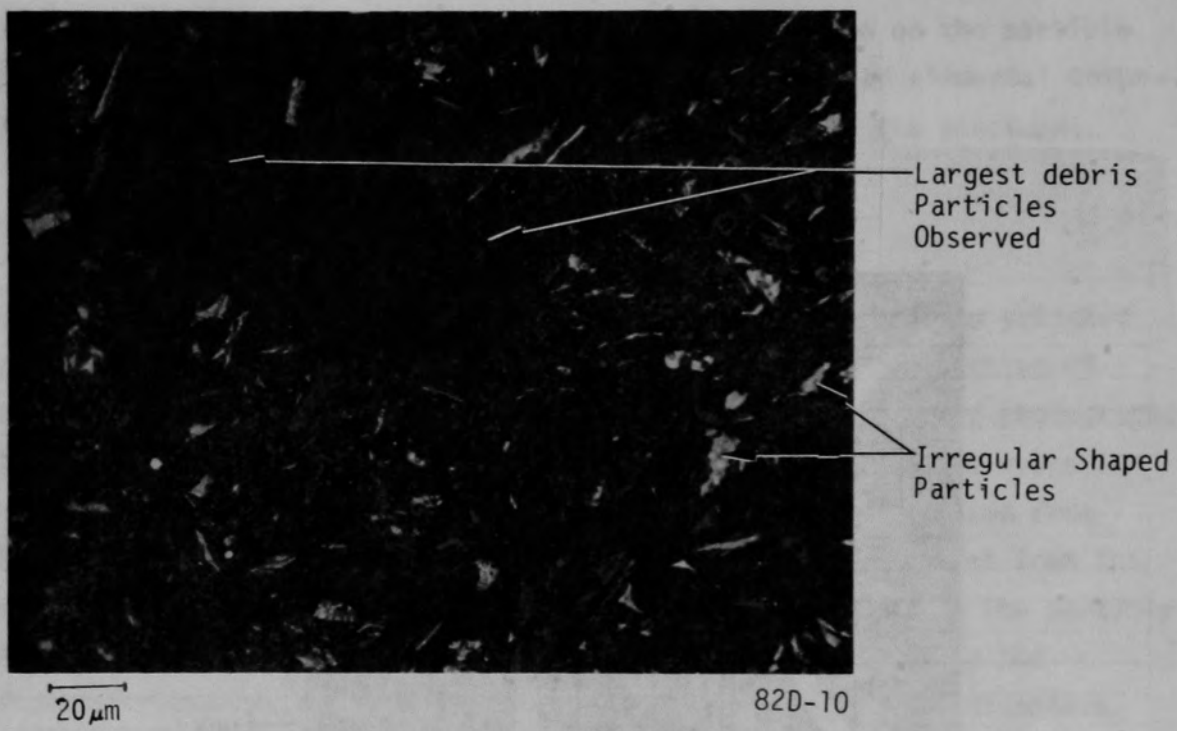


Figure B-5. Cross-section of Filter MUF-2B filter paper.



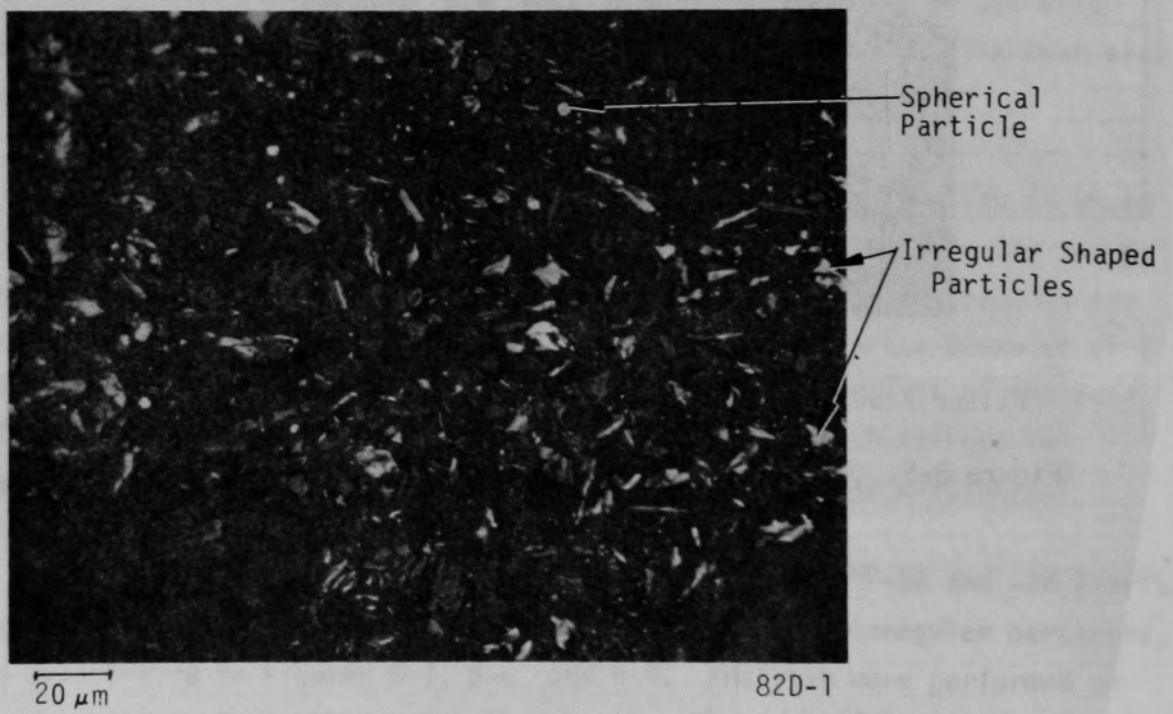


Figure B-6. Photomicrograph of the polished cross-section of Filter MUF-2B paper.

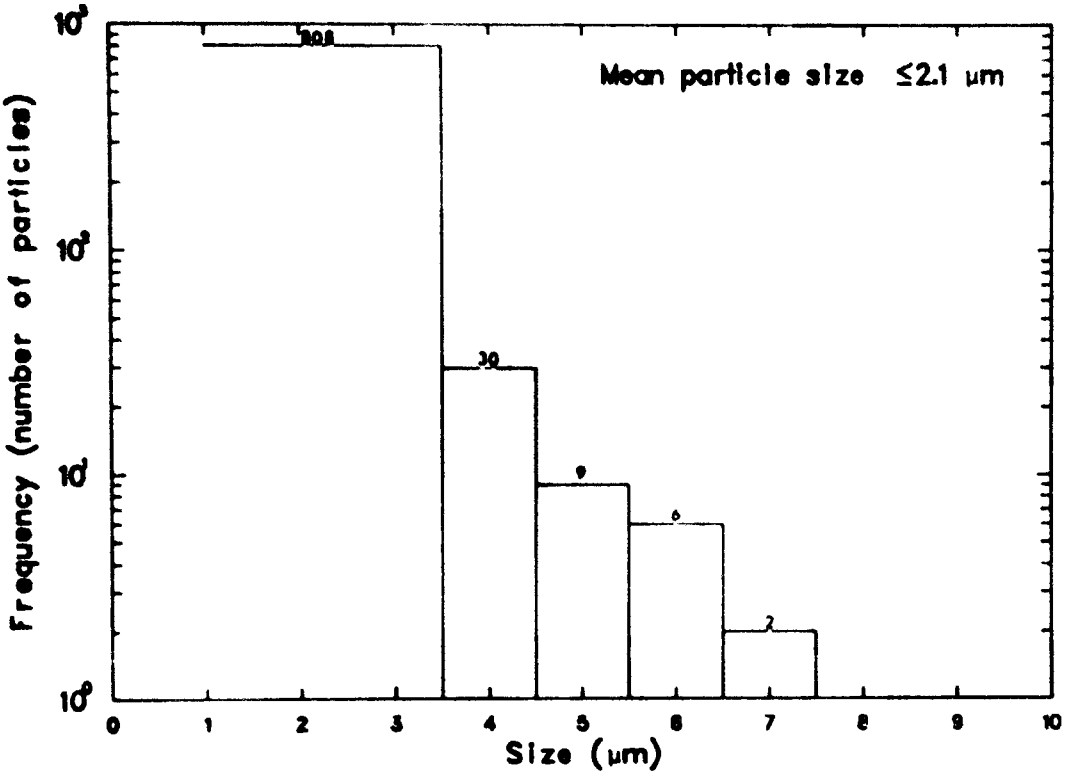
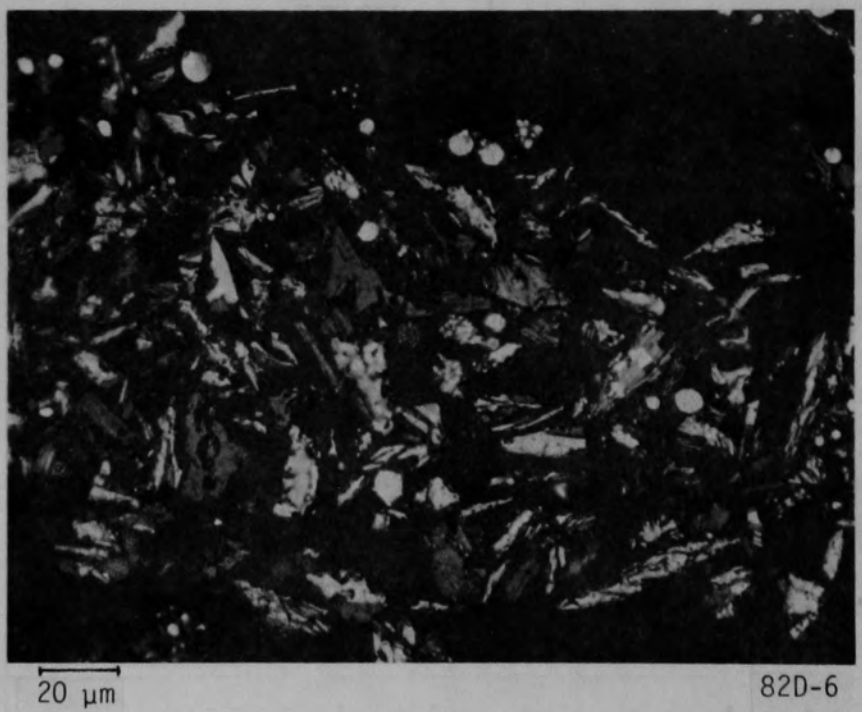


Figure B-7. Size distribution of spherical particles in Filter MUF-5B debris.

a. Stratified Particles



b. Mixture of Spherical and Irregular Shaped Particles

Figure B-8. Deposition pattern on Filter MUF-5B.

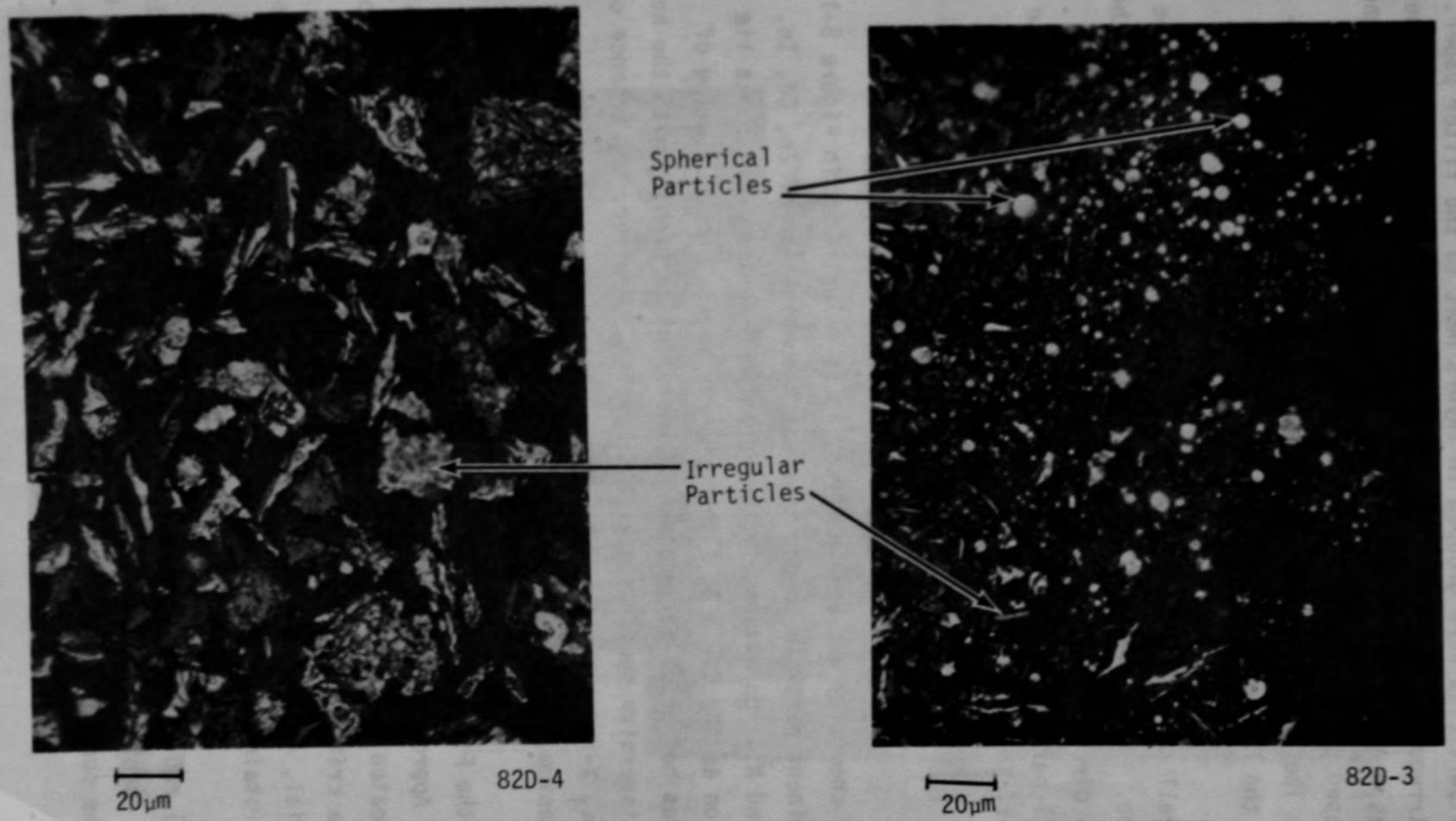


Figure B-9. Particulate patterns on the polished cross-section of Filter MUF-5B paper.

spherical and irregular shaped particles were observed. At most locations, the spherical particles were grouped in layers next to the filter paper, whereas the irregular shaped particles were deposited on top of the spherical particles. At other locations on the filter paper the spherical and irregular shaped particles were randomly deposited as illustrated in Figure B-8b. The irregular shaped and spherical particles at two other locations on the Filter MUF-5B filter paper are shown in Figure B-9.

The overall sphericity of the particles is a gross indicator of the particle shape. Sphericity is the ratio of the projected diameter to the diameter of a circle that just circumscribes the particle.[5] The mean sphericity all particles measured was about 0.61, thus, indicating quite irregular particles.

Particulate Chemistry

Particle chemistry was very diverse, as is illustrated in Figure B-10. The most prominent particle composition was a combination of Zr, Cd, In, Ag, Cr, Fe, and Ni. Unfortunately, the uranium M-series X-ray peaks lie in the same region as the Cd-In-Ag L-series X-ray peaks. Deconvoluting of these peaks was not practical because of a resolution problem with the hot cell energy dispersive spectroscopy (EDS) system. However, the absence of the uranium $L\alpha_1$ X-ray peak at 13.61 keV was assumed to indicate the absence of uranium.

Most of the Filter MUF-5B debris particles were formed from control rod failure. Approximately 61% of the debris was from the control material. As the rods heated up during the accident, the cadmium vapor pressure probably reached a critical pressure and ruptured the control rod cladding. The rod material, Ag-Cd-In, probably then melted and liquified the control rod cladding (stainless steel) and then the zircaloy guide tubes.

Some fuel rod material (uranium) did break up and deposit on the filters, but the quantity of fuel debris was minor compared to the quantity of debris from the control rods. Figure B-11 shows three particles that

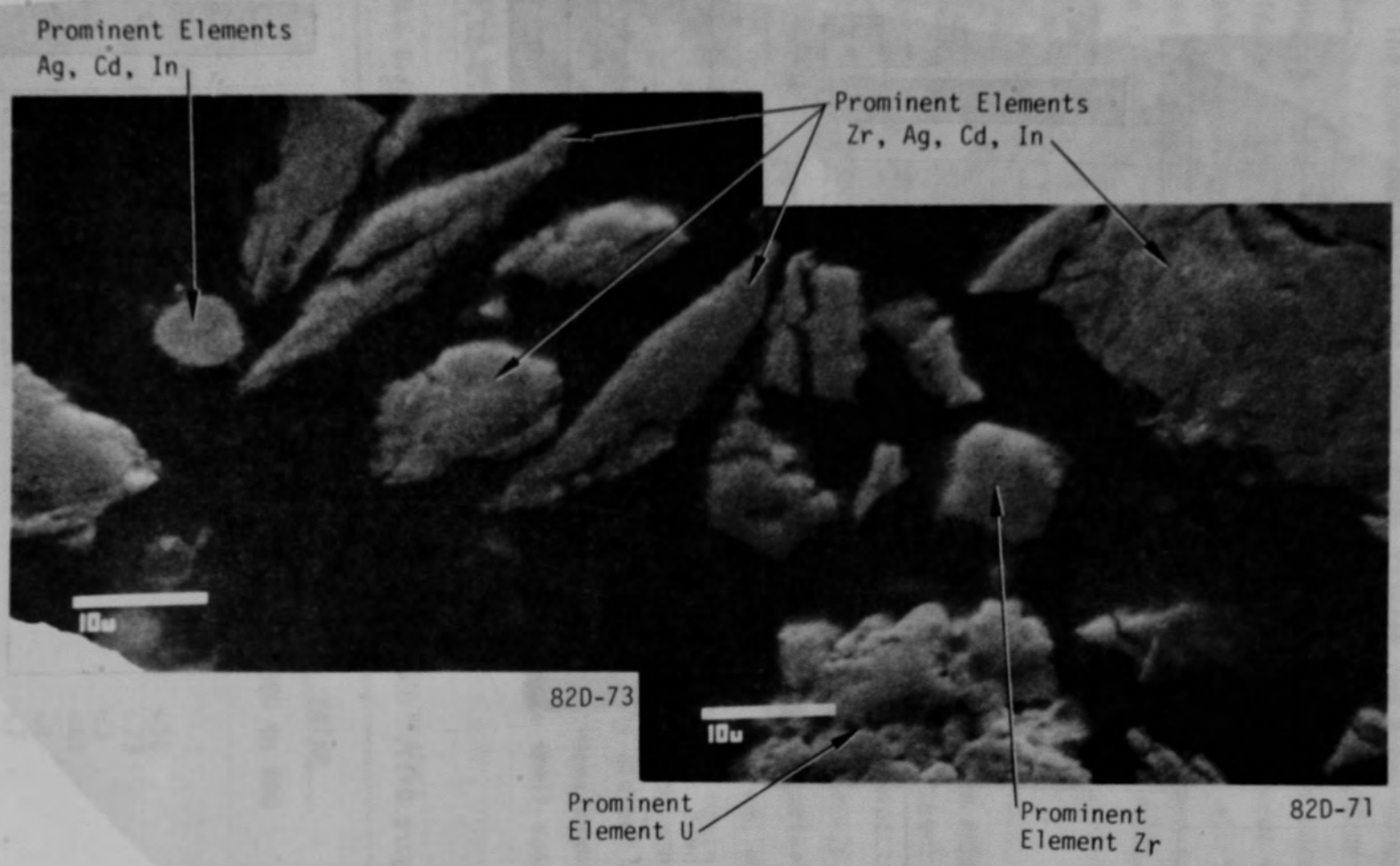


Figure B-10. SEM Photomicrographs of Filter MUF-5B debris particles.

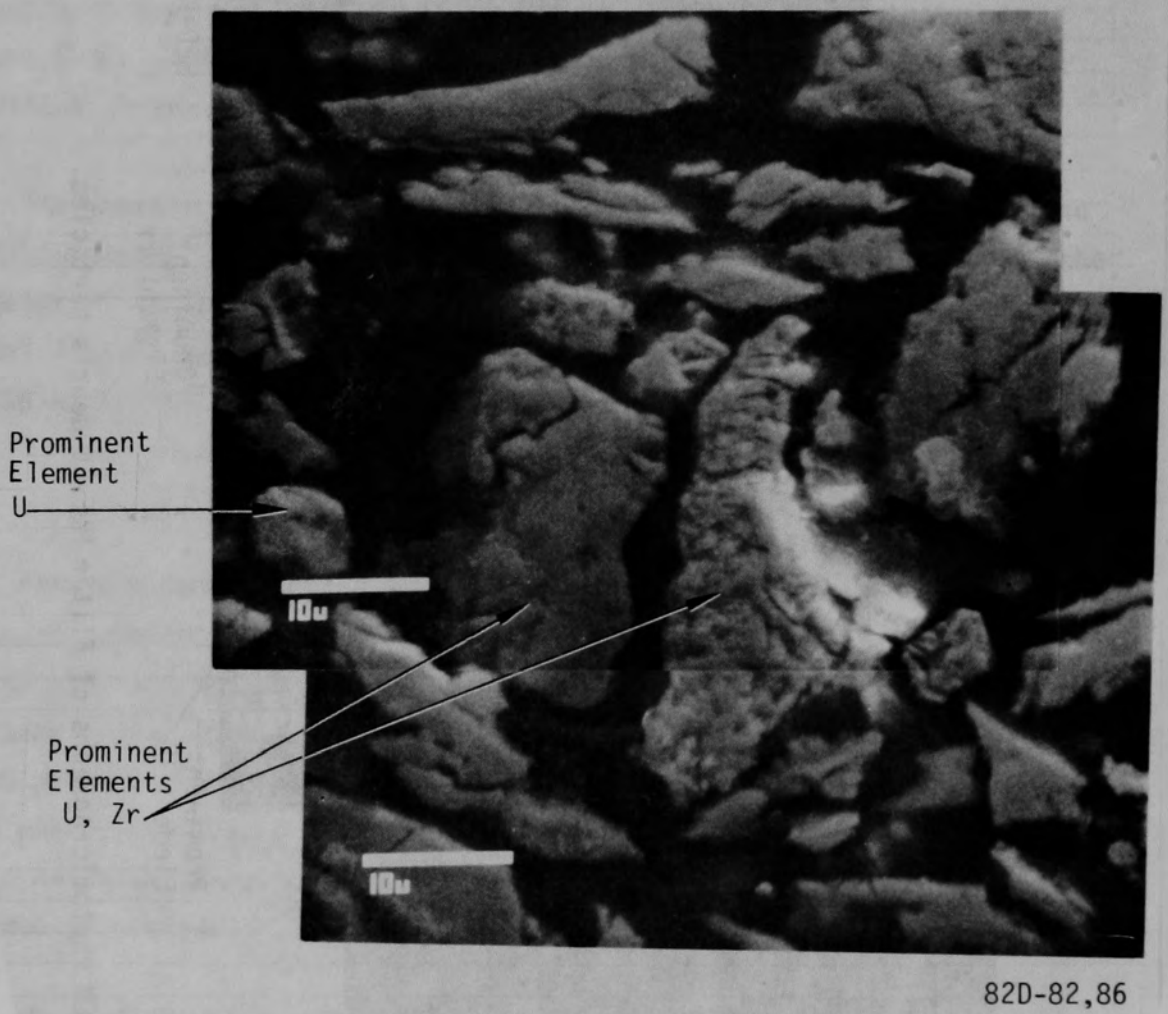


Figure B-11. Uranium particles in the Filter MUF-5B debris.

probably contain uranium. One particle is primarily uranium (UO_2), whereas the other two particles contain both uranium and zirconium.

The spherical particles found on the filters had X-ray spectra of Ag-Cd-In. It is possible that the spherical particles are Cd or CdO. This is suggested by the work of Adams and Tobias,[6] who simulated core-melt accidents on LWR fuel and control rods. They found the control rods burst at about 1678 K because of high cadmium vapor pressures. Their tests indicated that cadmium volatized and left the control rod and then formed CdO when oxygen was available. They also vaporized cadmium, and found the vapor to form small spherical particles, i.e., is, they found spherical particle structures very similar to that found in the filter debris.

We were requested to look for resin beads that may have entered the purification lines from the demineralizer tank. The resin beads were ion-exchange resins that cleaned the coolant lines of tramp radioactive elements. No intact resin beads were observed in the MUF-5B or -2B filter examination. Table B-8 provides the original physical characteristics of the resin beads. The preaccident resin bead size was >380 μm, which is much larger than any particle observed in the debris. The resin beads could have been reduced in size somewhat as the beads traveled through the reactor coolant system, but significant size reduction probably did not occur. Thus, there was no evidence of resin beads in the specimens examined. This observation is only tentative, however, because only a small sample of the debris was obtained.

TABLE B-8. PHYSICAL AND CHEMICAL CHARACTER OF RESIN BEADS

Physical Parameter	Value
Particle size (μm)	380 to 600
Composition (wt%)	
S	~16
O	~23
Li	~3
C	~47
H	~4
DivinylBenzene	~7

Radiation Background

Microanalysis of radioactive specimens was made difficult by the interference of low-energy gamma radiation and X-ray fluorescence radiation. This radiation was generated by bremstralung X-rays, beta particles, low-energy gamma radiation, and/or X-ray fluorescence from decay activation. The photo energy background spectrum from a MUF-5B debris specimen is shown in Figure B-12. Many of the background peaks were Kα X-ray peaks from major elements in the debris. Nuclide signatures were also evident. The ^{125m}Te gamma peak at ∿35.5 keV was in the background spectrum. The Tellurium Kα X-ray peak at 27.5 keV was, however, the most intense, followed by the Kβ X-ray peak at 31 keV, and then the gamma peak at 35.5 keV. Tellurium^{125m} is a daughter of ^{125}Sb and has a half-life of 58 days. The energy peak at ∿32 keV was probably the ^{137m}Ba Kα and Kβ X-ray peaks. Barium is the decay daughter of ^{137}Cs, the most prominent radioisotope in the debris. The other peaks shown in Figure B-12 were of similar origin. Quantitative analysis with X-rays generated from radioactive decay and low-energy gamma rays may not be feasible, but these troublesome background peaks could possibly provide useful qualitative information on fission products in the debris.

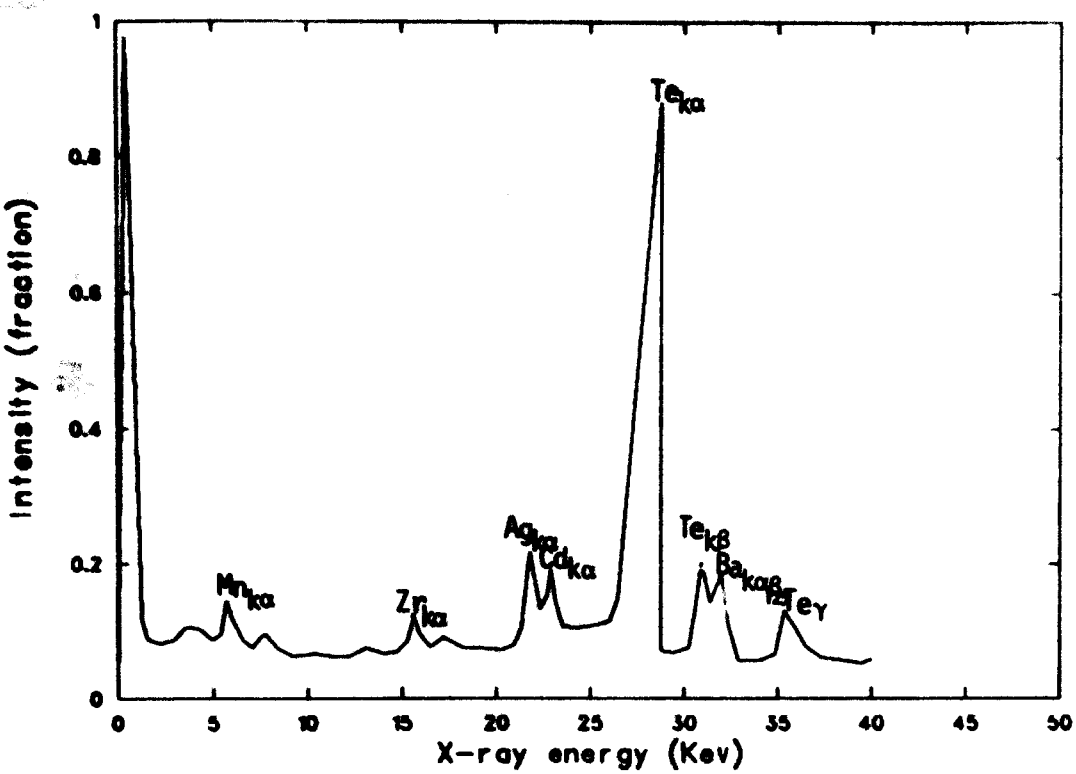


Figure B-12. Background radiation of debris from Filter MUF-5B.

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APPENDIX C

LOS ALAMOS NATIONAL LABORATORY ANALYSIS OF SAMPLE TMI-0104

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APPENDIX C

LOS ALAMOS NATIONAL LABORATORY ANALYSIS OF SAMPLE TMI-0104

Three Mile Island sample TMI-0104, which is homogenized debris from filter MU-F-5B, has been analyzed and used for procedure development.

Initially, a gamma scan of most of the sample was performed to determine if the gamma emitting isotopes were unevenly distributed in the sample. No indication of inhomogeneity was found.

A procedure was developed for dispersing the powdered sample and collecting it on mounts suitable for image analysis and electron microprobe characterization.

A portion of the sample was dispersed, mounted, and analyzed for particle size distribution using an image analyzer and elemental composition using an electron microprobe. The results of these analyses indicate a probable separation of the control rod chemical elements silver, indium, and cadmium during the course of the TMI accident. The particle sizing data are of a preliminary nature. Work is continuing in this area.

A spinning riffler sampling device was designed and built that would allow the division of the ~250 mg of sample into ten separate and representative fractions that could be handled easily and analyzed by various procedures.

Three sample fractions of the ten obtained from the spinning riffler were dissolved and analyzed by gamma spectroscopy. The results are in good agreement with those from previous work performed by Babcock and Wilcox and Idaho National Engineering Laboratory.

I. INTRODUCTION

This report presents the results of work performed by Los Alamos National Laboratory on TMI sample TMI-0104, which is homogenized debris from filter MU-F-5B.

Much of the work done on sample TMI-0104 has been aimed at procedure development with the intention of providing procedures that will be applicable to future samples. Procedures that have been developed include: (1) a method for sub-dividing samples without introducing segregation, (2) dissolving samples, and (3) dispersing the powdered samples for size analysis and electron microprobe analysis.

Data are presented for electron microprobe analysis, gamma spectroscopy, gamma scan, and particle sizing with an image analyzer. The particle-sizing data are of a very preliminary nature, but they are presented to show the nature of the information that can be obtained with the image analyzer.

Details of the work performed are presented below with some comments on the significance of certain observations.

II. EXPERIMENTAL AND RESULTS

A. Gamma Scan of the Homogenized Debris Sample from the MU-F-5B Filter

In order to determine if any gross inhomogeneity existed in the distribution of the gamma activity in sample TMI-0104, a gamma scan was made of the sample as received.* Qualitative data were also obtained on the gamma emitting isotopes in the sample.

The ~250 mg of homogenized debris was received in a ~1-cm-diameter by 2 cm-high plastic vial. To mount the sample for γ scanning, the container vial was placed in a larger snap-cap vial which was then placed in a stainless steel tube that was compatible with the gamma scanner mounts.

A diametral and an axial gross gamma scan were taken with a 1.27 mm primary collimator taking 0.051 mm steps. The results of the scans are shown in Figs. 1 and 2. No gross inhomogeneities are evident in the sample.

*Prior to γ-scanning a small grab sample had been removed from the sample vial.

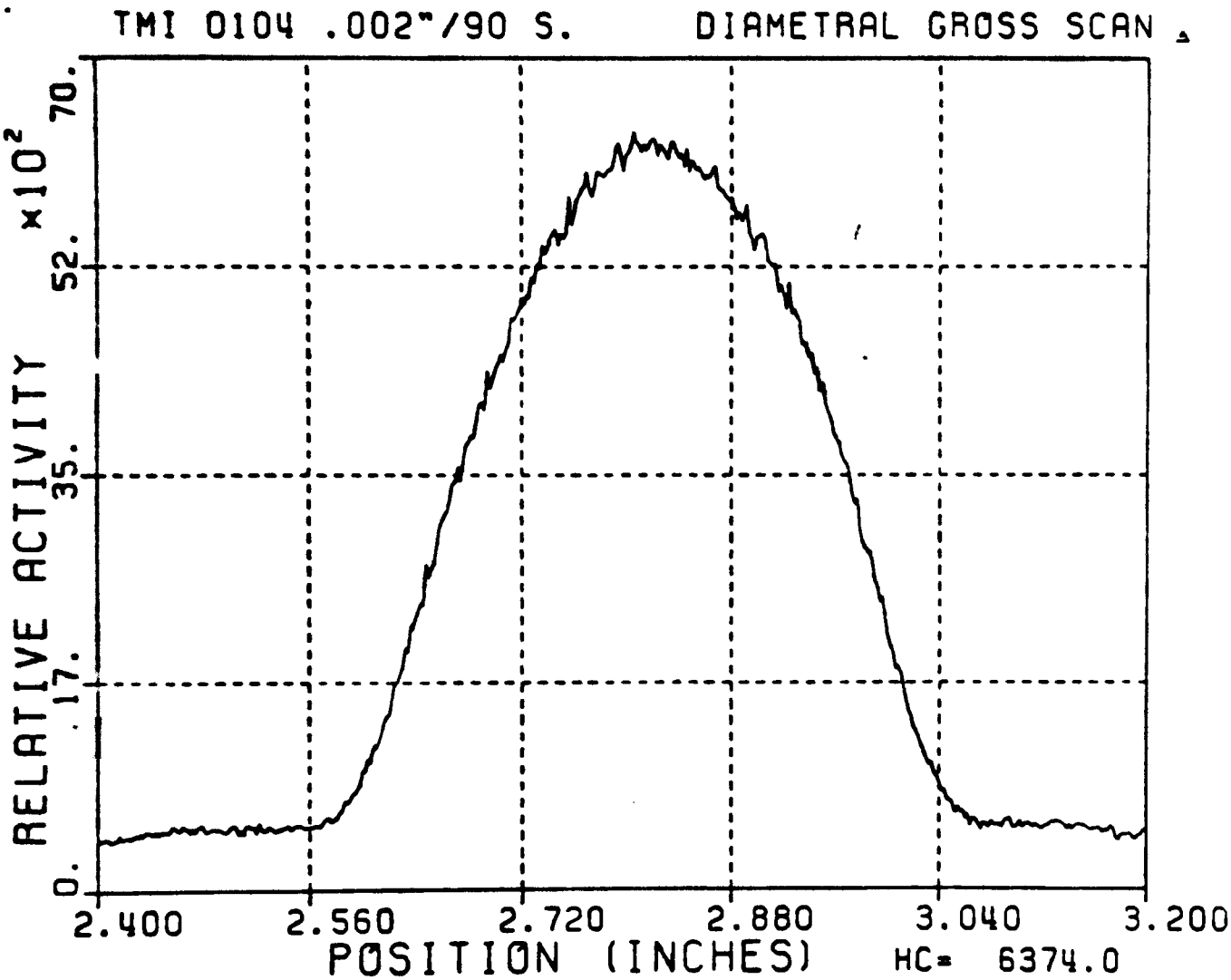


Fig. 1. Diametral gross gamma scan of sample TMI-0104.

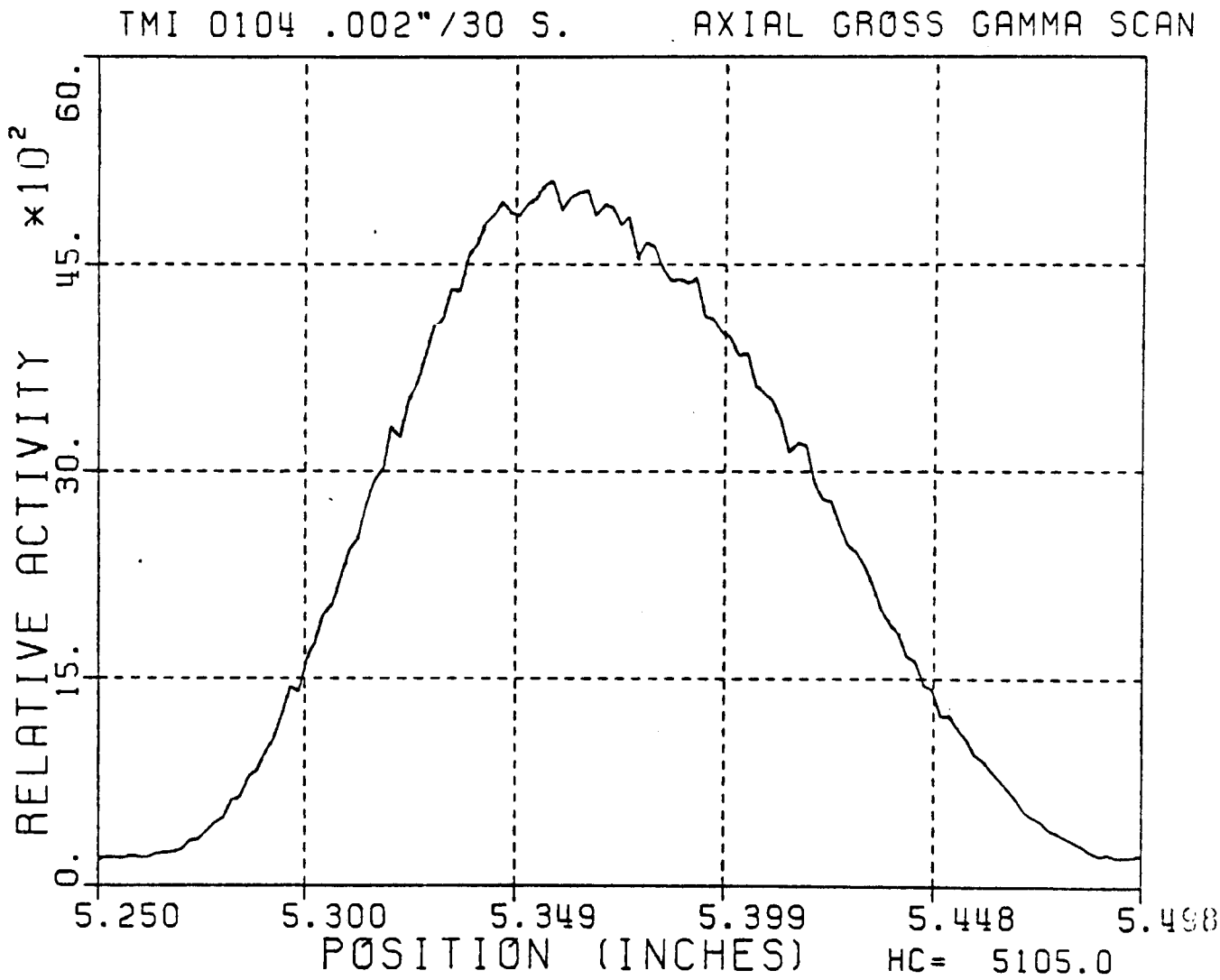


Fig. 2. Axial gross gamma scan of sample TMI-0104.

An axial multielement gamma scan of the sample was taken using a 1.27 mm primary collimator with a 7.62 mm secondary collimator taking 1.27 mm steps at 2 hours per increment. This scan provided the distributions of the gamma-emitting isotopes Cs-137, Co-60, Sb-125 and Cs-134. No unusual distribution of these isotopes were found.

An isotopic gamma spectrum was taken for 30 minutes using a 15.340 mm primary collimator with a 7.620 mm secondary collimator. The gamma energy peaks that were identified and the isotope to which they are attributed are given in Table I. The isotopes found correspond to those reported by B&W except for the shorter lived isotopes of Zr-95 and Cr-51 which have decayed to undetectable levels.

B. Preparation of Samples for Electron Microprobe and Image Analyzer Analysis

A metallographically mounted and polished piece of graphite was selected as a common sample mount for both the electron microprobe and image analyzer. The graphite provides an electrically conductive substrate for the sample that is suitable for the probe work.

Dispersing a powdered sample on the graphite mounts to give a good separation of particles free of agglomerates required several different attempts including: (1) dispersion wth iso-pentyl acetate; (2) dispersion in xylene using an ultrasonic bath; (3) smearing a mixture of the powered sample and iso-pentyl acetate on the mount with a glass slide. None of these approaches proved satisfactory.

Dispersion of the powder with an insufflator was then tried and adopted. A grab sample, visually estimated to be ~25 mg, was taken from the ~250 mg of homogenized debris and placed in a DeVilbiss-119 powder insufflator which had most of the sample reservoir filled with epoxy to reduce the volume to an appropriate size for the small sample. Graphite mounts were coated with a thin film of a solution of TEM replicating tape dissolved in xylene. After drying, the mounts were placed on the bottom of a 5 cm-wide × 25-cm-high × 30-cm.-long enclosure. All the mounts were placed near one end of the box. At the opposite end of the box, the outlet of the insufflator was placed in a small hole near the top of the box. Sample material was dispersed from the insufflator into the box. The cloud of powder settled to the bottom of the box and onto the mounts. This procedure produced well-dispersed samples that were suitable for analysis and almost free of agglomerates.

TABLE I

ENERGY PEAKS AND ASSOCIATED ISOTOPES IDENTIFIED IN SAMPLE NO. TMI 0104

Energy (keV)	Isotope
133.5	Ce-144 + Pr-144
696.5	
176.3	Sb-125
208.1	
380.4	
427.9	
463.4	
600.6[a]	
606.7[a]	
635.9	
671.4	
511.8	Ru - 106 + Rh - 106
621.9	
563.3	Cs-134
569.4	
604.7[a]	
795.8	
801.9	
1365.2	
661.6[b]	Cs-137
834.8	Mn-54
657.7[b]	Ag-110m
884.7	
937.5	
1173.2	Co-60
1332.5	

[a]The Sb-125 peaks at 600.6 keV and 606.7 keV are combined with the Cs-134 peak at 604.7 keV.

[b]The Ag-110m peak at 657.7 falls under the Cs-137 peak at 661.7 keV.

Preparation of the mounts as described above is expected to result in particle size segaration. Additional work to understand the problems of preparing mounts with representative samples is continuing.

C. Electron Microprobe Analysis

Forty-seven particles greater than 1 μm in diameter were analyzed for metallic element composition and size. The results of the analyses are presented in Table II. The data on particle diameter is given in units of micrometers (μm). The elemental composition is given as a normalized intensity, and is semiquantitative with an estimated accuracy of ± 20%. As an example, consider particle 20. The results give the metallic composition of the particle as being 31.2% Mg, 34.3% Si, and 34.4% Mo by weight. These results do not mean the particle contains only those elements; indeed the particle is probably largely carbon based material as indicated by the weak signal obtained. Consequently, drawing conclusions about the overall composition of the sample TMI-0104 from these data should be done with care.

Table III shows a histogram of the particle size distribution and Table IV provides a breakdown of the data in Table II according to element, number of particles, and composition.

Data from Table II indicate that the control rod alloy (80% Ag, 15% In, 5% Cd in 304 S.S. cladding) separated during the course of the accident. Silver occurs in particles with and without cadmium and indium, and cadmium and indium do not occur together in any of the analyzed particles. When silver does occur with cadmium or indium the composition bears no resemblance to control rod composition. It should be noted that the indium and cadmium x-ray lines are close together and some small amount of indium could be in the cadmium particles or vice versa; however, this effect is considered to be less than 5% in the normalized intensity.

Zirconium occurs with a wide variety of elements including the elements of the control rod, the elements of the 304 SS cladding, the elements of inconel and uranium fuel. Interestingly, zirconium does not occur as pure zirconium oxide or metal, which would give an intensity of ~100%. The highest intensity listed in Table II for zirconium is only 60.4%

TABLE II

ELECTRON MICROPROBE ANALYSIS OF TMI-0104[a]

Particle	Normalized Intensity Ratios in %												Diameter, (μm)
	Mg	Al	Si	Zr	Mo	Ag	Cd	In	Cr	Fe	Ni	U	
1	--	2.9	3.8	--	18.1	--	34.3	--	--	23.5	17.5	--	12.3
2	--	--	--	--	--	--	--	--	0.8	1.5	--	97.6	3.1
3	37.8	--	27.4	--	34.7	--	--	--	--	--	--	--	7.7
4	0.4	0.2	0.2	--	4.7	59.1	--	32.2	--	0.6	2.6	--	9.2[b]
5	38.0	--	18.4	--	43.5	--	--	--	--	--	--	--	12.6[b]
6	--	1.7	2.0	30.6	1.2	--	26.3	--	1.4	20.7	16.1	--	14.8
7	--	1.2	0.5	4.8	9.4	--	31.8	--	1.0	23.1	28.2	--	16.7
8	--	2.6	86.3	--	--	--	--	--	1.0	2.6	7.4	--	5.1
9	--	--	0.3	--	--	--	--	97.6	--	1.2	0.9	--	4.3
10	0.1	0.2	0.2	0.5	5.4	52.8	--	--	1.6	11.6	27.6	--	18.0
11	--	0.3	0.6	2.0	2.8	--	44.2	--	0.7	7.9	41.4	--	16.0
12	--	0.4	0.3	17.6	--	--	76.1	--	--	1.6	4.0	--	6.6
13	--	2.2	2.6	--	44.3	--	43.1	--	--	4.6	3.1	--	6.2[b]
14	--	--	7.0	--	18.8	--	--	43.2	6.1	14.1	--	--	19.2[b]
15	--	11.3	14.4	--	23.4	39.0	--	--	--	--	11.9	--	20.3[b]
16	--	--	99.9	--	--	--	--	--	--	--	--	--	1.7[c]
17	--	--	99.9	--	--	--	--	--	--	--	--	--	10.2[c]
18	--	0.4	0.2	47.0	--	43.1	2.2	--	--	5.4	1.7	--	5.6
19	--	7.0	6.4	--	22.4	--	28.8	--	--	17.7	17.7	--	4.2[b]
20	31.2	--	34.3	--	34.4	--	--	--	--	--	--	--	12.0[b]
21	--	0.5	0.4	--	11.2	19.4	--	18.3	6.2	11.4	32.5	--	3.1
22	--	--	0.1	--	0.7	20.8	78.4	--	--	--	--	--	6.7
23	41.5	12.4	--	--	46.1	--	--	--	--	--	--	--	10.7[b]
24	--	--	99.9	--	--	--	--	--	--	--	--	--	1.7[c]
25	--	0.3	0.2	0.9	2.5	13.9	67.8	--	1.0	4.1	9.4	--	24.8
26	--	--	--	0.4	0.7	15.6	81.7	--	--	0.4	1.1	--	6.4
27	--	0.1	--	--	1.7	--	2.7	--	0.6	84.0	10.9	--	6.7
28	--	1.6	0.6	0.8	19.7	--	65.5	--	2.3	8.1	1.4	--	9.4
29	--	0.2	--	60.4	--	16.8	--	6.5	0.7	4.1	9.4	--	7.0
30	--	--	--	--	--	--	--	--	--	0.2	--	99.8	4.9
31	--	0.3	0.1	6.0	1.3	14.1	73.8	--	0.8	2.1	1.5	--	6.4
32	--	2.1	1.0	8.8	18.6	--	37.4	--	1.0	20.6	10.4	--	2.0
33	--	1.2	0.7	--	18.5	--	51.8	--	1.8	7.6	12.5	--	4.0
34	--	5.1	4.0	7.4	15.3	--	30.7	--	3.3	15.6	18.6	--	5.6
35	--	1.2	1.4	1.5	14.2	--	27.3	--	1.5	28.1	24.8	--	9.0
36	33.0	--	60.9	--	--	--	--	--	--	6.1	--	--	9.4
37	--	1.3	0.7	3.1	8.3	--	--	27.0	4.8	35.4	7.8	11.5	4.4
38	26.6	--	73.4	--	--	--	--	--	--	--	--	--	10.0[b]
39	35.2	--	59.8	--	--	--	--	--	--	4.9	--	--	10.0
40	--	--	--	--	--	--	--	--	--	--	--	99.9	4.0
41	--	0.4	0.3	14.6	0.7	--	54.2	--	2.1	6.6	5.9	15.1	8.5
42	32.2	--	17.0	--	50.9	--	--	--	--	--	--	--	10.0[b]
43	--	--	3.4	13.1	-	--	--	--	--	5.2	3.8	74.5	9.0[b]
44	--	0.7	1.9	--	7.9	--	46.4	--	0.7	10.7	31.8	--	12.9
45	--	--	--	--	2.8	--	90.7	--	--	1.6	4.9	--	7.1
46	54.7	--	45.3	--	--	--	--	--	--	--	--	--	15.0[b]
47	--	3.6	2.3	--	15.5	--	36.2	--	3.4	16.2	22.8	--	10.0

[a]Particles less than one micrometer were ignored in this analysis.

[b]Low yield, ion exchange media.

[c]Particle moved during chemical analysis.

Particles 3,5,8,23, and 42 contained low concentrations of Ca.

Particles 8 and 13 contained low concentrations of Na.

TABLE III

PARTICLE-SIZE DISTRIBUTION FROM
ELECTRON MICROPROBE ANALYSES

Range in Micrometers	Number of Particles
1-5	********** (13)
5-10	********** (18)
10-15	********** (10)
15-20	**** (4)
20-25	** (2)

Average diameter of all 47 particles = 9.0 micrometers
Range 1.7 to 24.8. Only particles greater than 1 micrometer diameter were selected for microprobe analysis.

TABLE IV

BREAKDOWN OF ELECTRON MICROPROBE ANALYSIS ACCORDING TO ELEMENT, INTENSITY, AND NUMBER OF PARTICLES

Normalized Intensity Ratios in %	Number of Particles
Element: Mg	
<20	*(1)
20-40	*******(7)
40-50	**(2)
Element: Al	
<20	**********(21)
Element: Si	
<20	**********(17)
20-40	**(2)
40-60	**(2)
60-80	**(2)
>80	****(4)
Element: Zr	
<20	**********(13)
20-40	*(1)
40-60	*(1)
60-80	*(1)
Element: Mo	
<20	**********(22)
20-40	*****(5)
40-60	****(4)
Element: Ag	
<20	****(4)
20-40	**(2)
40-60	***(3)
60-80	*(1)
Element: Cd	
<20	**(2)
20-40	*******(7)
40-60	****(4)
60-80	*****(5)
>80	**(2)
Element: In	
<20	**(2)
20-40	**(2)
40-60	*(1)
60-80	
>80	*(1)
Element: Cr	
<20	**********(16)
Element: Fe	
<20	**********(24)
20-40	****(4)
40-60	
60-80	
>80	*(1)
Element: Ni	
<20	**********(26)
20-40	******(6)
Element: U	
<20	**(2)
20-40	
40-60	
60-80	*(1)
>80	***(3)

Six particles were analyzed that contained uranium. Three of the particles appear to be pure uranium oxide. The other three particles contain uranium with various other elements; but all three particles contain, U, Zr, Fe, and Ni. These data are not sufficient to confirm or reject the formation of a eutectic between the zircaloy-4 clading and the uranium fuel.

Aluminum is present in 21 of the 47 particles analyzed; however, it never has an intensity of >5.1% except on ion exchange media particles. If pieces of the burnable poison (pelletized Al_2O_3-B_4C) were present in the sample, there should be particles with near 100% intensity for aluminum. This lack of particles with high aluminum content could be due to the limited sample examined or reflect the limited amount of burnable poison material in the reactor core. It could also indicate that thermal shock and mechanical shock during the accident were not sufficient to fracture the material to produce micrometer-size particles.

Several particles of ion exchange media were present in the sample. These particles gave low yields of x-rays and generally contain large amounts of magnesium, silicon and molybdenum. The magnesium and silicon come from water clean up and the molybdenum, which is apparently soluble, is from inconel in the core.

Since only particles of >1 μm were sized, the data presented in Table III is obviously shifted toward higher average diameter; however, this information may be useful in determining if certain elements occur more frequently in certain particle sizes. From the limited data of this analysis, there is no obvious composition-size correlation.

D. Results of Particle-Size Analysis Using an Image Analyzer

Particle-size results will be of limited value for the sample TMI 0104 since the sample has been homogenized and it is not clear how seriously this operation altered the particle-size distribution. However, the sample was analyzed with the intentions of focusing mainly on the development of techniques that would be used on future samples.

Particle sizing was done with a Zeiss IM-35 metallograph coupled to a Hamamatzu image analyzer. Preparation of the samples for sizing is described in the section "Preparation of Samples for Electron Microprobe and Image Analyzer Analysis." It should also be recognized that these samples are probably not representative of the true sample.

Results of the particle-sizing effort are presented as a histogram in Fig. 3. The distribution appears to have a median particle diameter of ~3 μm. No effort has been made to determine the type of distribution, i.e. normal, log-normal, etc. One thing does become clear from the results in Fig. 3, and that is the very large number of particles that can be sized compared to other methods such as the electron-probe. The image analyzer should provide the best possible particle-size distribution data.

To date problems of dispersing the powdered samples and obtaining acceptable mounts for particle sizing have been solved. Present efforts are being directed at determining the number of sample areas that must be examined to get accurate size distribution results.

E. Division of TMI Samples Using Spinning Rifflers

Division of the powdered TMI samples into fractions that are of appropriate mass for analysis will be done using one or more spinning rifflers of appropriate size. Spinning rifflers were chosen because they satisfy the two principal requirements for sampling powders, i.e. sample a moving powder stream and sample frequently for short intervals to obtain a combined single sample.[1] At present a single spinning riffler has been fabricated. The device will handle samples of ~10 g or less.

Using this riffler, the ~225 mg of sample TMI-0104 was divided into 10 cuts. Three of these cuts, 2, 6, and 9 were dissolved and used for gamma spectroscopy analysis. Other cuts will be used for continued particle-size analysis methods development. The three cuts which were analyzed showed no discernable difference in composition indicating that the spinning riffler preformed properly.

F. Gamma Spectroscopy of Sample TMI 0104

Three of the 10 sample cuts of sample TMI-0104 were dissolved in concentrated HCl using the sealed-tube dissolution method described in Ref. 2. After dissolution, examination of the sealed tubes showed only pieces of filter paper and crystals of silver chloride. After transfer of each solution to 100-mL volumetric flasks some very small black particles could be seen. This may be carbon contamination from the dissolution procedure. The amount of black material in each flash was estimated to be less than a tenth of a milligram.

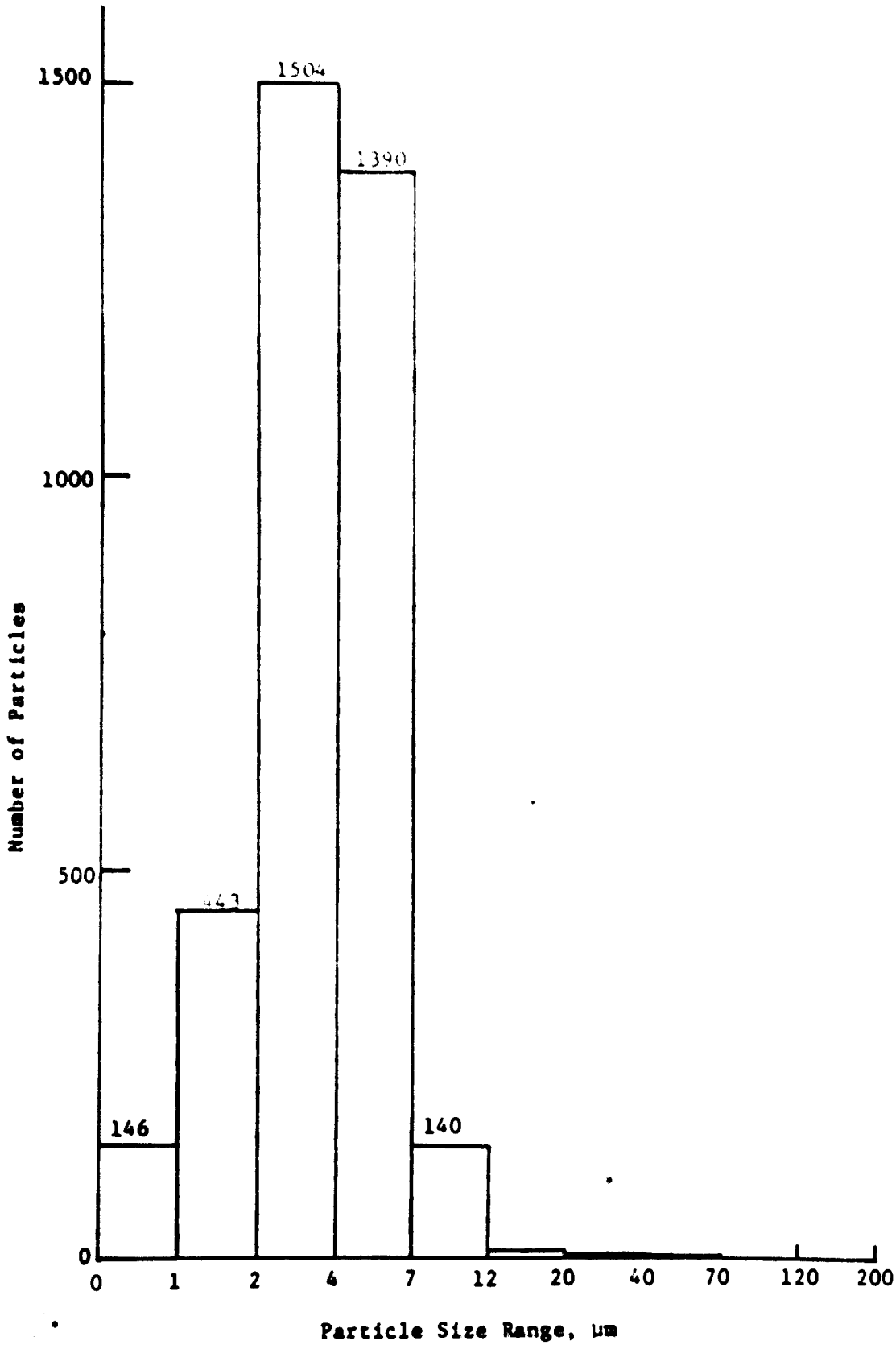


Fig. 3. Results of image analysis of TMI-0104 for particle size distribution.

The solutions were diluted to 100 mℓ and 2 mℓ of the dilution were analyzed using a Ge(Li) detection and a Camberra Series 85 multichannel analyzer. Table V contains the results of the analyses. The analyzer system was calibrated with a mixed isotope standard prepared by Amersham (product code QCY.44). Both the energy calibration and efficiency were determined. For calculations, gamma-ray yields were taken from the Nuclear Data Tables, Vol. 8, Sec. A, No. 5-6, January 1971.

The results of the analyses generally agree very well with analyses performed by B&W and INEL. The only unexplained discrepancy is the result for Ce-144 where the value obtained in these analyses is ~20% higher than results from the other two laboratories. The Ag-110m analysis is low for the obvious reason that AgCl precipitated from the solution of HCl.

III. CONCLUSION

Four important aspects of sample handling have been worked out:

(1) division of the samples to give representative fractions using the spinning riffler;
(2) dispersion of powdered samples using an insufflator;
(3) collection of dispersed particles on mounts suitable for particle-size analysis and probe analysis; and
(4) dissolution of the samples using the sealed tube method.

All of the methods are crucial to sample analyses. All of the methods should be applicable to future samples with little or no modification.

Particle-sizing procedures are still being evaluated with the objective of defining the minimum number of sample areas that must be examined to ensure that a representative size distribution is obtained for samples. The data that are presented in this report are not considered representative of the true distribution of sample TMI-0104 but are intended to show the type of data that will be available from future samples.

Electron microprobe data show that particles may have either simple compositions, such as uranium oxide, or complex compositions containing nine or more elements. The elemental ratios in the particles appear to bear little or no resemblance to the elemental ratios in the parent alloy. The most obvious example of this is the alloy of silver, indium, and cadmium which make

TABLE V

GAMMA SPECTROSCOPY ANALYSIS OF SAMPLE TMI 0104

Isotope	Sample Number[a]	Specific Activity (μCi/g)[b]	Average Specific Activity (μCi/g)[c]
Mn-54	2	8.0	
	6	9.0	9.0 ± 0.6
	9	10.0	
Co-60	2	222.7	
	6	240.6	228.7 ± 6.0
	9	222.7	
Ru-106	2	289.2	
	6	307.5	313.6 ± 16.2
	9	344.1	
Cs-134	2	259.5	
	6	266.7	266.7 ± 4.2
	9	273.9	
Cs-137	2	3540	
	6	3730	3606 ± 62
	9	3748	
Ce-144	2	527.6	
	6	573.6	550.6 ± 13.0
	9	550.5	
Sb-125	2	612.5	
	6	652.8	637.2 ± 12.5
	9	646.4	
Ag-110m	2	6.77	
	6	8.10	7.4 ± 0.5[d]
	9		

[a]The sample number refers to the cut of the sample taken for analysis (see Sec. II.E.).

[b]The ± values are estimates of the standard deviation of the mean.

[c]The specific activities are for 10/29/82.

[d]Silver chloride crystals formed in the samples and resulted low values for Ag-110m.

up the control rods. Cadmium and indium do not occur together in any of the particles. Silver occurs by itself and with cadmium or indium. This separation of silver cadumium and indium is evidence that the control rod alloy separated before the stainless steel cladding failed. Also, the lack of particles with high aluminum content may indicate that the burnable poison pellets of Al_2O_3-B_4C did not fail under the thermal and mechanical shock of the accident.

Gamma-spectroscopy of dissolved sample gave no unexpected results and generally agree with previous analyses by B&W and INEL.

References

1. T. Allen, Particle Size Measurement, Chapman and Hall Ltd., second edition, 1975.

2. C. F. Metz, G. R. Waterbury, "Sealed-Tube Dissolution Method with Applications to Plutonium-Containing Materials," Los Alamos National Laboratory report LA-3554 (July 1966).

APPENDIX D

PRELIMINARY RESULTS FROM THE ANL-E EXAMINATION OF TMI-2 FILTER SAMPLES

By

R. V. Strain

APPENDIX D

PRELIMINARY RESULTS FROM THE EXAMINATION OF TMI-2 FILTER SAMPLES

Five samples of material from TMI-2 makeup/letdown filters have been received at the Alpha-Gamma Hot-Cell Facility (AGHCF) at ANL-E for examination. The purpose of the examination is to characterize the debris from the filters by size, shape, and chemical composition. The planned examinations include:

- Visual examination of the samples
- Optical microscopy of mounted samples
- Scanning electron microscope examination of the samples
- Electron microprobe analysis of mounted samples
- Gamma spectroscopy of the samples

Preliminary examinations consisting of visual examination and optical and SEM microscopy have been performed on material from sample MUP-5B; and visual examination and qualitative gamma spectroscopy have been performed on a sample from MUP-2A. Preliminary results from these initial examinations are described below.

Visual Examination

Visual examination of samples from MUP-5B and MUP-2A, in the hot-cell showed them to consist of irregular rectangular pieces of fibrous material and fine particles. The pieces of fibrous material were thin slabs that varied in size from ~1/16 in. square to pieces 1/4 x 3/4 in. Visual examination of a small piece of the fibrous material in a glovebox indicated that it was composed of layers of the fine particles on an underlay of the fibrous filter material. The filter material itself appeared to be a fiberlgass mat.

SEM Examination

A piece of the filter material (~0.05 x 0.4 in.) and several of the small particles were placed on a sample stub using silver paint as an adhesive. This sample was vapor-coated with Au-Pd, after initial viewing in the SEM was hindered by electrical charging of the sample.

The piece of filter material that was placed on the sample stub is shown in Fig. 1. This piece will be referred to as the "large piece of filter material" in this description of the SEM examination. Several much smaller pieces of filter material and several clusters of particles were also included in this sample. The larger piece of filter material as shown in Fig. 1 appears to be a cross-section through the filter. A very high density of particles is present along the edge of the large piece toward the upper-left of the photo. The number of particles decreases fairly rapidly away from this edge, and very few particles are present in two thirds of the piece of the material toward the lower-right of the photo.

A typical area near this upper edge of the large piece of filter material is shown in Fig. 2. The large (sometimes angular) pieces of material and the strands shown in this photo are believed to be the filter itself. Non-dispersive X-ray analysis showed that the strands (fibers) contained silicon and they are assumed to be glass. The large, medium-gray, pieces produced no characteristic X-ray spectrum and are assumed to be low atomic number materials, probably a resin used to bond the fiberglass. The smaller particles are materials that were present in the TMI-2 core at the time of the accident. The photo in the upper-left of Fig. 3 shows the X-ray spectrum from the field shown in Fig. 2. The materials easily identified in this spectrum are listed in Table 1. The broad peak at ~3 Kev is composed of Ag, Cd, In, and possibly U. The other photos in Fig. 3 show X-ray spectra from the particles that are marked in the bottom photo in Fig. 2. The captions under these spectra indicate the most prominent element found at that particular location. Particle A was considered unique because of its high chromium content. Particle B appeared to be primarily silver, but the close proximity of the X-ray peaks for Ag, Cd, and In make the identification of the individual elements imprecise. The clear identification of silicon in the fiber and the strong presence of silicon in the fuel field spectrum indicate that the filter was constructed of fiberglass.

Another typical area of the large piece of filter material is shown in Fig. 4. This area is adjacent to the area shown in Fig. 2, but is closer to the upper-edge of the piece of filter. The number of particles per unit area is higher than that in the first area. The X-ray spectra of this field and individual particles are shown in Fig. 5. Again the materials present represent the control material (Ag, Cd, In), the fuel-rod cladding (Zr), control-rod cladding (Cr, Fe, Ni), and the silicon in the fibers. No uranium was apparent, based on the lack of prominent U-Lα peak at 13.6 Kev, but the level of detection is eroded by the presence of the control-rod materials.

An area at the upper edge of the large piece of filter material is shown in Fig. 6. In this area, almost none of the filter material itself is visible. This may represent a typical cross-section of the layer of particles that were deposited on the filter. A round particle (a) in the lower center of this photo was found to be high in U as shown by the X-ray spectrum shown in the lower photo in Fig. 6. The peak at 13.6 Kev was used to identify this particle as containing U. The other round particles were composed of the control material (Ag, Cd, In). The other particles were either high in zirconium or stainless steel components (Fe, Ni).

Another typical area of the large piece of the filter material is shown in Fig. 7. The full field spectrum shows that the particles in this photo are primarily composed of the control material and stainless with a smaller peak for Zr. The small round particle in the center of the photo appeared to be silver, and the relatively large rectangular particle was Zr. The medium gray background material is a low atomic number material.

A small piece of filter material is shown in Fig. 8. The round particle is primarily Ag, while the angular particles are mostly iron and nickel.

Individual clusters of particles are shown in Figs. 9, 10, and 11. The particle at the bottom center of the cluster in Fig. 9 was high in uranium. The other particles were high in Zr or Fe and Ni. The round particle in Fig. 10 was high in Ag, Cd, and In with the flake-like particles being high in Zr.

Some general impressions from this SEM examination are:

- The particles on this piece of filter were primarily stainless steel, zirconium, and Ag-Cd-In.
- A few particles were identified positively as containing uranium but uranium appeared to be a minor constituent. These results are somewhat uncertain because of the interference in the U Mα peaks.
- The filter itself appeared to be made of fiberglass.
- The control material (Ag-Cd-In) was found in the form of spherical particles (2 to 4 microns in diameter). In some cases the material had partitioned and the particles appeared to be pure Ag or Cd.
- The zirconium was in the form of either relatively large (2 microns) rectangular particles or small flakes.
- The stainless steel was generally found as very small (1 to 2 microns) rectangular particles.

One should keep in mind that this examination was of a 0.05 x 0.04 in. sample from the MUF-5b filter. Such a small sample may not be representative of other parts of the filter. Silver paint was not the best choice of an adhesive for this sample since it may be confused with the silver from the control rods in some cases. The presence of the silver paint was kept in mind when analyzing the spectra from various areas. In the future, an electrically conductive adhesive containing graphite rather than silver will be used.

Optical Microscopy

Two samples containing material from the MUF-5B filter were prepared for optical microscopy. One sample was prepared by placing a piece of the filter material (~1/4 x 1/2 in.) on edge in a standard metallographic mount using epoxy resin. The second sample was prepared by breaking-up a small piece of the material and mixing it in epoxy resin and then pouring that resin into a metallographic mount. These samples were then prepared using standard grinding and polishing practices.

A low magnification of the first sample (piece of filter) is shown in Fig. 12. It has not been determined whether the piece of filter represents a cross-section through the entire thickness of the filter or whether only a layer of the filter is present in the mount. Higher magnification photographs of this piece are shown in Figs. 13, 14, and 15. This sample shows that the debris is present on the filter in layers. In general the larger particles were found along the first layer near the top of the low magnification photo, but occasionally the larger particles are found elsewhere in this mounted sample. The larger particles range in size from 10 to 50 microns and appear

to consist of both metallic and ceramic materials. The smaller particles (1 to 5 microns) are found dispersed throughout the other layers in this mount. The filter material itself does not appear as a prominent feature in the optical microscopy of this sample. Apparently both the fibers and the rest of the filter have optical properties similar to the epoxy resin that was used as the mounting media. A resin-bonded fiberglass filter could be expected to react in this way to optical microscopy.

The second sample (broken-up piece of filter) is shown at low magnification in Fig. 16. The higher magnification photos of various locations on this mount (Figs. 17 and 18) primarily show clusters of the smaller particle. It is probable that the SEM sample consisted of a piece of filter that is shown as one of these clusters of small particles.

Some general impressions from the optical microscopy are:

- The method of sampling the filter material may have a significant influence on interpretation of the results. There were many more large particles in the sample where a relatively large piece of sample was mounted, as compared to the sample where the pieces of filter material were broken up.

- These large particles were either not present or not recognized in the SEM sample.

- The various materials from the core of the reactor may have characteristic shapes. The identification of the material in these samples will be pursued using the SEM and electron microprobe.

- Both ceramic and metallic particles are present on the filter.

Gamma Spectrometry

Several small pieces of the MUF-2A filter were placed in a small plastic vial for gamma spectrometry. The vial was placed near the GeLi crystal of the AGHCF gamma spectrometer. The sample vial was counted with no shielding or collimation. The gamma spectrum that was obtained during this counting period is shown in Fig. 19. The fission products that were identified from the spectrum were ^{144}Ce, ^{134}Cs, ^{137}Cs, ^{95}Nb, ^{144}Pr, ^{106}Rh, ^{125}Sb, and ^{95}Zr. This spectrum appears to be about what would be expected of fission products with 2 1/2 to 3 years decay time. The only activation product that was found was ^{60}Co.

Table I. Elements Present in the X-Ray Spectrum Shown in Figure 2

Element	Characteristic X-ray Energies					
	K_{α_1}	K_{α_2}	K_{β_1}	L_{α_1}	L_{α_2}	L_{β_1}
Silicon	1.740	1.739	1.832			
Chromium	5.414	5.405	5.946			
Iron	6.403	6.390	7.057			
Nickel	7.477	7.460	8.264			
Zirconium				2.042	2.040	2.124
Silver				2.984	2.978	3.151
Cadmium				3.133	3.127	3.316
Indium				3.287	3.279	3.487
Uranium	3.171*	3.337*		13.613	13.438	

M_{α_1} and M_{α_2} Lines.

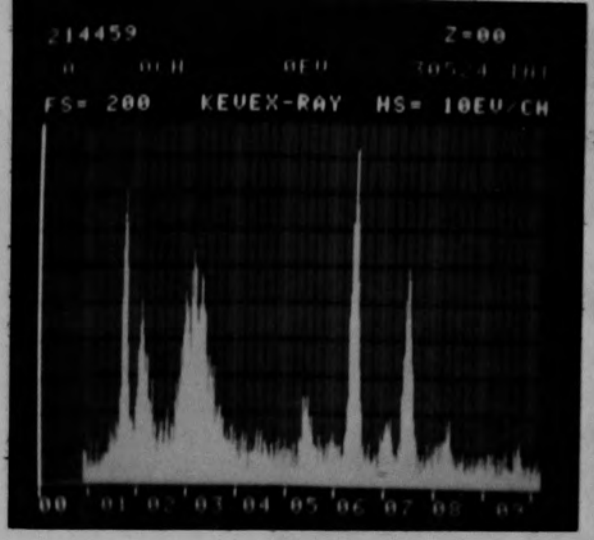
Large piece of filter material, 50X

Fig. 1.

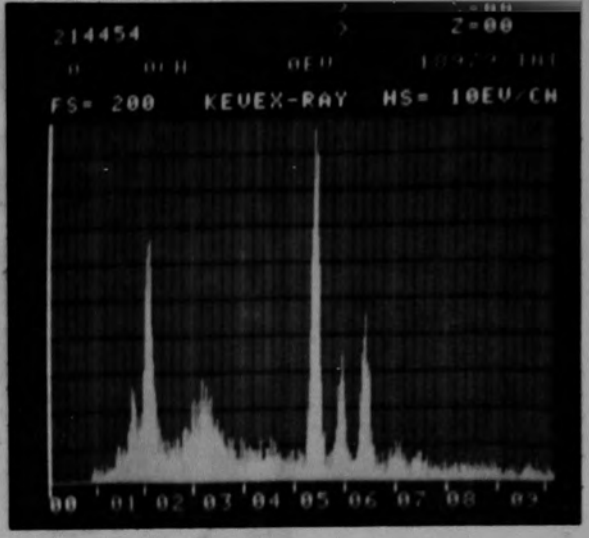
First typical area of large piece of filter, 1400X

Same area with the particles that were analyzed marked, * 1400X

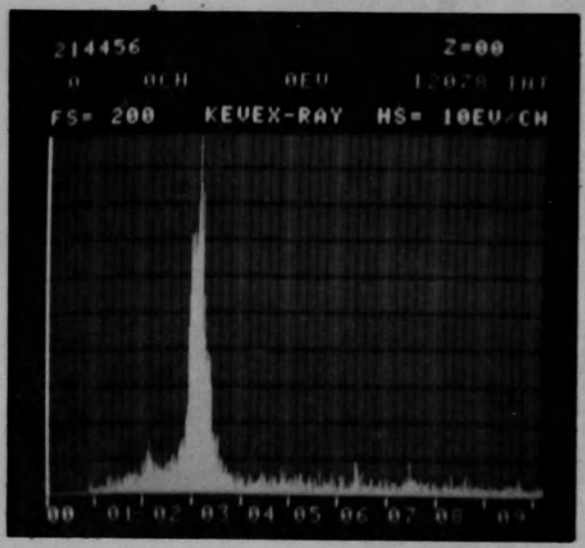
Fig. 2.



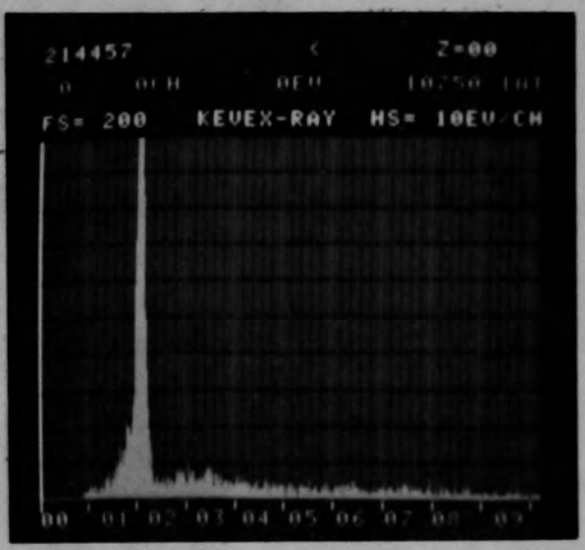
Full Field



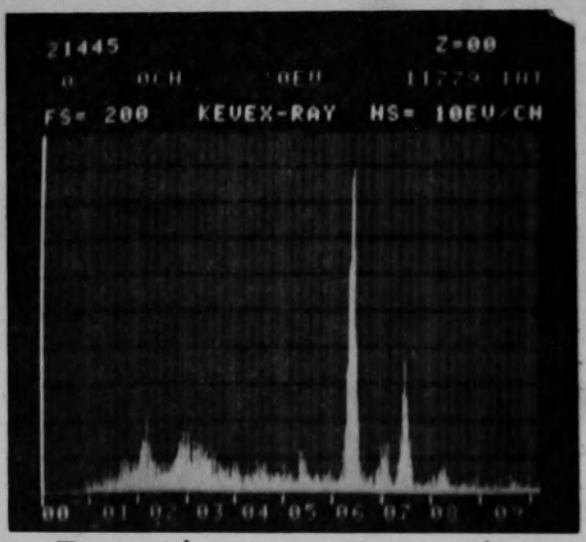
Particle A, Cr



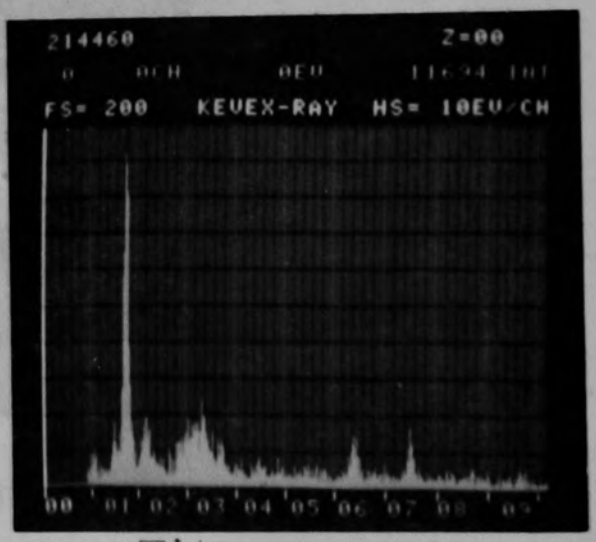
Particle B, Ag



Particle C, Zr



Particle D, Ni



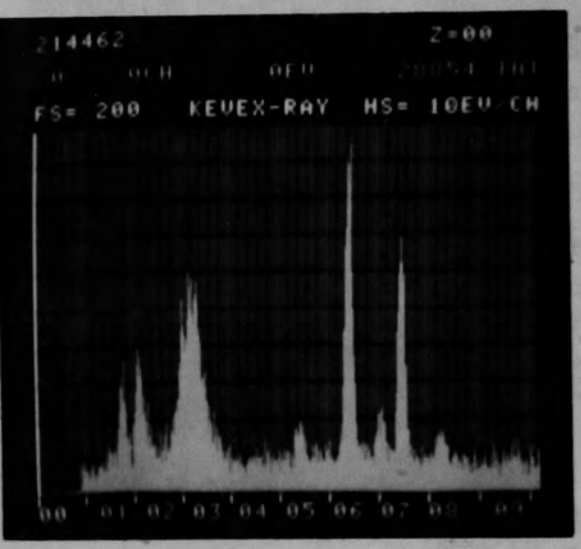
Fiber, Si

Fig. 3.

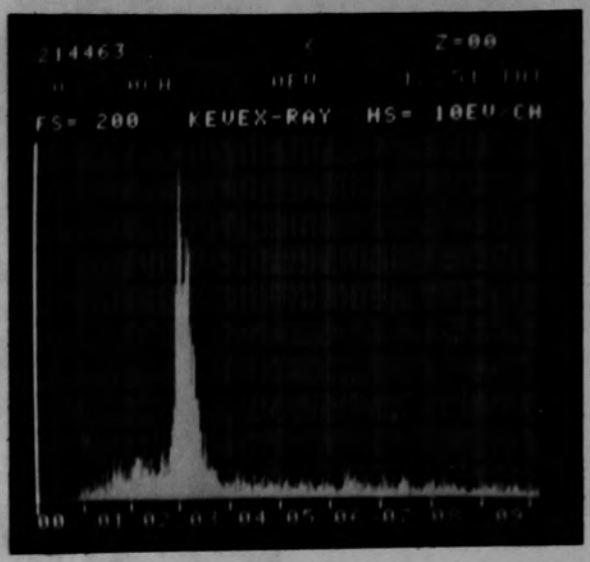
Second typical area of large piece of filter, 1400X

Same area with the particles that were analyzed marked, 1400X

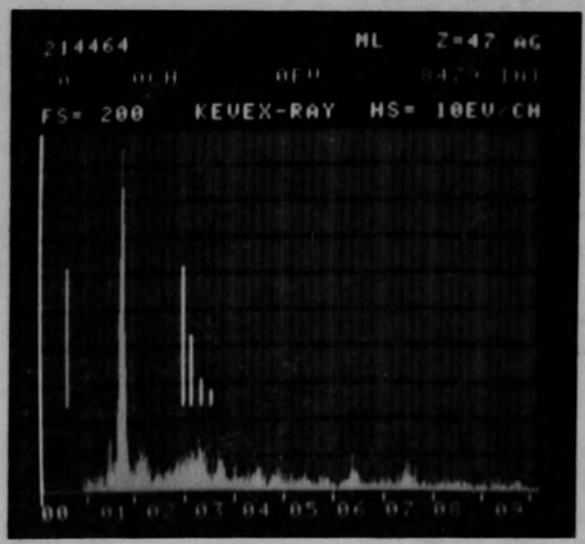
Fig. 4.



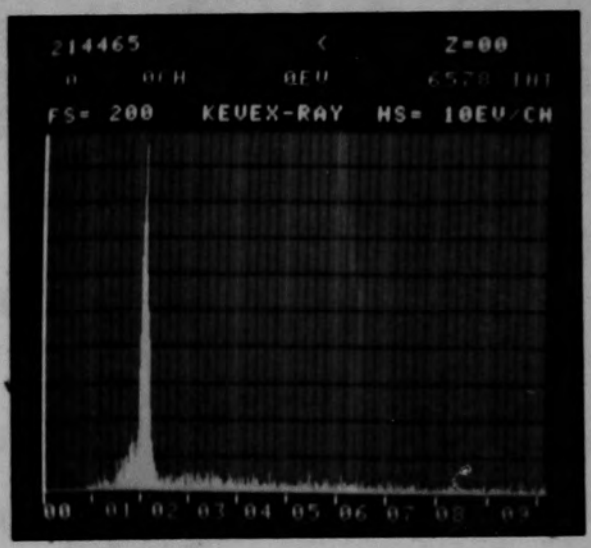
Full Field



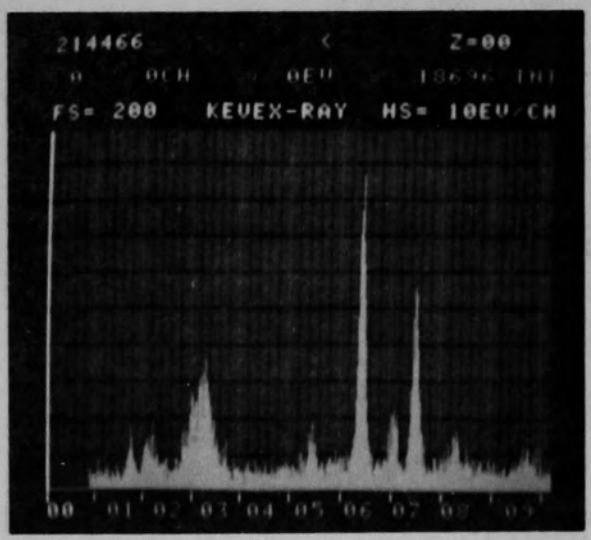
Particle A, Ag,Cd,In



Fiber, Si

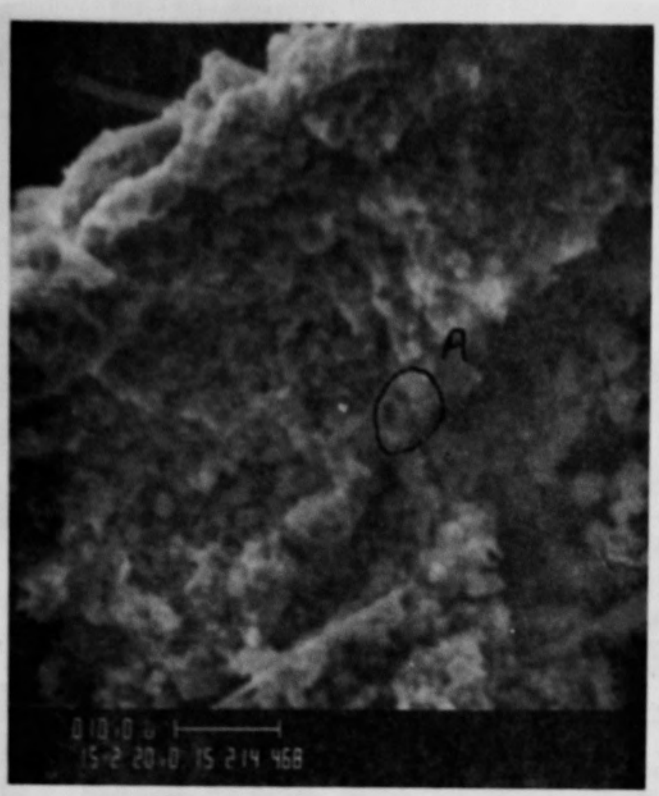


Particle C, Zr

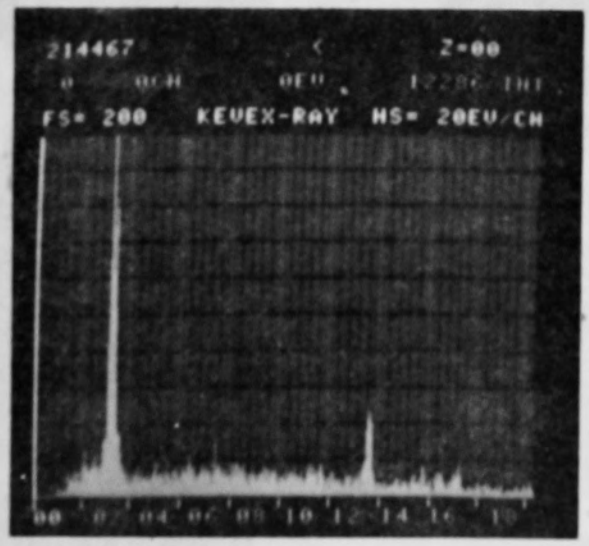


Particle D, Fe,Ni

Fig. 5.



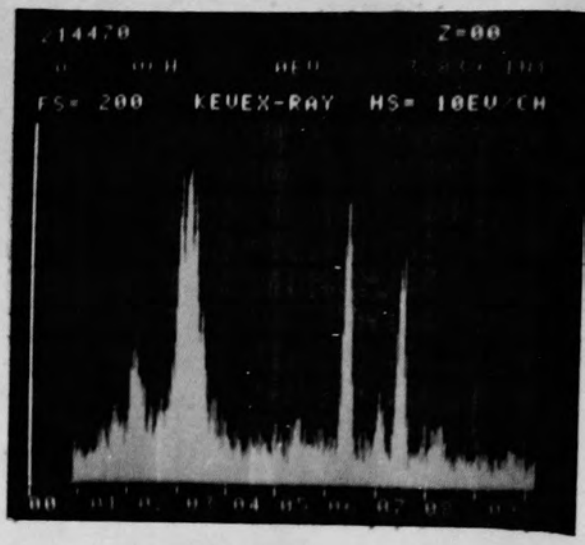
Area near the edge of the large piece of filter material, 1500X



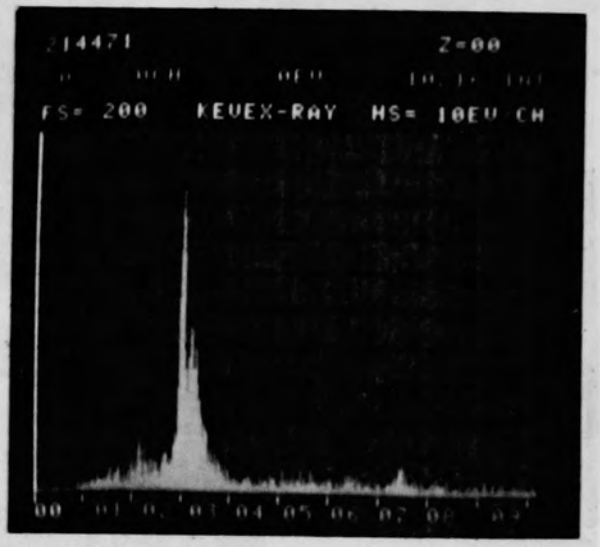
Particle A, high U

Fig. 6.

Area on the large piece of filter material, 2500X



Full field



Particle A, Ag

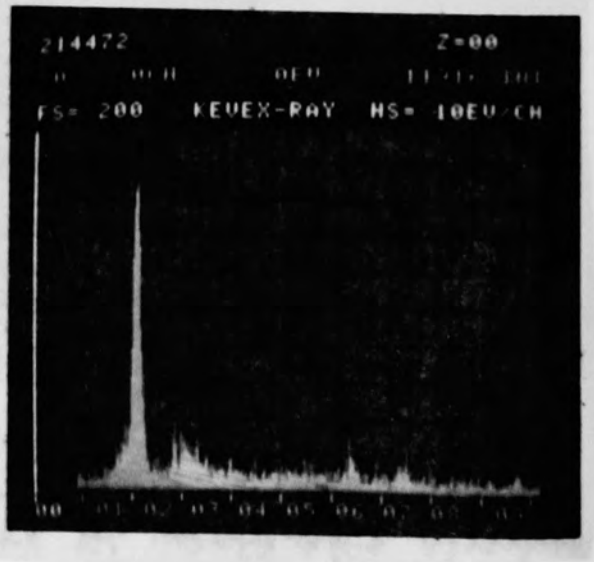
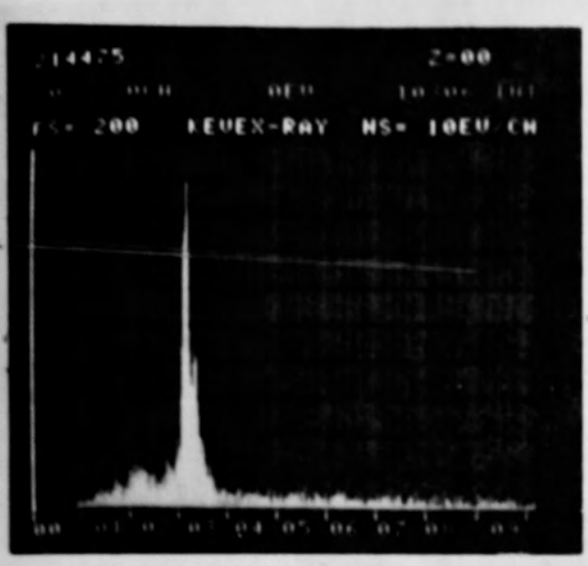
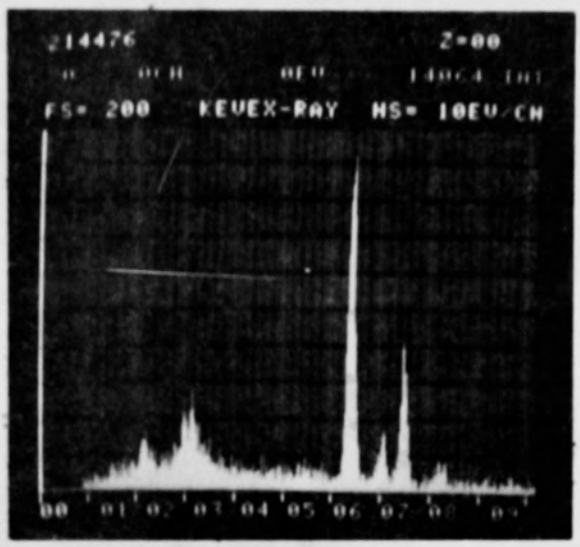


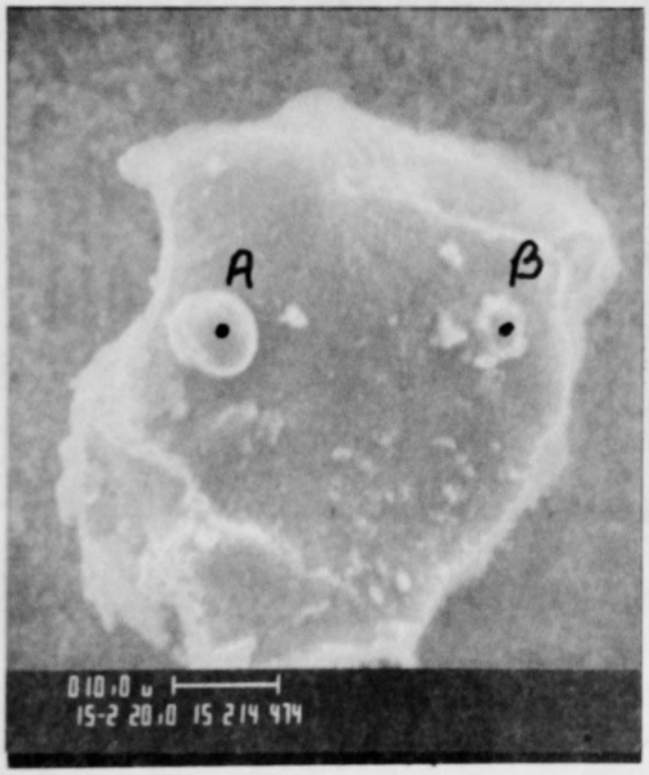
Fig. 7.



Particle A, Ag

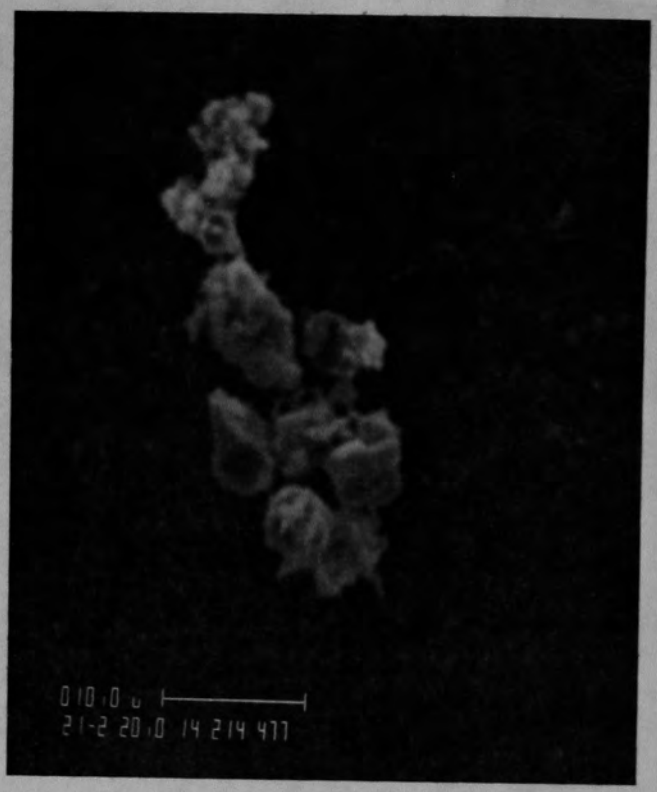


Particle B, Fe, Ni



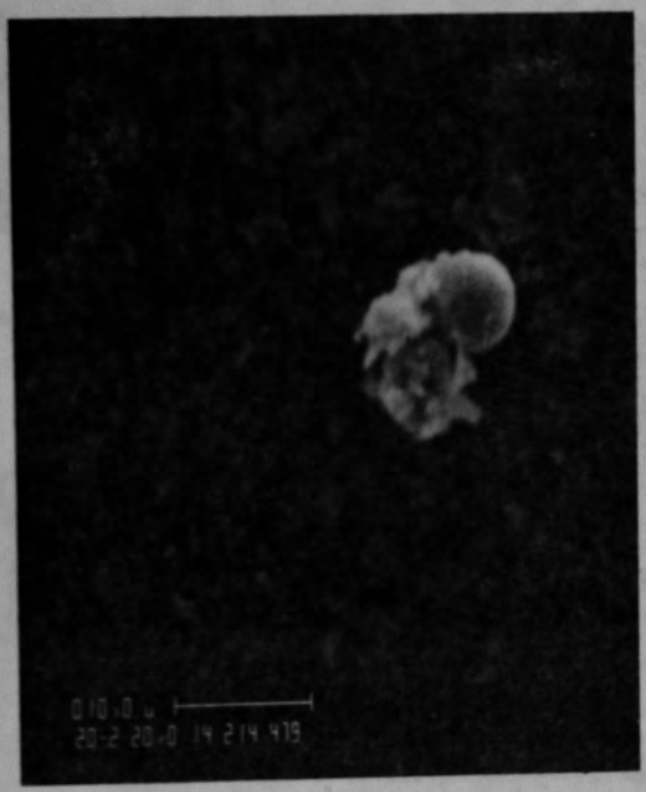
Small piece of filter material

Fig. 8.



Cluster of particles; high U. 1900X

Fig. 9.



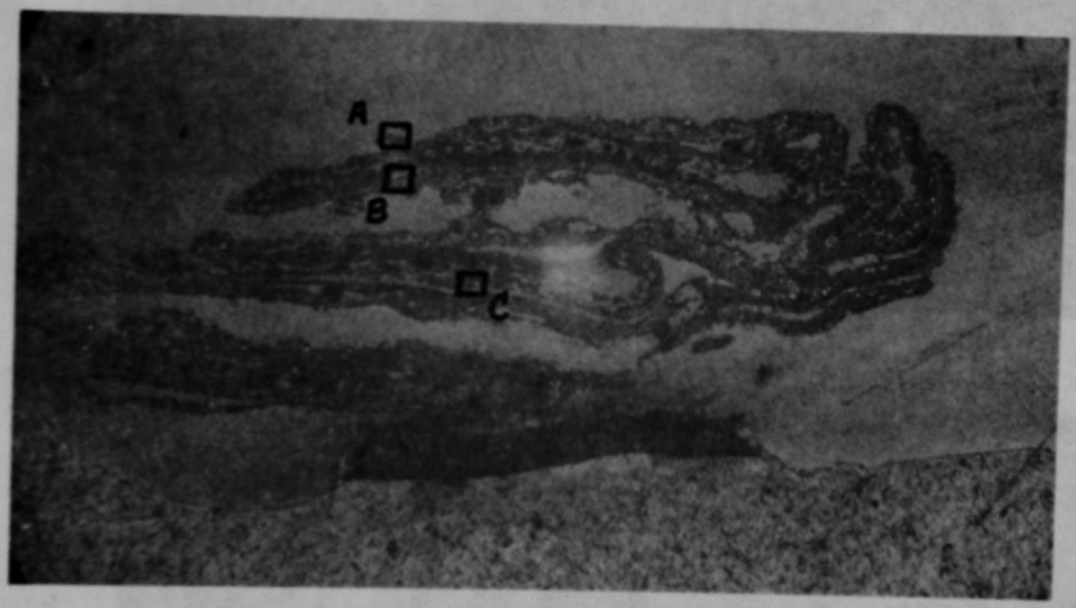
Cluster of particles; the round particle is high in Ag, Cd, In. 1800X

Fig. 10.

Cluster of particles; high Zr. 1800X

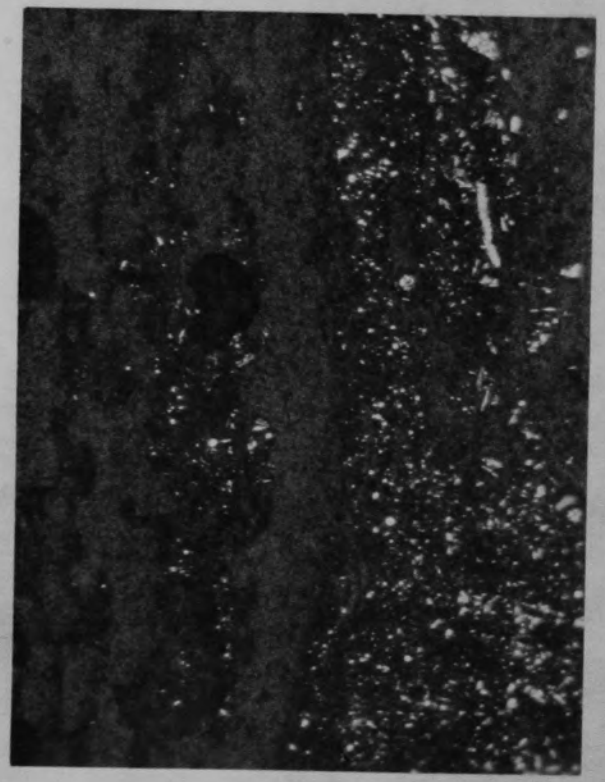
Fig. 11.

Mounted Piece of Filter Material from Sample MUF-5B. Neg. Neg. MSD-229864.



Piece of Filter Shown Above with the Area of Three of the Higher Magnification Photos Identified. Neg. No. MSD-229864.

Fig. 12

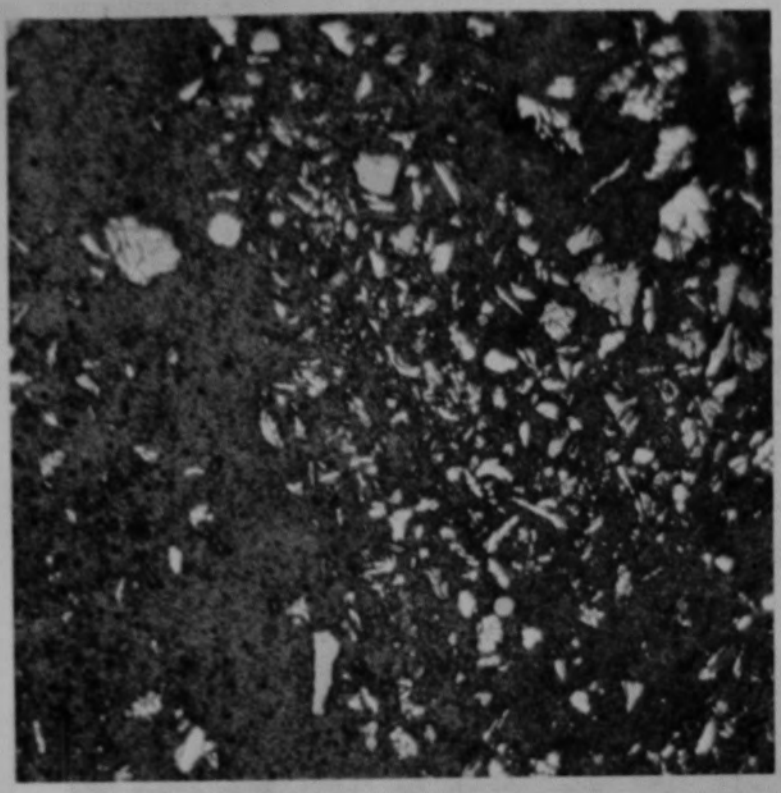


Neg. No. MSD-229875.

Neg. No. MSD-229874.

Neg. No. MSD-229873.

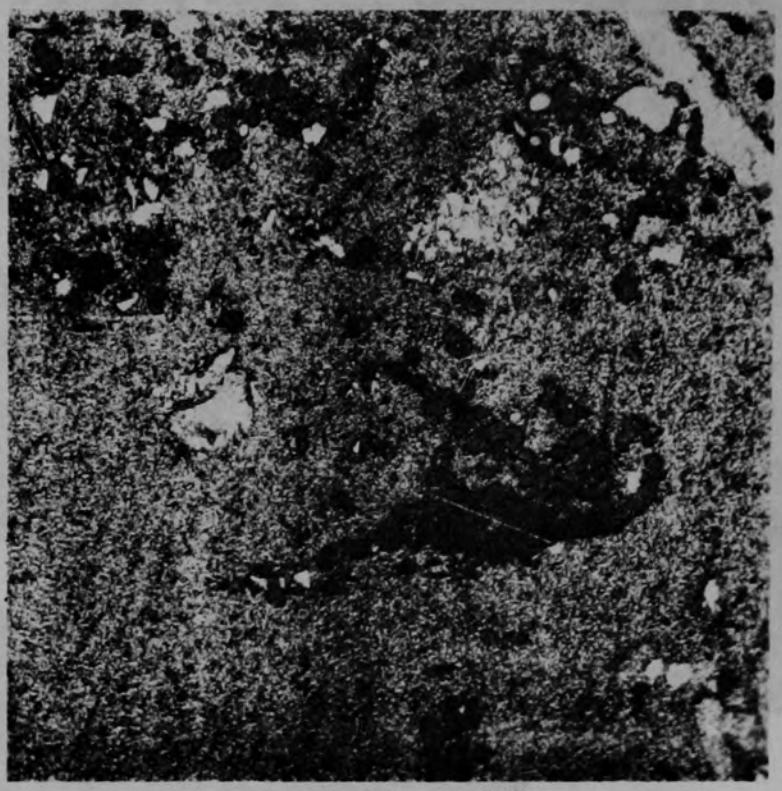
Fig. 13. Typical Areas of the Mounted Piece of Filter at Higher Magnification. 250X.



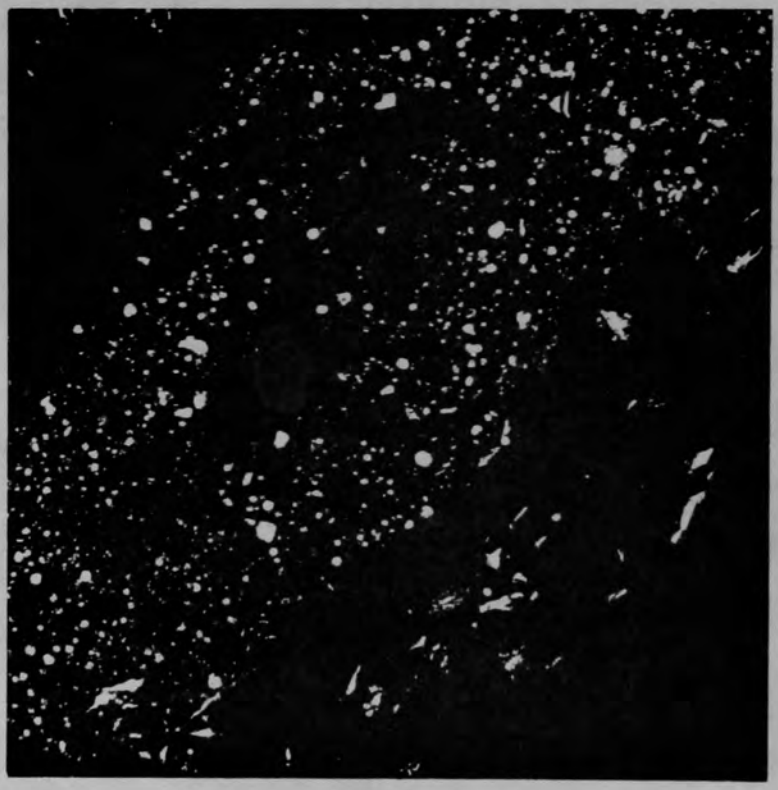
Neg. No. MSD-229868.

Neg. No. MSD-229870.

Fig. 14. Areas of the Piece of Filter Material with High Concentrations of Particles. 250X.



Neg. No. MSD-229869.

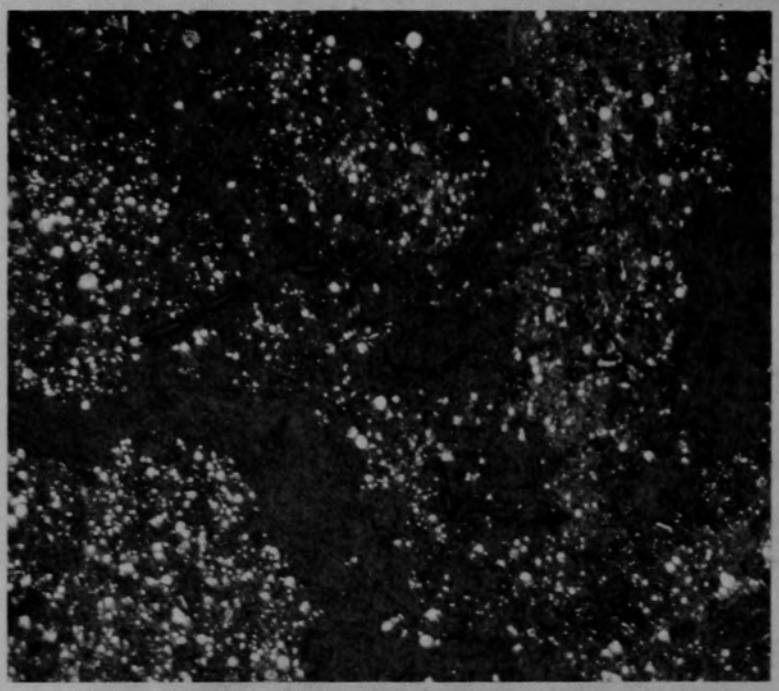


Neg. No. MSD-229872.

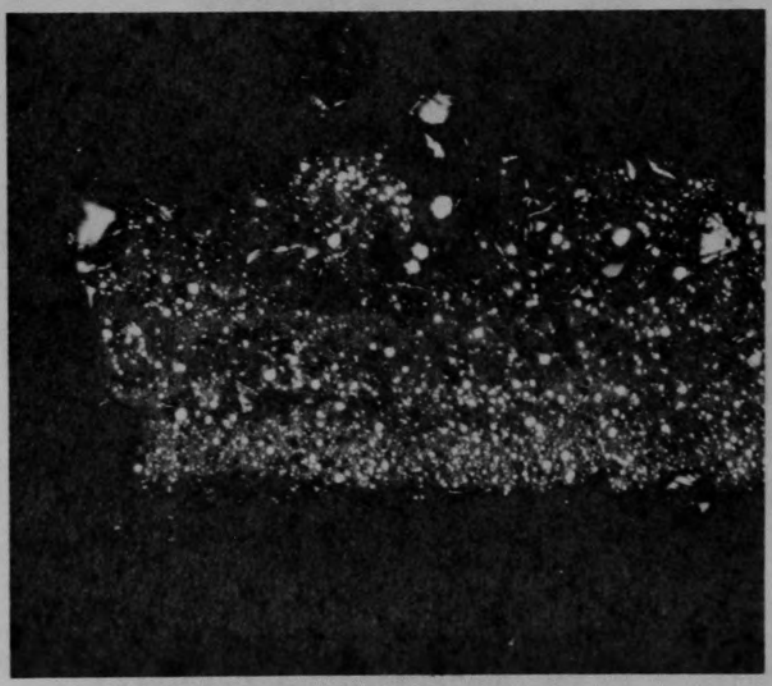
Fig. 15. Other Areas of the Mounted Piece of Filter. 250X.

Neg. No. MSD-229876.

Fig. 16. Low Magnification Photo of the Broken-up Piece of Filter Material from Sample MUF-5B. 10X. Neg. No. MSD-229876.

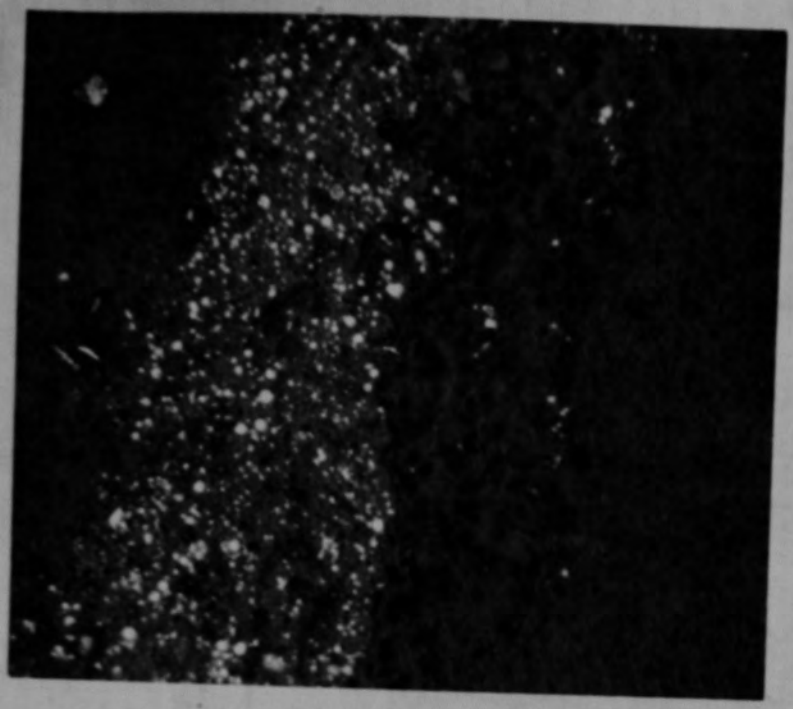


Neg. No. MSD-229884.

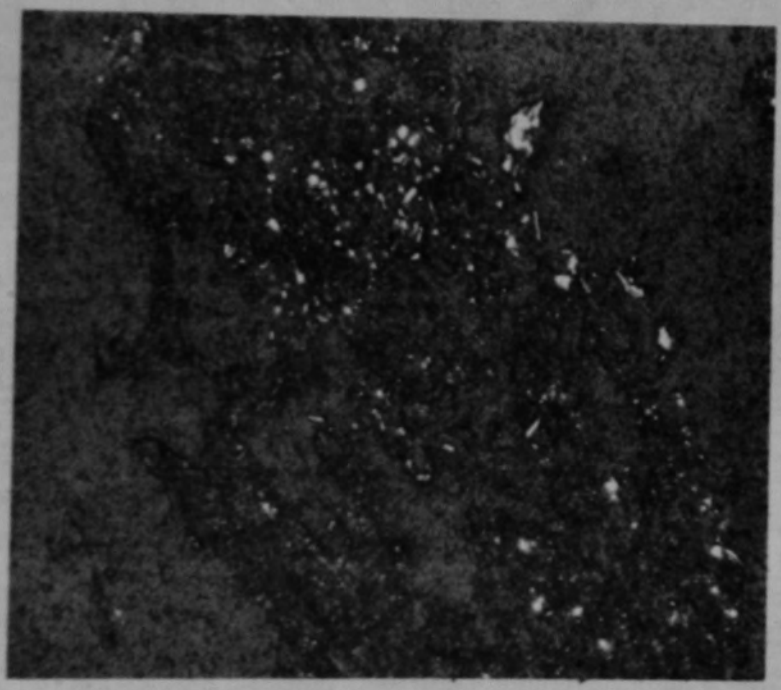


Neg. No. MSD-229882.

Fig. 17. Areas of the Broken-up Piece of Filter with High Concentrations of Debris.



Neg. No. MSD-229877.



Neg. No. MSD-229883.

Fig. 18. Other Areas from the Mount Continuing the Broken-up Filter Material.

11-29-82

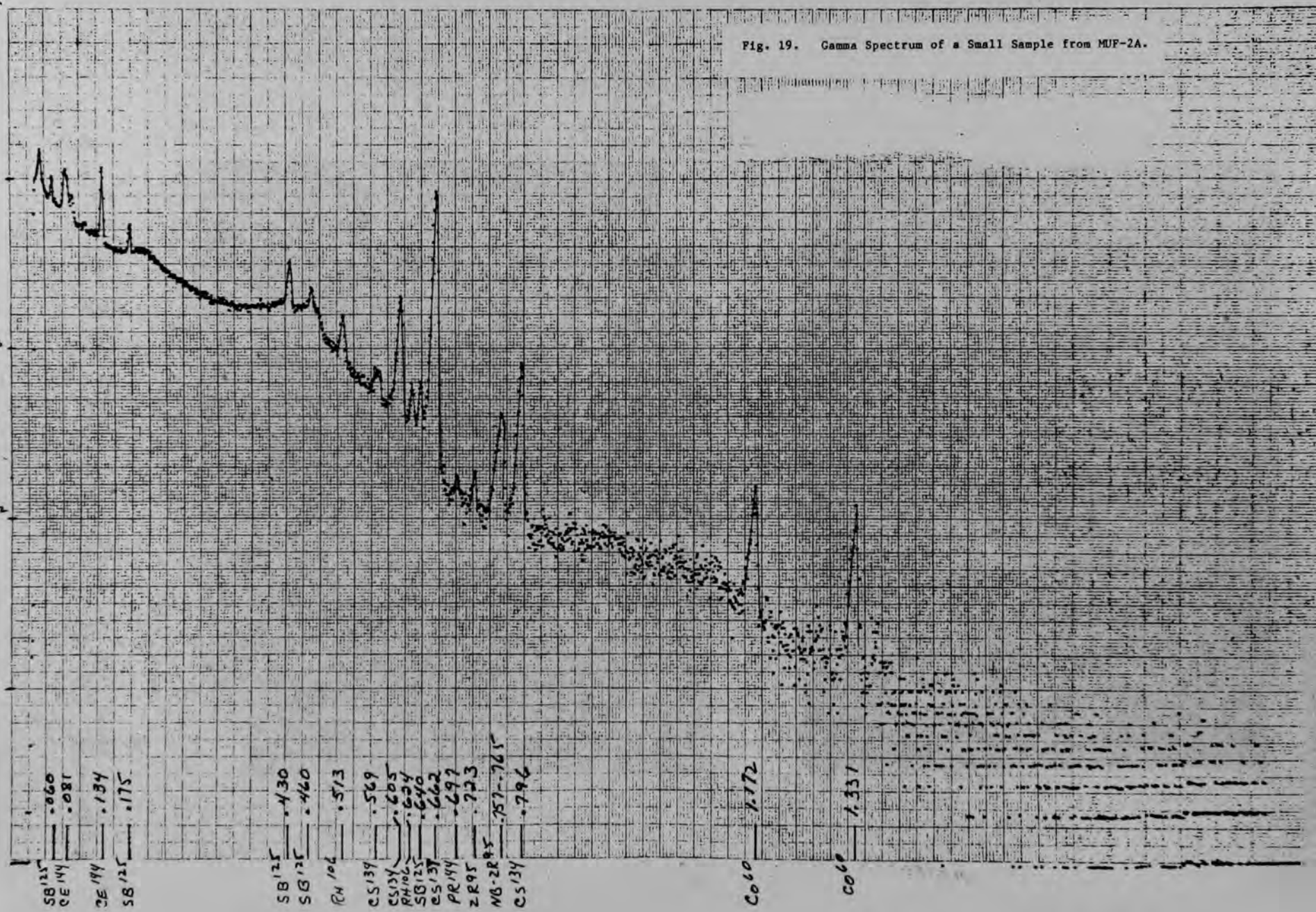


Fig. 19. Gamma Spectrum of a Small Sample from MUF-2A.

Fig. 19. Gamma Spectrum of a Small Sample from HRP-24.