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TMI-2 CORE DEBRIS GRAB SAMPLE QUICK LOOK REPORT

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Operated by the U.S. Department of Energy



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April 16, 1984

To: All holders of EGG-TMI-6531, Revision 1, TMI-2 Core Debris Grab Sample Quick Look Report, March, 1984

From: EG&G Idaho

ERRATA

Please make the following corrections to your copy of the subject Quick Look Report:

Page 26 Table 1

Sample weight for sample no. 1 should read "74" Sample weight for sample no. 5 should read "95"

EGG-TMI-6531 Revision 1

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SUMMARY

Examinations are being performed to acquire data on the extent and nature of the damage and the post-accident condition of the TMI-2 core. One of the tasks is the examination of the six core debris grab samples acquired during the fall of 1983.

Preliminary examination of five of the core debris samples has been completed at the INEL. This examination consisted of photo-visual and weighing of the as-received bulk samples. Additionally photo-visual, weighing, gamma spectroscopy and fissile determination were performed on eight select particles removed from these samples. This report presents and discusses the results from these measurements. Further analyses, primarily chemical and elemental, will be performed in FY-1984.

Observation of the debris samples showed fuel pellet fragments, shards of cladding or guide tubes, previously molten material, and fuel pellet surfaces glazed with previously liquified material. Five of the eight particles were composed primarily of fuel. Radionuclides 144 Ce, 106 Ru, and 154 Eu appeared to be associated with the fuel while 137 Cs and 125 Sb were released from the fuel and associated with the other nonfuel core materials.

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TMI-2 CORE DEBRIS GRAB SAMPLE QUICK LOOK REPORT

1. INTRODUCTION

On March 28, 1979, the Unit 2 pressurized water reactor at Three Mile Island underwent an accident that resulted in severe damage to the reactor's core. As a consequence of the TMI-2 accident, the Nuclear Regulatory Commission (NRC) has embarked on a thorough review of the causes and effects of severe core damage accidents. The nuclear community acknowledges the importance of examining TMI-2 in order to understand the nature of the core damage. Immediately after the TMI-2 accident, four organizations with interests in both plant recovery and accident data acquisition formally agreed to cooperate in these areas. These organizations, commonly referred to as the GEND Group--General Public Utilities, Electric Power Research Institute, Nuclear Regulatory Commission, and Department of Energy--are presently actively involve in reactor recovery and accident research. At present, the Department of Energy (DOE) is providing a portion of the funds in those areas where accident recovery knowledge will be of generic benefit to the U.S. light water reactor industry. In addition, DOE is funding acquisition and analysis of severe accident data (such as the examination of the damaged core).

The grab sample acquisition was performed primarily to provide data to support defueling of the reactor. Additionally, a TMI-2 Core Examination Plan has been prepared, which describes what technical/scientific data should be acquired during the TMI-2 core examination and how these data will be used to address specific reactor safety issues. One of the TMI-2 Core Examination Plan tasks is the examination of debris samples from the rubble bed.

Three samples each were acquired from the two core locations H8 and E9 (see Figure 1). The three samples were gathered at three different elevations, surface, 3-inches and 22-inches into the debris bed. Two different sampling devices were used to take the samples. One was a clamshell type tool used to take the surface samples. The other was a rotating tube

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Figure 1. TMI-2 core debris sampling locations.

device with doors cut into each side of two tubes. Figure 2 is a schematic showing how the sampling was performed. The examination of the debris grab samples has provided data on the physical and radioisotopic characteristics of the debris bed. Future work will address the debris chemistry. The information from the debris samples will aid in assessing the tooling and procedures required to defuel the TMI-2 reactor. In addition, these data will be used along with data from other tasks described in the Core Examination Plan to aid in defining the behavior of a commercial reactor core under the accident conditions that occurred at TMI-2.



Figure 2. TMI-2 core debris sampling schematic.

2. EXAMINATION SEQUENCE

The six grab samples were shipped to the Idaho National Engineering Laboratory (INEL). Five of the six grab samples were unloaded from the CNS-1-13C-II cask at the Test Area North and then transported in their 2R containers to the Test Reactor Area (TRA). The three inch deep sample from core location H8 was sent to B&W Lynchburg. The following examination steps were based on EGG-TMI-0001, "Implementation Plan for TMI-2 Core Debris Grab Samples Examiantion," (First Group of Samples) dated December 1983. The samples, in their sample cask, were removed from the 2R containers and placed in a laboratory hood at TRA. While in the hood, each sampler was removed from the cask and put into a preweighed container. Inside the sample cask, absorbent material had been placed to retain any residual water from the sample. This absorbent material was also removed from the cask and sent to the Radiation Measurement Laboratory for gamma counting. The sampler was then gamma counted using a mobile gamma ray spectroscopy system. This system, while not efficiency calibrated, did allow for isotopic identification and relative (qualitative) comparisons. The sampler was then weighed. Next the sampler was transferred to a glove box, where it was opened and the debris placed into a stainless steel tray. The debris was then visually inspected and photographed. At least one representative or interesting particle from each sample was then removed for gamma analysis and fissile determination. This report describes the visual examination and weighing of the samples, the fissile material and radiological measurements, and a discussion of the fission product inventory.

3. VISUAL EXAM AND WEIGHING

The visual exam was performed while the debris samples were in the glove box. The observations were hampered by the high radiation level with its resultant limited time for viewing. The radiation levels for the five samples, one inch away, ranged from 3 R/hr to 36 R/hr gamma using a teletector. Another complicating factor was the samplers were contained and shipped in a carbon steel cask and rust from the cask has possibly contaminated the samples. A schematic of the sample locations in the core and overall photographs of the samples are shown in Figure 3. Detailed photographs of the material in the samplers are presented in Figure 4 through 13. In general, fuel pellet fragments, shards of cladding or guide tubes, previously molten material, and fuel pellet surfaces glazed with previously liquified material (liquified fuel) were observed. A number of the pieces appeared to be covered with rust which had formed in the carbon steel cask. The debris Sample 1 (surface H8) is shown in Figure 4. A pile of very black. damp debris having a wide particle size range (greater than 1/4-inch to a fine debris) was obtained with this clamshell sampler. A large piece of fuel and cladding is apparent in the center of the photograph. This fuel rod remnant seems to be coated with previously liquid material.

The two deep (22-in.) rotating tube samples showed debris stratification when opened (see Figures 5 and 12). How much of that can be attributed to the actual core condition and how much may be an artifact of shipping or sampler operation is unknown. Both of the deep samples (Figures 6, 7, and 13) had a wide range of particle sizes (greater than 1/4-in. to fine debris). The E9 surface sample (Sample 4) was vastly different in appearance from the H8 Sample 1. This sample consisted of approximately 13 fairly large dry chunks of material (see Figure 8). The E9 sample 3-inches into the debris bed (Sample 5) contained several large pieces, probably fractured fuel pellets (Figures 9, 10, and 11). Additional visual information will be gathered from enlargements of the debris photographs.





Figure 3 Summary schematic showing TMI-2 core debris grab sample acquisition.



Figure 4. TMI-2 core debris sample #1 (surface-H8).



Figure 5. TMI-2 core debris sample #3 being removed from the rotating tube (22" deep-H8).



Figure 6. TMI-2 core debris sample #3 (22" deep-H8).



Figure 7. TMI-2 core debris sample #3 close-up (22" deep-H8).









Figure 10. TMI-2 core debris sample #5 (3" deep-E9).



Figure 11. Close-up of TMI-2 core debris sample #5 (3" deep-E9).



Figure 12. TMI-2 core debris sample #6 being removed from rotating tube (22" deep-E9).



Figure 13. TMI-2 core debris sample #6 (22" deep-E9).

Enlarged photographs of particles removed from samples are shown in Figures 14 through 20. Due to the inhomogeneity of the samples, the particles removed are not intended to be representative but are of general interest. The particle in Figure 19 resembles a curved piece of cladding or guide tube. The other particles do not have a definite structure and possible composition (fuel or nonfuel components) can only be inferred from the fissile material measurements.

Table 1 summarizes the results of the visual examination and the weighing. The sample weights are probably within ± 2 g; the large uncertainty due mainly to the air flow in the laboratory hood where the weights were measured.



Figure 14. Particle 1-1, removed from TMI-2 core debris sample #1 (surface-H8).



Figure 15. Particle 3-2, removed from TMI-2 core debris sample #3 (22 in. deep-H8).



Figure 16. Particle 4-1, removed from TMI-2 core debris sample #4 (surface-E9).



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Figure 17. Particle 5-1, removed from TMI-2 core debris sample #5 (3 in. deep-E9).



Figure 18. Particle 6-1, removed from TMI-2 core debris sample #6 (22 in. deep-E9).

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Figure 19. Particle 6-2, removed from TMI-2 core debris sample #6 (22 in. deep-E9).





Figure 20. Particle 6-3, removed from TMI-2 core debris sample #6 (22 in. deep-E9).

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Sample Number	Sampler Type	TMI-2 Core Location	Location of Sample in Rubble Bed	Sample Weight (grams)	See for Erron to Visual Characteristics
1	Clamshell	H8	307 ft 0 in. (surface)	474	A pile of very black, damp debris with a fairly wide range of particle sizes (dimensions ranging from 1/16 to 1/2 in.); several rounded surfaces; sporadic rust color throughout.
3	Rotating Tube	н8	22-in. into debris bed	147	Very black debris, slightly damp, wide range of particle sizes (dimensions 1/16 to 1/4-in.), small chunks to fine debris; similar to sample 1.
4	Clamshell	E9	306 ft 9 in. (surface)	14	Thirteen major chunks, dry, black with rust colored sides, basically sharp edges with one or two chunks having rounded edges; dimensions ranging from 1/4 to 3/8 in.
5	Rotating Tube	E9	3 in. into debris bed	-75 -95	Similar to Sample 4 with the following distinctions: many more pieces; greater size range (1/16 to 3/8 in.); some surfaces more reflective. Again, very dry.
6	Rotating Tube	E9	22-in. into debris bed	148	Very black debris, small chunks to fine debris, slightly damp, some pieces blackish gray. A couple of pieces resembled metal shards.

TABLE 1. SUMMARY OF PHOTO/VISUAL AND WEIGHING

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4. FISSILE MATERIAL AND RADIOLOGICAL MEASUREMENTS

Eight particles were removed for preliminary analyses. One particle was removed from Samples 1, 4, and 5; two particles from Sample 3 and 3 particles from Sample 6. The radiological measurements performed on the eight particles included delayed neutron analysis of the fissile material content and gamma spectrum analysis for measurement of gamma emitting radionuclides. The fission product to 235 U ratios were calculated for 125 Sb, 137 Cs and 144 Ce from these data and the measured sample weights. A comparison is made between the measured data decay corrected to March 29, 1979 and the ratios predicted using the ORIGEN code. The methods used to obtain the radionuclide and fissile material concentrations and the detailed results are discussed below in addition to the uncertainties associated with these analyses.

The fissile material (e.g., 235 U plus 239 Pu) measurements were performed at the Coupled Fast Reactivity Measurement Facility (CFRMF) by delayed neutron analysis. These data are presented in Table 2 along with the individual particle weights. The error quoted for the fissile material concentrations is a one sigma error associated only with the delayed neutron counting statistics and calibration using dilute samples. Other errors possibly associated with sample collection and transfer have not been included. It is expected that the measured fissile material concentrations are lower than the quantity actually present in the larger (greater than 0.5 gram) particles that have a significant ²³⁵U concentration (i.e., 3-2 and 4-1). This is because the large amount of 235 U present in the larger particles reduces the thermal neutron interrogating flux in the delayed neutron measurement. As a result, the measured fissile material concentration in the largest particle could possibly be increased by a maximum of 10% with other measurements being increased in proportion to the individual particle mass. An additional source of uncertainty in the data is the possible presence of boron on the surface of the samples. This element, which has a large neutron cross section, could reduce the apparent fissile material concentration by a small percentage (<10%) depending upon the thickness of the boron layer. In the future, analysis will be performed with calibration standards prepared for the size range of the particles and presence or absence of a boron layer will also be determined and a correction performed.

TABLE 2. FISSILE MATERIAL CONCENTRATIONS

Weight (grams)	Fissile Material (grams)	Fissile Material (wt %)
0.0181	<2(-6)	<0.01
0.0281 0.5239	$3.3 \pm 0.2(-4)^{a}$ 9.5 $\pm 0.6(-3)$	1.2 1.8 ^b
0.7638	1.37 <u>+</u> 0.09(-2)	1.8 ^b
0.182	4.0 + 0.3(-3)	2.2
0.2545 0.6256 0.1673	8.4 + 0.7(-5) 4.8 + 0.5(-5) 3.7 + 0.2(-3)	3.3(-2) 7.7(-3) 2.2
	Weight (grams) 0.0181 0.0281 0.5239 0.7638 0.182 0.2545 0.6256 0.1673	Weight (grams)Fissile Material (grams)0.0181<2(-6)

a. The notation used does not list the exponential (i.e., 3.3 \pm 0.2 (-4) = 3.3 \pm 0.2 x 10⁻⁴).

b. Measured concentration may be less than the actual value due to the mass of the sample. Based upon comparable samples, Sample 4-1 would be increased by 10% and Sample 3-2 by 7%.

The data listed in Table 2 indicate that five of the particles are composed mainly of fuel. The 235 U fuel enrichment for the TMI-2 core ranged from 1.98 wt% to 2.93 wt%. The enrichment at the E9 and H8 location was 1.98% and 2.64%, respectively. Four of the five particles have apparent enrichments in the range of the fuel in the TMI-2 core. These values would be higher if nonfuel materials are incorporated into the particles. Subsequent analyses will determine enrichment directly by measuring both 235 U and 238 U contents. Particles 1-1, 6-1, and 6-2 contained little 235 U (<1%) and therefore are composed of other materials present in the reactor core. Particle 6-2 is a curved piece of metal that appears to be a section of cladding or guide tube (see Figure 19). Both inner and outer surfaces of this material will be evaluated for chemical composition.

The principal gamma ray emitting radionuclides detected in the eight core debris particles are listed in Table 3. Figure 21 is a representative gamma spectrum for particle 4-1. These measurements were performed in a calibrated

LE 3. PRINCIPLE RADIONUCLIDE CONCENTRATIONS (microcuries/gm)

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	Gamma Ray	ingen in digen and an	H8 - 22	inches				E9-22 Inches	
tion in 144	Energy	H8 Surface	3-1	3-2*	E9 Surface	E9-3 Inches	6-1	6-2	6-3
TONUCTINE		1.08 + 0.05(0)	1.7 + 0.4(0)	b	4.8 + 0.4(0)	b	$2.1 \pm 0.3(-1)$	$1.8 \pm 0.3(-1)$	
•	835.1		1.46 + 0.02(+2)	A = 2 + 0.1(+1)	1.33 + 0.02(+2)	1.77 + 0.09(+1)	3.8 <u>+</u> 0.1(0)	2.46 + 0.09(0)	Þ
Co	1332.7	3.88 - 0.03(-1)			7.04 + 0.09(+2)	8.2 + 0.1(+2)	9.6 + 0.5(0)	2.18 + 0.06(+1)	1.02 • 0.03(+3)
6Ru	622.1	4.3 + 0.4(0)	3.71 + 0.08(+2)	1.58 - 0.02(+3)		6.6.4.0.3(+1)	3.63 + 0.04(+1)	2.98 + 0.03(+1)	1.20 • 0.07(+2)
550	427.9	1.19 <u>+</u> 0.01(+2)	1.62 + 0.03(+2)	8.7 + 0.3(+1)	7.8 - 0.2(*1)		6.6 + 0.1(0)	3.67 + 0.09(0)	1.73 ± 0.04(+2)
134 _{CS}	796.0	$9.3 \pm 0.7(-1)$	5.2 ± 0.1(+1)	$1.39 \pm 0.09(+1)$	2.22 + 0.07(+1)	3.1 ± 0.1(+1)		6 59 + 0 03(+1)	4.12 + 0.02(+3)
137 _{CS}	661.8	2.02 <u>+</u> 0.02(+1)	1.14 + 0.01(+3)	2.19 + 0.02(+2)	5.66 <u>+</u> 0.03(+2)	8.25 + 0.04(+2)	1.24 • 0.01(+2)		3.3 <u>+</u> 0.3(+3)
144 _{Ce}	2186.1	$2.0 \pm 0.4(+1)$	2.47 + 0.10(+3)	4.3 + 0.2(+3)	2.50 <u>+</u> 0.1(+3)	$3.0 \pm 0.2(+3)$	1.7 + 0.4(+1)	9.0 - 0.0(*1)	3.1 + 0.3(+1)
154 _{Eu}	1275	1.2 + 0.7(-1)	3.1 + 0.2 (+1)	6.4 + 0.3(+1)	1.9 + 0.1(+1)	2.5 <u>+</u> 0.2(+1)	1.4 + 0.5(-1)	1.1 + 0.1(0)	0, 1673
Sample Weight (grams)		0.0181	0.0281	0.5239	0.7638	0.182	0.2542	0.6256	

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a. Corrected for internal gamma ray attenuation by the sample (<20%).

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:. Not detected.

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Figure 21. Gamma ray spectra for particle 4-1 removed from TMI-2 core debris sample #4 (surface-E9).

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point source geometry at the Radiation Measurements Laboratory. The data, where appropriate, (i.e., particles 3-2 and 4-1) have been corrected for self shielding of the measured gamma rays by the mass of the sample. This correction was based on the 2186.1 KeV emission energy of the ¹⁴⁴Ce daughter (144 Pr) and a calculated attenuation factor for this energy ($\sim 9\%$) based on the density of the sample. These data were extrapolated to lower energies using the ¹⁵⁴Eu, ¹²⁵Sb and ¹⁴⁴Ce emission energies. Total corrections were less than 19%.

The data listed in Table 3 indicate several trends. The widest ranges for the radionuclide concentration data are for 144Ce, 106Ru, and ¹⁵⁴Eu. The ratio (i.e., the highest concentration divided by the lowest) is 252 for 144 Ce, 372 for 106 Ru, and 533 for 154 Eu. These data indicate that these fission products are not evenly distributed among the particles. The high concentrations for all three radionuclides are associated with Figure 21. Gamma ray spectra for particle 4-1 removed from TMI-2 core debris particles which have high fuel contents (i.e., 3-1, 3-2, 4-1, 5-1, and 6-3). ¹²⁵Sb has the smallest range of radionuclide concentrations (i.e., 3.0 (+1) - 1.6 (+2) μ Ci/q), a factor of 5.4, which indicates that it is dispersed more evenly between fuel and other core materials. For example, Sample 6-2 which appears to be a portion of zircaloy cladding or quide tube has the lowest 125Sb concentration but is within a factor of 2.6 of the 125Sb 134, 137_C concentration measured for Sample 4-1 which is principally fuel. have ratios of 186 and 204 respectively, which are not as high as the 144 Ce. 106_{Ru} and 154_{Eu} . The remaining radionuclides, 54_{Mn} and 60_{Co} are activation products and have concentration ratios of 27 and 60 indicating an activity dispersion similar to ¹²⁵Sb. These data are discussed further in the following comparison with the predicted fission product inventory.

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5. FISSION PRODUCT INVENTORY COMPARISON

The ratio of the fission products to the amount of 235 U in the particles may be calculated from the data in Tables 2 and 3. Table 4 presents the ratio of the the fission product to 235 U concentrations for the radionuclides 144 Ce, 137 Cs, and 125 Sb. For comparison purposes with ORIGEN code values, the fission product concentrations have been decay corrected to March 29, 1979. Footnote "a" lists the factors used to calculate these fission product concentrations in grams.

The TMI-2 core inventory as calculated by the ORIGEN code for March 29, 1979, is ratioed to the fission product/ 235 U ratio listed in Table 4 (e.g., $^{144}Ce/^{235}U$ measured/ $^{144}Ce/^{235}U$ calculated) and is presented in Table 5. It should be noted that the ORIGEN code calculations to date are only for core average fission product concentrations and actual core fission product inventories for specific locations may vary. In general, the measured $^{144}Ce/^{235}U$ ratio appears to follow the ORIGEN calculated ratio. Ratios greater than one indicate that there is more ^{144}Ce associated with a particle than would be calculated to be produced based on the average fuel irradiation. Although there is considerable scatter, the measured to calculated $^{144}Ce/^{235}U$ ratios average 1.06 for the five particles containing mainly fuel, indicating that $^{144}Ce/^{235}U$ ratios in those particles listed in Table 5 that the $^{144}Ce/^{235}U$ ratios in those particles probably have little meaning.

A significant fraction (generally >90%) of the 137 Cs has been released from the five particles that contain a significant fraction of fuel if they are representative of the ORIGEN derived core average inventory. Mass balance work to date has indicated 50 to 60% of the 137 Cs has been accounted for outside the core. The high 137 Cs ratio (22%) observed for the 6-3 sample is not explainable as yet. The high 137 Cs/ 235 U ratios for Samples 6-1 and 6-2, which have very little 235 U present indicate that the 137 Cs appears to concentrate on materials not associated with the fuel itself. 125 Sb (about 70% release for four of the particles that are

Sample Identification	144 _{Ce/} 235 _U	137 _{Cs/} 235 _U	125 _{Sb/} 235 _U
3-1	4.5(-3)	1.3(-3)	4.3(-5)
3-2	5.0(-3)	1.6(-4)	1.5(-5)
4-1	3.0(-3)	4.1(-4)	1.4(-5)
5-1	2.9(-3)	4.8(-4)	9.6(-6)
6-1	1.1(-3)	4.8(-3)	3.5(-4)
6-2	2.7(-2)	1.1(-2)	1.2(-3)
6-3	3.2(-3)	2.4(-3)	1.7(-5)

TABLE 4. FISSION PRODUCT/²³⁵U URANIUM RATIOS (at shutdown)^a

a. Fission product decay corrections to shutdown and specific activity.

Decay (half lifes)	Decay Factor	Specific Activity (µCi/g)
1.74	3.33	1.05(9)
0.16	1.115	8.65(7)
6.08	67.81	3.19(9)
	Decay (half lifes) 1.74 0.16 6.08	Decay (half lifes) Decay Factor 1.74 3.33 0.16 1.115 6.08 67.81

TABLE 5.	MEASURED	T0	PREDICTED	FISSION	PRODUCT/235U	CORE	INVENTORY R	ATIOS
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Sample Identification	144 _{Ce}	137 _{Cs}	125 _{Sb}
3-1	1.29	0.120	0.87
3-2	1.43	0.015	0.31
4 - 1	0.86	0.038	0.28
5-1	0.83	0.044	0.19
6-1	0.31	0.440	7.06
6-2	7.71	1.00	25.0
6-3	0.914	0.220	0.35
Core ratio ^a	3.5(-3)	1.09(-2)	4.95(-5)

a. Calculated from ORIGEN code fission product inventories and total $^{235}\mathrm{U}$ content in the core at shutdown.

	ORIGEN Core Concentration (grams)	
144 _{Ce}	7.7(3)	
137 _{Cs}	2.4(4)	
125 _{Sb}	1.09(2)	
235 _U	2.2(6)	

principally fuel) appears to behave similarly to the 137 Cs. However, based on the few samples measured, the 125 Sb association with nonfuel material appears to be greater than for 137 Cs.

6. CONCLUSIONS

Some preliminary conclusions may be drawn from the analyses of the debris samples to date. They are:

- Fuel pellet fragments, shards of cladding or guide tubes, previously molten material, and fuel pellet surfaces apparently glazed with previously liquid material were observed.
- 2. There is a wide range of materials present in the eight particles studied. Three of these particles contain very small fractions of ²³⁵U of which one appears to be a piece of cladding or guide tube material. The other five particles appear to be principally fuel.
- 3. Some radionuclides (i.e., ¹⁴⁴Ce, ¹⁰⁶Ru and ¹⁵⁴Eu) appear to be associated specifically with the uranium fuel.
- 4. The more volatile radionuclides, ¹³⁷Cs and ¹²⁵Sb, appear to have been released from the fuel and associated more with the nonfuel core materials.

