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GAMMA-RAY EMISSION TOMOGRAPHY EXAMINATION OF TMI-2 CORE SAMPLES[a]

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ABSTRACT

An examination method developed at the Idaho National Engineering Laboratory (INEL) is being used to determine the distribution of gamma-ray emitting radionuclides in samples of reactor fuel and prior molten materials. This paper presents the results of a gamma-ray emission tomographic examination performed on cross sections of samples of prior molten fuel material recovered from the TMI-2 reactor. The results indicate substantial relocation of fission products in the debris and the probable accumulation of specific radioactive species in various phases of the material. The observed behavior correlates with the results of microstructural examinations and elemental compositions obtained from SEM/WDX, electron microprobe, and radiochemical analyses.

INTRODUCTION

Examinations of prior molten reactor fuel have been performed to

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evaluate radionuclide retention and relocation in the material. Typically, these examinations have been performed by dissolution and radiochemical analysis of microcore samples; however, this method provides only a limited amount of information on the distribution of the radionuclides in the material. Sample materials with varying degrees of heterogeneity can provide misleading results and generally, only a few sample fractions can be examined because of cost. These effects introduce a significant uncertainty in the examination results and reduce the amount of information obtained from an examination program. The gamma-ray tomography system developed at the Idaho National Engineering Laboratory (INEL) provides quantitative information on the distribution of gamma ray active materials in examined samples.

The gamma ray tomography system provides either photographic depictions or isoconcentration curves of the distribution of radionuclides in the sample cross sections. These results are then compared with microstructural and elemental analysis results to evaluate the effects of fuel morphology, composition, and temperature on radionuclide distribution and relocation in the sample material. This paper presents a brief description of the gamma-ray tomography system and discusses the results of examinations performed on samples from the TMI-2 reactor core. The samples examined include a sample of prior molten debris from the lower reactor vessel head that reached an estimated maximum temperatures of 2800K, and examples of crust materials from the lower core debris bed at two elevations. The advantage of the gamma-ray tomographic method over the common technique of limited discrete sampling is demonstrated.

SYSTEM DESCRIPTION

The first generation INEL gamma-ray tomography system incorporates a scanning bed that translates and rotates the sample under a collimated hyperpure germanium (Ge) gamma-ray detector. The detector is highly collimated, and shielded. Figure 1 is a schematic showing the detector, collimator, and scanning bed. The gamma ray detector used for the

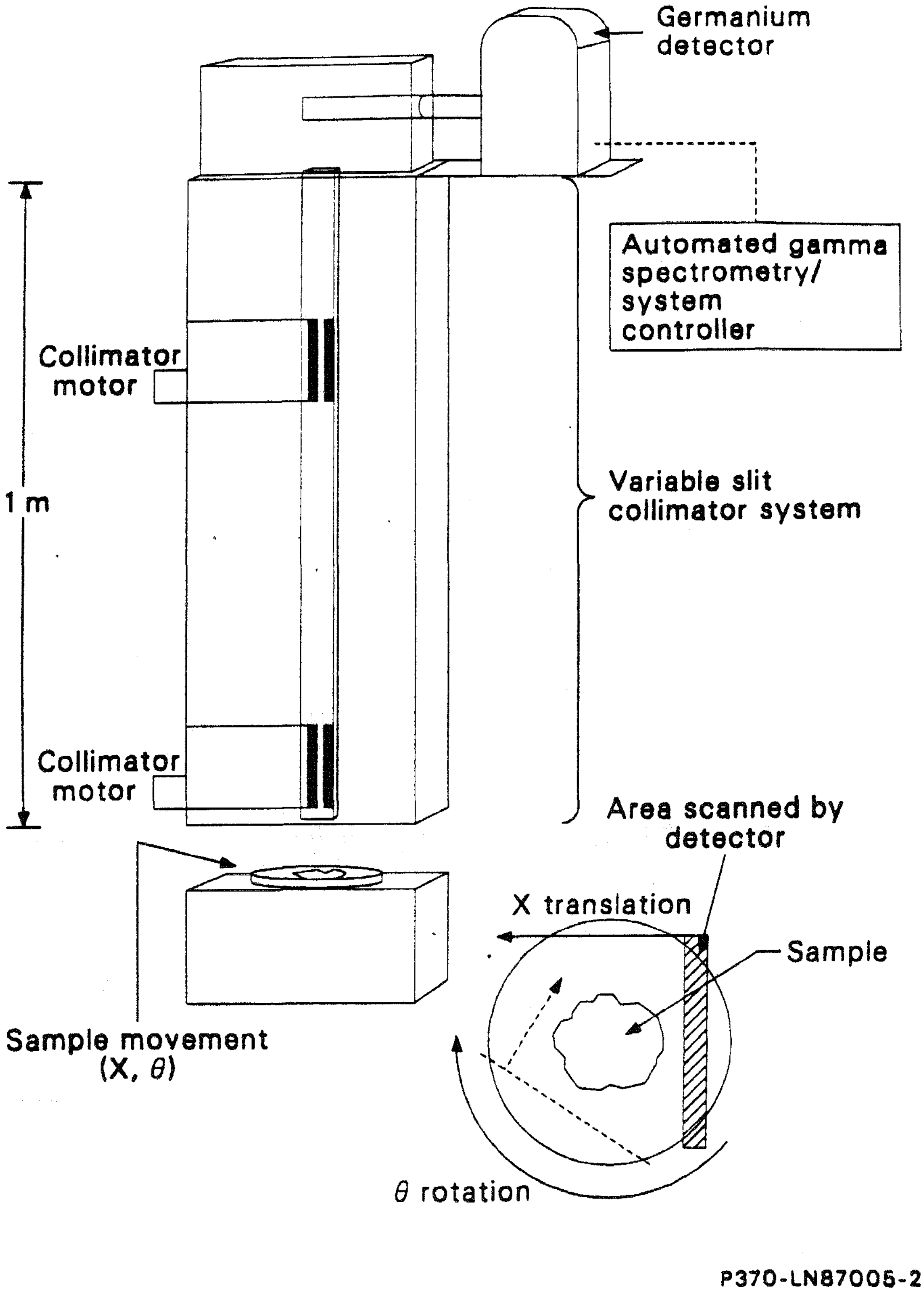

Figure 1. Gamma ray tomography system schematic

gamma-ray tomography system is a Princeton Gamma-Tech (PGT) hyperpure germanium detector with a tungsten backshield for use in high radiation fields. The collimator incorporates moveable upper and lower tungsten-faced blocks about 55 cm apart with a total tungsten thickness of 20 cm. The scanning bed combines rotational and translational motion to obtain the degrees of movement required for tomographic reconstruction.

The gamma spectroscopy system used for the tomography measurements was developed at the INEL for high count-rate applications[1]. This system is composed of a high count-rate data acquisition system, pulsers to correct for dead time losses and gain changes, and a PDP 11/44 computer for data processing, control, and spectral storage. The computer is interfaced to a CAMAC crate that controls the scanning beds. The PDP 11/44 appends the scan bed location (X, theta) to each associated spectrum.

Adequate characterization of a sample may require up to 1500 gamma ray spectra. These spectra are analyzed on a CYBER 176 computer using the gamma ray spectrum analysis code GAUSS VIII[2]. As the surface geometry and density of the material is variable, an algorithm was incorporated into the spectrum analysis program that uses the results of a specified multi-line gamma ray emitter (generally ^{154}Eu) to normalize the individual spectral results to a common basis for comparison purposes[3]. Upon completion of the gamma ray spectrum analysis, a sort routine is used to develop data files for individual radionuclides. These data files are then processed using a tomographic reconstruction routine (the Donner Algorithms for Reconstruction Tomography)[4] optimized for the expected sample geometries[5]. Both photographic reconstructions and isoconcentration curves are then generated depicting the distribution of radionuclides in the sample material.

EXAMINATION RESULTS

The first sample (particle 11-5-C) examined using the INEL gamma-ray tomography system was a 1-cm-thick cross section of a large particle (6.2

Figure 2. Particle 11-5-C sampling locations

cm dia.) that had relocated from the TMI-2 core as molten material and solidified on a pile of fuel debris located on the lower head of the reactor. Particle microstructures indicate a maximum temperature of about 2800K. Gamma-ray tomographic spectra were obtained at 36 angles and at 37 steps at each angle across the face of the sample (1332 gamma ray spectra). The collimator width was 0.19 cm and was equal to the translational distance between steps.

In addition to the gamma ray tomography examinations, individual samples were removed from a variety of locations in Particle 11-5-C for radiochemical examination. Figure 2 is a picture of the 11-5-C cross section showing where samples were removed for radiochemical and elemental examination. Table 1 lists the radiochemical and elemental examination results[1].

Figures 3 and 4 present overlays of the photograph of sample cross section 11-5-C with the tomographically developed isoconcentration plots for ^{60}Co and ^{137}Cs respectively. Similar plots have been developed for ^{106}Rh, ^{125}Sb, ^{144}Ce, and ^{154}Eu; however the two chosen are indicative of the information available through these measurements. Note that the isoconcentration plots for both ^{60}Co (a stainless steel activation product) and ^{137}Cs (a fission product) mirror the discrete radiochemistry data, revealing a generally higher concentration in the higher (7-9) sample numbers than in samples taken from the other side of the sample cross section. However, the information content of the tomographic reconstructions is clearly superior to the discrete samples.

Microstructural examinations were performed of the material in the porous (high sample number) and nonporous regions to evaluate microstructural differences between the two regions of the sample. Figures 5 and 6 are photomicrographs showing the microstructures of the two regions. In the nonporous region, the grain boundaries are relatively clean with little precipitated material present whereas in the porous region there are significant amounts of material in the grain boundaries

Figure 3. ^{60}Co distribution in particle 11-5-C

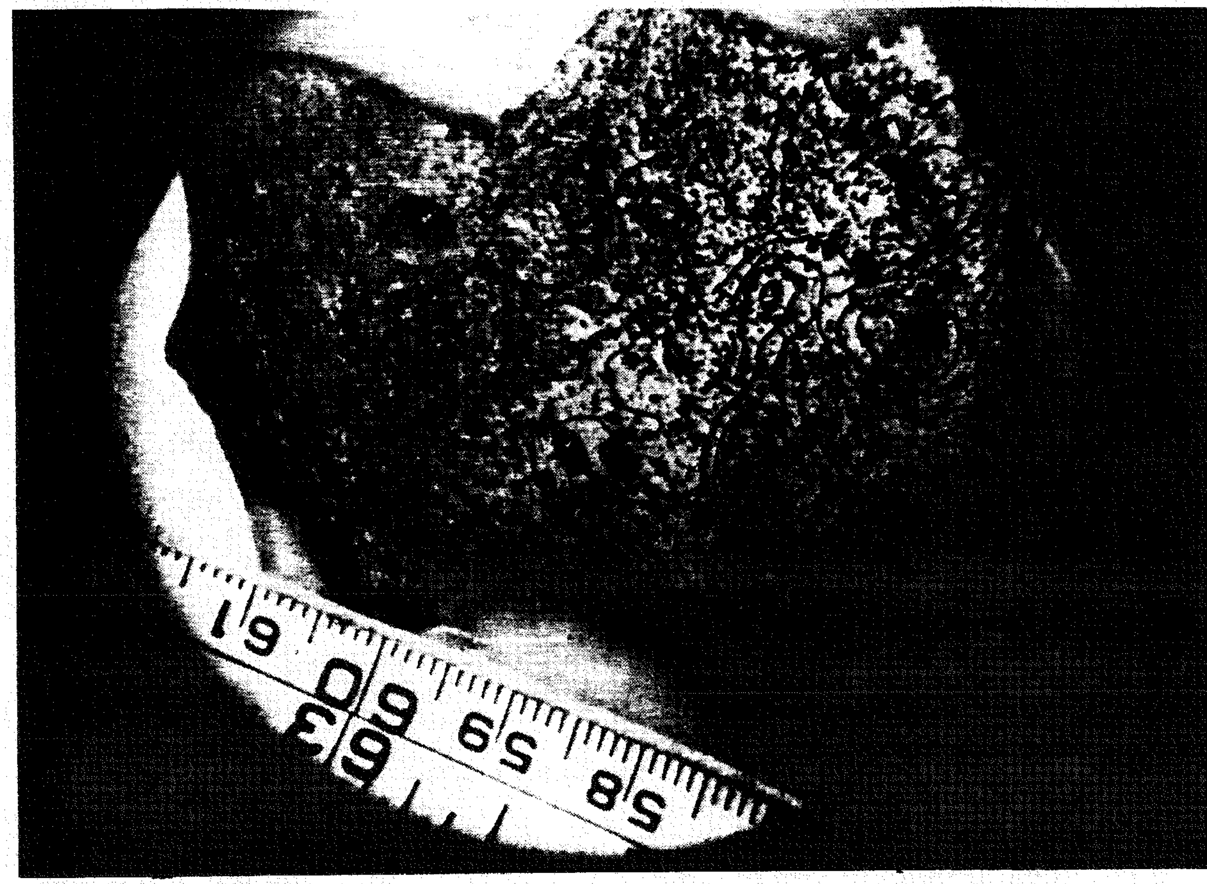

Figure 4. ^{137}Cs distribution in particle 11-5-C

Figure 5. Low porosity region - no grain boundary precipitates

Figure 6. High porosity region - metallic precipitates

Figure 7. Metal precipitates in grain boundaries with dendritic structure

and the metallography indicates the presence of a second phase surrounding the primary $(U,Zr)O_2$ grains. Electron microprobe examination of the grain boundary phase indicates the presence of Fe and Cr and some Ni oxides probably as eutectics with UO_2. The concentrations of Fe and Cr are indicated by the dark phases in the dendritic structures in Figure 7. These data suggest that the ^{137}Cs retention may be in the grain boundaries associated with the metallic phases.

Figure 8 shows the upper crust in the lower part of the TMI-2 reactor core. A large metallic globule is present at the edge of the crust. This crust was a coherent mass of ceramic type material with finely dispersed metallics salted in the matrix and a large metallic region. The data suggest that the metallic phases are located nearer the top surface of the plug above the ceramic matrix and the metallic fraction may have been deposited on the surface of the ceramic layer, filling cracks and voids in the material.

The metallurgical examinations indicate than in the metallic regions of the upper crust, the metallics (i.e., Fe, Cr, Ni, Ag, In, and other structural material constituents) are frequently found in the form of layers of different metallic alloys which have separated into layers with no orientation relative to the surface of the crust.

Radiochemical examinations of microcores from this crust sample indicates that the concentrations of the low volatiles and most of the medium volatiles (i.e., Ce-144, Eu-154, and Sr-90) are located mostly in association with the ceramic portions. Examination of the tomograms for the gamma ray emitters (Ce-144 and Eu-154) the data indicate that these radionuclides are not present in the metallic regions except at very low concentrations. However, the Sb-125 and Ru-106 radionuclides are expected to exhibt a greater range of concentrations in the upper crust as previous examinations have indicated that these radionuclides remain as metallics in the debris and tend to be scavenged by the metallic

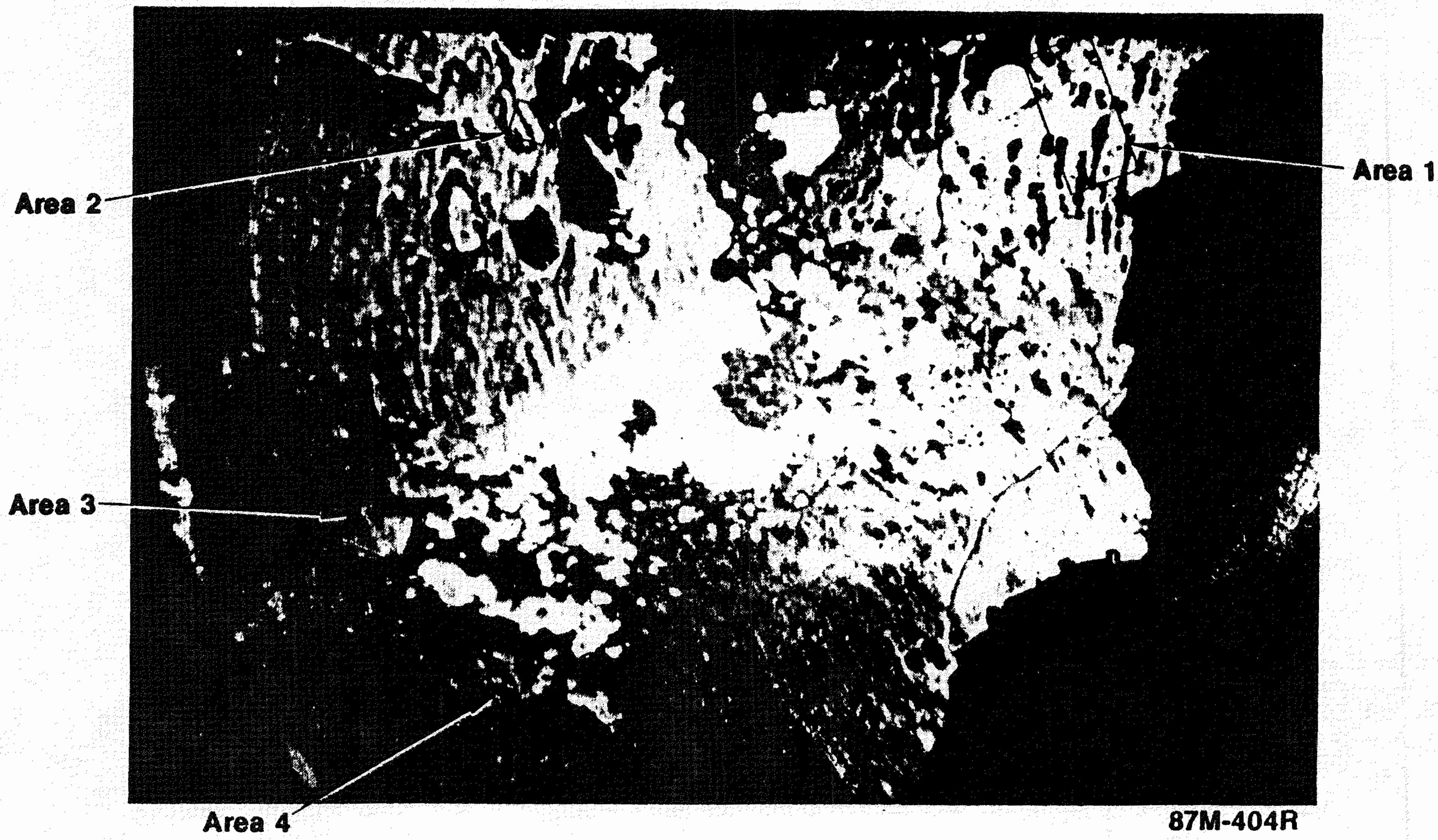

Figure 8. TMI-2 Upper Crust - Longitudinal cross section

structural materials. For Sb-125, comparisons were performed with the concentrations of elemental constituents of the crust and no correlations were observed with the exception that a general increase in metallic content, generally correlated with an increase in Sb-125 content. Examination of the Ru-106 and Sb-125 tomograms for this sample indicate that these radionuclides are concentrated near the top of the crust which suggests that their deposition was not controlled by scavenging effects entirely, but may have been affected by physical mechanisms such as retention of the material by a ceramic crust. Additional confirmation of this fact is that the Co-60, a neutron activation product is concentrated near the center of the particle, at a different location from the Ru-106 and Sb-125.

A cross section of the lower crust in the core was examined using the gamma ray tomography system. An example of this crust at the K9 core location is shown in Figure 9. The bottom of the crust is on the left side of the photograph and top crust (which has been partially removed) on the right side. Most of the material in the coolant channel between the fuel rods is metallic material with small inclusions of fuel material embedded in the matrix. The molten material flowed into pellet/pellet interfaces and cracks, and the metallic material next to the fuel was oxidized. These data indicate that the lower crust is composed of a prior molten, mostly metallic material containing fragments of fuel material (both intact and prior molten) which were swept down and around the intact fuel rods.

The gamma ray tomography examinations of this sample indicate that the inventory of the low volatiles have been retained in the fuel rod matrix and that in the flow channels there are relatively low concentrations of all fission products which suggests that the fission product content of the debris has not been concentrated in this part of the reactor core.

In summary, the gamma-ray emission tomography method provides significantly superior information content as compared to discrete

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Figure 9. TMI-2 Lower Crust - longitudinal cross section

sampling techniques. Examinations of Particle 11-5-C indicated the distribution of structural materials (^{60}Co) in the sample and the apparent association of the relatively volatile ^{137}Cs with the structural material components or porosity in the sample. The metallurgical examinations of the sample suggest that the volatile cesium is associated with metallic structural components located in the grain boundaries. The examination of the lower core crust layers indicates that the metallic radionuclides (Sb-125 and Ru-106) are concentrated in the metallic phases of the core and that the low volatile radionuclides are generally not affected by the high temperatures produced during the accident.

SUMMARY

The INEL gamma-ray tomography system has been used to evaluate the distribution of fission products in a sample of prior molten material from TMI-2. The results to date are preliminary and additional interpretation is now being performed. Gamma-ray emission tomography is a promising technique offering greater information content than limited discrete sampling. This technique, coupled with limited discrete sampling, lead to the following observations concerning the TMI-2 samples:

- Activation products, such as ^{60}Co, are concentrated in areas rich in structural materials.

- ^{137}Cs was more strongly correlated with grain boundary inclusions than with fuel material, implying a release from the hot fuel matrix and trapping in grain boundary material.

- The radionuclides that are expected to remain as metallics (Sb-125 and Ru-106) are retained in metallic regions of the core possible by scavenging or a physical retention method.

- o There is no apparent correlation between Co-60, an activation product of the structural materials and the distribution of the metallic radionuclides Ru-106 and Sb-125.

- o The low volatile radionuclides (e.g., Ce-144 and Eu-154 are retained in the ceramic matrix at high temperatures and are not mobile to the metallic regions.

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