

SUMMARY OF TMI-2 CORE SAMPLE EXAMINATIONS^a

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ABSTRACT

Core debris samples were obtained from the upper plenum debris, the central consolidated region, standing fuel rod assemblies, and the lower plenum debris to determine the temperatures that were reached during the TMI-2 accident. The materials interactions that occurred, bulk composition of the materials, oxidation of the materials, and retention of fission products in the core materials were also determined. The results of the metallurgical examinations and the bulk compositions are described in this paper.

INTRODUCTION

The TMI-2 Sample Acquisition and Examination is being conducted by the Department of Energy and the Nuclear Regulatory Commission to provide information to complete the understanding of the TMI-2 accident, and to help resolve severe accident and source term technical issues. Following the accident, the TMI-2 core region consisted of an upper void region almost entirely surrounded by damaged, but standing, peripheral fuel assemblies. Directly below the void region was a region of loose debris resting on a hard crust. The crust completely surrounded a centrally located, consolidated region that was previously molten. A region of intact standing fuel rods extended upwards from the bottom of the core to the bottom of the crust surrounding the previously molten region. About 19,100 kg of molten core material relocated to the lower plenum region of the reactor vessel. Samples have been obtained from the upper debris bed; the standing fuel assemblies surrounding the upper void region; the

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centrally located, previously molten region and surrounding crust; the standing fuel assemblies supporting the lower crust of the previously molten region; and, the previously molten debris that relocated to the lower plenum region. These samples have been subjected to extensive metallurgical and radiochemical examinations to determine elemental composition, oxidation state, materials interactions, peak temperatures, and fission product retention. Selected results of these examinations are the subject of this paper.

UPPER CORE FUEL ASSEMBLIES AND DISTINCT COMPONENTS

The upper void region, illustrated in Figure 1, was approximately 1.5 m deep with an overall volume of 9.3 m³. Forty-two of the original 177 fuel assemblies were standing at the periphery of the core void. Only two of these fuel assemblies contained more than 90 percent of their full-length cross sections with the majority of the fuel rods intact. Non-destructive exams were performed for fuel assembly and control rod and burnable poison rod assembly upper end fittings from this region. Both non-destructive and destructive examinations were performed on fuel and control rod upper ends. The examinations that were performed are listed in Table 1. Results of the examinations of these components indicated large radial and axial temperature gradients, and differences in cooling behavior of control material existing over short distances. Peak temperatures of the upper spacer grids were 1500 to 1700 K. Previously molten control material had flowed upwards into the plenum spring region of control rods. Also, in the upper end of a fuel rod that had broken off, a previously molten mixture of fuel, cladding, and control and structural material had migrated upwards into the fuel cladding gap for several centimeters. This indicates that at some time during the accident the lower ends of the upper core region were immersed in liquid core materials.

TABLE 1. TMI-2 DISTINCT CORE COMPONENT EXAMINATION PROGRAM

<u>Component</u>	<u>Non Destructive exams</u>				<u>Destructive exams</u>		
	<u>insitu CCTV</u>	<u>photo graph</u>	<u>neutr radio</u>	<u>gamma scan</u>	<u>metal ograph</u>	<u>chem comp</u>	<u>radio chem</u>
Fuel Assembly Upper End Fittings	X	X					
Control and Burnable Poison Rod Assembly Upper End Fittings	X	X					
Fuel Rod Upper Ends	X	X	X	X	X	X	X
Control Rod Upper Ends	X	X	X	X	X	X	X

UPPER PLENUM DEBRIS

The loose debris bed below the core void that was resting on a crust ranged in depth from 0.6 to 1 m, and consisted of whole and fractured fuel pellets, control rod spiders, end fittings and resolidified material² totaling about 26,600 kg. Samples obtained from the upper debris bed for examination are shown in Figure 2. The debris bed was heterogeneous, exhibiting a broad range of oxidation and material interactions, but was well mixed. Ninety percent of the sample particles were between 1 and 5 mm in their maximum dimension. Most of the debris bed consisted of shattered and resolidified material that contained regions of previously molten (U,Zr)O₂, indicating temperatures greater than 2700 K. There were also a few samples of previously molten material that were almost pure UO₂, indicating temperatures up to 3100 K. Metallographic examinations of individual particles indicated that most of the debris bed had remained at temperatures below 2000 K, or was exposed to high temperatures for only a short time. The bulk composition of the loose debris bed was primarily uranium, zirconium and oxygen with significant depletion of silver, indium, and cadmium control material, Zircaloy cladding, and structural material. Thus, the materials that are the first to melt and/or form eutectics and relocate are depleted from the upper debris bed. The upper debris bed consists of the remnants of highly oxidized fuel rods that shattered when they were cooled, and that were depleted of control, structural and cladding material before cooling.

Figure 3 displays the major melting and interaction points of core materials versus a temperature scale. The determination of whether these phenomena have occurred make it possible to bound peak temperatures. The silver, indium, cadmium control material melting range is between 1073 and 1100 K. Zircaloy transforms from the alpha to the beta phase between 1105 and 1245 K. Zirconium (from the cladding) forms eutectics with iron and nickel (from stainless steel control rod cladding or Inconel grid spacers) at 1200 K but the solid diffusion rate is too slow to be of concern until the temperature reaches 1500 K. Inconel, stainless steel and Zircaloy melt at 1660, 1720, and 2030 K, respectively. The dissolution of UO₂ by zirconium begins at 2030 K.

CENTRAL PREVIOUSLY MOLTEN REGION

The large, crust-enclosed previously molten, consolidated region below the loose debris was approximately 3 m in diameter, 1.5 m thick at its center, and 0.25 m thick at its periphery, and was comprised of about 32,700 kg of material. The previously molten region inside the crust consisted of a mixture of structural, control, and fuel material that reached temperatures of 2700 K, and possibly as high as 3100 K during the accident³. The bulk of the previously molten region was determined to be a mixture of both ceramic and metallic phases; however, examples of entirely ceramic and metallic particles were also

present. An example of an entirely ceramic particle from the central region is shown in Figure 4 which is a back scattered electron (BSE) image. The BSE image represents a composite average of the atomic number of the elements in a particular phase where the degree of brightness is proportional to the atomic number of the element (e.g., the high atomic number uranium is brighter than the lower atomic number iron). Examination of this ceramic particle indicated that the light regions were uranium and zirconium oxides, and the dark regions were mixtures of iron, chromium, and aluminum oxides. An entirely metallic particle from the central, previously molten region is shown in Figure 5. The metallic matrix of this sample consisted of a nearly dendritic structure of iron-nickel-chromium with circular inclusions containing silver-indium-tin. Although some of the Ag-In-Sn inclusions contained voids, many contained a spherical particle of chromium oxide (Cr_2O_3).

An example of a particle from the central, previously molten region that contained both metallic and ceramic phases is shown in Figure 6. The ceramic phase is primarily oxides of zirconium and uranium with small amounts of iron and nickel oxides, and is similar to the entirely ceramic particles found both in the central region and in the upper debris bed. The bulk composition of the metallic phases in the central region (which comprised about 15% of the central region) consisted principally of Fe-Ni and Ag-In phases. Thus, the central region contained much more metal than the upper debris bed where no pure metallics were found. Metals from the stainless steel cladding of the control rods, the silver, indium, cadmium control materials, and the Inconel spacer grids (only the Inconel contains molybdenum), existed in abundance in the central, previously molten region.

UPPER CRUST

The upper crust consisted of two phases, a ceramic phase containing mostly fuel material components, and a metallic phase (which comprised about 25% of the upper crust) containing mostly structural and control rod material components. No intact fuel material was present. The peak temperature of the upper crust was at least 2700 K. A representative sample of the upper crust from core position K9 is shown in Figure 7. In this figure, the lighter colored phases are the metallic and the darker phases are the ceramic. The ceramic material in the upper crust was similar to the ceramic material in the central region and the ceramic material in the upper debris bed. However, the bulk composition of the metallic in the upper crust differed from the metallic in the central region. The metallic composition of the upper crust consisted primarily of three phases, namely Fe-Ni, Ag-In-U, and Ni-Sn. The upper crust contained considerably more metallic uranium than the central region and considerably less cadmium.

PERIPHERAL CRUST

The peripheral crust was similar to the upper crust except that there were remnants of intact fuel pellets that may have fallen into the peripheral crust from the upper debris bed, and the metallic phases in the crust were rich in zirconium as compared with the upper crust and the central region. The bulk composition of the metallics in the peripheral crust was similar to the lower crust, except the principal element in the peripheral crust was zirconium. Three phases existed in the metallic portion of the peripheral crust; namely Fe-Zr-Ni-Cr, Ag-In, and Zr-Ni-In. As is the case for the metallics in the upper crust and the previously molten central region, the constituents of the metallic region, with the exception of uranium, zirconium and cadmium, are all several times the core average. The high percentage of zirconium in the peripheral crust implies that this crust was formed at a time in the accident when the molten zirconium and control and structural material was flowing downward and freezing.

LOWER CRUST

The lower crust consisted of vertical fuel pellet stack columns surrounded by previously molten material as illustrated in Figure 8. Apparently during the early part of the accident, structural and control rod material flowed down the coolant channels, dissolved the Zircaloy cladding and flowed into the pellet/pellet interfaces and cracks in the pellets and froze forming the lower crust. The molten material was composed of a mixture of metallic structural and control rod components with relatively small (100 to 200 micron) UO_2 inclusions. It is clear that the eutectic between zirconium and iron and zirconium and nickel formed, and therefore, the previously molten metallic in the lower plenum reached at least 1400 K. Also, rounded edges of ceramic UO_2 particles that were carried downward in the melt indicate the onset of dissolution requiring that temperatures in the melt approach 2200 K. The metallic material in the lower crust contained an even higher percentage of zirconium than the peripheral crust. The composition of the three phases of the metallic material in the lower crust were Zr-Fe-Ni-Cr, Ag-In, and Zr-Ni-In.

STANDING FUEL ASSEMBLY STUBS

The standing fuel assembly stubs below the previously molten region varied in length from about 0.2 to 1.5 m, with the longer fuel assembly stubs located at the core periphery. Metallographic examinations indicate that the fuel rod stubs reached relatively low temperatures (below 1100 K) during the accident. The standing fuel assembly stubs supporting the previously molten region taken together with the intact peripheral fuel assemblies surrounding the upper void region comprised 44,500 kg.

Although no significant damage occurred to the fuel rod stubs, zirconium rich metallic material, similar to that in the lower crust, flowed downward inside of an empty instrument tube in Assembly G8. Thus, empty instrument guide tubes provide an escape path for the relocating molten metallic material and some of this material may have reached the lower plenum by this means.

LOWER PLENUM DEBRIS

The debris resting on the lower head accumulated to a depth of 0.75 to 1 m above the lowest head elevation, and to a diameter of 4 m. The spatial distribution of the lower plenum debris was neither uniform nor symmetric, which may be the result of debris redistribution caused by hydraulic forces during operation of the A-loop reactor coolant pump after relocation of the debris to the lower plenum. Particle sizes varied from large "rock" (up to 0.2 m) to granular particles (less than 0.1 mm). The particles were previously molten ceramic, $(U,Zr)O_2$, homogeneous and very porous⁴. A lower plenum ceramic particle is shown in Figure 9. These ceramic particles were similar in composition to the ceramic particles in the upper debris bed and the previously molten central region and to the ceramics in the upper and peripheral crusts. The peak temperature of the lower plenum debris varied between 2700 and 3100 K. Additional large volume samples from the lower plenum region are to be analyzed to confirm the results of radiological measurements that indicate that iodine, ruthenium and antimony exist in much lower concentrations in the lower plenum debris than in the upper debris bed.

CONCLUSIONS

Based on the metallurgical examination of TMI-2 core samples, the temperatures reached in the upper debris bed, the upper crust, the peripheral crust, the central previously molten region and the lower debris bed were all between 2700 and 3100 K. The ceramic material in these regions was similar, and consisted primarily of oxidized uranium and zirconium with lesser amounts of iron and nickel oxides precipitated on the grain boundaries of the U-Zr-O matrix. The temperature of the lower crust was greater than 1400 K and probably close to 2200 K.

The metallic material in the various parts of the central consolidated region varied depending on the region. The lower and peripheral crusts contained zirconium rich metallics that were principally zirconium, iron, nickel, and uranium with lesser amounts of silver, chromium, tin, indium, molybdenum and cadmium. Metallics in the central, previously molten region were primarily iron and nickel, with lesser amounts of silver, tin, chromium, molybdenum, indium and cadmium. Metallics in the upper debris bed contained a large fraction of uranium and were composed principally of iron,

uranium, nickel and zirconium with lesser amounts of silver, chromium, tin, indium, molybdenum, and cadmium. Only trace amounts of oxidized metallics were found in the upper and lower debris beds. These two regions were significantly depleted of both control and structural materials.

The temperatures and compositions of the various regions of the core are consistent with the following materials interactions that have been determined from both in- and out-of pile experiments⁵⁻⁷. As the temperature increases the silver, indium and cadmium control material melt at about 1100 K but are contained within the stainless steel cladding. The first melt to form that is free to flow downward results from the eutectic interaction between the Inconel spacer grids and the Zircaloy fuel rod cladding and Zircaloy guide tubes and stainless steel cladding of the control rods at about 1500 K. Once this first liquid is formed, additional eutectic (such as between the Inconel and Zircaloy reaction products and the stainless steel control rod cladding which would allow the already molten silver, indium and cadmium to join in the eutectic formation sequence) forms more rapidly due to enhanced atomic mobilities in the liquid as opposed to the solid state. The combination of liquid cladding, structural and control materials is then capable of flowing downwards. The temperature increases at a much faster rate when the oxidation of the zirconium in the cladding accelerates at around 1700 K and at about 2200 K dissolution of the UO_2 in the fuel rods begins. At about this same temperature, the stainless steel control rod cladding melting point is reached allowing control rods to fail at locations other than at spacer grids, releasing much more molten silver, indium and cadmium which interacts with the Zircaloy control rod guide tube as does the molten stainless steel. The complex mixture of liquid uranium, zirconium and tin from the fuel rods and the structural and control materials then flows downward apparently carrying UO_2 particles with the melt. When this complex mixture reaches the lower part of the core that is covered with water, it freezes forming a lower crust which grows to the periphery of the core and supports the upper part of the core which continues to increase in temperature. These interactions are consistent with the formation of the lower crust in the TMI-2 core, the depletion of control and structural materials in the upper plenum debris, the formation of the central molten region in the core, and the bulk compositions that were found in the various ceramic and metallic regions of the core. The absence of control and structural materials in the lower plenum debris samples is consistent with relocation of ceramic material from the central molten region of the core.

It is clear that the integrity of the lower head of the reactor pressure vessel was challenged by the relocation of 19,200 kg of molten core material to this region. The U. S. Nuclear Regulatory Commission, in partnership with the Organization for Economic Co-Operation and Development, has initiated a program to obtain and examine samples from the TMI-2 lower reactor pressure vessel head and from debris located adjacent to the lower head.

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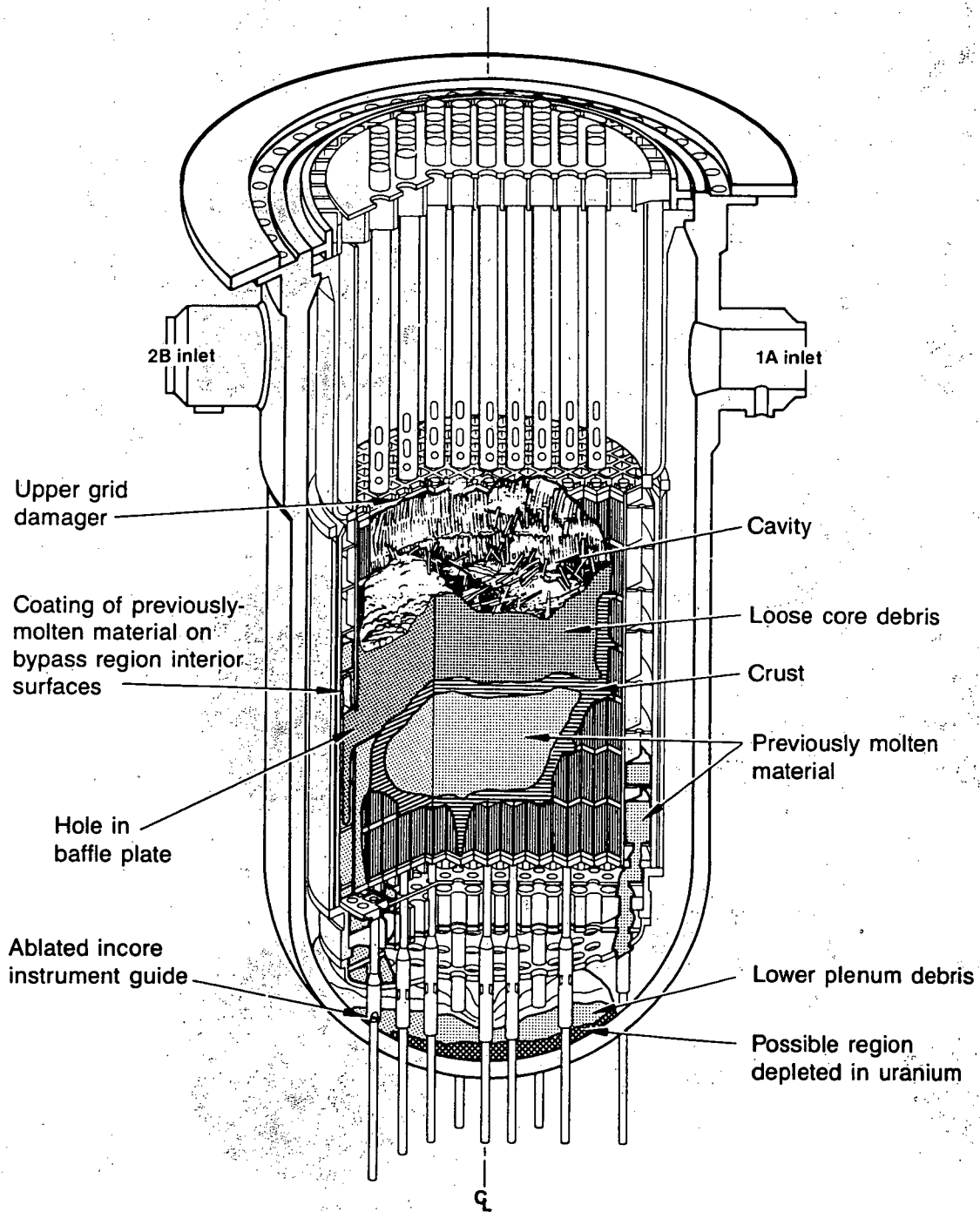


Figure 1. TMI-2 end state core condition.

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TMI-2 Core Debris Grab Samples

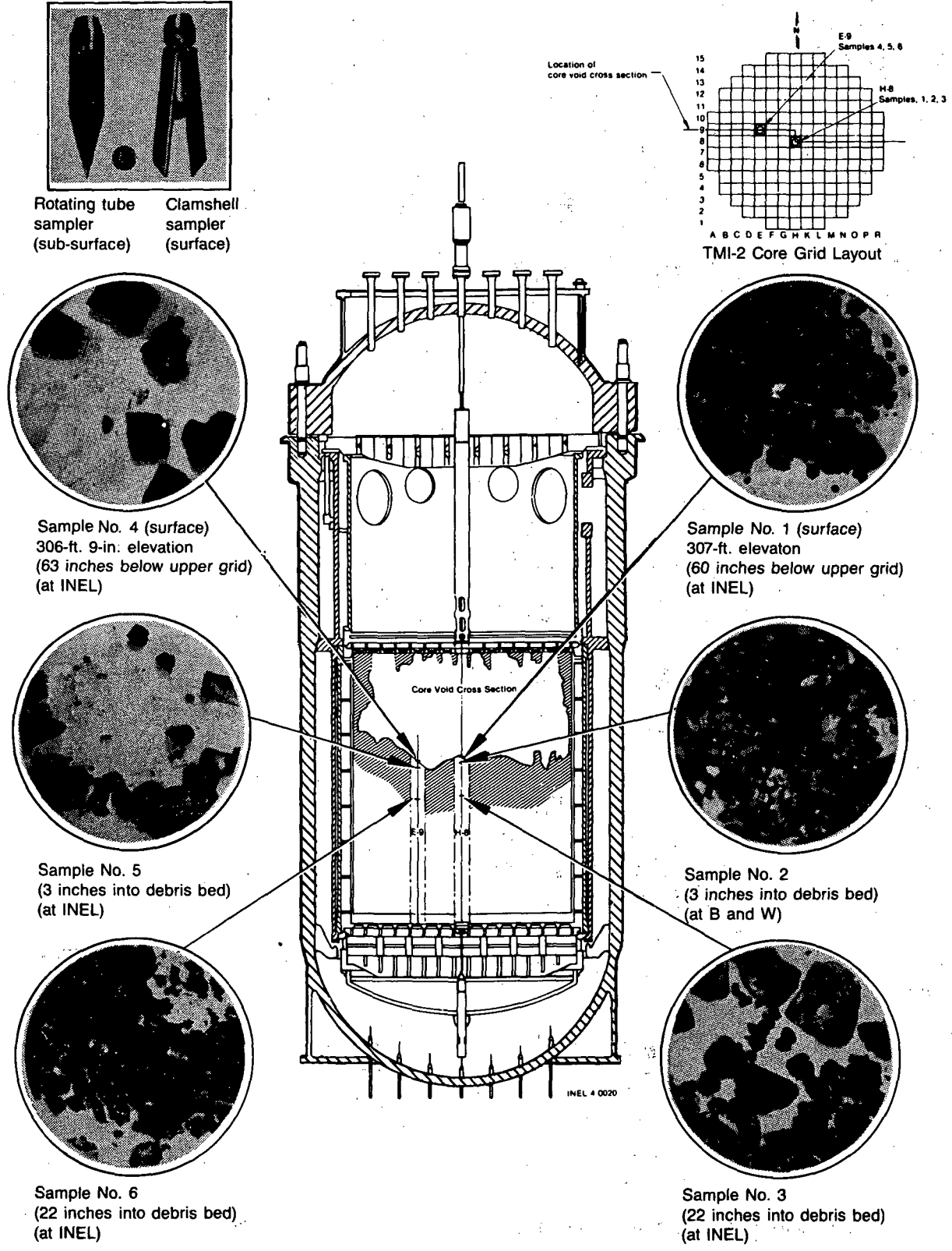
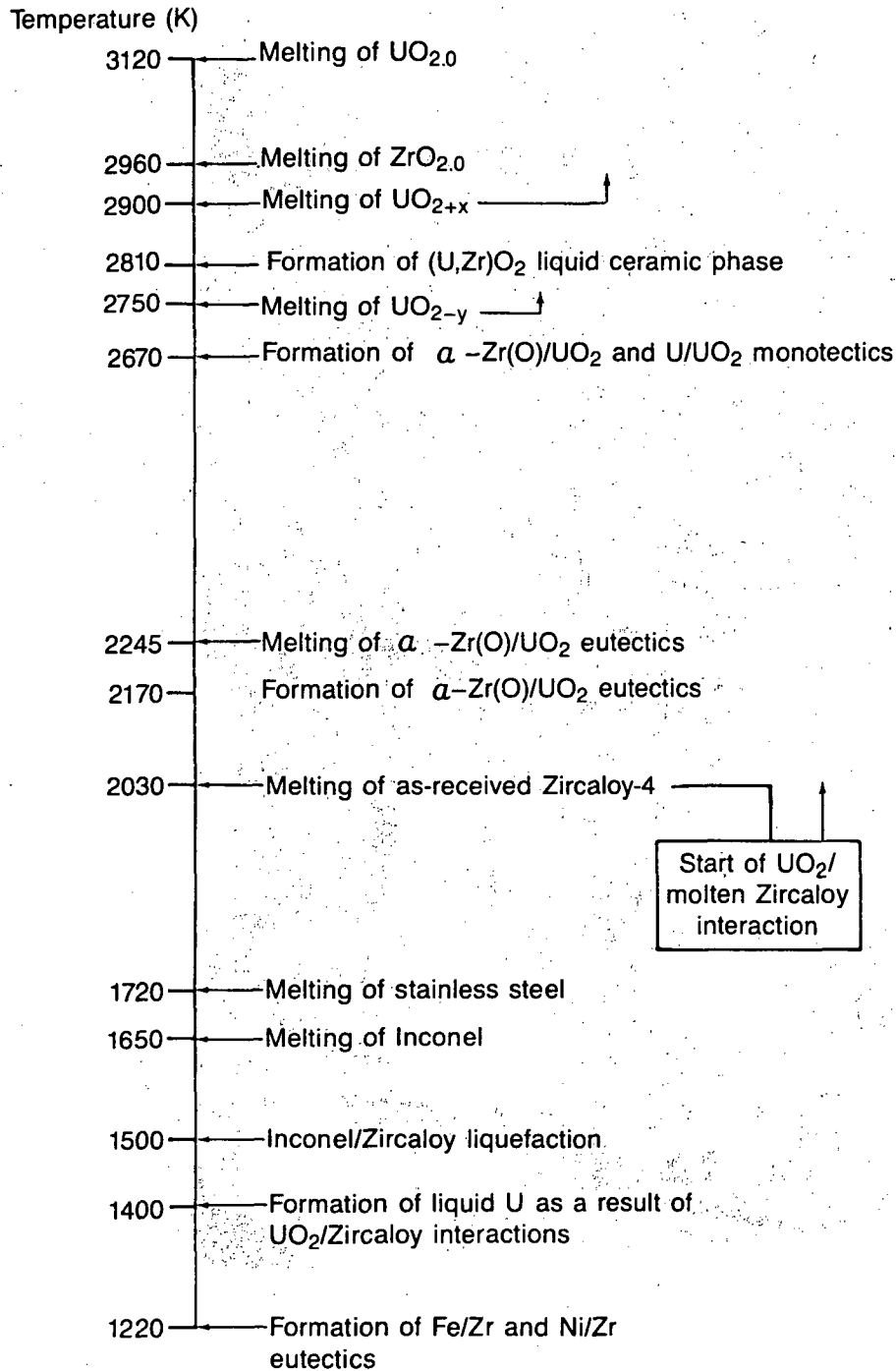


Figure 2. TMI-2 core debris grab samples.



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Figure 3. Temperature indicators for nuclear core materials.

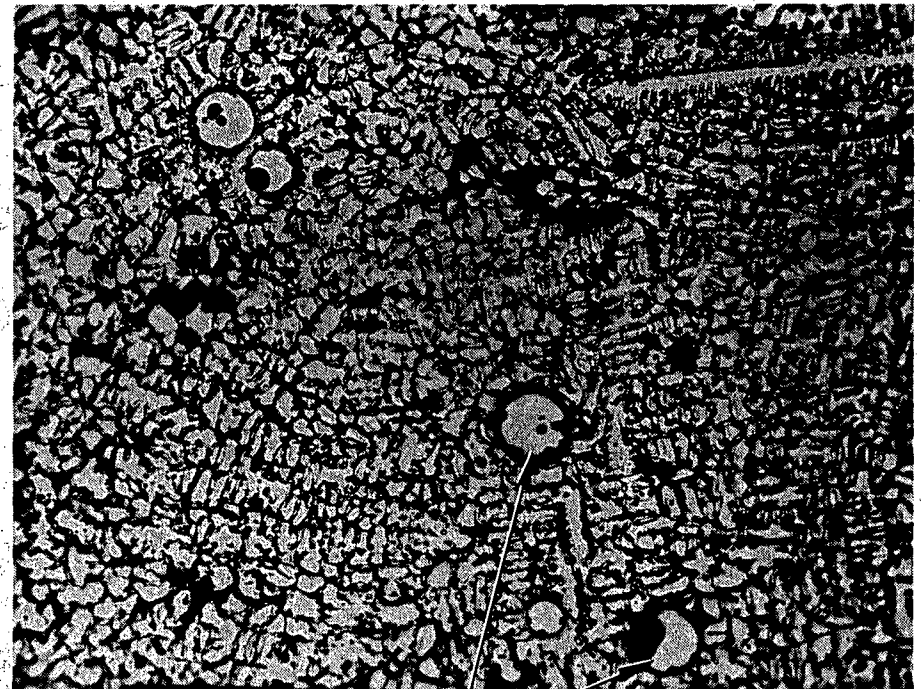


Examination
area

Figure 4. Core location K9 — ceramic particle (BSE image).



87M-94

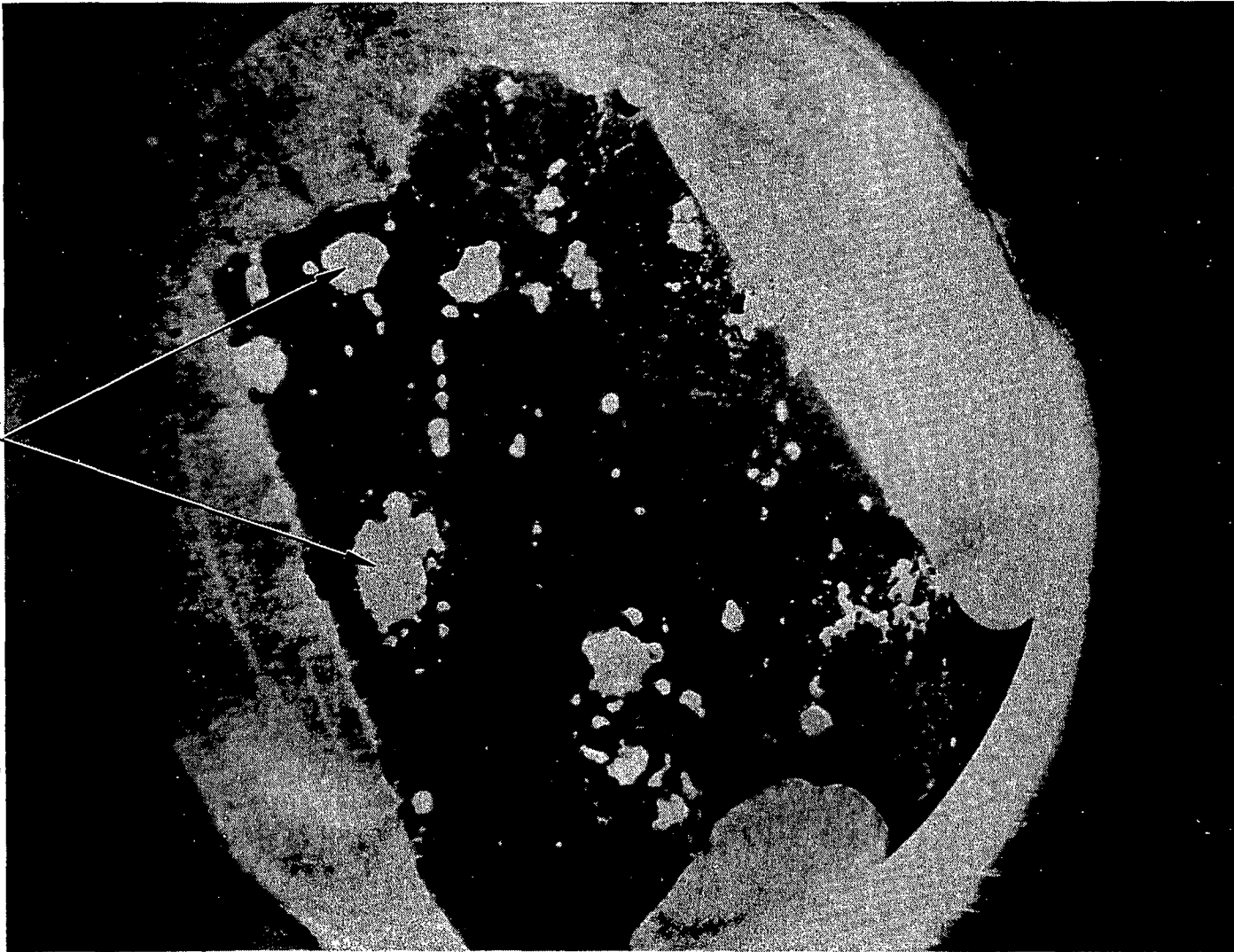


Silver inclusions

87M-327

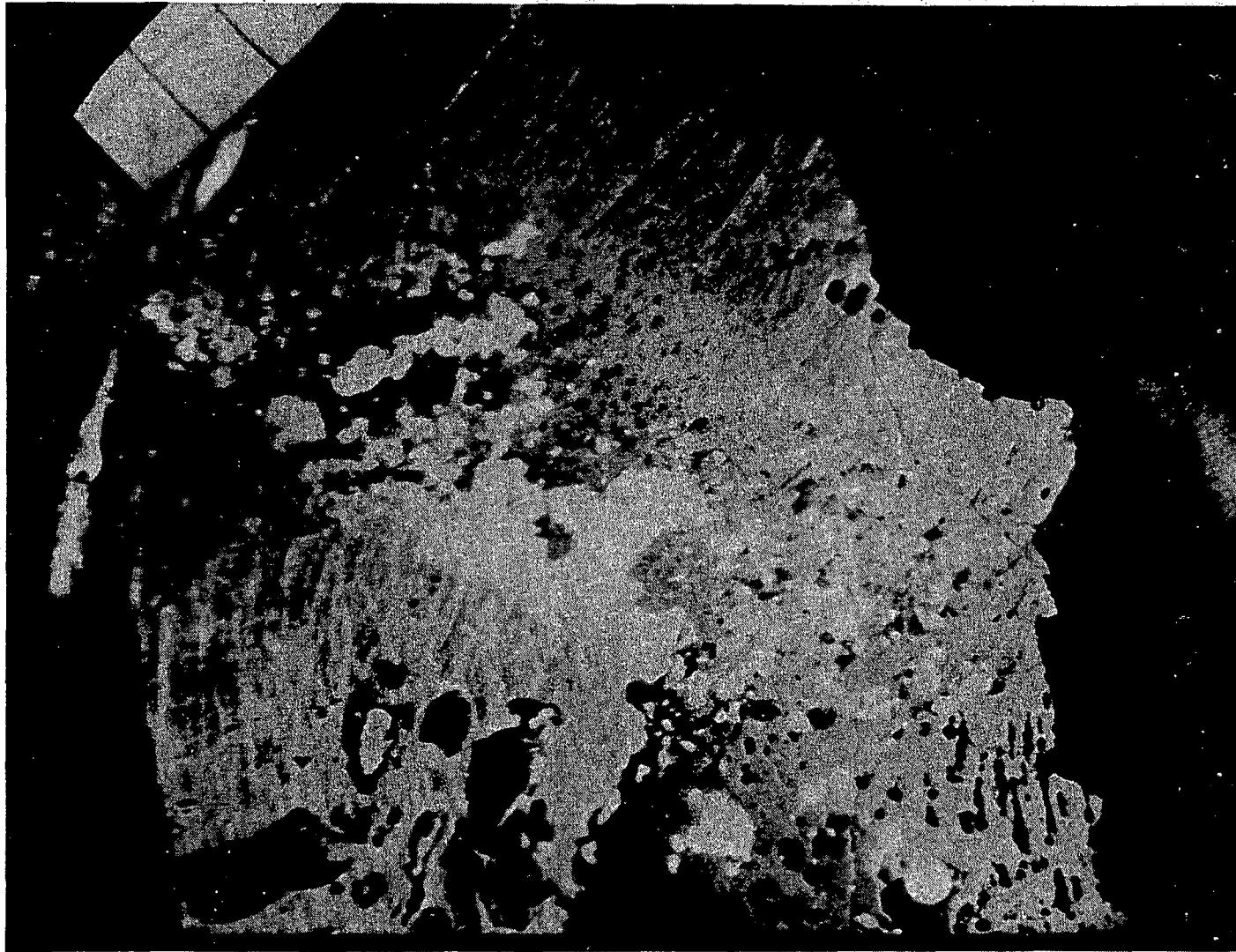
Figure 5. Core location 09 — metallic particle.

Metallic
inclusions



87m-100R

Figure 6. Ceramic particle with metallic inclusions (core location G12)



87m-404R

Figure 7. Core location K9 — upper crust.

Core Material Interaction and Cladding Dissolution

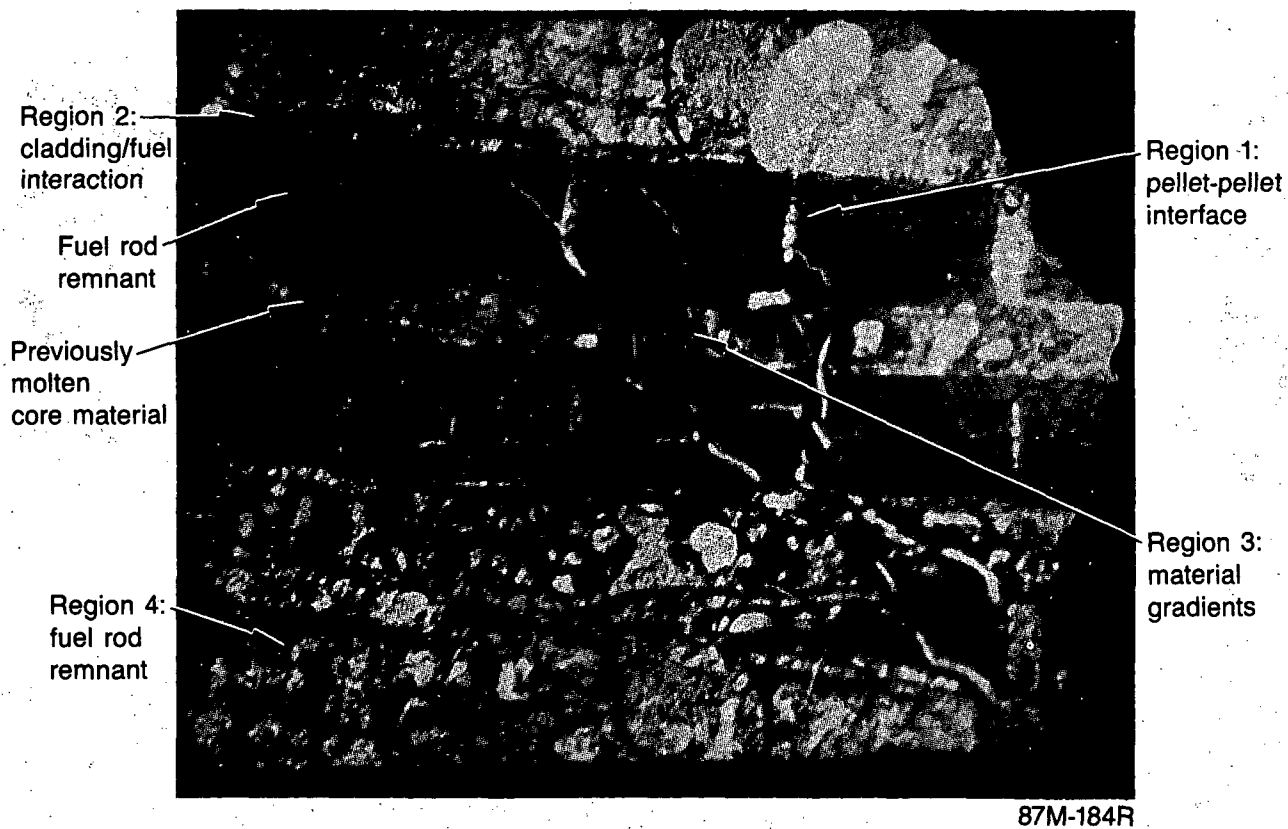
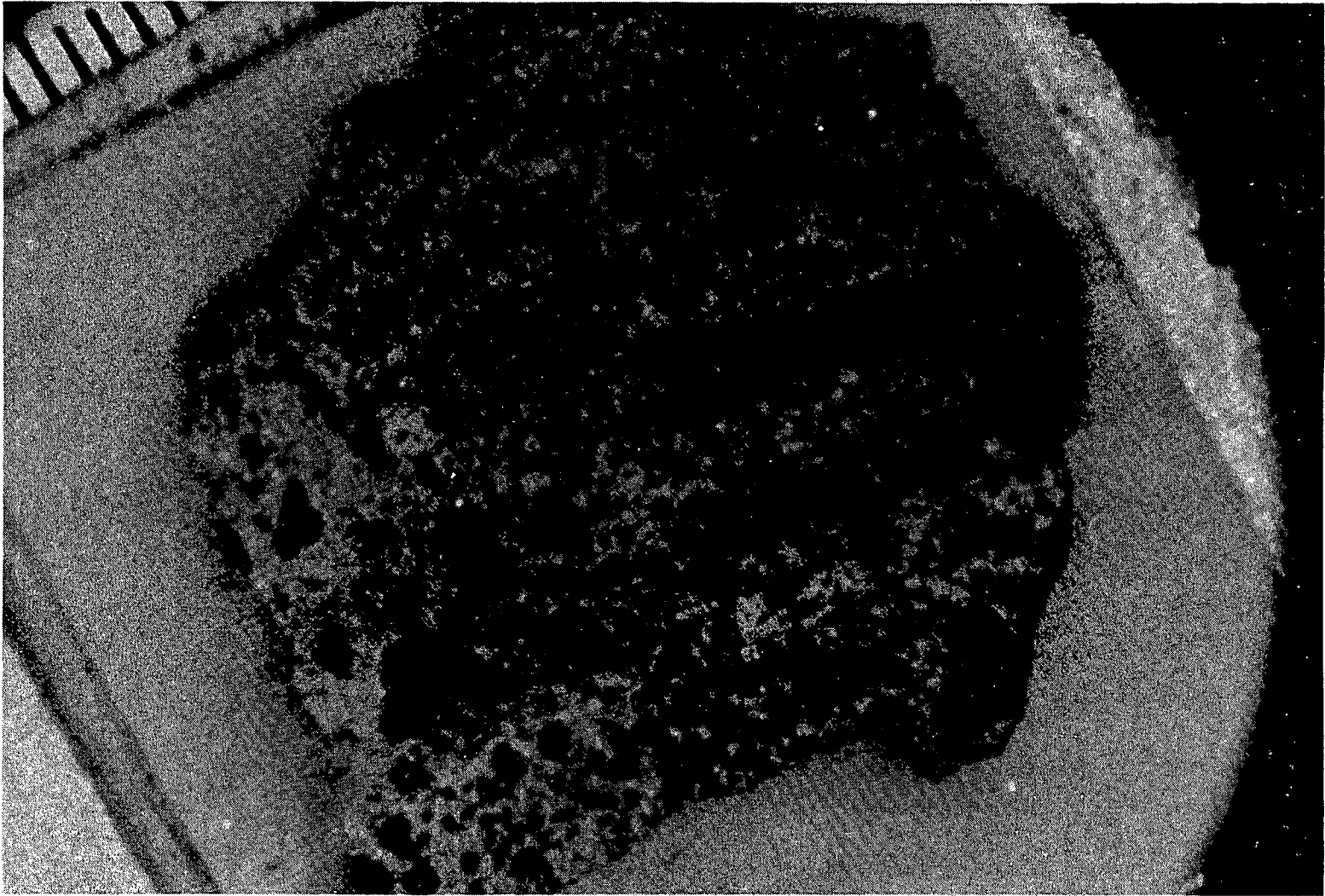


Figure 8. Lower crust from core position K9 illustrating previously molten metallic material frozen around fuel rods.



11-5-C Sampling Locations

Figure 9. Cross-section of lower plenum particle 11-5.