A Brief Review of Past INL Work Assessing Radionuclide Content in TMI-2 Melted Fuel Debris: The Use of ^{144}Ce as a Surrogate for Pu Accountancy

D. L. Chichester
S. J. Thompson

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Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

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EXECUTIVE SUMMARY

This report serves as a literature review of prior work performed at Idaho National Laboratory, and its predecessor organizations Idaho National Engineering Laboratory (INEL) and Idaho National Engineering and Environmental Laboratory (INEEL), studying radionuclide partitioning within the melted fuel debris of the reactor of the Three Mile Island 2 (TMI-2) nuclear power plant. The purpose of this review is to document prior published work that provides supporting evidence of the utility of using ^{144}Ce as a surrogate for plutonium within melted fuel debris. When the TMI-2 accident occurred no quantitative nondestructive analysis (NDA) techniques existed that could assay plutonium in the unconventional wastes from the reactor. However, unpublished work performed at INL by D. W. Akers in the late 1980s through the 1990s demonstrated that passive gamma-ray spectrometry of ^{144}Ce could potentially be used to develop a semi-quantitative correlation for estimating plutonium content in these materials.

The fate and transport of radioisotopes in fuel from different regions of the core, including uranium, fission products, and actinides, appear to be well characterized based on the maximum temperature reached by fuel in different parts of the core and the melting point, boiling point, and volatility of those radioisotopes. Also, the chemical interactions between fuel, fuel cladding, control elements, and core structural components appears to have played a large role in determining when and how fuel relocation occurred in the core; perhaps the most important of these reaction appears to be related to the formation of mixed-material alloys, eutectics, in the fuel cladding.

Because of its high melting point, low volatility, and similar chemical behavior to plutonium, the element cerium appears to have behaved similarly to plutonium during the evolution of the TMI-2 accident. Anecdotal evidence extrapolated from open-source literature strengthens this logical feasibility for using cerium, which is rather easy to analyze using passive nondestructive analysis gamma-ray spectrometry, as a surrogate for plutonium in the final analysis of TMI-2 melted fuel debris.

The generation of this report is motivated by the need to perform nuclear material accountancy measurements on the melted fuel debris that will be excavated from the damaged nuclear reactors at the Fukushima Daiichi nuclear power plant in Japan, which were destroyed by the Tōhoku earthquake and tsunami on March 11, 2011. Lessons may be taken from prior U.S. work related to the study of the TMI-2 core debris to support the development of new assay methods for use at Fukushima Daiichi. While significant differences exist between the two reactor systems (pressurized water reactor (TMI-2) versus boiling water reactor (FD), fresh water post-accident cooing (TMI-2) versus salt water (FD), maintained containment (TMI-2) versus loss of containment (FD)) there remain sufficient similarities to motivate these comparisons.

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A Brief Review of Past INL Work Assessing Radionuclide Content in TMI-2 Melted Fuel Debris: The Use of ^{144}Ce as a Surrogate for Pu Accountancy

1   INTRODUCTION

This report serves as a literature review of work performed at Idaho National Laboratory, and its predecessor organizations Idaho National Engineering Laboratory (INEL) and Idaho National Engineering and Environmental Laboratory (INEEL), studying radionuclide partitioning within the melted fuel debris of the reactor of the Three Mile Island 2 (TMI-2) nuclear power plant. The purpose of this review is to document prior published work that provides supporting evidence of the utility of using ^{144}Ce as a surrogate for plutonium within melted fuel debris. When the TMI-2 accident occurred no quantitative nondestructive analysis (NDA) techniques existed that could assay plutonium in the unconventional wastes from the reactor. However, unpublished work performed at INL by D. W. Akers in the late 1980s through the 1990s demonstrated that passive gamma-ray spectrometry of ^{144}Ce could potentially be used to develop a semi-quantitative correlation for estimating plutonium content in these materials.

The generation of this report is motivated by the need to perform nuclear material accountancy measurements on the melted fuel debris that will be excavated from the damaged nuclear reactors at the Fukushima Daiichi (FD) nuclear power plant in Japan, which were destroyed by the Tōhoku earthquake and tsunami on March 11, 2011. Currently, multiple research and development programs are underway to develop comprehensive, quantitative assay systems for analyzing the waste material that will be taken from these reactors. However, lessons may also be taken from prior U.S. work related to the study of the TMI-2 core debris to help inform this research and support the development of new assay methods for use at Fukushima Daiichi. While significant differences exist between the two reactor systems (pressurized water reactor (TMI-2) versus boiling water reactor (FD), fresh water post-accident cooing (TMI-2) versus salt water (FD), maintained containment (TMI-2) versus loss of containment (FD)) there remain sufficient similarities to motivate these comparisons.

The prior work cited in this report, and Dr. Akers unpublished prior work, can provide 'lessons learned' that may prove useful towards the answering the material accountancy challenges at FD.

1.1   Special Nuclear Material Accountancy of the Waste Materials from TMI-2

A challenge faced by GPU Nuclear Corporation (GPUN), the owner of TMI-2, during the post-accident cleanup activities was to comply with United States Nuclear

Regulatory Commission (NRC) regulations in section 10 of the U.S. Code of Federal Regulations part 70 (10 CFR 70), which required that all special nuclear material (SNM) at the plant be accounted for to the nearest gram weight.[1] The waste was destined for shipment to INEL, and the Department of Energy (DOE) also had similar requirements for receipt of the TMI-2 wastes. Many plans were proposed for achieving this goal. However, ultimately no formal SNM accountability on a gram basis was required at the time of material shipments from TMI-2 to INEL.

The NRC granted the TMI-2 plant owner an exemption wherein each shipment was covered by a DOE/NRC Form 741.[1] The overall plan centered on having GPUN perform a survey of post-defueling survey to determine the total quantity of SNM left at TMI-2 after all wastes were removed. This survey was then used to establish the basis for the quantity of SNM left at TMI-2. Separately, computer calculations were used to estimate the total SNM inventory in TMI-2 at the time of the accident.   The difference between these two values, less losses due to radioactive decay, were then used to estimate the SNM inventory present in the reactor waste materials ultimately shipped to INEL and elsewhere. This was the basis for the "one reactor core" concept. GPUN shipped "one reactor core" to INEL, less small amounts of material left at the facility that could not be removed from the structure or which were trapped in filters or other process equipment as "hold-up" losses.

1.2   Report Layout

Following this introduction, section 2 of this report provides a review of prior work published by research staff at INL dealing with the assessment of fission product locations and inventories in different materials and location of the TMI-2 reactor system. The goal of this section is to succinctly collate many different sources of information pertaining to this subject. In order to retain the original intent of the authors of these sources much of the text is taken directly, word for word, from the original publications. The start of each subsection in section 2 contains the references from which the subsection is extracted. (Note: For clarity and ease of reading, quotation marks have been omitted from this review report.)

The third section of this report attempts to extract the salient information from the prior work that is directly related to assessing the feasibility of using ^{144}Ce as a surrogate for Pu for post-accident melted fuel-debris plutonium accountancy. Section 4 then provides a summary of the available information on this topic.

2   SUMMARY OF PRIOR INL WORK RELATED TO ASSESSING TMI-2 FISSION PRODUCT LOCATIONS AND INVENTORIES

The following subsections are presented in chronological order according to the year the source reference documents were published.

2.1 Gamma-Ray Mapping of TMI-2 Fuel Debris

In 1988 INEL published the results of a research project[*] that developed a new method to map the 2-D spatial distribution of gamma-ray emitting radionuclides in high density reactor fuel and prior-molten materials.[2,3] The goal of this project was to demonstration the collection of this type of data and to show its usefulness for evaluating radionuclide retention and transport in fuel-debris materials. Prior to this project this type of data was collected by mechanical sampling followed by dissolution and radiochemical analysis.

2.1.1 Gamma-Ray Mapping System Description

The fuel-debris gamma-ray mapping system consisted of a high-resolution, high-rate, high-purity germanium (HPGe) gamma-ray detector, a slit collimator, and a scanning bed. The collimator was comprised of tungsten collimator blocks separate by about 55 cm, with a total tungsten thickness of approximately 20 cm. The detector was a located approximately 100 cm from the furthest collimator block; a tungsten shield was located behind the detector as a protective measure when analyzing high-dose-rate materials. Measurements were taken by placing a sample on the scanning bed and then taking successive gamma-ray measurements, translating and rotating the sample in-between each scan. A schematic of the system is shown in Figure 1. Gamma-ray spectral data was automatically analyzed and parsed using INL's GAUSS spectrum analysis code[†]; these results were then used as the input into a separate software code (the Donner Algorithms for Reconstruction Tomography) for final analysis and data-map generation. The system also included a photographic imaging system, allowing the final gamma-ray data to be overlaid as isoconcentration curves depicting radionuclide distributions onto a photograph of the test item.

[*] Note: The published titles of the papers describing this work use the phrase "tomographic examinations" in their titles. However, this project did not involve tomographic imaging as it is understood today.
[†] The GAUSS spectral analysis code is still maintained and used at INL, it is the basis for INL's current gamma-ray analysis software tools.
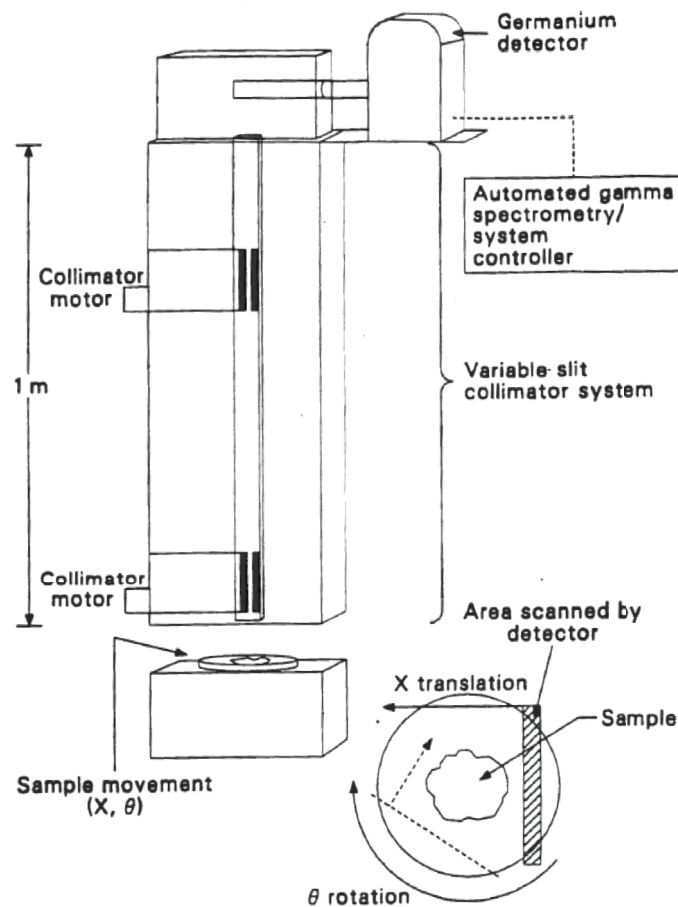
Figure 1 INL gamma-ray mapping system schematic.[2]

2.1.2   Use of the Gamma-Ray Mapping System

Reference 2 presents quantitative data of the use of the gamma-ray mapping system for the analysis of one TMI-2 sample (particle ID# 11-5-C) obtained from the lower reactor vessel head. This sample is an example of prior molten fuel (U/Zr) that reached approximately 2800 K (a temperature determined based on microstructural data of the sample). The report makes reference to the analysis of a second sample from the upper part of the reactor core, in an area where molten material flowed down and around relatively intact fuel rods, but no data is presented.

When the measurements were performed six radionuclides were measurable in these two samples.

- ^{60}Co: produced by neutron activation analysis of structural materials and expected to remain with, and be transported by, structural materials after melting.

- ^{106}Ru and ^{125}Sb: low volatile and moderate volatile, respectively, fission products expected to remain as metallics in the TMI-2 core rather than form oxides (due to large energy requirements for oxidation).

- ¹³⁷Cs: a highly volatile fission product that significantly contributes to the dose of some TMI-2 fuel debris but which is found in widely varying concentrations in different samples according to local heating and transport histories.
- ¹⁴⁴Ce(Pr) and ¹⁵⁴Eu: relatively non-volatile fission products that are expected to be retained quantitatively within fuel debris materials and provide an indicator of fuel mobility with the core.

Sample particle 11-5-C was a cross section of a large debris particle (6.2-cm diameter) that left the core as molten material and solidified on to the pile of fuel debris located on the lower head of the reactor. A photograph of the cross-section of this particle is shown in Figure 2. The sample gamma-ray measurements were taken over 37 steps across the face of the sample for 36 different angular rotations (a total of 1332 gamma-ray spectra). The spatial resolution of the distribution reconstructions was 0.19 cm, which was equivalent to the step distance.
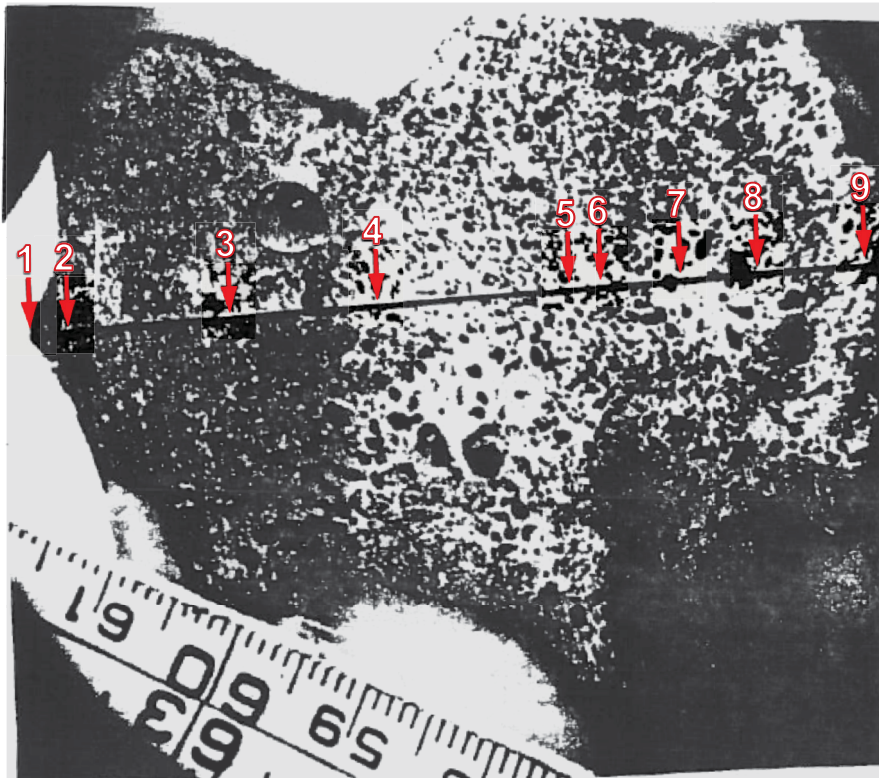
Figure 2 Photograph of the cross-section of particle 11-5-C used for gamma-ray scanning. Also shown are the locations where radiochemical analyses were made.[2]

In addition to gamma-ray scanning, samples were removed from this particle for radiochemical examination. Samples were taken from nine locations in particle 11-5-C, the sample location are indicted in Figure 2. The results of these analyses are presented in Table 1.

Table 1   Radionuclide and elemental concentration in reactor vessel lower head sample.[2]

|	Location/Concentration								
Radionuclide*	1	2	3	4	5	6	7	8	9
^{60}Co	<3.6E 0	4.3E 0	4.8E 0	6.8E 0	9.5E 0	9.9E 0	1.09E+1	1.58E+1	1.45E+1
^{90}Sr	1.10E+4	2.87E+4	–	3.25E+3	1.04E+3	4.69E+3	5.45E+3	7.6E+3	5.86E+3
^{106}Ru	9.5E 0	8.6E 0	1.26E+1	3.11E+1	1.72E+1	1.71E+1	2.44E+1	2.93E+1	2.88E+1
^{125}Sb	<2.2E 0	<2.0E 0	<2.4E 0	<3.7E 0	<5.1E 0	<6.2E 0	<7.6E 0	9.9E 0	1.00E+1
^{129}I	<8.2E−6	<2.0E−6	2.38E−6	7.72E−6	–	3.03E−5	2.89E−5	3.15E−5	2.15E−5
^{134}Cs	<5.5E−1	<2.2E 0	3.3E 0	7.1E 0	1.29E+1	2.21E+1	2.52E+1	3.06E+1	2.75E+1
^{137}Cs	2.55E+1	6.75E+1	1.12E+2	2.36E+2	4.36E+2	7.29E+2	8.63E+2	1.03E+3	9.27E+2
^{144}Ce	3.75E+2	3.70E+2	3.41E+2	4.03E+2	3.89E+2	4.40E+2	4.01E+2	4.33E+2	4.42E+2
^{154}Eu	4.81E+1	4.59E+1	4.38E+1	4.35E+1	4.09E+1	4.16E+1	3.88E+1	3.73E+1	3.51E+1
Element**									
Cr	–	2.5E−1	3.4E−1	3.7E−1	5.8E−1	5.9E−1	8.2E−1	1.11	1.14
Fe	–	8.7E−1	1.18	1.25	1.83	2.31	2.88	2.89	3.11
Mn	–	3.9E−2	5.3E−2	4.6E−2	6.3E−2	7.8E−2	<1.1E−1	8.8E−2	9.1E−2
Mo	–	6.0E−2	<5.0E−2	6.6E−2	5.2E−2	<1.0E−1	2.0E−1	<6.0E−2	1.5E−1
Ni	–	<4.8E−2	<6.0E−2	6.6E−2	1.7E−1	<1.2E−1	<2.3E−1	3.3E−1	2.4E−1
U	–	6.66E+1	6.29E+1	6.12E+1	6.45E+1	6.57E+1	6.37E+1	6.71E+1	6.04E+1
Zr	–	1.25E+1	1.15E+1	1.15E+1	1.25E+1	1.21E+1	1.11E+1	1.35E+1	1.31E+1

* μCi/gram on April 1,1986
** weight percent

P631−WHT−588−01

Examples of the results of this system for inspecting particle 11-5-C are presented in Figure 3 for ^{60}Co and in Figure 4 for ^{137}Cs. (*Note: these photos are taken from the best available copy of the original images from reference 2. Unfortunately, it is very difficult to recognize and follow all of the isoconcentration lines in these images.*)   In Figure 3 high concentrations of ^{60}Co are located on the right side of the image, in agreement with radiochemical sample locations 8 and 9.   In general, these locations were found to correspond to higher-porosity areas of the sample, suggesting a relationship between the degree of porosity and the structural material content in the fuel debris. Analysis of the radiochemistry results for ^{137}Cs and Figure 4 indicate a direct correlation between ^{137}Cs retention, porosity, and elemental content. Referring to Table 1, a similar correlation exists for ^{129}I.   More detailed analysis presented in reference 2 suggests that ^{137}Cs retention may be occurring in Fe and Cr grain boundaries of the fuel debris associated with the metallic phases.

Figure 3 Distribution of ^{60}Co in particle 11-5-C.[2]
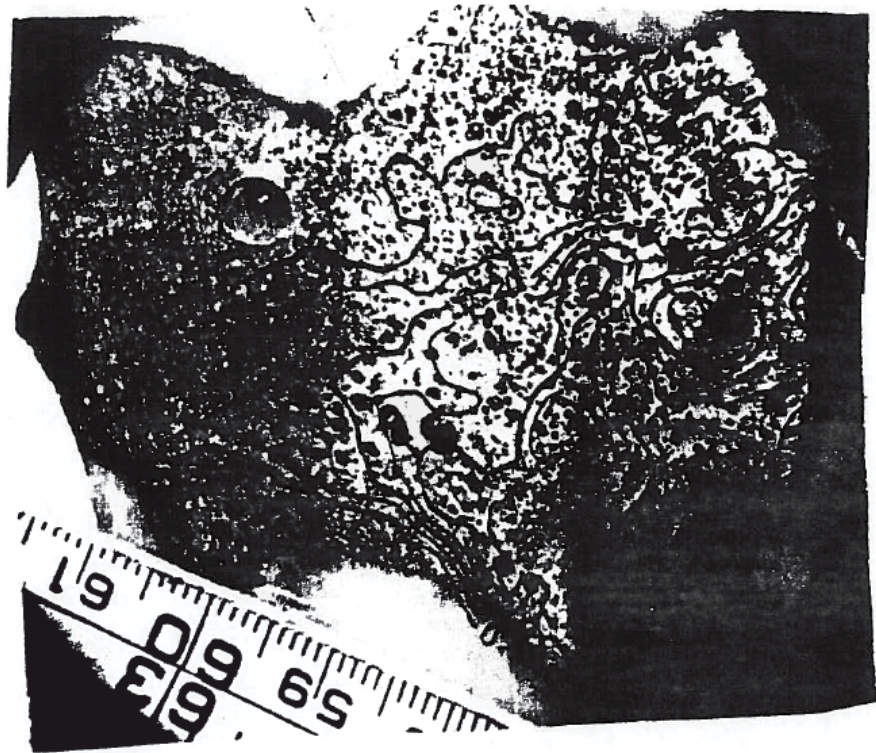
Figure 4 Distribution of ^{137}Co in particle 11-5-C.[2]

2.2 TMI-2 Core Debris Examinations: Physical and Microstructural Conditions

In 1990 INEL published the results of analyses of TMI-2 core debris samples to determine the temperatures that were reached during the accident, the material interactions that occurred, the bulk composition of the materials, the oxidation of the materials, and the retention of fission products in the core materials.[4] The key findings of this report are the following.

- **Temperature**
  - o 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 2800 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.
  - o The temperature of the lower crust was greater than 1400 K and probably close to 2200 K.

- **Composition**
  - o 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.
  - o Metallics in the central, previously molten region were primarily iron and nickel, with lesser amounts of silver, tin, chromium, molybdenum, indium and cadmium.
  - o 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.
  - o Only trace amounts of oxidized metallic were found in the upper and lower debris beds. These two regions were significantly depleted of both control and structural materials.

- **Interactions**
  - o As the temperature increases the silver, indium and cadmium control material melt at about 1100 K but are contained within the stainless steel cladding.
  - o 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 joint 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.

- o The temperature increases at much faster rate when the oxidation of the zirconium in the cladding accelerates at around 1700 K. 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.
- o The complex mixture of liquid uranium, zirconium and tin from the fuel rods and the structural and control materials 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
- o 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.

2.3 TMI-2 Core Debris Examinations: Radiochemical Analysis

In 1990 INEL published the results of analyses of TMI-2 core debris samples performed to characterize the distribution of core materials and fission products. Principal subjects included the relocation of core materials out of the reactor vessel, the formation of complex core material interaction products, cesium retention in prior molten material, and the retention of tellurium, antimony, and ruthenium in association with core materials.[5] The key findings of this report are the following.

- **Composition**
  - o During the accident much of the core structural materials relocated within the core volume. Zirconium and structural materials relocated to form the crusts and metallic inclusions present in the lower part of the reactor core. Almost half the zirconium in the upper part of the core relocated to help form these structures in the damaged core.
  - o The data suggests that other structural materials present in the core, primarily the Inconel grid spacers, contributed to the formation of these metallic crusts that surrounded the molten high temperature ceramic core.
  - o In the process of forming the crusts the metallic fission products ruthenium and antimony partitioned from the fuel material and relocated to the metallic crusts and inclusions with the structural constituents.

o In addition to the structural materials the less volatile control rod materials, silver and indium, also concentrated in the metallic crusts.

- **Fission Product Distributions**
    o The fission product distribution and retention data indicate no release of low volatiles from the prior molten fuel, and that none was released from the reactor vessel except as particulate material carried out of the vessel by the reactor coolant.
    o Strontium-90 is primarily retained in the fuel material similar to the low volatiles although there is some evidence of mobility within the fuel melt.
    o The inventory data indicate that the ^{106}Ru and ^{125}Sb were retained in the lower core with the bulk of the inventory located in the central core region crusts and metallic inclusions at concentrations up to 20 times those found in intact fuel.
    o For the high volatiles, the ^{85}Kr data indicate accountability of approximately 90% of the core inventory with relatively low uncertainties as compared to the more reactive species such as cesium and iodine.
    o The ^{129}1 and ^{137}Cs data indicate almost complete retention in the reactor building with only small releases to the auxiliary building where the activity was retained principally in the reactor coolant bleed tanks and the letdown demineralizers.

2.3.1   TMI-2 Fission Product Inventory – Low Volatile Elements

The distribution of fission products in the TMI-2 reactor system was estimated based on representative examples of each group of fission products that were measurable at the time the fission product measurements were made (4-7 y after the accident). The low volatility fission products include elements from the noble metals, some rare earths and actinides, tetravalents, and early transition elements. Generally, the oxides of these elements have low volatilities; however, some (e.g., LaO or CeO) have lower boiling points that do the elements. The only radionuclides from this group which were measurable during the core examination program were cerium/praseodymium-144, europium-154, and europium-155. The isotope ^{144}Ce is produced principally by direct beta decay from fission, whereas the europium radionuclides are primarily produced by neutron activation of fission produced species. The radionuclides produced by neutron activation have greater associated uncertainties with their data as the neutron activation makes their production dependent on neutron flux and spectrum, and consequently, on the core location of the fuel material.

A listing of the distributions of the three low-volatility fission products measured in different parts of the TMI-2 reactor system is provided in Table 2. The low volatiles were not released from the fuel and only a small amount was physically transported to the reactor coolant system as intact or melted fuel. As might be expected, the fuel material distribution is similar to the mass distribution, as it is the primary constituent present in the reactor vessel. The ^{154}Eu and ^{155}Eu data indicate similar trends to those seen for the

^{144}Ce, except for variations in the measured retention values for ^{154}Eu which are due to the relatively large uncertainty associated with radionuclides produced by neutron capture. The ^{144}Ce inventory was found completely within the solid parts of the core and the resolidified ex-vessel debris, including the intact fuel (30%), sub-core debris and crust areas (51.3%), and in-core debris intermixed with intact fuel and core structures (24.1%). No significant evidence of ^{144}Ce transport via water or steam to the balance of the reactor system was observed.

Table 2 Low-volatility fission product distribution in the TMI-2 reactor system.[5]

Fission product repositories	Fission product distribution Percent of inventory [a]		
	^{144}Ce	^{154}Eu	^{155}Eu
Ex-vessel			
Containment atmosphere, basement, and tanks	0.01	[b]	[b]
Reactor coolant system	[b]	[b]	[b]
Auxiliary building	[b]	[b]	[b]
In-vessel			
Upper reactor plenum	[b]	[b]	[b]
Upper core debris – A	26	30	24
– B [c]	20	19	19
Upper crust region	1.4	2.0	1.6
Consolidated region	24	32	22
Lower crust	5.9	7.9	5.1
Intact fuel rods	30	30	30
Upper core support assembly	3.4	4.5	[d]
Lower core support assembly	4.7	6.3	[d]
Lower head-reactor vessel	16	21	[d]
Total	105	122	110 [d]

a) Percentage of the total amount of the fission product inventory calculated from comparisons with ORIGEN2.

b) Insignificant amount (< 0.1 wt%) based on the measurement data.

c) Two sets of bulk sample measurements were performed on the upper debris bed. The A series was performed on samples from near the center of the core at a variety of depths whereas the B series were bulk samples from near the bottom of the debris bed. For the totals, the B series data were used.

d) Measurements not performed for this radionuclide at this core location. The total shown value in parenthesis is a total which assumes the same distribution as ^{154}Eu for the repositories where measurements were not performed.
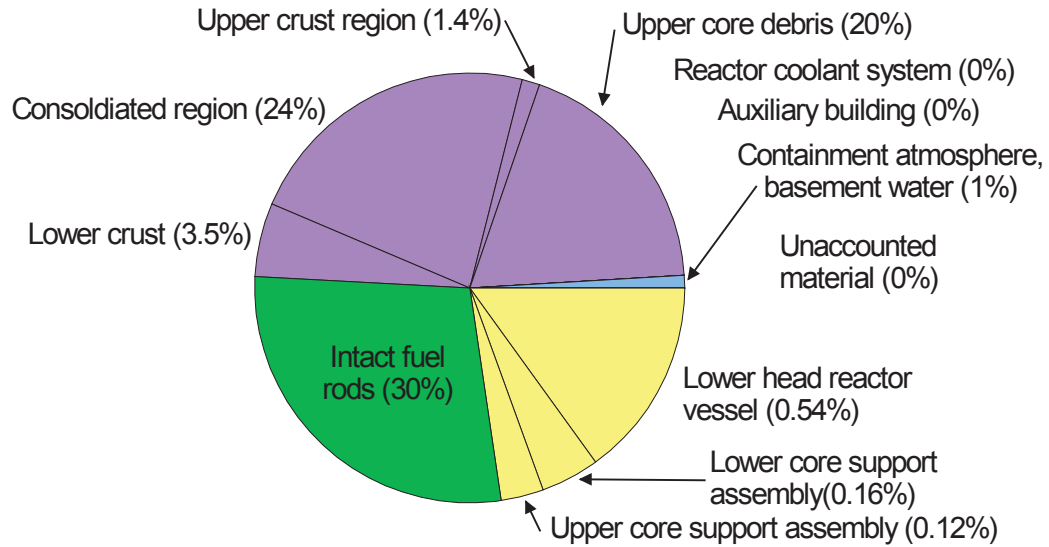
Figure 5 End state ^{129}I distribution in the TMI-2 reactor system.[5]

2.3.2   TMI-2 Fission Product Inventory – Medium Volatile Elements

The medium volatility fission products measurable at the time the core examinations were made were ^{90}Sr, ^{106}Ru, and ^{125}Sb. The strontium is expected to be present as an oxide in the melted fuel whereas the ruthenium and antimony are expected to remain as metallic due to high oxidation potentials that are not obtainable in a steam environment. A listing of the distributions of the three medium-volatility fission products measured in different parts of the TMI-2 reactor system is provided in Table 2.

Table 3   Medium-volatility fission product distribution in the TMI-2 reactor system.[5]

Fission product repositories	Fission product distribution Percent of inventory [a]		
	^{90}Sr	^{106}Ru	^{125}Sb
Ex-vessel			
Containment atmosphere, basement, and tanks	2.1	0.5	0.7
Reactor coolant system	1	[b]	0.2
Auxiliary building	0.1	[b]	0.7
In-vessel			
Upper reactor plenum	[b]	[b]	[b]
Upper core debris – A	23	14	13
– B [c]	19	16	24
Upper crust region			
ceramic	0.73	0.8	0.5
metallic	[b]	3.8	7.8
Consolidated region			
ceramic	8.3	2.2	3.1
metallic	[b]	9.0	6.9
Lower crust			
ceramic	4.5	5.7	7.4
metallic	[b]	24	36
Intact fuel rods	30	30	30
Upper core support assembly	3.9	0.23	0.22
Lower core support assembly	5.3	0.32	0.30
Lower head-reactor vessel	18	1.1	1.0
Total	93	94	119

a) Percentage of the total amount of the fission product originally present calculated from comparisons with ORIGEN2.
b) Insignificant amount (<0.1 wt%) based on the upper plenum measurements.
c) Two sets of bulk sample measurements were performed on the upper debris bed. The A series was performed on 16 cm 3 samples from near the center of the core at a variety of depths whereas the B series were bulk samples from near the bottom of the debris bed. The data provide a range. For the totals, the B series data were used.

The measurement data for ^{90}Sr, the radionuclide in this category that exhibited the least mobility, show that that only a small amount of this radionuclide (< 3.5% of the core inventory) was released from the reactor vessel. Almost all of the strontium released was retained in the reactor building with only 0.1% released to the auxiliary building where it was retained in the reactor coolant bleed tanks. In the reactor vessel, the distribution is similar to the fuel material distribution and micro-examinations indicate that the ^{90}Sr was retained in the prior molten fuel material. The examination data indicate

that there was minor solubility of the ^{90}Sr in the metallic region samples.[6]   However, the data indicate that this phenomena is restricted to the reactor vessel and that there is almost complete accountability for the ^{90}Sr, similar to the less volatile ^{144}Ce.

The ^{106}Ru data follow a similar distribution to that observed for ^{125}Sb and indicates that much of the ^{106}Ru is found in association with metallic samples at concentrations 4 – 12 times those of intact fuel. Of particular importance to the accident scenario is the timing of the transport of the metallic material to form the crusts and the metallic inclusions in the consolidated region.   These data suggest that the metallic materials had to have been transported past the melting fuel when these radionuclides were being released to allow the accumulation of these radionuclides in the metallic melt.

High concentrations of ^{125}Sb are present in the prior molten metallic portions of the upper and lower crusts and the consolidated region.   The metallic sample concentrations of ^{125}Sb are 6 – 20 times those found in intact fuel. These data and the metallurgical data suggest that the ^{125}Sb remained in a metallic state. The inventory data indicate that 62% of the core inventory of ^{125}Sb was retained in the lower core.

2.3.1   TMI-2 Fission Product Inventory – High Volatile Elements

The highly volatile radionuclides measurable at the time measurements were made were some noble gases (primarily ^{85}Kr), cesium (^{137}Cs) and iodine (^{129}I).   These radionuclides were present as gases during the high temperature portion of the accident and were primarily released from the reactor vessel when the reactor fuel melted. Their distributions through the reactor system are presented in Table 4. Specific details of the distribution of ^{129}I and ^{137}Cs are shown in Figure 6 and Figure 7.

The TMI-2 measurements indicated that significant relocation and transport of about half of the high-volatile radionuclides occurred, moving these materials to the reactor coolant system and consequently, to the reactor building basement. The inventory data for ^{85}Kr is reported to be accurate to within 5-10. Examinations for noble gases in the upper debris bed, namely ^{85}Kr, indicate retentions similar to those measured for ^{129}1 (approximately 6% of core inventory). These data are a relatively accurate measure of the retention of noble gases in the TMI-2 system and indicates retention of almost the entire core inventory.

Table 4 High-volatility fission product distribution in the TMI-2 reactor system.[5]

Fission product repositories	Fission product distribution Percent of inventory [a]		
	^{137}Cs	^{129}I	^{85}Kr
Ex-vessel			
Containment atmosphere,	[b]	[b]	54
basement water	47	(47) [c]	[b]
Reactor coolant system	3	1	[b]
Auxiliary building	5	7	[b]
In-vessel			
Upper reactor plenum	[b]	[b]	[b]
Upper core debris – A	5.3	5.9	6
– B [d]	4.3	5.3	[b]
Upper crust region	0.41	0.27	[b]
Consolidated region	0.77	2.1	[b]
Lower crust	1.4	3.5	[b]
Intact fuel rods	30	30	30
Upper core support assembly	0.46	0.12	[b]
Lower core support assembly	0.63	0.16	[b]
Lower head-reactor vessel	2.1	0.54	[b]
Total	95	97	91

a) Percentage of the total amount of the fission product originally present calculated from comparisons with ORIGEN2.

b) Insignificant amount (< 0.1 wt%) based on the measurement data.

c) Wide range of concentrations and quantity of debris would indicate percentage of total inventory that would be greater than the core inventory. Consequently, the ^{137}Cs inventory is considered representative of the quantity of iodine deposited in the reactor building.

d) Two sets of bulk sample measurements were performed on the upper debris bed. The A series was performed on 16 cm a samples from near the center of the core at a variety of depths whereas the B series were bulk samples from near the bottom of the debris bed. The data provide a range. For the totals, the B series data were used.
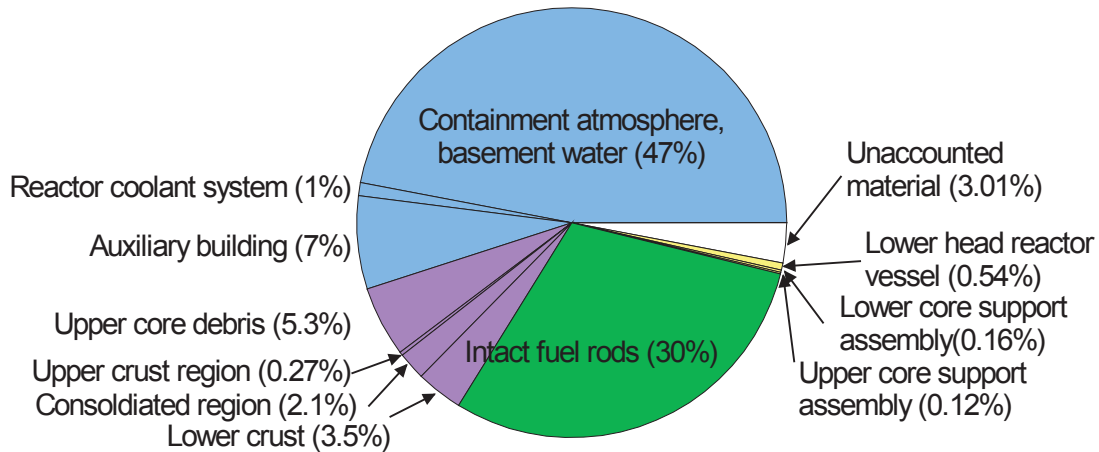
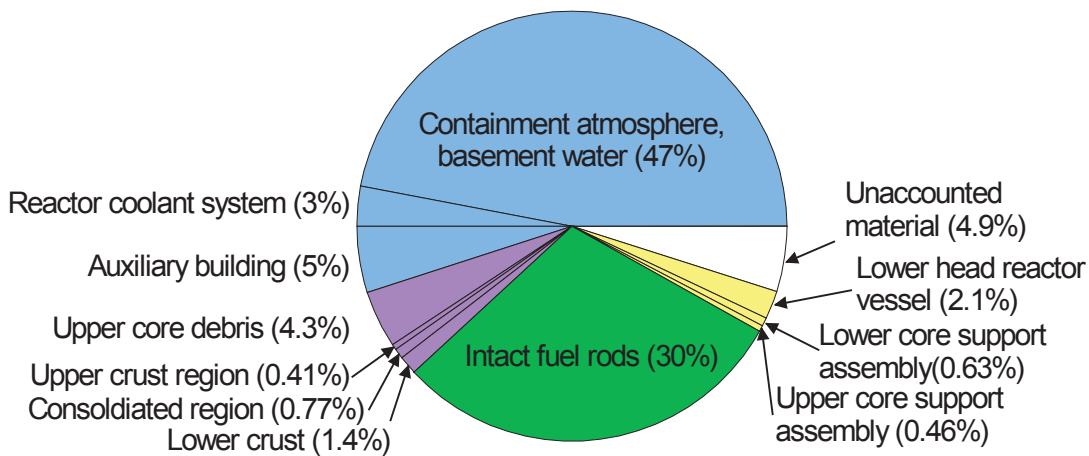**Figure 6 End state [129]I distribution in the TMI-2 reactor system.[5]**

Figure 7 End state ^{137}Cs distribution in the TMI-2 reactor system.[5]

The uncertainties associated with the [129]I data are significantly greater than those associated with the noble gas data as [129]I is highly reactive and plates out on surfaces making accurate assessments of the inventory difficult. Measurements of radioiodine concentrations in the reactor building basement, the major repository for iodine outside the reactor core, indicates that radioiodine reacted with particulate debris in the reactor building basement water and concentrated in the loose debris deposited on basement surfaces. Since there was a wide range of iodine concentrations in the basement sediment, and since the quantity of sediment in the basement is not well known, an assumption was made to estimate the same retentions for iodine in the basement as was observed for cesium. This was justified on the premise that the transport of these two radionuclides in solution would be similar, while also recognizing that cesium is less reactive than iodine and that much of the ^{137}Cs remained in solution, was more easily measurable, and had lesser uncertainties associated with its data.

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Other key findings of the high-volatile fission product distribution in the TMI-2 reactor system include the following observations.

- Similar distributions were indicated for both ^{129}1 and ^{137}Cs in the TMI-2 reactor vessel.

- The data for the upper core debris indicates that about 20% of the original inventories of both ^{129}1 and ^{137}Cs were retained in partial fuel pellets and deposited on surfaces in the debris bed. In the lower parts of the core almost all ^{129}1 was released from the molten fuel regions and there was complete retention in the partial fuel assemblies beneath and around the molten region.

- Outside the reactor vessel, the 137Cs data indicate almost complete retention of the cesium in the reactor building with 5% released to the auxiliary building. This activity was retained in the reactor coolant bleed tanks and the letdown demineralizers. The ^{137}Cs data listed in Table 4 indicate low retention in the prior molten material with almost all ^{137}Cs being released from the fuel material. However, data from the lower core region indicates the retention of up to 20% of the original inventory of the fuel retained in the prior molten material.

- There is evidence from autoradiography and gamma spectroscopy examinations that cesium was retained in association with structural materials deposited in grain boundaries in the fuel.[2]

- The total inventory of ^{137}Cs present in the lower core is quite low at about 2.6%, this indicates significant transport and release of ^{137}Cs to the reactor vessel from this part of the reactor core.

2.3.2 General Summary of TMI-2 Core Debris Radiochemical Analyses

The following overall observations were made from the TMI-2 core debris radiochemical analyses.[5]

- The composition data indicate that during the accident much of the core structural materials relocated within the core volume.
    - o Zirconium and structural materials relocated to form the crusts and metallic inclusions present in the lower part of the reactor core.
    - o Almost half the zirconium in the upper part of the core relocated to help form these structures in the damaged core.
    - o The data suggests that other structural materials present in the core, primarily the Inconel grid spacers contributed to the formation of these metallic crusts that surrounded the molten high temperature ceramic core.

- In the process of forming the crusts, metallic fission products, ruthenium and antimony, partitioned from the fuel material and relocated to the metallic crusts and inclusions with the structural constituents.

- In addition to the structural materials, the less volatile control rod materials, silver and indium also concentrated in the metallic crusts.
- The fission product distribution and retention data indicate no release of low volatiles from the prior molten fuel and that none was released from the reactor vessel except as particulate material carried out of the vessel by the reactor coolant.
- Strontium-90 was primarily retained in the fuel material, similar to the low volatiles, although there is some evidence of mobility within the fuel melt.
- The inventory data indicate that the ^{106}Ru and ^{125}Sb were retained in the lower core with the bulk of the inventory located in the central core region crusts and metallic inclusions at concentrations up to 20 times those found in intact fuel.
- For the high volatiles, the ^{85}Kr data indicate accountability of approximately 90% of the core inventory with relatively low uncertainties as compared to the more reactive species such as cesium and iodine.
- The ^{129}1 and ^{137}Cs data indicate almost complete retention in the reactor building with only small releases to the auxiliary building where the activity was retained principally in the reactor coolant bleed tanks and the letdown demineralizers.

2.4 TMI-2 Core Debris Examinations: Second Look

In 1994 INEL published a second set of analyses reviewing the TMI-2 core debris materials, this time focusing on the debris from the damaged reactor's lower head area.[7] This paper further-refined observations made in the previous paper on the subject (references 4 and 5) and included additional information about the partitioning and transport of the rare-earth elements Ce and Eu within the core.

The key findings of this report *pertaining to radionuclide transport* in the TMI-2 reactor core are the following.

- Radiochemical analyses of the TMI-2 core lower-head debris indicate that the debris was composed of approximately 70 wt% uranium, 13.75 wt% zirconium, and 13 wt% oxygen.
  - o The remaining constituents are the elemental constituents of stainless steel and Inconel core components that probably melted during the relocation of debris to the lower head.
  - o The composition accounts for approximately 97 wt% of the debris.
- The examinations suggest that much of the high-volatile radionuclide content had volatilized out of the debris before the solidification of the molten debris and thus left primarily medium- and low-volatile components in the debris bed.
- The small amount of interconnected porosity and the nonreactive nature of the solidified ceramic indicate that leaching and other release mechanisms were insignificant.

- **Low-volatility fission products:**
  - The concentrations of Ce measured in the companion samples indicate that nearly all this radionuclide was retained.
  - Similarly, analyses suggested that most of ^{154}Eu was also retained in the core debris. However, considerable uncertainty remains in post-accident estimation of the starting inventory of ^{154}Eu in the TMI-2 core prior to the accident.
- **Medium-volatility fission products:**
  - Analysis of the data showed that ^{90}Sr retention in the TMI-2 lower-head core debris area ranged from 48 to 96%, which indicates that this radionuclide was mobile and was not fixed in the fuel melt with the low-volatile radionuclides.
  - The low retention of ^{125}Sb in the companion samples probably resulted from the partition of metallic antimony (unoxidized because of the high potential required to oxidize the element) from the oxidic uranium melt in the upper core region. As a consequence, the melt that relocated to the lower head was low in ^{125}Sb content. (In previous core examinations, high concentrations of ^{125}Sb were found in metallic samples from the upper core region.)[8]
- **High-volatility fission products:**
  - The radionuclide ^{137}Cs was measurable in all samples at retentions substantially lower than those predicted with ORIGEN2 for undamaged fuel. However, higher retentions (18%) were found in the northeast quadrant of the lower-head area.
  - It is not known why high levels of this radionuclide, as well as medium- and low-volatile radionuclide concentrations, existed in the northeast region.
- A summary of the key measured fission products found within different quadrants of the TMI-2 core lower-head area is presented in Table 5.

Table 5 High-volatility fission product distribution in the TMI-2 reactor system.[7]

	Radionuclide retention, %		
Radionuclide	Southeast (1-9)	Southwest (1-11)	Northeast (1-12)
^{90}Sr	48	47	96
^{125}Sb	1.9	1.1	5.6
^{137}Cs	3.6	1.3	18
^{144}Ce	91	85	97
^{154}Eu	83	84	80

Retention is calculated on the basis of the uranium content of the sample material as determined from the elemental analysis results. Results have been corrected for burnup and show a reduction of almost a factor of 2 in the inventory of ^{154}Eu and ^{125}Sb. Radionuclide concentration data are in reference 9.

3   POTENTIAL USE OF ^{144}Ce AS AN INDICATOR OF PLUTONIUM IN DAMAGED-CORE FUEL DEBRIS

In intact fuel ^{137}Cs is often used as a burnup indicator and, as such, a measurement analogue for plutonium in gamma-ray based NDA measurements.[10] However, in breached fuel subject to accident conditions such as those that occurred at TMI-2, the utility of ^{137}Cs for this purpose is dramatically reduced. This is due to the high-volatility of the element cesium and the facts that when fuel melts it changes phases, undergoes mixing, is exposed to high pressure and temperature water and steam, and is exposed to aggressive physical agitation promoting mixing and dissolution. This was shown above in Figure 7, where over 50% of the estimated TMI-2 ^{137}Cs inventory was found to be transferred from the fuel and fuel debris to other parts of the reactor's containment as contaminated water and air.

In contrast, analysis of the debris from the TMI-2 core and reactor has shown that the low-volatility radionuclide ^{144}Ce tends to move and relocate with the bulk fuel even while in the molten phase of an accident. Cerium remains nonreactive during this process, and also following the fuel transition from liquid back to the solid phase. Because of this, the use of ^{144}Ce as an analogue for plutonium in support of NDA safeguards material accountancy measurements has been proposed.

3.1 ^{144}Ce Decay and Gamma-Ray Emissions

Thermal-fission of ^{235}U results in the production of isobar fission products with A = 144 with a probability of approximately 5.5%.[11] The radioisotope ^{144}Ce has a half life of 284.9 days.[12] It decays via β^- emission to ^{144}Pr; low-energy gamma-rays of 133.5 keV and 80.1 keV are produced in this transition 23.5% of the time while 76.5% of these decays produce no gamma-rays. This decay scheme is presented in Figure 8. Due

to the low energy of these radionuclides, and the very high intensity of low-energy photon background associated with spent fuel, it is not practical to measure the gamma rays associated with the decay [144]Ce without radiochemical separation.
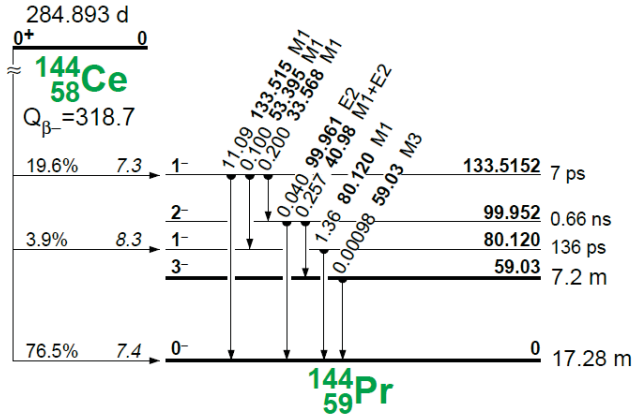

Figure 8 Decay scheme for [144]Ce.[12]

The radioisotope [144]Pr (praseodymium) has a half life of 17.28 m.[12] It decays via β⁻ emission to [144]Nd. While the branching ratio for gamma-ray production in this reaction is low, there are three gamma rays with energies greater than 500 keV and production probabilities greater than 1%. The decay scheme of [144]Pr is shown in Figure 9, the energies and yields of the gamma rays produced during the decay of [144]Pr to [144]Nd are shown in Figure 10. The half-life of the isotope [144]Nd is >2.3 × 10^{15} years. No gamma-ray emissions are practically observed from its decay in spent fuel.

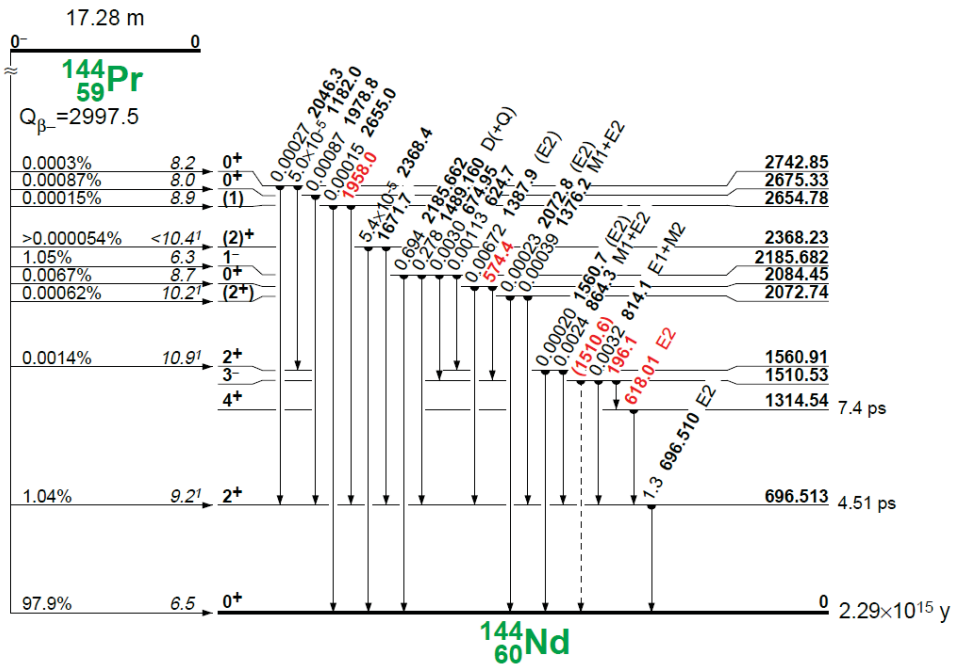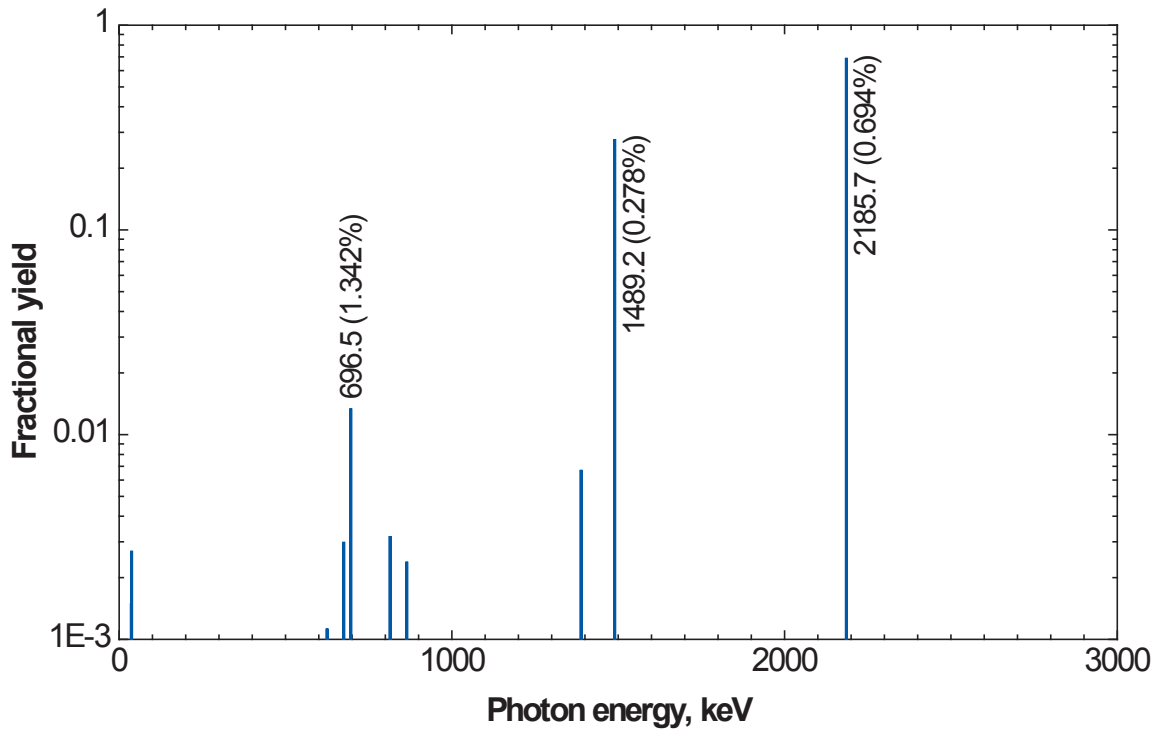**Figure 9 Decay scheme for [144]Pr.[12]**

Figure 10 Most prominent photons from the decay of ^{144}Pr to ^{144}Nd.[13]

For comparison, thermal-fission of ^{235}U produces the isobar 137 with a yield of 6.19%.[11] The decay of ^{137}Cs yields the short-lived isomer ^{137m}Ba, which decays to the ground state of ^{137}Ba with the emission of a 661.7 keV gamma ray with a probability of 85.1%. Normally in spent fuel, the very-strong gamma-rays from the decay of ^{137}Cs (and ^{134}Cs) dominate a gamma-ray spectrum. However, in spent fuel aged more than a few hundred days, the high-energy 2185.7 keV gamma-ray from ^{144}Ce decay is plainly evident. In fuel aged more than a couple of years, the lower energy 696.5 keV gamma rays from the decay chain of ^{144}Ce are also clearly observed. Examples of these observations are presented in Figure 11.
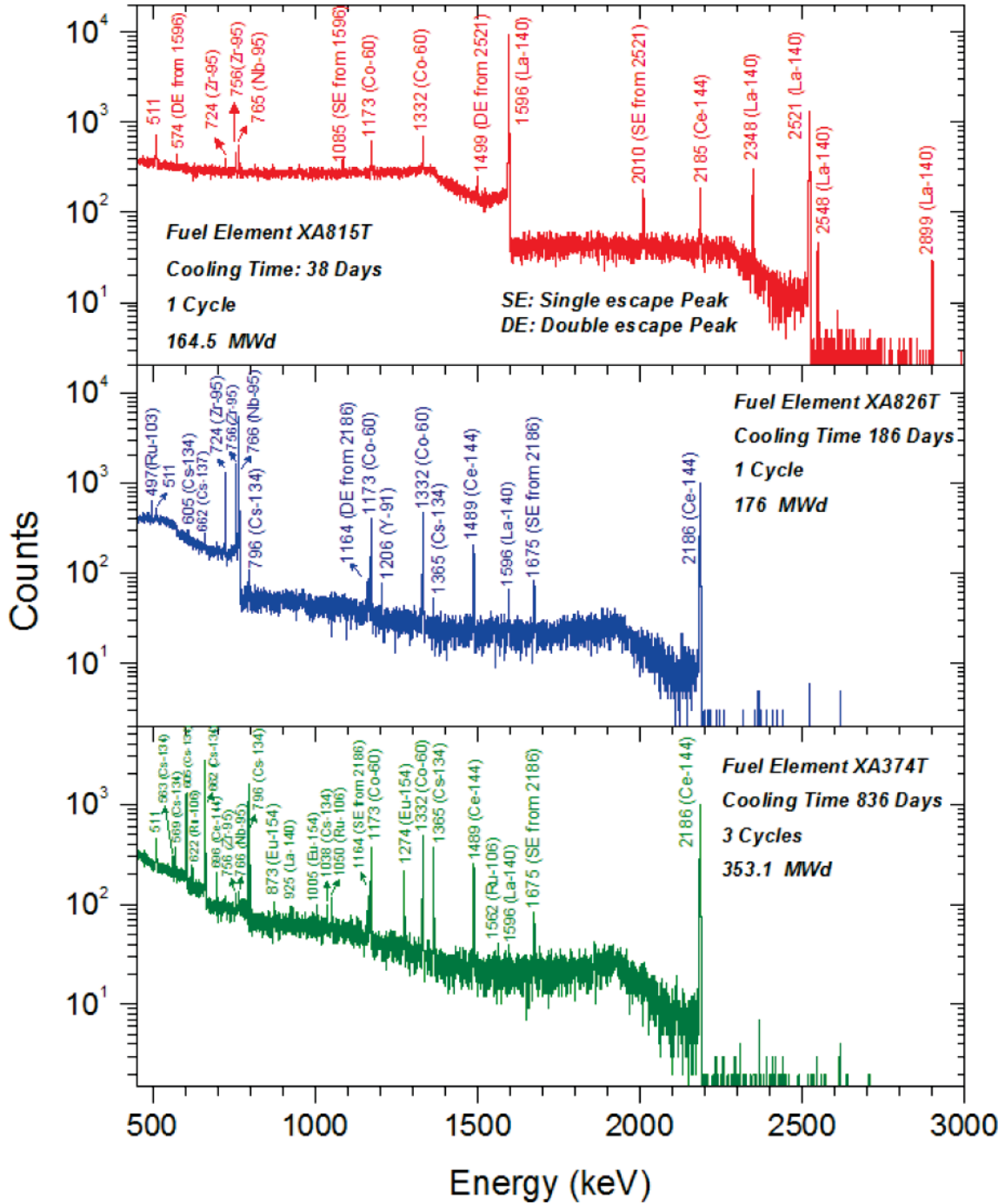
Figure 11 Gamma-ray spectra from spent fuel INL's Advanced Test Reactor (ATR) aged for 38 days (red/top), 186 days (blue/middle), and 836 days (green/bottom).[10]

3.2 Comparing ^{144}Ce and Pu

Work at INL focused on studying materials from the TMI-2 reactor system over the past two decades suggests that gamma-ray spectrometry of ^{144}Ce in melted fuel debris can be used as a surrogate for assaying plutonium.[14] Unfortunately, no empirical results have been published to demonstrate this technique, or to quantify its performance. Absent this evidence, it is therefore necessary to examine supporting evidence that might help substantiate the efficacy of this concept.

- **The utility of surrogates**
  - o Elemental surrogates have been successfully used to track the presence of other radioisotopes at TMI-2.[15]
  - o Cobalt-60 was found to effectively serve as a surrogate for ^{63}Ni transport at TMI-2.
  - o Cobalt-60 was found to effectively serve as a surrogate for ^{99}Tc ^{63}Ni transport at TMI-2.
  - o Antimony-125 was found to effectively serve as a surrogate for ^{129}I ^{63}Ni transport at TMI-2.
  - o Cerium oxide is well known as a useful chemical surrogate for plutonium oxide in fuel-cycle research and development.[16]

- **Melting and boiling points**
  - o The melting and boiling points of metallic plutonium are comparable to those of cerium, both of which are significantly higher than most other fission products in spent fuel (Figure 12).

Figure 12 Melting and boiling temperatures of some elements in spent fuel.

Volatility	Element	Melting Point, °C	Boiling Point, °C
Low	Cerium	795	3443
	Europium	826	1529
Medium	Strontium	777	1382
	Ruthenium	2334	4150
	Antimony	630.6	1587
High	Cesium	28.4	671
	Iodine	113.7	184.3
	Krypton	-157.5	-153.2
	Plutonium	*639.4*	*3228*

- **Reactivity and oxidation**
  - o Cerium is reactive in air, quickly forming an oxide. However, cerium oxide is highly nonreactive. The melting temperature for cerium oxide is 2400 °C while its boiling point is 3500 °C.

o Plutonium is reactive in air, quickly forming both oxides and hydrides. However, in the presence of water, or water vapor, hydride formation is inhibited in favor of oxidation. The melting temperature for plutonium oxide is 2400 °C while its boiling point is 2800 °C.

- **Minimal discovery of Pu in the TMI-2 demineralizers**
  o Prior reports indicate that only trace quantities of cerium were found in the water or atmosphere of the TMI-2 reactor system.
  o Similarly, very small quantities of plutonium were found in the TMI-2 water or atmosphere. This is demonstrated, for example, by the low amount of transuranic content found in the TMI-2 demineralizers.[17]

- **Minimal discovery of transuranic content (compared to the core inventory) in TMI-2 non-core wastes**
  o Prior reports of TMI-2 wastes consistently refer to ^{90}Sr and ^{137}Cs as the two most significant radionuclides pertaining to the disposal of waters, purification resin columns, and sludges from TMI-2.[18]

However, the use of cerium as an absolute surrogate for plutonium in post-accident reactor debris cannot be taken to be an always-useful tool. Prior reporting has suggested, for example, that cerium may be present in some instances where little or no transuranic material exists. This was found to be true in lower-activity conditions where trace quantities of transuranic would be estimated.[19]

4   SUMMARY

Following the nuclear reactor accident at TMI-2 in 1979 significant effort was spent towards studying the accident. These activities were multidisciplinary in nature and included chemistry, chemical engineering, metallurgy, mechanical engineering, and nuclear engineering. An important part of this research involved destructive radiochemical analyses and nondestructive analyses (primarily based on gamma-ray spectrometry) of the melted fuel debris from the reactor. Because of this work sophisticated post-accident narratives have been developed that describe how the accident evolved over time, and when, where, why, and how fuel and damaged fuel debris moved and was relocated throughout the TMI-2 reactor system.[20]

The fate and transport of radioisotopes in fuel from different regions of the core, including uranium, fission products, and actinides, appear to be well characterized based on the maximum temperature reached by fuel in different parts of the core and the melting point, boiling point, and volatility of those radioisotopes. Also, the chemical interactions between fuel, fuel cladding, control elements, and core structural components appears to have played a large role in determining when and how fuel relocation occurred in the core; perhaps the most important of these reaction appears to be related to the formation of mixed-material alloys, eutectics, in the fuel cladding.

Because of its high melting point, low volatility, and similar chemical behavior to plutonium, the element cerium appears to have behaved similarly to plutonium during the evolution of the TMI-2 accident. Anecdotal evidence extrapolated from open-source

literature strengthens this logical feasibility for using cerium, which is rather easy to analyze using passive NDA gamma-ray spectrometry, as a surrogate for plutonium in the final analysis of TMI-2 melted fuel debris. Unpublished prior work related to this topic communicated by one of the original NDA measurement specialists tasked with performing the original inventory assessments for the TMI-2 melted fuel debris (Dr. D. W. Akers) substantiates the usefulness of this technique. Further work is needed to record and publish this prior work. Also, further work is needed in order to develop upper and lower uncertainty estimates for the applicability of using Ce, specifically ^{144}Ce, for this purpose, and to delineate the strengths, weaknesses, and limitations of the approach.

The TMI-2 reactor accident bares superficial similarities to the FD reactor accident that occurred in Fukushima, Japan, in 2011. It is possible the use of cerium as a surrogate for plutonium inventory at FD might also provide a convenient approach to support nuclear material accountancy. However, to validate this approach it is clear that a substantial multidisciplinary research program will be needed to address the unique conditions found in the FD reactors and to account for the differences in accident progress and evolution that occurred there.

5 OTHER LITERATURE RELATED TO THIS TOPIC BUT NOT CITED HERE

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