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TECHNIQUES AND METHODS IN NUCLEAR MATERIALS TRACEABILITY*

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TECHNIQUES AND METHODS IN NUCLEAR MATERIALS TRACEABILITY*

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ABSTRACT

HEU AND PLUTONIUM OF UNKNOWN ORIGIN

HEU SAMPLE Isotopic signatures and correlation methods were applied to determine the origins of HEU and plutonium samples reported as illicit trafficking in nuclear materials. In the HEU case, the isotopic concentrations reported in weight percent (w/o) were 1.078, 87.768, 0.210, and 10.944 for U-234, U-235, U-236, and U-238, respectively. The isotopic signature of U-236 at the product level of about 0.210 percent indicates that in the enrichment process the initial feed U-236 concentration could have ranged in the 0.004 to 0.007 percent level, and the depleted uranium stream could have U-235 tails ranging between 0.2 and 0.3 percent. The U-236 feed concentration level is indicative of uranium-reprocessing streams from very low-burnup fuels. The reported U-234 and U-236 isotopics indicates that the HEU sample is the product of a gas diffusion isotopic separation process of a uranium product stream from reprocessed low-burnup fuels.

PLUTONIUM SAMPLE The measured concentration of the Pu-242 isotope at the level of 0.75 weight (w/o) percent in the plutonium vector, 0.17, 87.56, 10.78, 0.809, 0.688 percent of Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, respectively, as reported in the plutonium sample, is indicative of natural uranium fuel irradiated in the extreme outer regions of the rector core. In the outer regions of the core, the spectrum is softer, the Pu-239 generation is much reduced, and the transmutations of the higher plutonium isotopes are increased. The reported Pu-241 concentration at the 1 percent level compared to the normally anticipated level of about 2 percent, which is consistent with the low burnup of natural uranium, indicates that the sample is representative of irradiated natural uranium fuel reprocessed some 15 years ago. However, in the outer-region of the core, the Pu/U ratio is extremely low (low flux level) and for Pu-239 at about 87 percent, the burnup is also very low. The amount of plutonium produced in this region could therefore be insignificant.

INTRODUCTION

The nonproliferation community is currently addressing concerns that the access to special nuclear materials may increase the illicit trafficking in weapons-usable materials from civil and/or weapons material stores and/or fuel cycles systems. Illicit nuclear traffic usually involves reduced quantities of nuclear materials perhaps as samplings of a potential protracted diversionary flow from sources to users. To counter illicit nuclear transactions requires the development of techniques and methods in nuclear material traceability as an important phase of a broad forensic analysis capability. The International Conference on Nuclear Smuggling Forensic Analysis held during November 7-9, 1995 at Lawrence Livermore National Laboratory (Ref. 1), involved a broad spectrum of international experts including intelligence and law enforcement representatives to discuss techniques for handling and analyzing nuclear materials in illicit nuclear trafficking incidents.

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TRACEABILITY OF NUCLEAR MATERIALS OF UNKNOWN ORIGIN The isotopic signatures and correlation methods developed at Argonne were applied to determine the origins and/or the traceability of the highly-enriched uranium (HEU) and plutonium oxide samples reported as illicit trafficking in nuclear materials.

HEU OF UNKNOWN ORIGIN The analysis of the HEU reported in Nucleonics Week, February 16, 1995 and in Ref. 2, indicated the existence of traces of the U-236 isotope.

The isotopic concentrations reported in weight percent were 1.078, 87.768, 0.210, and 10.944 for U-234, U-235, U-236, and U-238, respectively. Referring to the U-235 and U-236 correlation curve in Fig. 1, the concentration of the U-236 isotope would be expected to range at a level of several weight percent, for HEU fuel irradiated from 93 percent to 87.7 weight percent in an HEU light water research reactor. The reported trace level of U-236 suggested an enrichment process product with a trace concentration of U-236 in the feed uranium stream.

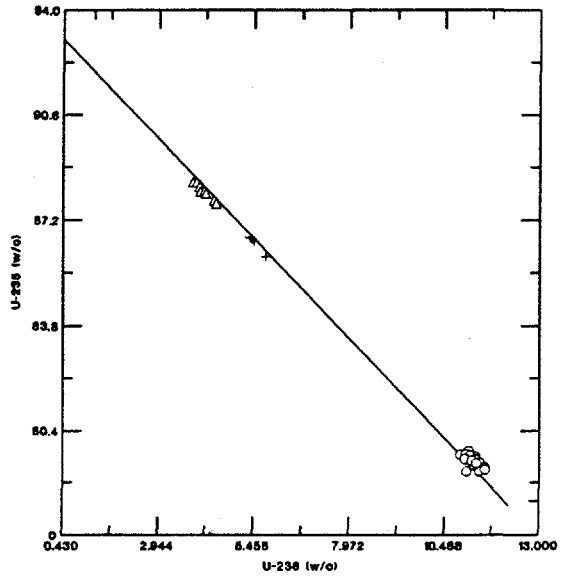

FIGURE 1. CALCULATED CORRELATION (solid curve) FOR HIGHLY-ENRICHED URANIUM, LIGHT-WATER RESEARCH SYSTEMS AND REPROCESSING DATA FOR TWO FOREIGN (Δ, +) AND ONE DOMESTIC (o) REACTOR

Minor uranium isotopic compositions for reactor fuels had been investigated to determine the variations in the concentration levels usually found in the specifications of enriched uranium fuels used in research facilities. Scoping studies of uranium isotopes from gaseous diffusion enrichment processes (Refs. 3 and 4) using a reference simple cascade model were compared with the compositions of the stock uranium fuels used in the zero-power-critical (ZPR) facilities at ANL (see Fig. 2).

The U-234, U-235, and the U-236 weight percents for uranium oxide or metallic fuels, as a function of U-235 enrichment ranging from 1.8 to 93 w/o were examined to determine the dependence on uranium enrichment in a reference gas diffusion process.

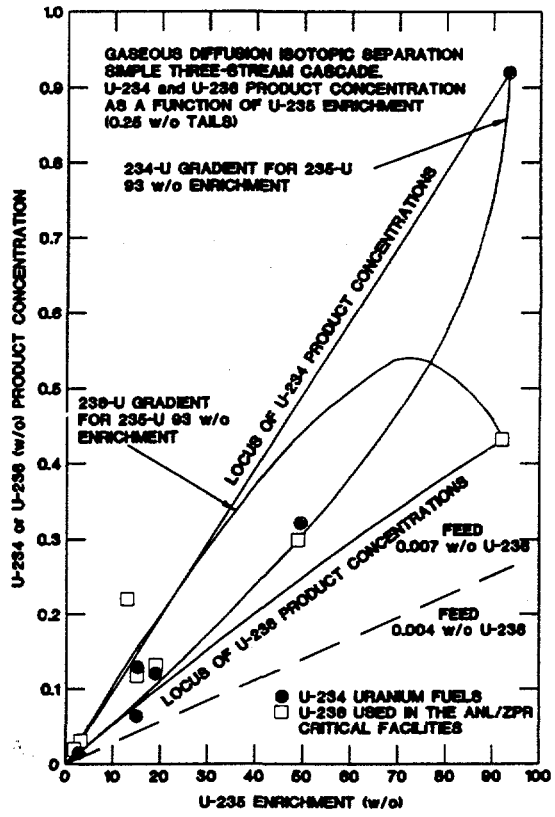

FIGURE 2. GASEOUS DIFFUSION ISOTOPIC SEPARATION

The isotopics of the feed uranium stream assumed initial concentrations of U-236 at 0.004 and 0.007 percent. The U-234

concentration exhibited an increasing enrichment functional relationship with respect to the U-235 enrichment, and the U-236 concentration increase exhibited a negative curvature. The isotopic ratios for low-enriched LEU and highly-enriched HEU uranium fuels indicates that the isotopic signature measurements made at the product and tail steams of the enrichment process can become important data validation procedures for enrichment plants. The results in Fig. 2 indicate that if the side withdrawals are small with respect to the product rate, then the concentrations approach the cascade gradients. This mode of cascade operations could explain, in part, the many different isotopic specifications found in the inventories of enriched fuels used in reactor facilities.

For the isotopic signature of U-236 at the product level of about 0.210 percent, the correlation in Fig. 2 indicates that at the U-235 enrichment product level of 87.7 percent, the initial feed in the enrichment process could have contained U-236 concentrations at about the 0.004 to 0.007 percent level. The depleted uranium stream could have U-235 tails ranging between 0.2 to 0.3 percent. The above concentration levels are indicative of uranium reprocessing product streams from very low-burnup fuels.

The U-234 concentration is indicative of a product stream from a typical gas diffusion enrichment process with a natural uranium feed. Consequently, the reported U-234 and U-236 isotopics seem to indicate that the HEU sample is an enrichment product stream of a gas diffusion (or a gas centrifuge) isotopic separation process of a uranium product stream from reprocessed low-burnup fuels.

PLUTONIUM OF UNKNOWN ORIGIN The interdiction of a plutonium sample was reported in Nucleonics Week, May 25, 1995 (see also Ref. 5). The Euratom's Transuranium Institute (ITU) in Karlsruhe performed the chemical and physical analysis of the 363 g plutonium sample. Argonne was invited by Dr. Lothar Koch of the ITU to communicate Argonne's analytical methods and interpretation of the measured plutonium isotopic data.

The measured concentration of the Pu-242 isotope at the level of 0.68 percent in the plutonium vector, 0.17, 87.56, 10.78, 0.809, and 0.688 percent of Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, respectively, was not internally consistent with the expected core-averaged batch processed isotopic concentration. The core averaged isotopics for either graphite or D_2O type reactors indicates that the Pu-242 content of 0.688 percent is high for the Pu-239 content ranging between 85 to 90 percent level. The typical core averaged plutonium isotopics for a range of Pu-239 concentrations are listed in Table I.

TABLE I
NOMINAL PLUTONIUM CONCENTATIONS FROM LOW-BURNUP REACTOR

	GRAPHITE				D_2O	
Pu-238	0.024	0.088	0.096	0.188	0.016	.022
Pu-239	91.46	87.46	80.83	75.99	90.35	87.74
Pu-240	7.08	10.26	15.13	18.46	8.56	10.72
Pu-241	0.99	2.06	3.62	4.65	0.99	1.4
Pu-242	0.063	0.132	0.422	0.70	0.07	0.12

The isotopics are representative for a range of initial U-235 enrichments from natural uranium to 2 percent and a range of burnup to establish trends. For a Pu-239 concentration level of about 87 weight percent, the expected value for the Pu-242 concentration is expected to be about 0.12 to 0.13 percent for either the graphite or the D_2O type of reactor. Neutron spectral effects are secondary in the case of the reactor core-averaged isotopic concentrations. However, the reactor core regional dependence of the plutonium isotopic concentrations as a function of burnup differ significantly between the inner core region and the outer core region. The typical reference functional behavior of the production and depletion of the Pu-239 and Pu-242, are included in Figs. 3 and 4, respectively. The functional relationships for the inner core region (which approximate core-averaged values) for the Pu-239 isotope illustrates that the concentration level of about 87 percent occurs in the burnup range of 1900 to 2100 Mwd/Mt.

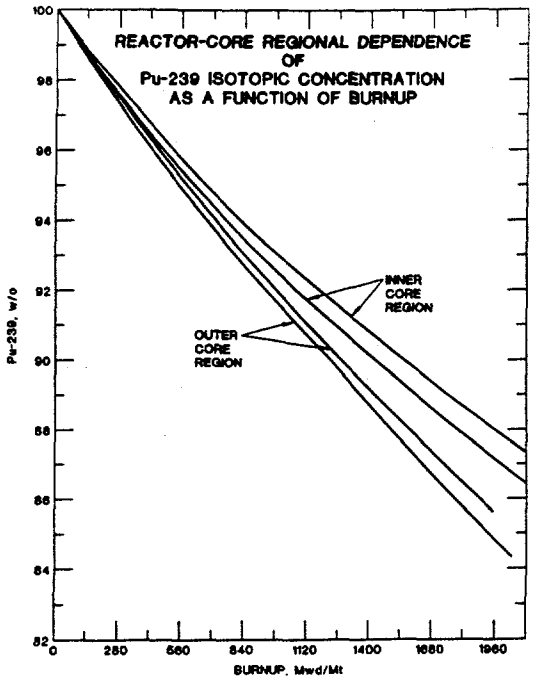

FIGURE 3. Pu-239 ISOTOPIC CONCENTRATION VERSUS BURNUP

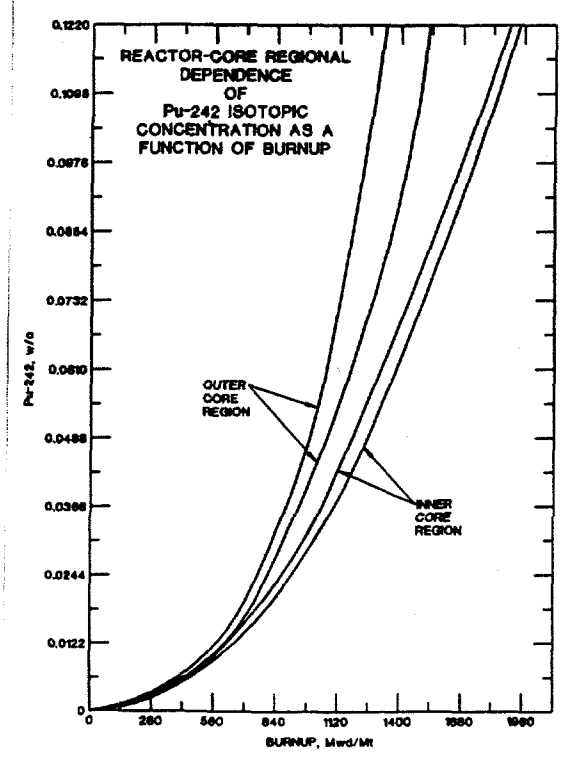

FIGURE 4. Pu-242 ISOTOPIC CONCENTRATION VERSUS BURNUP

Referring to the Pu-242 correlation in Fig. 4, the inner core isotopic concentrations could be expected to range at the level of 0.12 to 0.13 or more which are consistent with the data listed in Table I. However, the correlations in the outer core region indicates that the concentrations of the Pu-242 isotope can be expected at levels much greater than 0.12 to 0.13 percent and to approach the 0.6 to 0.7 percent level for burnups in the range of 1500 to 1700 Mwd/Mt at the extreme outer core regions. In the outer regions of the core, the neutron spectrum is softer, the Pu-239 generation is reduced, and the transmutations of the higher plutonium isotopes are increased.

The Pu-240 and Pu-241 isotopic generation and depletion have similar functional relationships. The reported Pu-241 concentration at the 1 percent level compared to the normally anticipated level of about 2 percent or more, which is consistent with low burnup of natural or slightly-enriched uranium, indicates that the sample is representative of irradiated uranium fuel reprocessed some 15 years or more ago. The reported plutonium isotopics of the interdicted sample are indicative of low-burnup uranium fuel irradiated in the extreme outer regions of the reactor core. In the outer region of the core, the Pu/U ratio is low (low flux level), and for Pu-239 at about 87 percent, the burnup is also low. The amount of plutonium produced in this region would also be low.

SUMMARY

A more detailed study would be required for a more definitive nuclear forensic analysis. The above scoping investigation does emphasize that the isotopic signatures and correlation techniques can be implemented to establish a basis for declared measurement data validation in nonproliferation and/or safeguards concerns relating to the synergism of potential anomalies and traceability of nuclear material flows and inventories.

Nuclear forensic analysis capability in source attribution assessments is an important phase of a nonproliferation program addressing illicit trafficking in nuclear

materials. Isotopic signatures and correlation techniques can be used to establish the initial source (origin) of nuclear material, enrichment process products, reprocessing processes, fuel fabrication products and/or blending processes to gain knowledge in the many different isotopic specifications associated with nuclear fuel cycles and nuclear material exchanges.

The database and/or information databank and interpretation methods would form, in part, the technical foundation of the forensic analysis effort. The domestic collaborative effort would involve laboratories with specific expertise and experienced analysts in each subsystem of the fuel cycle. The international nature in the trafficking of nuclear materials establishes the need for international cooperation and collaboration in nuclear forensics via informal communication and cooperative programs. International cooperative initiatives would include exploring the procedures for expanding and exchanging databases, analytical chemistry methods needed, and analysis algorithms for the forensic interpretation of the measured technical data obtained in the interdiction of an illicit nuclear material trafficking incident. Establishing the existence of forensic analysis methods and techniques in the traceability of nuclear material flows and inventories, could introduce a significant level of deterence in diversion via illicit trafficking by virtue of detection, and identification of the origin, the trafficking route, and the end-use.

REFERENCES

1. S. Niemeyer, Conference Synopsis, "International Conference on Nuclear Smuggling, Forensic Analysis," Lawrence Livermore National Laboratory (November 7-9, 1995)

2. F. Sus, J. Krtil, and M. Beran, "Basic Analytical Data on a Significant Illicit HEU Shipment Secured in Prague," Proceedings of the ESARDA Symposium, Aachen, Germany, p. 589 (May 9-11, 1995).

3. S. Blumkin and E. Von Halle, "The Behavior of the Uranium Isotopes in Separation Cascades," Oak Ridge Gaseous Diffusion Plant Report No. K-1839, Part 1-5 (January 19, 1976).

4. P. Persiani, "Nonproliferation Analysis of the Reduction of Excess Separated Plutonium and High-Enriched Uranium," Proceedings of the 36th Meeting of the INMM, Palm Desert CA, p. 1176 (July 9-12, 1995).

5. L. Koch, "Methodology of Nuclear Fingerprints to Identify Origin and Intended Use of Seized Materials," Conference Synopsis, International Conference on Nuclear Smuggling, Forensic Analysis, Lawrence Livermore National Laboratory (November 7-9, 1995).