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TECDOC No. 1730

Application of Nuclear Forensics in Combating Illicit Trafficking of Nuclear and Other Radioactive Material

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APPLICATION OF NUCLEAR FORENSICS IN COMBATING ILLICIT TRAFFICKING OF NUCLEAR AND OTHER RADIOACTIVE MATERIAL

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Marketing and Sales Unit, Publishing Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
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fax: +43 1 2600 29302
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For further information on this publication, please contact:

Detection and Response to Malicious Acts Section
International Atomic Energy Agency
Vienna International Centre
PO Box 100
1400 Vienna, Austria
Email: Official.Mail@iaea.org

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FOREWORD

As a scientific discipline, nuclear forensics poses formidable scientific challenges with regard to extracting information on the history, origin, movement and processing of nuclear and other radioactive material found to be out of regulatory control. Research into optimized techniques is being pursued by leading nuclear forensic research groups around the world. This research encompasses areas including evidence collection, analytical measurements for rapid and reliable categorization and characterization of nuclear and radioactive material, and interpretation using diverse data characteristics or the 'science of signatures' from throughout the nuclear fuel cycle.

In this regard, the IAEA recently concluded the Coordinated Research Project (CRP) entitled Application of Nuclear Forensics in Illicit Trafficking of Nuclear and Other Radioactive Material. The CRP seeks to improve the ability of Member States to provide robust categorization and characterization of seized material, reliable techniques for the collection and preservation of nuclear forensic evidence, and the ability to interpret the results for law enforcement and other purposes. In accordance with broader IAEA objectives, the CRP provides a technical forum for participating institutes from Member States to exchange technical information to benefit national confidence building as well as to advance the international discipline of nuclear forensics.

This CRP was initially planned in 2006, commenced in 2008 and was completed in 2012. Three research coordination meetings (RCM) were convened at the IAEA in Vienna to review progress. K. Mayer of the European Commission's Institute of Transuranium Elements was the chairperson of the first RCM held in December 2008. C. Larsson of Defence Research and Development Canada chaired the second RCM in May 2010. F. Dimayuga of Atomic Energy of Canada Limited, Chalk River Laboratories, chaired the third RCM held in April 2012. The leadership of the chairpersons was essential to establishing the technical viability of nuclear forensics at the IAEA and with the Member States.

The IAEA wishes to thank Member State experts for the organization of the CRP and for the preparation of this publication. The IAEA responsible officer for this publication was D.K. Smith of the Division of Nuclear Security.

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SUMMARY

Nuclear forensics uses measurements of isotopic, chemical and physical data characteristics (i.e., signatures) of samples to determine the origin and history of seized materials and may link these materials to individuals, places, and events important to a law enforcement investigation or a nuclear security vulnerability assessment. The objective of this CRP was to advance nuclear forensics state–of–practice to aid such investigations and assessments. Through diverse research topics that incorporated field based methods and evidence collection, laboratory based instrumentation and procedures, as well as modeling and interpretation, the technical work delivered procedures for enhanced categorization and characterization of nuclear and other radioactive materials seized out of regulatory control, techniques to preserve forensic evidence, and solutions to strengthen nuclear forensics as part of Member State's nuclear security infrastructure. Conclusions demonstrated i) the viability of nuclear forensics as a technical discipline that can facilitate key international partnerships between Member States, ii) the importance of sequencing nuclear forensic analyses to ensure the integrity of the ensuing forensic investigation, iii) the importance of modeling to establish predictive nuclear forensic signatures where samples are not available, iv) the need for Member States to develop a national response plan that includes nuclear forensics as well as means of handling evidence contaminated with radioactive material, and v) recognition for further peer reviewed research into nuclear forensics particularly in the area of identifying signatures to develop a national nuclear forensics library to aid in the interpretation of analytical results. The results from the present work were incorporated as part of a new CRP (J02003) entitled “Identification of High Confidence Nuclear Forensic Signatures for the Development of a National Nuclear Forensics Library” that focuses on the identification of priority nuclear forensic signatures in the context of the development of a national nuclear forensic library.

INTRODUCTION

The security of nuclear and other radioactive material has been of concern since reports to the IAEA in the early 1990’s of the loss and seizure of materials to include highly enriched uranium, plutonium, and other highly radioactive sources. Reports of nuclear and other radioactive material out of regulatory control persist. As of the end of 2012, Member States had declared to the IAEA’s Incident and Trafficking Database (ITDB) 2331 incidents involving nuclear or other radioactive material that have been trafficked, stolen, lost, or diverted from authorized controls.

To address this issue, the IAEA has prioritized the development of nuclear forensics to enable Member States to address the security requirements of radioactive materials. Nuclear forensic science, referred to as nuclear forensics, is the comprehensive scientific analysis of nuclear or other radioactive material, or of evidence contaminated by radioactive material, to provide insight to material origin and history in support of investigations. The development nuclear forensics occurs within the State’s obligation under international law, including requirements that State’s create punishable or criminal offences for the unauthorized possession of nuclear or other radioactive material. All Member States need to be properly oriented to nuclear forensics since illicit trafficking is a trans–boundary problem. Nuclear forensics is an important component of the Office of Nuclear Security’s 2014–2017 Nuclear Security Plan. Nuclear forensics has been recognized in IAEA General Conference resolutions relating to nuclear security including the 57th IAEA General Conference in 2013.

National response to the occurrence of nuclear or other radioactive material out of regulatory control is foreseen to return forensic evidence that may be used in a criminal proceedings as well as information regarding potential vulnerabilities affecting a State’s nuclear security regime. For these reasons, nuclear forensics remains an important part of the IAEA nuclear security programme.

This CRP assists in strengthening the nuclear forensics capability of Member States to characterize seized items while preserving forensic evidence in view of utilizing the evidence in support of nuclear attribution[1].

Nuclear forensic science involves an iterative and deductive approach, in which the results from one analysis are used to guide the selection of subsequent analyses. Given that some of the analytical tools used in radioactive material analysis are destructive, the proper selection and sequencing of analyses is critical. This sequencing will be determined by the ultimate goals of the investigation, leads in the case, the potential nuclear forensic signatures (physical, chemical, elemental, isotopic) enabling precise interpretation, the amount of sample available for analysis, and analytical methods for measuring nuclear forensic signatures. It is important to emphasize that all sampling and analyses have to be performed with due regard for preservation of evidence and maintaining an evidentiary chain of custody.

Nuclear forensics can support a State's ability to assess whether material encountered out of regulatory control is or is not consistent with nuclear or other radioactive material produced, used, or stored within the State. Within the context of nuclear forensics, signatures are the characteristics of a given sample of nuclear or other radioactive material that distinguishes that material from other nuclear or radioactive material. Analysis of these signatures may aid in the identification of the origin and processing history of the material. Much of the research and development in nuclear forensic interpretation centres on the understanding of signatures determined by discovery using an empirical approach through the systematic analysis of nuclear and radioactive material as well as predictive modeling based on the chemistry and physics of nuclear processes.

Signatures are unique combinations of physical, chemical, elemental, and isotopic data characteristics. To obtain these characteristics, both non–destructive and destructive analytical methods are employed (e.g., gamma ray spectroscopy, x–ray fluorescence and x–ray diffraction, electron microscopy, and mass spectrometry). Often, the interpretation of these signatures, and the identification of questioned material, relies on the comparison with known materials. To aid in comparison, a national nuclear forensics library enables a State to reliably assess whether nuclear or other radioactive material encountered out of regulatory control originated from within that State or not. Such a library consists of comprehensive descriptions, and in some cases samples, of nuclear and other radioactive material produced, used or stored within a State. In some cases, due to the high levels of radioactivity, as may be the case for spent nuclear fuel, experimental data may be supplemented with data obtained from models that simulate production processes within a nuclear reactor.

International cooperation is critical to ensuring that technical advancements in nuclear forensics are shared for maximum benefit for all States. The IAEA, through a programme of development of technical guidance, outreach and training, assists States to promote the awareness and understanding of nuclear forensics as well as their development of the technical capability. This goal is aligned with the objectives of other international groups focussed on nuclear forensics, including the Nuclear Forensics International Technical Working Group (ITWG) and the Global Initiative to Combat Nuclear Terrorism (GICNT) Nuclear Forensics Working Group.

The IAEA's programme of CRPs offers a mechanism for collaborative international scientific research and confidence building. The work of this CRP "Applications of Nuclear Forensics in Illicit Trafficking of Nuclear and other Radioactive Material" was conducted within this framework to evaluate a wide array of nuclear forensic analytical methods, further the 'science of nuclear forensic signatures', promote a means of nuclear forensic interpretation, and position nuclear forensics as a component of a national response plan to nuclear security events.

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[1] Nuclear attribution (as defined in the Nuclear Security Series 2 "Nuclear Forensics Support") is a process to identify the source of nuclear or radioactive materials used in illegal activities, to determine the point-of-origin and routes of transit of such material, and to ultimately contribute to the prosecution of those responsible.

DESCRIPTION OF THE COORDINATED RESEARCH PROJECT

Nuclear forensics provides information on the origin, history and manufacturer of nuclear and other radioactive material outside of regulatory control and is an important tool in nuclear security infrastructure. Objectives of the IAEA CRP J02001 "Application of Nuclear Forensics in Illicit Trafficking of Nuclear and other Radioactive Material" were to:

- Combat illicit trafficking of nuclear or other radioactive material through improved procedures and techniques for nuclear forensics categorization and characterization of seized material;
- Develop techniques to preserve forensic evidence contaminated with radioactive material;
- Optimize nuclear forensic analysis;
- Provide nuclear forensics support to requesting States.

In keeping with broader IAEA objectives, this CRP provided a scientific forum for participating institutes from Member States to exchange technical information for national confidence building as well as to advance the international discipline of nuclear forensics. This CRP consisted of four financial contracts and three collaborative agreements with leading institutes and practitioners involved in nuclear forensics. This CRP was conceptualized in 2006 and the research programme commenced in 2008. During the conduct of the CRP, three Research Coordination Meetings were convened in December 2008, May 2010, and April 2012. The CRP J02001 concluded its three–year funding cycle in 2011.

As listed in Table 1, the projects conducted under this CRP dealt with a variety of nuclear forensic research topics:

- Instrumentation and field work / sample collection;
- Laboratory methods / procedures and techniques;
- Modeling and interpretation.

The participants consider the CRP mechanism to be an extremely valuable tool to address international technical needs in nuclear forensics. In light of the outcome of this CRP, the participants recommend that future CRPs be initiated to address the identification of signatures and the development of analytical methods and tools for interpretation, including assistance to States in the development of a national nuclear forensics library.

TABLE 1. CONTRACTS AND AGREEMENTS: IAEA COORDINATED RESEARCH PROJECT ON "APPLICATION OF NUCLEAR FORENSICS IN ILLICIT TRAFFICKING OF NUCLEAR AND OTHER RADIOACTIVE MATERIAL"; (C) = FUNDED CONTRACT AND (A) = AGREEMENT

Research institution	Chief Scientific Investigator	Title	Research topic*
IDEA System GmbH, Germany	H.R. Doerfel	Identification, localization, and categorization of RDD (C)	1
Korea Institute of Nuclear Safety, Republic of Korea	S.C. Kim	The development of IT-based in-situ mobile response supporting system for deterring illicit trafficking of nuclear and radioactive materials (A)	1
Australian Nuclear Science and Technology Organisation, Australia	T. Evans M. Colella,	Exploiting critical evidence contaminated with alpha emitting radionuclides (A)	2
Institute for Transuranium Elements, European Commission	M. Wallenius, K. Mayer	Procedures and techniques for nuclear forensic investigations (A)	2, 3
Centre for Energy Research, Hungarian Academy of Sciences, Hungary	L. Lakosi	Development of nuclear forensics methods and techniques for combating illicit trafficking of nuclear and other radioactive material (C)	2
Department of Electrical and Computer Engineering, Democritus University of Thrace, Greece	G. Nikolaou	Determination of the origin of unknown nuclear material through an isotopic fingerprinting method (C)	3
Instituto de Pesquisas Energéticas e Nucleares, Brazil	J. De Souza Sarkis	Establishment of procedures and techniques for nuclear forensic investigations Part II – workshop on nuclear forensics (C)	2, 3

*Research topic:

1. Instrumentation and field work / sample collection;
2. Laboratory methods / procedures and techniques; and
3. Modeling and interpretation.

PROJECT REPORTS

The following Sections provide an abbreviated report of each of the projects conducted under this CRP, highlighting specific findings and recommendations from the research programme. For further information on any of these projects, including final research reports submitted by the principal investigators, please contact the IAEA Office of Nuclear Security (http://www-ns.iaea.org/security/).

IDENTIFICATION, LOCALIZATION, AND CATEGORIZATION OF RDD

H.R. DOERFEL
IDEA System GmbH
Karlsruhe, Germany
Email: info@idea-system.com

INTRODUCTION

The focus of this CRP was the development of innovative gamma ray detection software for the non–destructive, in–situ assessment of source radiation characteristics associated with a radiological dispersal device (RDD). The IDEA ILC software enables the evaluation of gamma spectra collected with portable detector systems such as the Spectral Radiation Scanner (by ATOMTEX), the Quantifier (by SARAD), or the identiFINDER (by Flir) at various locations around single radiation sources or distributed contamination, respectively, with the location of the measurements being determined by global positioning system (GPS) techniques.

The software allows both the identification and categorization of high activity (approximately 5 MBq minimum) radioactive sources out of regulatory control and the assessment of an extended contamination field resulting from the detonation of a RDD or from a nuclear accident.

BACKGROUND OF THE PROJECT

The source distribution is reconstructed by adopting iterative procedures as applied in nuclear medical diagnostics. The reconstructed source distribution is displayed in a geospatial environment provided by internet service providers or other on-line based map providers. Based on the source distribution, the radiation field is calculated. In the case of open sources, such as distributed contamination, the potential activity concentration in the ambient air due to evaporation and/or formation of aerosols is calculated. This information is then used for calculating both the potential external and internal exposure due to the source(s). Expanding upon other source identification and reconstruction methods currently available, IDEA ILC allows for the assessment of the potential exposure at the site without performing measurements at the source, i.e., the measurements can be performed at a safe distance from the source and the radiation field is reconstructed using the source distribution in the environment.

When radioactive explosive devices are involved, qualified response personnel must first render these devices safe. For planning of such operations, it is crucial to categorize the suspected radioactive material and to assess any potential exposure of the response personnel. IDEA ILC provides this initial categorization without disturbing the device and any associated evidence. In addition, the software can predict the potential exposure at any location around the source and thus identify the potential health risk to first responders, law enforcement personnel, and the public. Lastly, the software enables response planning to minimize exposure of the first responders by incorporating dose rate distribution in the vicinity of the source.

DESCRIPTION OF THE SOFTWARE

The software includes special spectrum handling tools for improvement of accuracy and sensitivity. The spectrum condensation tool (Fig. 1) transfers the original spectrum to a standardized energy band spectrum with the width of energy bands being proportional to the width of the full energy peaks of the original spectra. This procedure results in a better expression of the full energy peaks and improves the detection sensitivity of the radiation spectrum.

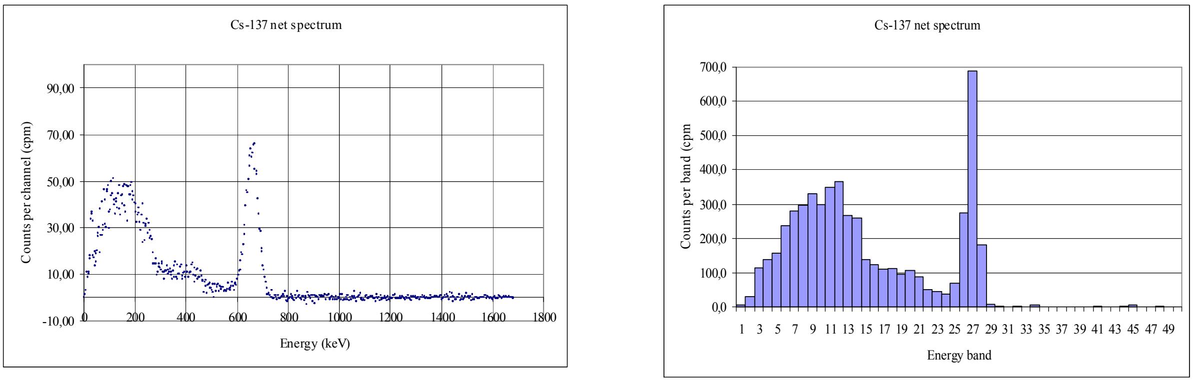


FIG. 1. Condensation of gamma–spectra for improvement of sensitivity.

Another tool utilises the influence of forward scattering of photons on the condensed spectra (Fig. 2). It has been shown that the ratio of the count rate in the peak maximum to the count in the valley below the peak is governed by the mass of material in between the source and the detector. This correlation is used for correction of absorption and thus for improvement of accuracy.

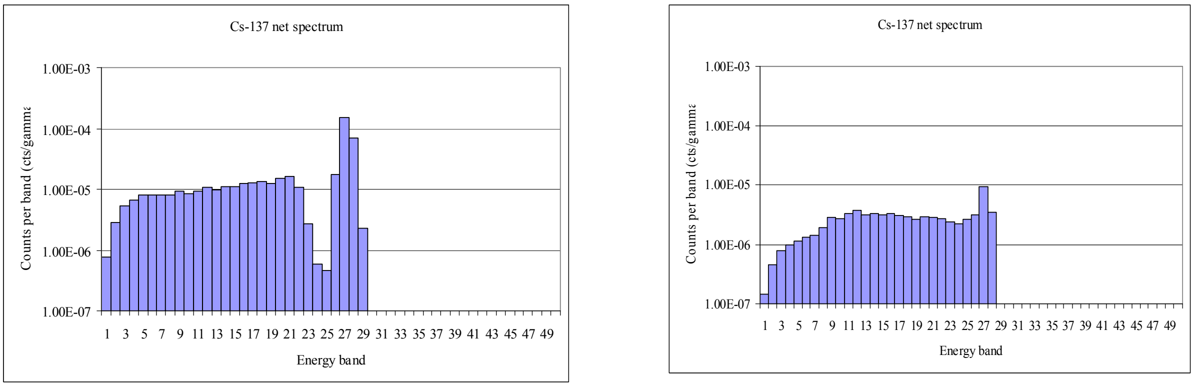


FIG. 2. Influence of forward scattering on condensed gamma–spectra (left: Cs-137 source without absorber; right: Cs-137 source behind 5 cm iron absorber).

In the present version 1.1 the IDEA ILC software provides the following features.

Software evaluation modes:

- Real time radiation evaluation during data collection on the ground (monitoring on foot or by vehicle), offshore (ship) or airborne (helicopter or airplane with manual input of altitude over ground);
- Retrospective evaluation of compiled data.

Software evaluation procedures:

- Condensation of measured spectra in energy bands according to the energy resolution of the detector system;
- Matching of the measuring geometry with geographic mapping (maps from online search engines or other providers);
- Definition of dynamic investigation frames (on-line evaluation) or frames where the source is assumed to be located (retrospective evaluation);

- Iterative reconstruction of the source location or source distribution, within the investigation frames for any energy band;

- Visualization of the reconstructed source location/distribution in the environment with a special color code for the involved radionuclides;

- Automatic correction for absorption using the forward scattering method (available for ^{137}Cs, to be developed for other gamma-emitting radionuclides).

Software outputs:

- Activity distribution of any identified radionuclide in terms of becquerel (Bq) (single source) or Bq/m^2 (distributed sources or contamination, respectively) in the map;

- External dose for any location to be selected on the map.

Additional provisional software outputs include (i) internal dose due to inhalation of radionuclides by re-suspension and (ii) soil contamination calculated in terms of Bq/kg, both derived from the reconstructed ground contamination. These outputs, however, represent only rough estimates and must be elaborated in more detail in future versions of the software. Various visualizations using the IDEA ILC software in radiation source identification (Fig. 3) simulated source reconstruction (Fig. 4, Fig. 5) and spatial contamination (Fig. 6, Fig. 7) scenarios follow.

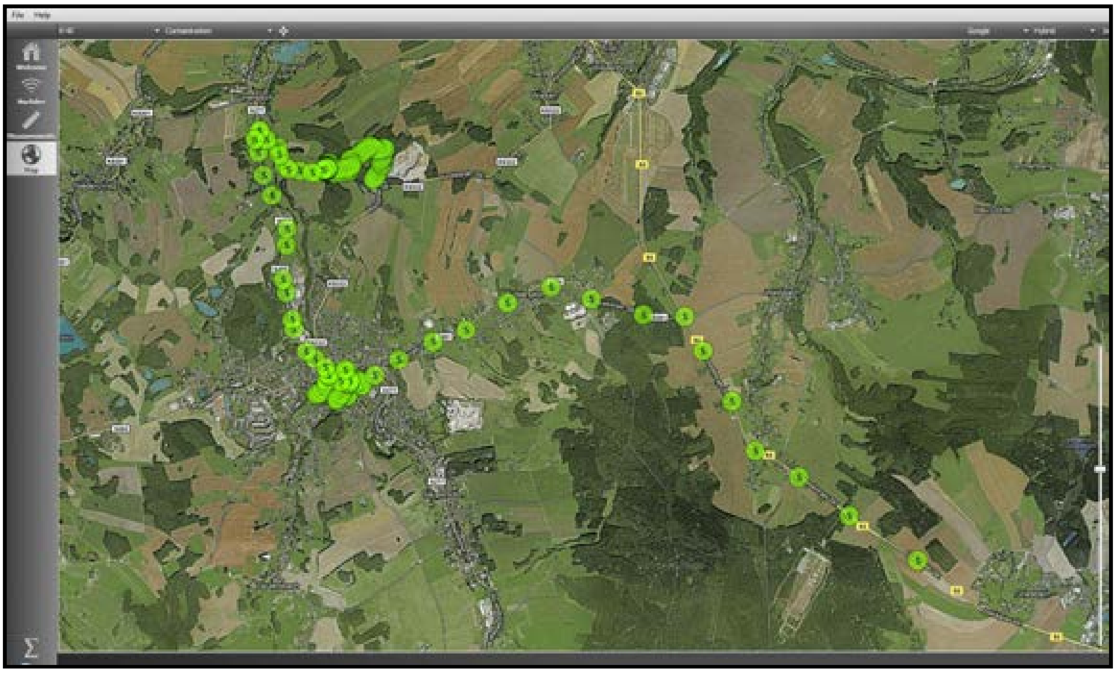


FIG. 3. Display of measurements (incorporating Google hybrid map); "S" are points contributing to radiation source identification.

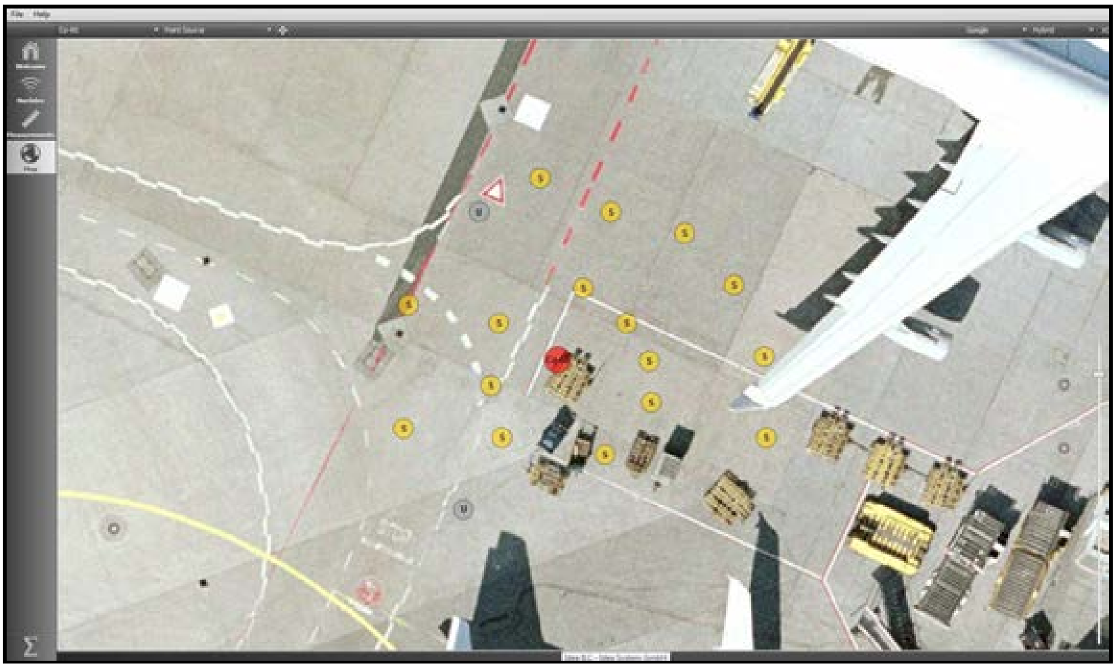
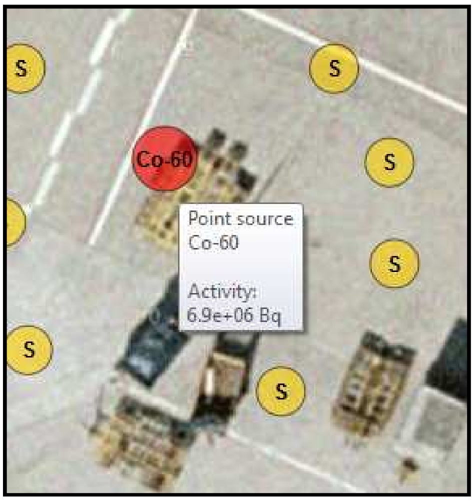


FIG. 4. Display of reconstructed point radiation source (artificial scenario at Frankfurt, Germany airport); "S" represent points contributing to source identification; "U" are points not contributing to the reconstruction of the source.



Point source
Co-60

Activity:
6.9e+06 Bq

FIG. 5. Display of activity of reconstructed radiation point source (artificial scenario at Frankfurt, Germany airport); "S" represent points contributing to source reconstruction; "U" are points not contributing to the reconstruction of the source.

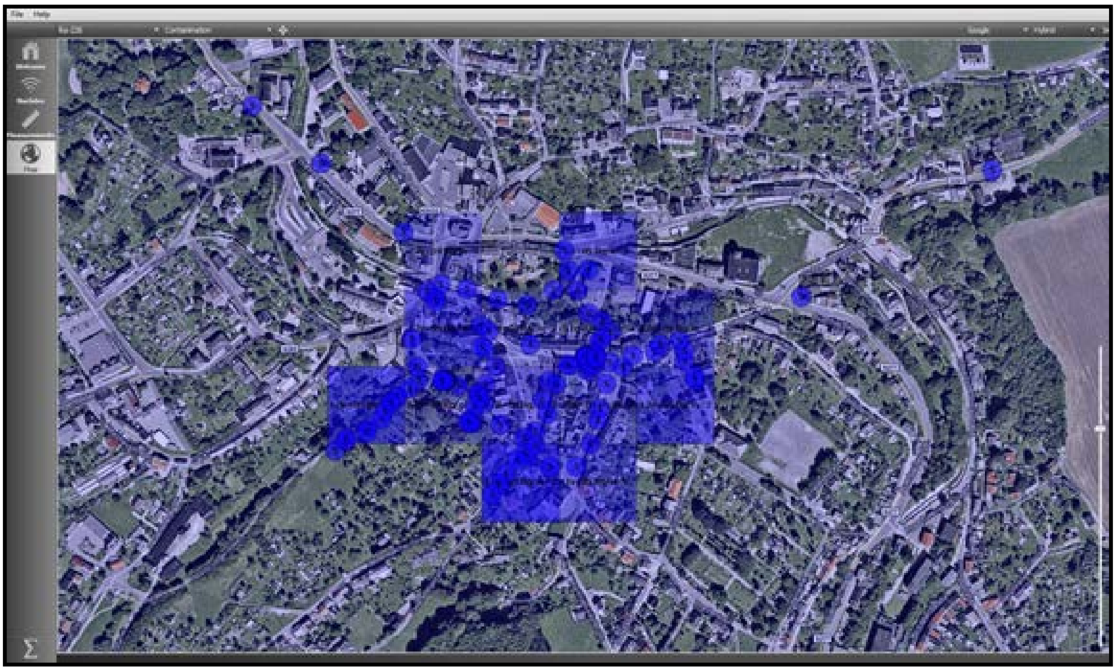


FIG. 6. Display of reconstructed area spatial contamination (measurement of ^{226}Ra in Kirchberg, Saxonia, Germany).

Contamination field

Size:
139.95mx139.95m

Activity:
1e+05 Bq/m^2

FIG. 7. Display of detailed information about the reconstructed area contamination (measurement of ^{226}Ra in Kirchberg, Saxonia, Germany).

CONCLUSIONS

The software provides a tool for identification, localization and characterization of high activity (approximately 5 MBq) points and distributed gamma-emitting sources based on measurements with any kind of portable gamma-spectrometric detector systems in connection with GPS devices.

The quality of the source location and source activity results depends primarily on the accuracy of the coordinates of the measuring position provided by the GPS device. If the uncertainty of the GPS coordinates is less than 3 meters, the uncertainty of the source location is less than 1 meter and the uncertainty of the source activity is less than 20%. However, if adjacent buildings or other large structures at the measurement site disturb the collection of GPS data, the uncertainty may be much higher, particularly with respect to the location of the source.

Based on the current experience, a new generation of portable gamma-spectrometers have been designed that may be coupled with smartphones with GPS and movement sensors for reliable assessment of measuring position in any environment, even inside buildings. The software is implemented as an application on the smartphone and the results of the measurements can be transferred online to any interested person or to a central server compiling the results of various detector systems in nuclear security event response or a nuclear accident.

Special applications may be implemented for well-defined measuring problems including the determination of the total activity of uranium that incorporates the isotopic enrichment. The measurement is based (i) on the assessment of the effective absorption of the gamma-radiation by comparing the 63 and 92 keV peaks of ^{234}Th and (ii) the assessment of the enrichment by comparing the peaks of ^{235}U and ^{234}Th.

Further recommendations include adaptation of the software to locate radioactive sources hidden or buried underground, a robust programme of field testing to involve a variety of detection scenarios, training for responders on how to use the detection software routinely with high confidence as part of the response to a nuclear security event, as well as improving the limits of detection of the software and the gamma ray detector.

THE DEVELOPMENT OF IT-BASED IN-SITU MOBILE RESPONSE SUPPORTING SYSTEM FOR DETERRING ILLICIT TRAFFICKING OF NUCLEAR AND RADIOACTIVE MATERIALS

S.C. KIM
Korea Institute of Nuclear Safety
Daejeon, Republic of Korea
Email: k110ksc@kins.re.kr

INTRODUCTION

The Republic of Korea is a global leader in the development of nuclear energy. South Korea presently has 20 operating nuclear power reactors (conventional pressurized light water reactors and heavy water reactors) in 4 nuclear power plants that supply 17,000 MWe or almost 40% of the country's electricity. South Korea presently can supply up to 95% of electrical capacity as needed. Scheduled new builds to 2016 include eight more plants with a total capacity of 26,000 MWe. Accompanying the growth of nuclear power is a national commitment to a comprehensive nuclear security infrastructure.

The Republic of Korea has an effective framework for countering nuclear and radiological terrorism. Ministerial roles and responsibilities are integrated into a national response plan. The framework includes environmental monitoring, emergency characterization, emergency management, consequence management, and includes alert notification to the IAEA. This infrastructure is data intensive involving feeds from the nuclear power plants and 70 environmental monitoring stations. In South Korea there is an additional focus on tracking radioactive sources and nuclear materials through a source tracking system. Radiation detection portals are now operating. In addition the Republic of Korea maintains an active nuclear weapons test radiation monitoring program.

INFORMATION TECHNOLOGY (IT) BASED TOOLS FOR RESPONSE TO ILLICIT TRAFFICKING OF NUCLEAR AND OTHER RADIOACTIVE MATERIAL

The CRP agreement between the IAEA and the Korea Institute of Nuclear Safety (KINS), Republic of Korea represents a full-spectrum nuclear emergency management system that integrates several individual systems for monitoring and response to nuclear security events. The system uses a web-based IT-tool with command and control (C&C), video conferencing, measurement visualization, and geographic information system modules. This tool, named TETRA, was developed during the project to enable detection and response to radioactive materials out of regulatory control the result of a radiological accident or radiological terrorism. The technology integrates internet-based monitoring using feeds from portable radiation detectors, 24/7 radiological monitoring, a global positioning system (GPS), real-time scene video, a geographic information system (GIS) interface, and a video-conference feature that allows all responsible national agencies to be fully interconnected (Fig. 1). The system was deployed at several recent high profile national level events in the Republic of Korea.

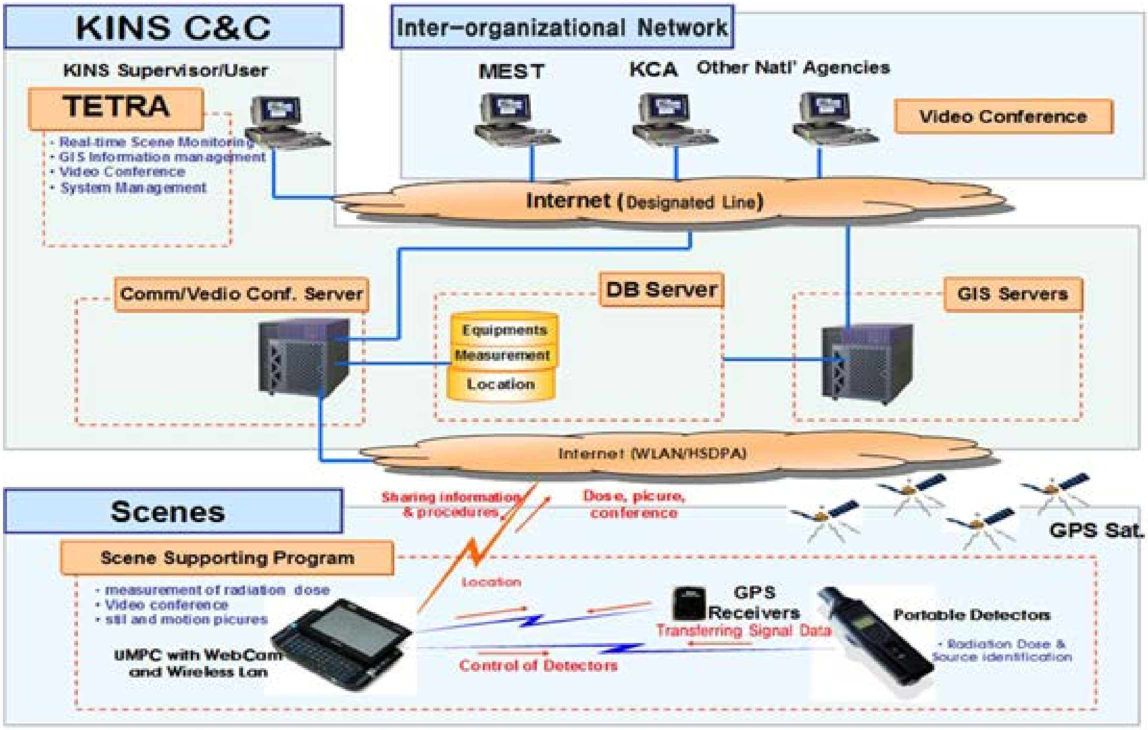


FIG. 1 Schematic of the TETRA full-spectrum Republic of Korea nuclear emergency management system that integrates monitoring and response to nuclear security events to prevent illicit trafficking of nuclear and radioactive material.

NUCLEAR FORENSICS CAPACITY BUILDING

South Korea is committed to developing a robust nuclear forensics capability to track nuclear and radioactive materials with technical support from the IAEA. Nuclear forensics is positioned to be a key technical capability of the South Korean national response plan to address nuclear and radioactive materials out of regulatory control. To do this, the Republic of Korea recognizes the importance of building dedicated receival and laboratory facilities to analyze evidence contaminated with radioactive material, training personnel, and establishing procedures to respond to nuclear security events including those for the collection and transportation of evidence, analysis at specialized nuclear forensic laboratories, and resulting high confidence interpretation of multiple nuclear forensic signatures (e.g., questioned versus known samples) to guide national level response.

Through its long history of commitment to monitoring radioactivity throughout South Korea, KINS maintains a strong network of analytical laboratories that can be utilized for a comprehensive national programme in nuclear forensics. Existing capabilities include alpha spectroscopy (for uranium and plutonium), beta spectroscopy and gamma spectroscopy laboratories, an inductively coupled plasma mass spectrometer and a radiochemical separations laboratory. More than 500 environmental samples are processed annually in these facilities. Nuclear forensic analysis will be developed in the context of the existing framework including regular procedures and analytical standards. To meet the goal of establishing a national capability in nuclear forensics, it is imperative to raise awareness of the discipline through all relevant state organizations.

CONCLUSIONS

The Republic of Korea is presently developing a national framework of nuclear forensics including a national technical center. The IAEA is supporting South Korea’s efforts towards establishing the necessary infrastructure associated with the national programme including training, written guidance, analytical best practices, and encouraging collaboration with other international partners. To this end, the Republic of Korea hosted an IAEA regional training workshop on “Radiological Crime Scene Management and Introduction to Nuclear Forensics” at KINS in Daejeon in August 2011. This workshop gathered experts from Cambodia, China, Indonesia, Malaysia, Mongolia, Philippines, Singapore, Thailand, United Arab Emirates, Vietnam, and the Republic of Korea to enhance awareness and the participants' understanding of the capabilities required for radiological crime scene management and an introduction to nuclear forensic analysis and interpretation as part of a State's national response plan. Through a combined programme of technical assistance in nuclear forensics, training in awareness and methods, research, development of a nuclear forensic laboratory, and international partnerships, the Republic of Korea is establishing a nuclear forensic capability commensurate with its international leadership in the development of civilian nuclear power.

EXPLOITING CRITICAL EVIDENCE CONTAMINATED WITH ALPHA EMITTING RADIONUCLIDES

T. EVANS, M. COLELLA
Australian Nuclear Science and Technology Organisation
New South Wales, Australia
Email: tee@ansto.gov.au

INTRODUCTION

In a situation where radioactive materials are used in a malevolent act it is highly likely that evidence needed to link a perpetrator to the crime will be contaminated with and/or exposed to radiation. This research addresses fundamental issues concerning the processing of evidence contaminated with radioactive material by characterizing the combined effects of alpha radiation and decontamination processes on trace evidence. To date, many research efforts on nuclear forensics have focused on gamma emitting radioisotopes from industrial radionuclide sources including ^{192}Ir, ^{60}Co and ^{137}Cs. Here we report on the challenges inherent in performing the safe, efficient, and effective analysis of evidence exposed to or contaminated with alpha emitting radioisotopes.

Although alpha emitting radioisotopes do not pose an external radiation hazard, there is a significant internal radiation hazard associated with this type of exposure. A traditional forensic laboratory is not outfitted with the necessary radiological controls or worker health physics protection to handle radioactive materials and therefore cannot receive evidence contaminated with radioactive material. Mitigating the hazard of working with radioactive contamination whilst maintaining the integrity of the evidence requires the cooperation of radiation safety specialists working in concert with their law enforcement counterparts.

In addition we consider the impact of high-energy alpha particles on forensic evidence including hairs, fibers, and fingermarks. We assessed existing methods as well as developed new methods for handling and processing traditional evidence contaminated with alpha emitting radioactive material. Two glove boxes were refurbished and commissioned for this work. Although the examination of contaminated evidence in the restricted environment of a glove box was technically challenging, it proved successful for the techniques investigated.

Trials included development of techniques for decontaminating and examining evidence contaminated with radioactive material. The feasibility of these approaches was demonstrated using a proof-of-concept study.

In general, the impact of alpha radiation on trace evidence was negligible in the lower dose range of less than 250 kGy. However in the higher dose range of 250–1000 kGy alpha radiation inflicted damage on forensic trace evidence that may affect the ability to analyze and interpret certain evidence types.

HAIRS AND FIBERS

Using a number of analytical techniques including scanning electron microscopy (SEM), microspectrophotometry (MSP), and Fourier transform infrared spectroscopy (FTIR), the surface and chemical structure of hairs and fibers were found to be largely resistant to exposure to alpha radiation. Minimal effects were observed in dyed Caucasoid hair as well as acrylic, cotton, and wool fibers. Nylon and polyester fibers also showed minimal alpha radiation-induced damage.

Figure 1 compares the backscattered SEM image of a non-irradiated and 1000 kGy alpha irradiated acrylic fiber.

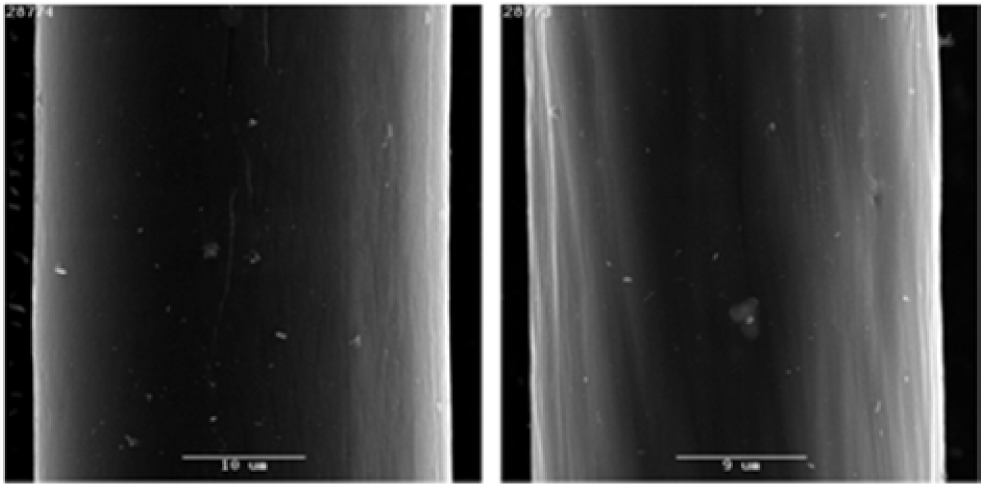
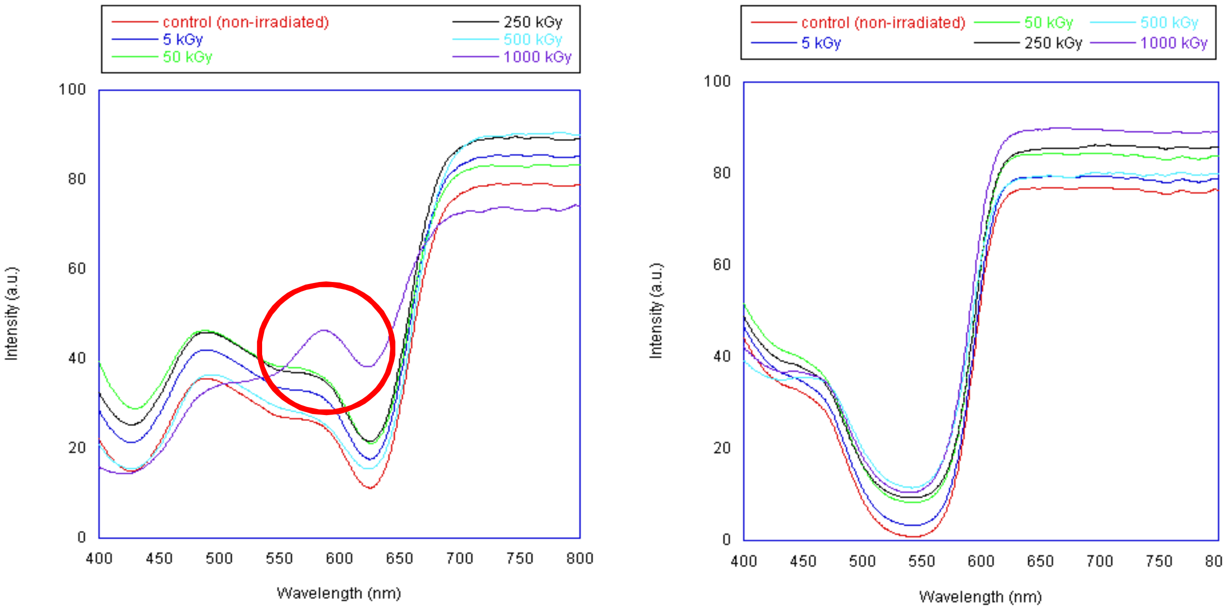


FIG 1. Backscattered electron images of an acrylic fiber which was not irradiated (left) and which underwent 1000 kGy alpha irradiation (right). Scale bar at bottom of image.

Figure 2 illustrates the occurrence of radiation damage, measured by MSP, to black acrylic fibers after 1000 kGy alpha irradiation (circled in red) compared with red acrylic fibers that were unaffected by a comparable exposure.



Black acrylic

Red acrylic

FIG. 2. MSP spectra of black (left) and red (right) acrylic fiber as a function of alpha radiation dose.

The contamination and subsequent decontamination of fibers produced no impact on their characteristic properties used for forensic purposes, with the exception of blue cotton. Further investigation of blue cotton is warranted to determine whether this is an inherent variation or due to the decontamination treatment.

FINGERMARKS

Alpha radiation exposure was detrimental to the development of latent fingermarks (i.e., fingerprints) when doses were greater than 250 kGy. Exposure of substrates to alpha radiation prior to fingermark deposition and post fingermark deposition impeded the ability to recognize identifiable features required for fingermark comparison (Fig. 3).

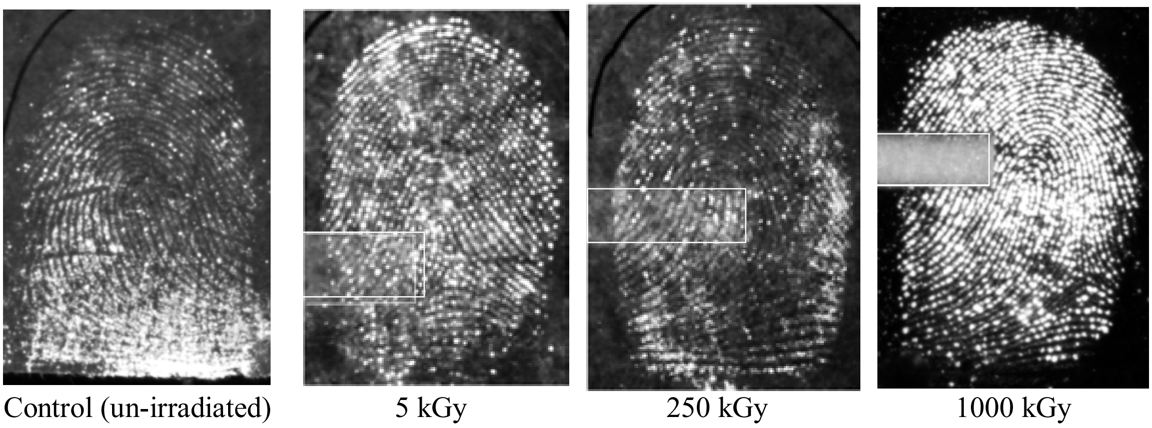


FIG 3. Developed (using IND-Zn) fingermarks on paper as a function of alpha radiation dose.

A summary of the fingermark development techniques investigated and their applicability as a function of alpha radiation dose is provided in Table 1. At the extreme dose of 1000 kGy, none of the applied development techniques were successful.

TABLE 1. FINGERMARK DEVELOPMENT TECHNIQUES AND THEIR APPLICABILITY AS A FUNCTION OF ALPHA RADIATION DOSE (✓ INDICATES DEVELOPMENT, ✗ INDICATES IMPAIRED DEVELOPMENT)

	Alpha irradiation dose delivered to latent fingermark				
Development Technique	5 kGy	50 kGy	250 kGy	500 kGy	1000 kGy
1,8-Diazafluoren-9-one (DFO)	✓	✓	✓	✓	✗
Indandione (IND)-Zn	✓	✓	✓	✗	✗
Ninhydrin	✓	✓	✓	✗	✗

The decontamination of fingermarks contaminated with radioactive material was detrimental to the development and enhancement of sufficient detail for comparative purposes. Subsequently, two bench–top fingermark development and enhancement techniques were introduced to the glove box to enable handling and processing of items without the need for decontamination. Cyanoacrylate fuming with Rhodamine 6G (R6G) stain solution and indanedione–zinc (IND-Zn) were successfully modified for their use in the restrictive environment of the glove box. An essential modification to the glove box was the inclusion of a variable wavelength light source (Polilight®) and camera system to allow image enhancement and capture in–situ.

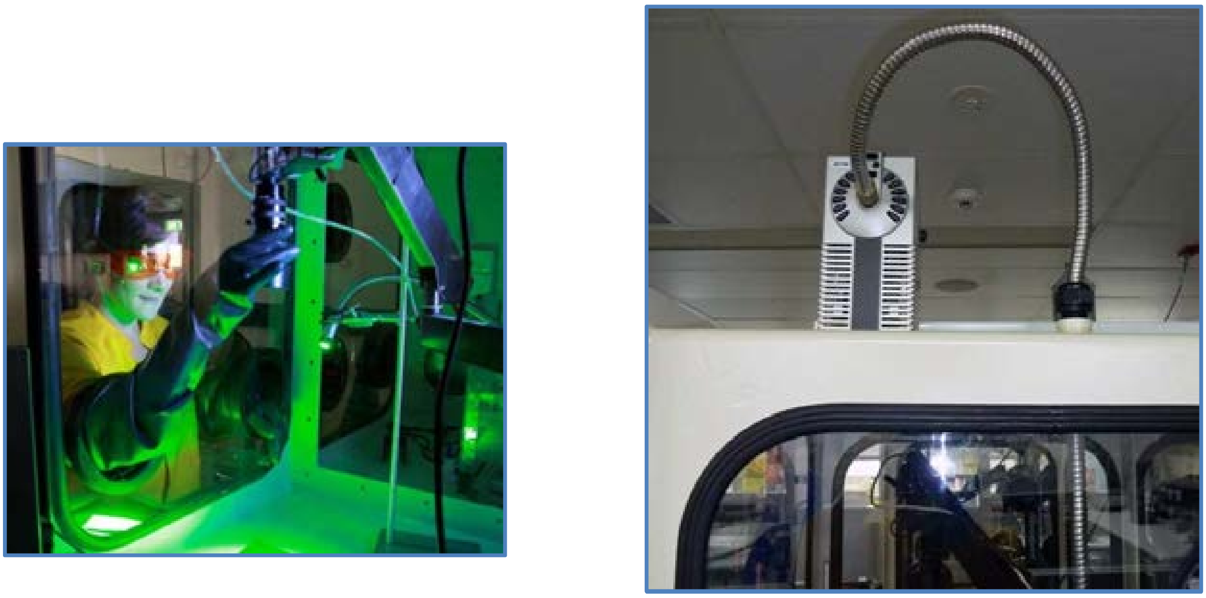


FIG. 4. Polilight® PL500 light guide and imaging system inside the glove box (left) and Polilight® PL500 unit external to the glove box (right).

DNA

The application of two commercially available DNA extraction kits (ChargeSwitch® and DNA IQ™) in the glove box was successful for processing of DNA from alpha contaminated evidence. Further work investigating the feasibility of searching and extracting from a wider range of evidence types is warranted.

CONCLUSIONS AND RECOMMENDATIONS

A nuclear security event involving alpha emitting radionuclides poses a serious health risk to the public and responders. It is vital that scientific researchers and law enforcement personnel work together in the forensic response to such an incident, utilizing techniques that are both scientifically valid and legally admissible. The primary goal of this project was to determine the efficacy of techniques for the analysis of traditional forensic evidence that has been exposed to or contaminated with alpha emitting radioactive material. The outcome of this work reiterates the need for heightened awareness when considering forensic evidence and interpreting data that has come from items that may have been exposed to or come in contact with an alpha emitting radioactive material.

Hair is generally resistant to degradation by alpha radiation, and thus forensic examination of such evidence is valid. The exception to this is dyed hair, with some degradation of the dye observed at radiation doses greater than 50 kGy.

The chemical structures of fibers of a wide range of types and colors are resistant to degradation by alpha radiation; the application of forensic techniques targeting this component of the fibers remains valid. Some degradation of dyes utilized on fibers occurs as a result of alpha radiation exposure, and thus care should be exercised when undertaking comparisons dependent on dye properties. Decontamination of fibers contaminated with radioactive material should be carried out prior to their analysis in a traditional forensic laboratory. Decontamination should be carried out in a glove box as dictated by the type and quantity of contaminant; this decision requires a risk assessment.

Further experiments incorporating a range of contamination levels to fully validate in–glove box decontamination procedures should be conducted as part of a future follow–on research program. Development of latent fingermarks should be undertaken in the glove box when evidence is contaminated with alpha emitting radioisotopes. Cyanoacrylate fuming with either basic yellow 40

(BY40) or R6G dyes is suitable for the development of fingermarks on non–porous surfaces. IND–Zn is a suitable technique for the development of fingermarks on porous surfaces in the glove box environment. A smaller dedicated fuming chamber with a hot plate (for example) is required for the containment and generation of the cyanoacrylate fumes. Impacts to the development of fingermarks were identified in some circumstance at doses greater than 250 kGy. Further experiments for the optimization of these techniques by forensic examiners should be undertaken.

Extraction of DNA with a commercially available solid phase extraction in a glove box is feasible in cases where evidence has been contaminated with alpha emitting radioisotopes. Further experiments to confirm its applicability for a range of radioisotopes are required. Measures for reducing the likelihood of DNA contamination during glove box DNA extraction should be implemented including installation of an ultraviolet (UV) light, autoclaving of consumables, and the institution of protocols for the separation of samples.

PROCEDURES AND TECHNIQUES FOR NUCLEAR FORENSICS INVESTIGATIONS

M. WALLENIUS, K. MAYER, Z. VARGA
Institute for Transuranium Elements
European Commission
Joint Research Centre
Karlsruhe, Germany
Email: Maria–S.Wallenius@ec.europa.eu

INTRODUCTION

The experience gained at the Institute for Transuranium Elements (ITU) during 20 years of nuclear forensic analysis is provided in Part 1 of the report to assist new or less experienced laboratories in the conduct of a nuclear forensic examination. Part 2 of the report compiles research conducted over the past three years on new characteristic parameters and signatures that provide improved insight to the origin of seized nuclear materials.

PART 1. NUCLEAR FORENSICS GUIDANCE

Nuclear forensics must be considered as part of the radiological crime scene response to a nuclear security event. Without appropriate and representative sampling, the analysis performed later in the nuclear forensics laboratory has little value if the sampling has not been performed correctly (e.g., sampling vials or tools are contaminated with radioactive materials or cross–contamination occurs from adjacent samples or other extraneous materials). This report focuses on the steps following the sampling to include nuclear material analysis in the laboratory, interpretation of the analytical data, and lastly, attribution using reference data or comparison samples.

Preliminary analysis

Nuclear forensic analysis starts with visual inspection of the sample. The sample will be photographed, however the package should not be opened before non–destructive gamma spectrometry analysis, which is used to confirm the on–site categorization and gain possible additional information (e.g. fission and daughter products) using high resolution gamma spectrometry (HRGS) in the controlled laboratory environment. In the case of a solid sample (e.g., metal or ceramic pellet), its geometry will be measured using a ruler or caliper. For powder samples optical microscopy analysis will be performed to obtain the coarse magnification (on sub–millimeter range) of the individual grain structure.

Non–destructive analysis

Non–destructive analysis (NDA) should be performed before any destructive analysis (DA) is done. This is especially important for a small sample in order not to consume it before as much information as possible – to include valuable evidence – has been acquired. The most common NDA technique is HRGS, the same technique that is used for the advanced categorization of seized samples. Besides serving as a preliminary analytical technique for the isotopic composition analysis of U and Pu, it can be used, when calibrated accordingly, for quantitative analysis with less precision than destructive methods but with a minimum sample size.

For plutonium (Pu), there are two other NDA techniques that can be used for the quantification of the mass of Pu. These are neutron coincidence counting (NCC) and calorimetry. In order to calculate the Pu mass in the sample, one needs to know the isotopic composition of Pu (and ^{241}Am for calorimetry).

Electron microscopy is a versatile and powerful technique for the microstructural characterization of solid samples. When scanning electron microscopy (SEM) analysis is coupled with energy dispersive

X–ray spectrometry (EDS) or wavelength dispersive X–ray spectrometry (WDS), the morphology, surface structure and elemental composition and distribution can be determined with a resolution of a few nanometers. Even better resolution (i.e., sub–nanometer) can be achieved using transmission electron microscopy (TEM).

X–ray diffraction (XRD) can be used to determine the crystal structure of material and its molecular composition.

Destructive analysis

Destructive analysis is usually classified into two types, namely mass spectrometric and radiometric techniques. Mass spectrometric techniques, such as thermal ionisation mass spectrometry (TIMS), inductively coupled plasma mass spectrometry (ICP–MS), glow discharge mass spectrometry (GDMS) or secondary ion mass spectrometry (SIMS) are used typically to measure isotope ratios of long–lived nuclides in a variety of sample matrices. Mass spectrometry can be also used to quantify the elements of interest using isotope dilution analysis (IDA).

Destructive radiometric techniques, such as alpha spectrometry (AS) or liquid scintillation counting (LSC) measure the emitted alpha, and in the latter case also the beta radiation, which can be used for the identification and quantification of the radioactive isotopes. To determine the elemental concentration of uranium (U) or Pu the examiner can use X–ray fluorescence (XRF) and K–edge or L–edge densitometry (KED or LED), which measure the emission of the characteristic x–rays of the element.

Other types of techniques, which are destructive, in addition to radiometric or mass spectrometric techniques include ion chromatography (IC) that can be used to measure anion concentrations or infrared spectroscopy (IR) that can be used to determine the molecular compound of uranium in uranium intermediate products, such as "yellow cakes".

Volumetric methods, such as potentiometric titration, are well established and often considered the most precise methods for U and Pu assay determination.

Data interpretation

Nuclear forensics interpretation of the analytical data is the intermediate step between the sample analysis and the attribution of the sample to determine its origin. Data interpretation utilizes various statistical and visualization tools depending on the measured parameter. These include simple statistics (based on uncertainty evaluation), advanced statistics (e.g., cluster analysis, principal component analysis), graphical evaluation and subject matter expertise. Before the investigated sample can be, however, attributed to a certain origin (e.g., production facility, uranium mine), the evaluated data need to be, in most cases, compared against known data (e.g., in databases that are part of a national nuclear forensics library).

Attribution using reference data

The attribution of nuclear or other radioactive material to include its origin, production method, transit route and the point where regulatory oversight was lost is a complex task. All of the aforementioned tasks cannot be solved exclusively by nuclear forensic analysis, but also require traditional forensics and associated documentation of nuclear material movements to determine the transit route and the last legal owner, respectively. Nuclear forensic analysis can provide essential information about the origin and the production method of nuclear material. However, in the absence of reference data for comparison, obtaining conclusions may be more difficult. At ITU, case studies demonstrate the utility of the ITU's nuclear materials database (i.e., containing information on commercial reactor fuels of several European and Russian nuclear fuel manufacturers) and comparison samples to provide information on the origin and history of nuclear materials.

PART 2. POTENTIAL NEW CHARACTERISTIC PARAMETERS FOR NUCLEAR FORENSICS

In order to improve the confidence in determining the origin determination of seized nuclear materials, the search for new characteristic parameters is a priority. These characteristic parameters, also referred to as signatures, yield information about the history of nuclear material. The signatures can also provide insight to how the nuclear material was produced or which kind of process was used. In the case of uranium, the signatures can aid in where the uranium originated including the type of uranium deposit from which the ore was mined.

The approach for this complex study was first to identify those signatures that are indicative of the uranium source material, i.e. uranium ore. Uranium ores are typically very inhomogeneous (containing U rich parts plus parts which are rich in other elements, i.e. the gangue); for this reason, ores were considered being less ideal samples for this study. The next product in the processing of uranium after extraction as U ore, is uranium ore concentrate (UOCs), also referred to as "yellow cake". UOCs are produced in industrial scale at uranium mills and they are considerably more homogeneous compared to U ores. Therefore, UOCs were selected for use in this study.

Impurities

Rare earth element (REE) patterns often remain unaltered during deposition of uranium bearing sediments or during metamorphism, thus they provide information on the history, petrogenesis and provenance of geologic samples. Variations in the REE pattern of UOC samples may reflect differences in the geologic origin of the uranium deposit [1, 2, 3]. The behavior of the valence-variable Ce (+4) and Eu (+2) often results in positive or negative anomalies compared to the other REE and may be exploited to obtain information on the conditions of formation. This information can be utilised in nuclear forensics.

Anions (e.g., F^-, Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-}) are found in trace levels in UOC as a result of the many mineral acids used in the uranium concentration and refinement process. For instance, sulphuric and nitric acids are commonly used for the leaching or dissolution of the uranium ore.

The majority of the investigated UOC samples had a nitrate concentration less than 1 ppm, while few samples had significantly higher (well above 10 ppm) nitrate content. Significantly higher concentrations were attributed to the process–related origin of the nitrate. High nitrate content was found in those samples where nitrate solution was used for stripping during processing steps. If nitric acid was used only for the dissolution of the ore, the nitrate content in the UOC was not significantly higher.

Fluoride is usually not added intentionally to the material during UOC production, however some chemicals may contain elevated levels of fluoride. Another possible source of fluoride in the UOC is presence of fluoride bearing minerals. Several fluoride–bearing minerals are often found in U ores, such as apatite, fluorite, micas, and amphiboles. In addition to that, igneous rocks can contain up to 1000 ppm F^- mainly from the fluoroapatite mineral $Ca_5(PO_4)_3F$. Fluoride in UOC is mainly derived from the U ore mineralogy (feed material) introduced into the UOC production. Fluoride is commonly associated with uranium in some minerals, mainly those of volcanic origin. If phosphate is used as the uranium feed (uranium recovered as a by–product), an elevated level of fluoride is expected, as fluoride is a major component of the feed present at per cent level in the starting material. For this type of deposit, the fluoride concentration was found to be significantly higher than the average.

Radiogenic isotope ratios

Lead (Pb) isotopes of ^{206}Pb, ^{207}Pb and ^{208}Pb are the final decay products of the ^{238}U, ^{235}U and ^{232}Th series, respectively. Therefore, uranium products often contain lead in trace levels. Depending on the age of the uranium deposit, the initial U/Th ratio in the deposit and on the weathering conditions, large differences in the lead isotope ratios in uranium material may be expected [4]. Besides the isotope ratio comparison between samples from different mines, lead isotopes can be utilized to estimate the age of the ore deposit.

The variation of the $^{87}Sr/^{86}Sr$ ratio in minerals is due to the decay of long–lived ^{87}Rb (half–life, 4.8×10^{10} years) to ^{87}Sr and the highly variable Rb/Sr ratio in nature. The $^{87}Sr/^{86}Sr$ isotope ratio is widely used for geologic age determinations and during the past decade more extensively in origin assessment (e.g., ivory, human bone, or food products) and was chosen as a possible indicator of geologic provenance because of its geochemical characteristics. $^{87}Sr/^{86}Sr$ can provide quantitative variations between different UOC samples, however the differences are considerably smaller compared to lead isotopic ratios. The measurements require a very precise isotope ratio measurement technique (e.g., multi–collector ICP–MS), but the results for strontium also have the advantage over the lead isotope signature as the within mine variations are less pronounced compared to measurements of lead isotope ratios [4].

The $^{143}Nd/^{144}Nd$ ratio varies in nature due to the decay of ^{147}Sm to ^{143}Nd and variations (0.510–0.513) have been recorded. The $^{143}Nd/^{144}Nd$ ratio depends mainly on the age of the neodymium bearing minerals. However, it is less prone to weathering and it is expected to be very robust signature. Significant differences in this ratio between mines have been observed; within mine differences are even smaller than in the case of strontium. However, to establish the real potential of this signature, more samples need to be analysed.

Minor uranium isotopes

Natural variation in the $^{234}U/^{238}U$ isotope ratio, which is the consequence of preferential leaching of ^{234}U from a uranium bearing mineral lattice following radioactive decay, can be effectively used to discriminate some U ore concentrates. Although, the range of natural variation in the $^{234}U/^{238}U$ isotope ratio is small, it is detectable using a high precision measurement method (e.g. multi–collector — TIMS). However, there may be overlap in the $^{234}U/^{238}U$ isotope ratios when comparing large numbers of samples.

^{236}U was also detected in a few UOCs, which is unusual as the detection limit of TIMS ($\sim10^{-10}$) is below the typical ^{236}U abundance in natural uranium samples. The ^{236}U found in these samples is most likely a cross–contamination from sub–sampling or sample preparation. However, the very low $^{236}U/^{238}U$ ratios (down to 10^{-14}) can be measured by accelerator mass spectrometry (AMS). This technique was applied in our study to a few selected UOC samples. The obtained results for UOCs were not always consistent between respective ore samples indicating within mine inhomogeneity in the $^{236}U/^{238}U$ ratio of the U ores [5]. The same phenomenon was observed also for the $^{234}U/^{238}U$ ratio of UOC samples from the same mine. Additional studies are needed to understand these variations.

Chemical composition

Typically, UOCs contain a high fraction of natural uranium (> 65 wt% U) and involve a wide range of chemical compounds, such as ammonium uranate (AU), sodium diuranate, uranyl hydroxide, uranyl peroxide or an oxide form, if the material has been calcined. UOCs are typically recovered by precipitation of uranium using a variety of precipitants after purification by solvent extraction or ion exchange.

Fourier–transform infrared spectrometry (FTIR) can be used for the identification of uranium ore concentrates. Besides the chemical compound of U, FTIR can be used to detect the main anionic impurities in the UOCs [6]. A combination of the knowledge of the UOC's chemical form and its impurity content may yield information about the production of the UOC. This data can be utilized in the evaluation of potential production locales for the UOCs.

OUTLOOK

The research objectives were to identify nuclear forensic signatures that can be characteristic of the place of origin of uranium or the production processes used in uranium production, and several promising candidate signatures have emerged. However, in order to ascertain the robustness and persistence of these signatures, further research is needed into how the signatures behave in other U products representative of later stages of the nuclear fuel cycle (e.g., uranium fluorides and uranium oxides).

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DEVELOPMENT OF NUCLEAR FORENSICS METHODS AND TECHNIQUES FOR COMBATING ILLICIT TRAFFICKING OF NUCLEAR AND OTHER RADIOACTIVE MATERIAL

L. LAKOSI, E. SZELES, TAM C. NGUYEN, J. ZSIGRAI, ZS. STEFANKA, R. KATONA, ZS. VARGA, T. BIRO
Centre for Energy Research,
Hungarian Academy of Sciences
Budapest, Hungary
Email: lakosi.laszlo@energia.mta.hu

BACKGROUND

The majority of illicit nuclear and other radioactive material trafficking incidents in Hungary occurred during the 1990s, when both radioactive sources and nuclear material were confiscated from smugglers or found in metal scrap shipments. The smuggled nuclear materials were typically depleted, natural and low enriched uranium (DU, NU, LEU) pellets and also rods, originating from RBKM, VVER-1000, and CANDU nuclear reactors.

The Governmental Decree 17/1996 (I. 31.) delegated the identification, categorization and characterization of seized nuclear materials to the Institute of Isotopes (Hungarian abbreviation: IKI). The institute has the expertise and capabilities to perform basic measurements for characterization of seized nuclear material to support criminal investigations. The Institute of Isotopes is now organized as the Centre for Energy Research. Scientists are conducting research and development to further improve credibility, enhance applicability, and explore new indicators to improve nuclear forensics methods in Hungary.

Prompt identification and analysis of seized nuclear material in situ (e.g. those encountered at a border checkpoint) and maintenance of the integrity of forensic evidence favours the use of non–destructive analysis (NDA). According to national regulation, a requirement is to determine — in addition to the geometric and mechanical characteristics — the isotopic composition, the total amount of nuclear material and, as far as possible, matrix as well. In order to perform these tasks, high–resolution gamma–spectrometry (HRGS) is utilised. The on–site NDA is followed by detailed studies, in order to characterize — as fully as possible – any confiscated material by laboratory NDA in conjunction with destructive analysis (DA) methods.

Joint research

The γ–spectrometric method was tested in a series of demonstration exercises where on–site seizure of nuclear material was simulated. Exercises commenced in 1999 and were conducted regularly through 2008 with the participation of national authorities and observers from the International Atomic Energy Agency and European Commission's Joint Research Centre — Institute of Transuranium Elements (JRC–ITU).

OBJECTIVES

The Institute of Isotopes is developing a comprehensive nuclear forensics analytical capability that will enable Hungary to provide information on the origin and history of radioactive material outside of regulatory control and enable effective national and international response.

For the first year, the work plan included:

- Development of a method for determination of the age and isotopic composition of uranium bearing material seized in illicit trafficking incidents;

- Development of mass spectrometric technique for analysis of isotopic composition and comparison of results with non–destructive assay.

The work plan for the second year comprised three parts:

- Determination of the capabilities and limitations of HRGS for forensic analysis of uranium; in particular, identify isotopic ratios of NU, LEU and high–enriched uranium (HEU) samples with their associated measurement uncertainties to be considered as the forensic signature of the material;
- Preparation of the procedures for determination of the age of uranium–bearing nuclear material using a gamma spectrometric technique;
- Comparison of DA and NDA methods and development of guidelines for establishing forensic signatures of nuclear material.

The programme of the third year required:

- That the HRGS results be fully compared with atom based mass spectrometry detection technologies including inductively coupled plasma mass spectrometry to bench–mark this data for a range of uranium bearing materials of interest to nuclear forensics.
- Research to include scanning electron microscopy, x–ray diffraction analysis, laser ablation micro–analysis, and imaging analysis to determine distinguishing physical characteristics of uranium including grain size, shape and sorting.

Materials investigated in this study include: i) powder (oxide and other U compounds) assayed by DA and NDA methods; ii) certified DU, NU, LEU, HEU reference materials assayed by DA and NDA methods; iii) oxide and metallic HEU materials from the Nuclear Forensics International Technical Working Group (ITWG) round robin (RR) exercises assayed by DA and NDA methods; iv) seized DU, NU, LEU oxide fuel pellets of CANDU, RBMK, VVER–440, VVER–1000 types assayed by NDA methods; v) seized fuel rods (broken), VVR–SM fuel rods and assemblies, and EK–10 fuel rods assayed by NDA methods, vi) sealed sources assayed by NDA methods; and vii) U–ores assayed by DA and NDA methods.

CHARACTERIZATION OF URANIUM-BEARING MATERIAL OF UNKNOWN ORIGIN BY HRGS

NDA methods are routinely used in the Institute of Isotopes for quantitative assay of U–containing nuclear materials. The peak ratio method using intrinsic calibration for U isotopic abundance measurements as well as a relative method for mass determination of nuclear materials in the form of pellets or powder are used, based on a standard U calibration set, applying attenuation correction. Besides isotopic composition, physical form, and chemical impurities, the date of production of the material is important as well. Furthermore, identifying reprocessed uranium is an important clue in the characterization of nuclear material of unknown origin. In order to determine whether an interdicted uranium sample was prepared from reprocessed material, traces of fission or activation products are examined. One of the nuclides frequently used for this purpose is ^{236}U, which can easily be quantified using mass spectrometry. With gamma spectrometry ^{236}U can only be detected in weapons grade uranium, with a large margin of uncertainty (e.g., approximately 15%). Another nuclide which can only be found in reprocessed uranium is ^{232}U. This nuclide can be easily quantified by low–background gamma spectrometry, regardless of the ^{235}U enrichment.

Accordingly, the main steps of the analysis follow:

Determination of: 1) isotopic composition, including the control of the radioactive equilibrium between parent and daughter isotopes; 2) amount of U–isotopes; 3) total U–content (based on the two previous measurements); 4) age (production date – time of the last radiochemical separation) of the material; and 5) ^{232}U content.

The performance of the first three of the aforementioned methods was also tested under field conditions, using portable detection instrumentation.

Identification of isotopic composition

The isotopic composition / ratio (“isotopics”) is determined on the basis of the peak–ratio technique. By relying on peak ratios, there is no need to know absolute detection efficiencies. Allowing the enrichment to be determined without use of standards or without determination of geometry–dependent calibration constants, the relative efficiency (intrinsic) calibration method is based on the use of a relative efficiency curve as a function of energy. It is determined from the same spectrum as the measured activity ratios. Accordingly, the isotopic ratio can be obtained from the measured intensity ratio, the relative efficiencies taken at the corresponding energies and from the values of the decay constants and emission probabilities taken from the literature. The analyst determines several gamma intensities from individual U–isotopes, and normalizes the data to a common (relative) efficiency curve which, at the same time, accounts for the attenuation of different energy gamma rays in the absorbers and in the sample as well.

Gamma spectra of nuclear materials are routinely analysed in the 0–300 keV region by the advanced Multi–Group Analysis (MGA) commercial computer code, using intrinsic calibration. One module of this software, the “MGAU” (version 4.0) is used for determining U isotopic ratios, which provides reliable results in the range of 0.05 to 95% ^{235}U abundances [1,2]. Measuring times of about 15–60 minutes are usual for determining ^{235}U enrichments within 1.5–10% uncertainties [3]. A MGAU fit of the spectrum of a certified reference sample is shown in Fig. 1.

In addition to using automated spectrum evaluation, “manual” procedures were developed. For determining ^{238}U content, the unknown and reference samples are assayed by a 150 cm^3 coaxial high purity germanium (HPGe) detector, in some cases in a low–background counting chamber with a wall thickness of 20 cm and inner size of 120 × 60 × 120 cm (height × width × length). A relative efficiency curve is shown in Fig. 2, fitted for ^{232}U, ^{235}U, ^{238}U, ^{237}Np, ^{214}Bi line energies (the latter is necessary for U age dating, following) from a single spectrum.

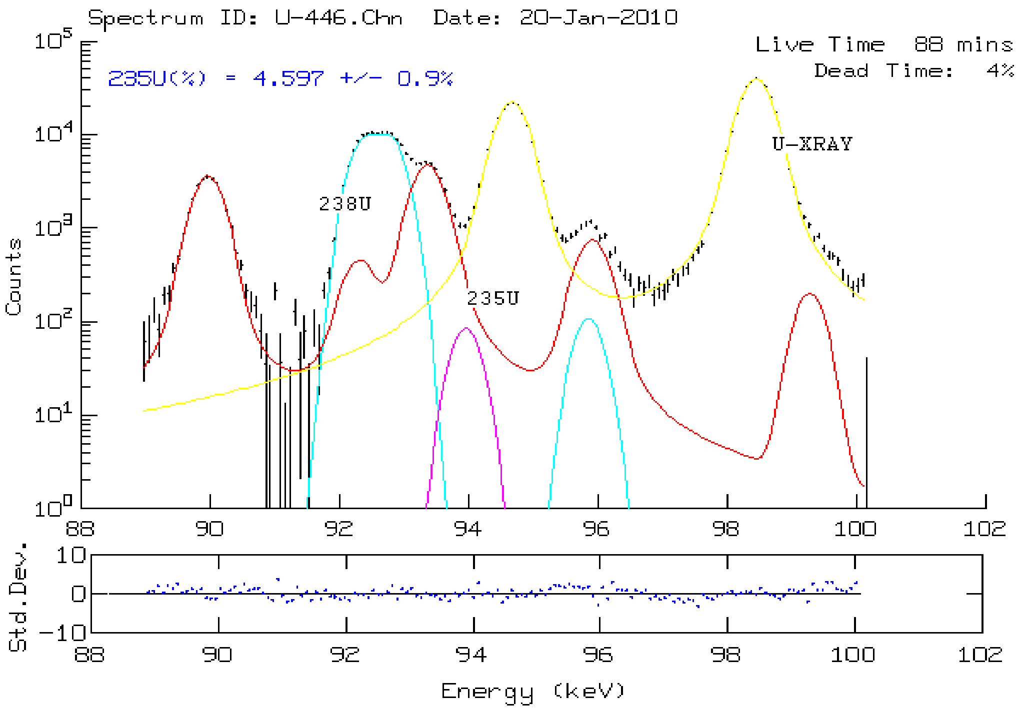

FIG 1. MGAU fit of the peaks of a 4.6% enriched certified reference sample in the low–energy region used for enrichment determination. Fitting errors are shown at the bottom.

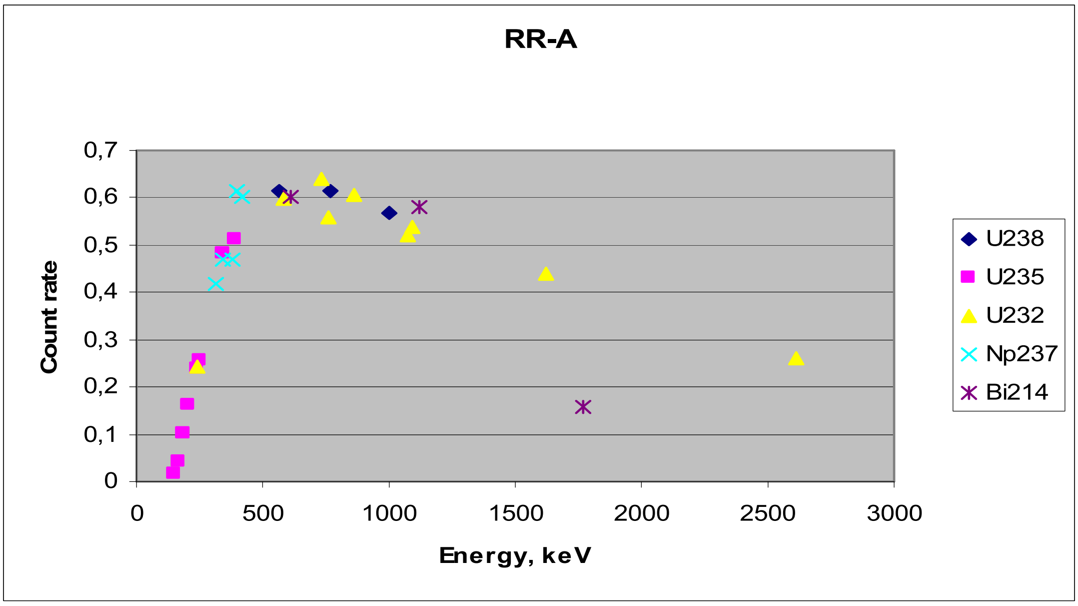

FIG 2. Relative efficiency curve taken from the spectrum of an ITWG round robin sample by the 150 cm^3 coaxial detector.

Low energy spectra of ^{235}U and the minor isotopes ^{234}U and ^{236}U are recorded by applying planar HPGe detectors. Relative efficiencies of such a detector can be seen in Fig. 3.

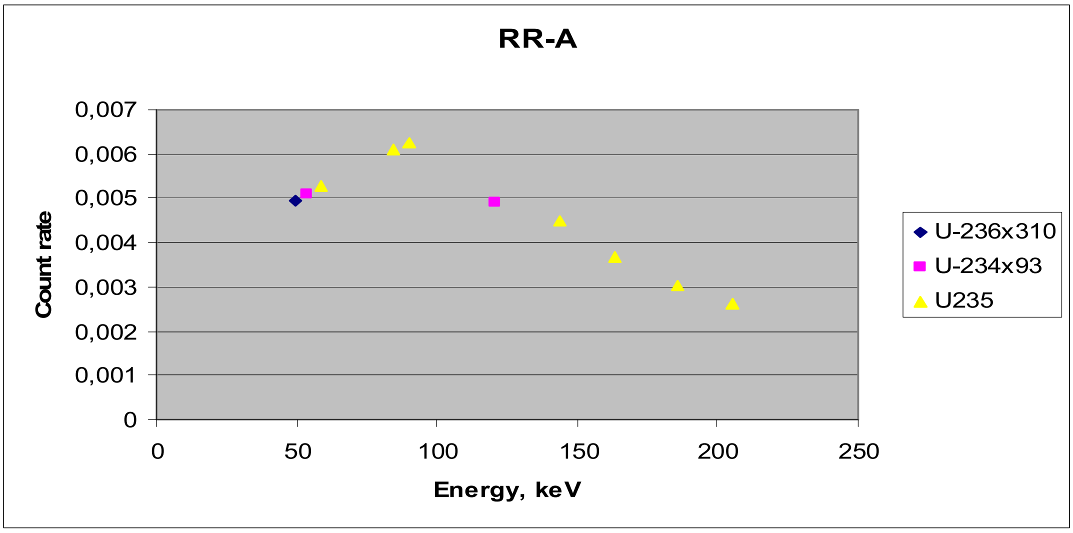

FIG 3. Relative efficiency curve taken from the spectrum of an ITWG round robin sample by a planar detector.

To a 2 g 90% enriched oxide sample we applied a reference material of known isotopic composition consisting of 90% ^{235}U enrichment for calibration. Measurements for four samples of approximately 0.5 g, 1 g, 1.5 g and 2 g were carried out with each material. The count rates per unit mass at different energies were fitted to the function that models the law of self–absorption within the sample. Values obtained for ^{235}U abundance do not depend on the thickness of the sample. Using relative efficiency calibration, for a 10 g 36% enriched oxide sample we achieved ±0.03 and ±0.02% relative uncertainties of ^{235}U and ^{238}U isotopic abundances, respectively.

The 49.37 keV gamma energy line was identified for ^{236}U decay. Although this peak is very close to the 49.55 keV peak of ^{238}U, in the case of 90% enrichment this does not disturb the evaluation of the 49.37 keV peak. Namely, in the observed sample the isotopic ratio of ^{238}U is so low, that the intensity of its peak is less than the background level i.e. it is below the detection limit. Therefore, the ^{238}U peak can be neglected when evaluating the peak intensity at 49.37 keV. The ratio ^{236}U/^{235}U was evaluated by comparing the 49.37 keV yield to the relative efficiency curve constructed from the yields of the 58.6 keV, 84.2 keV, and 90 keV peaks of ^{235}U. Then the ^{236}U content was calculated to be $(5.1 \pm 0.8)\times10^{-3}$ g/g total sample mass. This corresponds to 16% relative uncertainty in determining the isotopic abundance of ^{236}U.

Two approximately 5 g metallic HEU (90%) samples were also investigated. There was no reference material available for calibration so the isotopic composition was determined applying intrinsic calibration where a relative efficiency is determined from the same spectrum as the measured activity ratios. The spectrum of a 93% enriched metallic U sample from an ITWG round robin exercise (ITWG RR 2010) can be seen in Fig. 4, measured by the 150 cm^3 coaxial detector. The ^{232}U lines hint that the material contains reprocessed U and its intense gamma lines enable a relative efficiency calibration over a wide energy interval from 238.6–2614 keV. A relative efficiency function was constructed for each sample based on the presence of ^{232}U in them. As can be seen in Fig. 2, gamma lines of ^{237}Np were also identified. The former is further evidence – beside the presence of ^{232}U – that the sample contains reprocessed material.

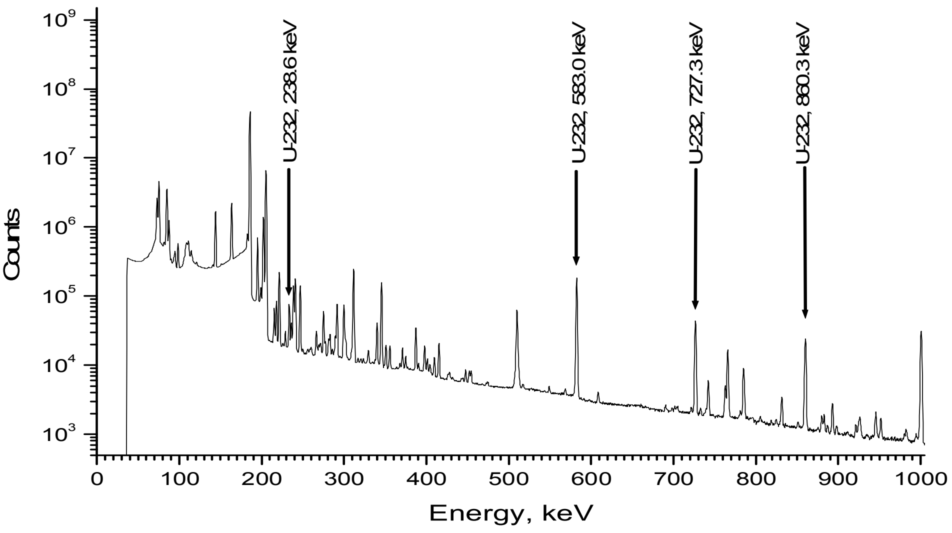

FIG 4. Spectrum of a 93% enriched 5 g metallic U sample from an ITWG round robin exercise (2010). ^{232}*U lines are available for intrinsic (relative efficiency) calibration for the whole energy range.*

The relative uncertainty in determining ^{238}U in metallic samples is much higher than that established for an oxide sample. The measurement was complicated by the reduced intensity due to the larger self–absorption in the high density U metal; additional measurements were not possible due to the unavailability of a reference sample. The improved results for ^{236}U are due to measuring cut fragments of the materials dissolved in nitric acid (i.e., destructive analysis), thus eliminating the intense Compton tail in the low energy region of the spectra of the dense samples. More precise results can be achieved in assaying less enriched samples.

Determining total uranium content

Unlike the enrichment measurements, which can be considered a more or less routine task, determination of the total U mass requires a sample–specific approach. The reason for this is that the attenuation correction needed for calculating total mass depends on the matrix, the cladding, and the geometry of the material. Additionally, the sample cannot be infinitely thick with respect to the assay gamma energy. For our purposes we were unable to use low–energy lines, since their intensity gets saturated due to the self–absorption even within the dimension of a single pellet, i.e. the intensity would be practically independent of the size and the U–content of the sample. For this reason, the 1001 keV line of ^{234m}Pa (a daughter product of ^{238}U) is used for the U–mass determination.

Applying a relative method, the mass of ^{238}U (M_{238}) was found from the count rate (C) in the 1001 keV peak, using calibration standards. If the sample is homogeneous in its composition and density, one can write $M_{238} = KC/F$, where F is the attenuation correction factor, characteristic of the sample material and geometry, while K depends on the detection efficiency and the γ–yield. In the case of relative measurements, however, K is a calibration constant, determined by using five certified reference sources (EC Nuclear Reference Material Set 171). The sources are encased in 7 cm diameter cans, each containing 200 g U_3O_8 powder in 2 cm thickness, of enrichment ranging from 0.32–4.46%. A common counting geometry was employed along the axis of the cans. If the radii of both the detector and the sample are much smaller than the source–to–detector distance, then the deviation of the detected γ–rays from the direction parallel to the system axis is negligible. In this case the setup can be approximated with a one–dimensional model. The attenuation correction factor was approximated by a standard method.

Several materials were selected for measurement. Pellets to be measured were chosen randomly from each material as a representative set and were arranged, next to each other, into a hexagonal lattice.

Samples of unidentified chemicals available in powder form were also examined. Both sample types were placed into the same type of container; the batches were composed of two types of the pellets each consisting of 19 pellets each. The pellets were aligned to the direction of the detector along their longitudinal axes, forming flat cylindrical (or rather hexagonal) disks 5.7 cm in diameter.

The uranium content of the seized fuel rods was determined by comparison with pellets already measured and arranged in rod form to serve as a reference sample. The rod was aligned perpendicularly to the detector axis. The count rate of the standard samples was measured and the calibration constant was determined for 10 cm, 20 cm, 30 cm and 60 cm source–to–detector distances. With the current geometry, 20 cm is the minimum distance for the one–dimensional model. The attenuation coefficient of the unknown samples was determined by placing a standard ^{152}Eu point source behind the examined material and measuring the transmission of the γ–rays of the source through the samples. From the ratio of the measured intensity of the 1001 keV line to the calculated F, the mass of ^{238}U can be determined using the calibration. From this and from the measured isotopic ratios the total U amount was easily calculated. On the basis of this procedure the U–content of pellet– or powder–form samples was determined to an accuracy of 2%. The results were good at a distance of 20 cm and were acceptable at a distance of 10 cm.

Uranium age determination by gamma spectrometry

The daughter/parent ratio as a function of decay time is widely used for determining the age of radioactive samples. In the case of uranium, age dating determination is somewhat difficult because the relevant isotopes (^{234}U, ^{235}U, ^{238}U) have very long half–lives, so only small amounts of daughter nuclides accumulate as in–growth. In contrast, the age of nuclear materials is, at most, merely a few decades, which is very short compared to the long half–lives of the parent isotopes. Therefore, one would expect that the daughter nuclides could only be quantified after destructive chemical separation, followed by mass–spectrometric or alpha–spectrometric techniques. However, it has been demonstrated that the daughter/parent activity ratio for ^{214}Bi / ^{234}U can be obtained by directly measuring the count rates of the relevant gamma peaks of ^{214}Bi and ^{234}U by low–background gamma spectrometry. Based on this, we developed a new method for uranium age dating using gamma–spectrometry [3,4]. This method is non–destructive and does not require the use of reference materials with known ages.

^{234}U decays through ^{230}Th to ^{226}Ra, which in turn decays to ^{214}Bi through three short–lived nuclides. The time needed for secular equilibrium between ^{226}Ra and ^{214}Bi is about 2 weeks, so it can be assumed that the activities of ^{226}Ra and ^{214}Bi are equal at the time of the measurement. Therefore, using the law of radioactive decay, the activity ratio ^{214}Bi/^{234}U at time T after purification of the material can be calculated with a good approximation as

$$\frac{A_{Bi214}}{A_{U234}} = \frac{A_{Ra226}}{A_{U234}} = \frac{1}{2}\lambda_{Th230}\lambda_{Ra226}T^2 . \qquad (1)$$

The activity ratio ^{214}Bi/^{234}U can be determined in several ways from the gamma spectra of a sample. A reliable method uses a reference material of approximately like age, enrichment, and form, as the investigated sample. Another approach does not require any reference materials but uses the absolute efficiency of the detector determined by “point–like” standard sources [3]. The activities of ^{214}Bi and ^{234}U in the sample are measured in an efficiency–calibrated geometry. In an alternate approach, a relative efficiency calibration is used to determine the activity ratio ^{214}Bi/^{234}U, without the use of any standard or reference materials [4].

The intensity of the peaks of ^{234m}Pa can be accurately measured within a reasonable timespan (1–2 days), if the relative ^{235}U abundance of the material is less than approximately 90% and provided that a sufficient amount of the investigated material is available (depending on detector efficiency). The relative efficiency calibration is applicable to samples of arbitrary shape and chemical form (e.g. fuel rods). Measurements of the HEU metal and oxide powder, HEU and LEU reactor fuel rods, LEU pellet and powder were made in the enrichment range from 4.4–90%.

It should be noted that for lower ^{235}U abundances, the amount of ^{234}U (and therefore of ^{214}Bi) is lower as well, so the corresponding activity is more difficult to measure and the uncertainty caused by the variation of the natural background becomes greater. In addition, a Compton background caused by the peaks of ^{238}U daughters is also present in the spectrum, disturbing the evaluation of the ^{214}Bi peaks. A lower limit on the ^{235}U abundance of the material exists that allows the age to be determined by gamma–spectrometry, depending on the amount and the age of the material, detector efficiency and background level. For example, with our current equipment, the age of 10 g of uranium oxide powder having natural uranium isotopic composition can only be measured if it is more than approximately 70 years old. Conversely, it has been estimated that with an average well type detector, if 10 grams of uranium oxide is used for the measurement, the lower limit for age–dating could be around 30 years for natural uranium, and as low as two years for 90% enriched HEU. The least enriched uranium to be dated by HRGS was an oxide enriched to 5%; the age of which was determined to be 54 ± 7 years. The youngest sample was 6.7 ± 0.7 year–old metallic U of 90.8% enrichment.

Age results of the U samples assayed at IKI are plotted as a function of the activity ratio ^{214}Bi/^{234}U in Fig. 5.

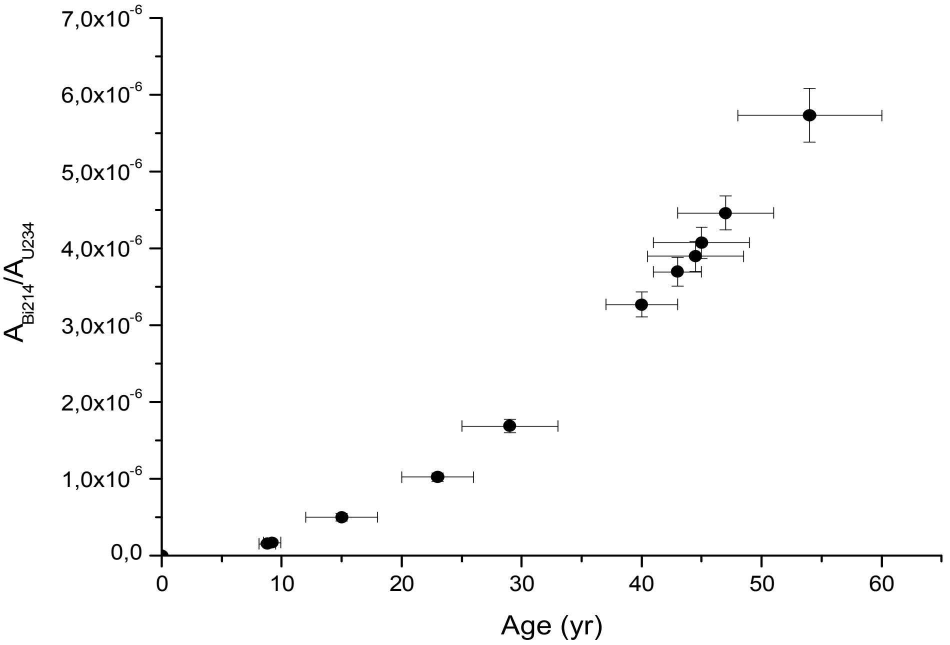

FIG. 5. Age results measured at the Institute of Isotopes by high resolution gamma ray spectroscopy.

The activity of ^{214}B in LEU material is low, hence the fluctuation of the natural background due to atmospheric ^{222}Rn, predecessor of ^{214}Bi, has a substantial influence on the result. In order to reduce this effect, the sample to be measured was tightly surrounded by polystyrene foam (“Styropor”). In addition, N_2 vapor was directed around the sample through a pipe from the cryostat of the detector to displace the air from its surroundings with the assumption that any possible radon activity in liquid nitrogen has already decayed by the time of the measurement. Systematic control of the 609 keV peak in the background spectrum confirmed this expectation when performing measurements in the counting chamber.

Identifying reprocessed uranium by HRGS

HRGS is capable of revealing whether or not uranium (of any enrichment) has been reprocessed based on the detection of ^{232}U relative to mass spectrometry that indicates the presence of ^{236}U. Uranium-232 activity was quantified by low–background gamma spectrometry, irrespective of the ^{235}U enrichment [5], from the activity of its descendants, ^{212}Bi and ^{208}Tl. Their activities were determined relative to ^{238}U, using a relative efficiency curve (like the one in Fig. 4) for each sample. The activity ratio ^{232}U/^{238}U was measured in a low–background counting chamber using the 150 cm^3 coaxial germanium detector. For the study a set of certified reference materials (CRM) and several other samples were used, with ^{235}U enrichments in the range of 0.23–90%. The ^{232}U content in the samples is plotted in Fig. 6 as a function of the enrichment. Seized enriched fuel pellets and the ITWG round robin samples indicate these samples incorporate reprocessed material. This is supported by the ^{237}Np found in the ITWG round robin samples.

As can be seen, however, ^{232}U can be found in all the enriched and even depleted CRM samples. The ^{232}U content in those not reprocessed ("virgin") materials may be due to the ^{232}U contamination in the enrichment facilities.

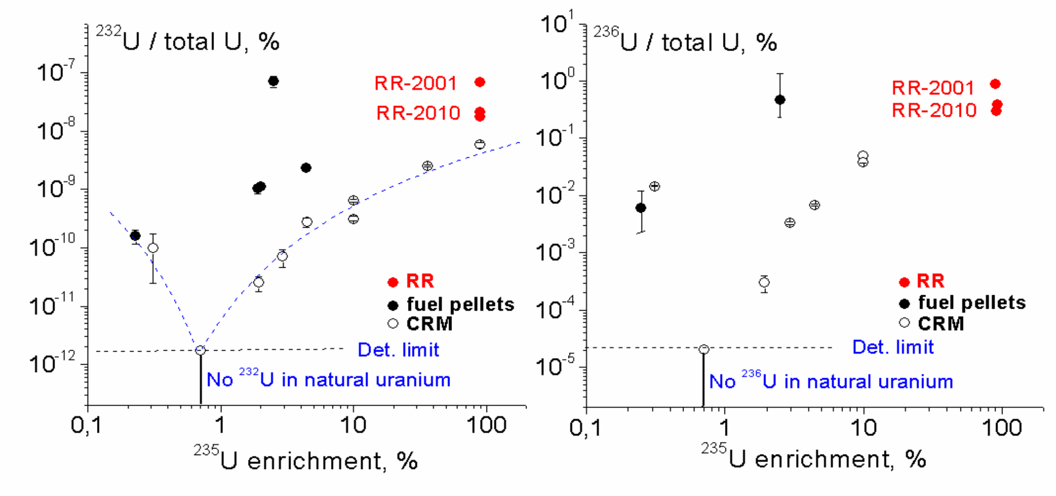

FIG 6. ^{232}U and ^{236}U content of several reference and various other materials as a function of ^{235}U enrichment.

For certified reference material samples, uranium isotope ratio measurements made by mass spectrometry were also available. The mass spectrometry results showed that in those samples in which a higher amount of ^{232}U was found, there is also much more ^{236}U than in the other ones, confirming that these samples contain reprocessed uranium.

For comparison, ^{236}U content of these materials is plotted in Fig. 6 as well. The ^{236}U content was provided for the CRM samples in the certificates, while fuel pellets and round robin samples were analysed for ^{236}U content by ICP–MS.; a similar pattern of isotope distributions is revealed. It would appear that ^{236}U content in virgin material is also due to the contamination in the enrichment facilities. Preliminary results show strong correlation between the two minor isotopes, indicating a 236–to–232 ratio of about 10^7, in accordance with values in the literature [6]. More data is clearly needed.

The 0.6% ^{236}U content in a seized reactor fuel pellet (Fig. 6) is evidence of the presence of reprocessed material in the fuel, which may have serious consequences in depletion calculations.

MASS SPECTROMETRIC ASSAY OF URANIUM AND TRANSURANIUM ELEMENTS USING VARIOUS SAMPLE INTRODUCTION METHODS

At present, ICP–MS is the most frequently used inorganic mass spectrometry technique for concentration and isotope ratio measurements down to femtogram/gram level. This powerful analytical technique is also increasingly used for the measurement of long–lived radionuclides, including uranium and plutonium (Pu). The ICP–MS technique offers a new possibility for fast, simple and relatively inexpensive destructive analysis of nuclear samples [7, 8].

As a DA method isotope dilution (ID) ICP–MS measurements are very precise and give useful results in the case of isotope ratios and isotopic concentrations in ultra–low level. However the solution based ICP–MS technique, is usually time consuming and requires chemical sample preparation.

ICP–MS combined with a laser ablation sample introduction unit offers the possibility for a near non–destructive analytical method exploiting the high sensitivity and accuracy of the ICP–MS technique.

Analysis of single particles by laser ablation ICP–MS (LA–ICP–MS)

The LA–ICP–MS method is a quasi non–destructive analysis that is used for age determinations and isotopic composition determinations of small single particles of HEU; nanogram amounts of a sample are sufficient to make the measurements.

A novel methodology was developed for the measurement of the isotopic composition measurement of single particles. A summary of isotopic analysis results from individual uranium particles by LA–ICP–sector-field (SF) MS is listed below [9]:

- A technique was developed for U–oxide particles with different enrichment and lateral dimensions (down to 10 microns)
- Application for low–abundance (i.e., minor) isotopes (^{234}U, ^{236}U)
- Achieved a typical precision of: 0.2–5% RSD for ^{235}U/^{238}U
- Evaluation for analysis of sub–micron particles (JRC Institute for Reference Materials and Measurements; Nuclear Signatures Interlaboratory Measurement Evaluation Programme)
- Location of particles for fission track and scanning electron microscope/ energy dispersive spectrometry (SEM/EDS)

HEU samples were analysed by isotope dilution ICP–MS (ID–ICP–MS) and LA–ICP–MS for comparative purposes. ID–ICP–MS requires sample dissolution. Results from the two ICP–MS methods were also compared to the results of gamma–spectrometric measurements. The ID–ICP–MS technique was chosen as the reference to compare LA–ICP–MS against due to the homogeneity of the dissolved sample and resultant superior signal stability over the course of the measurement.

For ID–ICP–MS sample preparation, 300–500 mg of each sample were weighed into a plastic tube and dissolved in 9 mL of 10M ultra–pure nitric acid heated to 90°C for 6 hours. Solutions were diluted before introduction into the ICP–MS.

The analyses were performed using double–focusing magnetic sector ICP–MS equipped with a single electron multiplier (ELEMENT2, Thermo Electron Corporation, Bremen, Germany). Measurements using liquid sample introduction were carried out in low–resolution mode (R=300) with a low–flow microconcentric nebulizer operated in a self–aspirating mode connected to a stable introduction system spray chamber in order to improve isotope ratio precision. The equipment is installed in a clean room (Class 100 000) in order to mitigate the risk of cross contamination among samples. The sample preparation laboratory is of Class 10 000, with a Class 100 laminar flow hood. Tuning and mass calibration of the instrument by LA–ICP–MS measurements was verified by the analysis of the NIST 612 reference material.

For LA–ICP–MS sample introduction, a Nd:YAG laser was employed (New Wave model UP–213; Fremont, California USA). The ablated material is transported by argon as a carrier gas into the plasma. Using a NIST 612 glass reference standard, the laser was optimized to maximize $^{238}U^+$ intensity and minimize the UO^+/U^+ ratio. Laser ablation measurements were carried out using an UP–213 system. For the measurements line scan was used, scan speed: 5µm s^{-1}, repetition rate: 10 Hz, laser beam diameter: 95 µm, laser energy: 80% (0.28 mJ). Figure 7 is a typical laser ablation signal from a HEU particle.

The spectra during the laser ablation measurements were recorded in chromatographic mode. The $^{230}Th/^{234}U$ intensity ratio was calculated by the integration of the isotope signals over the selected ranges. For the integration the built–in software of the ELEMENT2 instrument (Thermo Electron Corporation, software Version 2.41) was used. For the analysis three parallel measurements were used at different sampling positions.

The calculation of combined uncertainties of calculated age values was performed by the use of commercially available GUM Workbench software as specified by the ISO/BIPM guide.

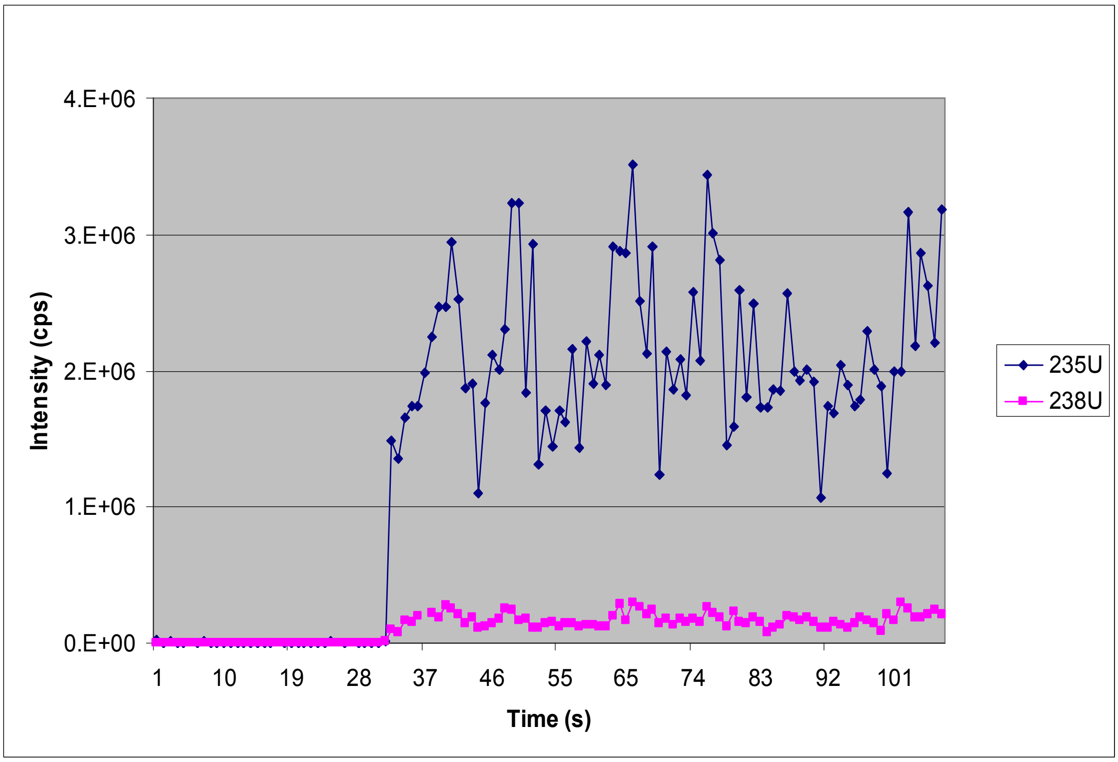

FIG 7. A typical laser ablation signal of the HEU particle.

Table 1 presents a summary of the analysis data of a single HEU metallic particle by ID–ICP–MS, LA–ICP–MS and HRGS.

TABLE 1. RESULTS OF A SINGLE HEU–1 METALLIC PARTICLE SAMPLE USING DA AND NDA TECHNIQUES AVAILABLE AT HUNGARY'S INSTITUTE OF ISOTOPES. UNCERTAINTIES ARE GIVEN WITH A COVERAGE FACTOR K = 2 (CONFIDENCE LEVEL APPROXIMATELY 95%)

Samples: HEU–1				
Nuclide	HRGS	ID–ICP–MS	LA–ICP–MS	
			Bulk analysis	Particle analysis
U-232 g/g	1.2E-10±1.5E-11	-	-	-
U-234	0.01±0.0003	0.0042±0.0002	-	-
U-235	0.926±0.01	0.925±0.009	-	0.926±0.03
U-236	0.003±0.0003	0.0035±0.0002	-	-
U-238	0.061±0.01	0.061±0.0065	-	0.065±0.004
Pu-239 pg/g	-	63.46±3.54	-	-
Pu-240	-	8.44±0.548	-	-
Pu-241	-	5.55±0.588	-	-
Age (years)	7.3±0.7	7.59±0.29	7.66±0.34	-

Results from HRGS analysis are in good agreement with mass spectrometric measurements. Solution based (ID–ICP–MS) uranium isotopic composition results are in good agreement with results from bulk and particle analysis by LA–ICP–MS.

Results show that DA and NDA methods all together as complementary techniques are applicable for characterization of HEU.

Analysis of uranium–bearing materials by LA–ICP–MS

Uranium oxide pellets of various enrichments and other materials confiscated in Hungary as well as samples of ITWG round robin exercises were also analyzed. For the destructive analyses the known amounts of uranium–oxide materials were dissolved in 6M ultrapure nitric acid while heating slightly in a water bath. The uranium oxide powders (the ITWG round robin HEU sample and the laboratory–prepared natural uranium–oxide powder) were pressed hydraulically at a pressure of 20 MPa into a disk–shaped pellet with a diameter of 5 mm and an approximate thickness of 1 mm. Compression time was approximately 3 minutes. Nuclear fuel samples could be directly analyzed by LA-ICP–MS. The spectra during the laser ablation measurements were recorded in chromatographic mode. The ^{230}Th/^{234}U intensity ratio was calculated by the integration of the isotope signals over the selected ranges. For the integration the built–in software of the ELEMENT2 instrument (Thermo Electron Corp., software Version 2.41) was used. For the analysis three parallel measurements were used at different sampling positions. An example spectra from the LA–ICP–MS is presented in Fig. 8.

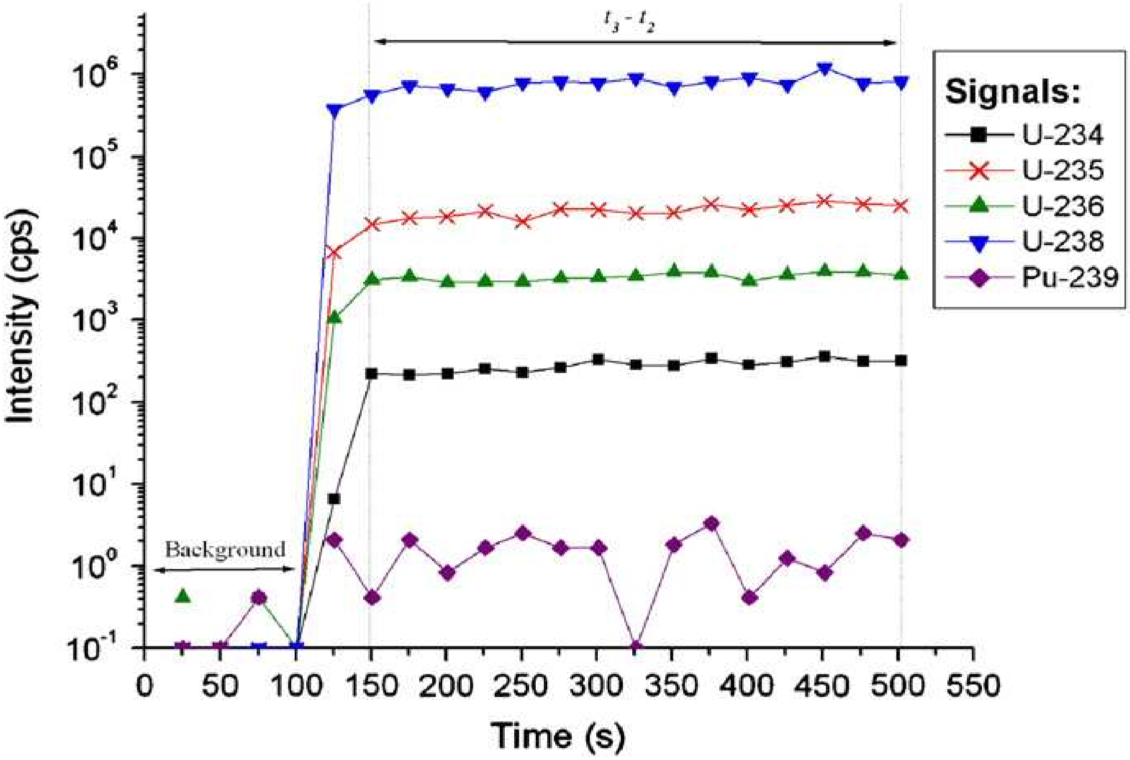

FIG. 8. A typical LA–ICP–MS measurement of a LEU sample in medium resolution mode (R=4000).

Analytical results are shown in Table 2 in the case of HEU samples originated from the ITWG round robin exercise. Results of IKI are in good agreement with other results from various laboratories that participated in the ITWG round robin exercise in 2001.

TABLE 2. RESULTS OF THE MEASUREMENTS OF ISOTOPIC COMPOSITION OF THE ITWG HEU ROUND ROBIN SAMPLE (2001). ALL RESULTS ARE SHOWN IN MASS PERCENT. HUNGARY'S INSTITUTE OF ISOTOPES ANALYSES ON THE LAST LINE OF TABLE. ALL PARTICIPATING LABORATORIES ARE SHOWN BY THE ANONYMOUS CODED NAMES; THE APPLIED ANALYTICAL METHODS ARE SHOWN IN PARENTHESES.

Laboratory (Analytical method)	^{234}U	^{235}U	^{236}U	^{238}U
Azores (HRGS, ICP-MS, TIMS))	0.97	89.99	0.68	8.37
Borneo (ICP-MS)	0.85 ± 0.15	86.7 ± 1.5	0.57 ± 0.08	11.9 ± 0.9
Chatham (TIMS)	0.960 ± 0.001	89.94 ± 0.06	0.643 ± 0.003	8.462 ± 0.006
Galapagos (TIMS)	0.96	89.89	0.68	8.47
Mindanao (TIMS)	0.96 ± 0.40	89.91 ± 0.11	0.678 ± 0.23	8.443 ± 1.29
Tobago (ICP-MS)	1.05 ± 0.07	89.37 ± 1.8	0.69 ± 0.05	8.88 ± 0.2
Tonga (TIMS)	0.967 ± 0.001	89.99 ± 0.002	0.679 ± 0.001	8.362 ± 0.005
Trinidad (MC-ICP-MS)	0.995 ± 0.075	90.01 ± 0.35	0.673 ± 0.030	8.365 ± 0.033
Average	*0.964 ± 0.055*	*89.475 ± 1.140*	*0.661 ± 0.039*	*8.906 ± 1.220*
IKI (LA-ICP-SFMS)	0.965 ±0.050	90.032 ± 1.300	0.681 ± 0.040	8.322 ± 0.250

Summary of isotopic composition results from the ITWG 2001 HEU round robin sample

For validation and cross–checking of the LA–ICP–MS method, a joint analysis project was carried out between the IKI and the JRC-ITU, which is the center of nuclear forensic applications in Europe. In the frame of the joint analysis, uranium oxide pellets from three different batches, corresponding to three different seizures in Hungary, were investigated.

Through joint analysis with the ITU, the isotopic composition of a seized NU and a LEU oxide pellet were analysed at IKI by HRGS and LA–ICP–SFMS. The data was generally consistent with that obtained at ITU by several mass spectrometers, namely by multi-collector (MC)–ICP–MS, TIMS, isotope dilution (ID)–MS, and by HRGS as well. Data are shown in Table 3–4. Comparative results from the NU material show that for ^{235}U IKI LA–ICP–MS results agree within 0.5% with ITU TIMS results, which is generally regarded as a benchmark technique for enrichment determination. With regard to the LEU material, IKI results agree within 0.67% of ITU TIMS.

TABLE 3. ISOTOPIC COMPOSITION OF A SEIZED NATURAL URANIUM OXIDE MATERIAL USING DESTRUCTIVE AND NON–DESTRUCTIVE ANALYTICAL METHODS AT THE INSTITUTE FOR TRANSURANIUM ELEMENTS AND BY HRGS AND LA–ICP–MS AT HUNGARY'S INSTITUTE OF ISOTOPES. DATA ARE SHOWN IN WEIGHT PERCENT, THE VALUES IN BRACKETS DENOTE TWO STANDARD DEVIATIONS OF THE MEAN VALUES (2σ)

	MC-ICP-MS	TIMS	Solution nebulization (SN)-ICP-MS	HRGS (ITU)	HRGS (IKI)	LA-ICP-MS
U-234	0.00515(9)	0.00494(31)	0.00579(55)	0.005(8)	0.007(2)	0.00524(10)
U-235	0.71213(65)	0.71121(41)	0.71100(85)	0.70(10)	0.716(18)	0.7083(43)
U-236	nd	nd	0.0012(22)	nd	nd	nd
U-238	99.2827(6)	99.2839(20)	99.2820(12)	99.30(10)	99.278(18)	99.2865(44)

nd: not detected

TABLE 4. ISOTOPIC COMPOSITION OF A SEIZED LEU MATERIAL. THE INVESTIGATION WAS CARRIED OUT USING DESTRUCTIVE AND NON-DESTRUCTIVE ANALYTICAL METHODS AT THE INSTITUTE FOR TRANSURANIUM ELEMENTS AND BY HRGS AND LA-ICP-SFMS AT HUNGARY'S INSTITUTE OF ISOTOPES. DATA ARE SHOWN IN WT%, THE UNCERTAINTIES IN BRACKETS DENOTE TWO STANDARD DEVIATIONS (2σ)

	MC-ICP-MS	TIMS	SN-ICP-SFMS	HRGS (ITU)	HRGS (IKI)	LA-ICP-SFMS
U-232					3.2(9)x10^{-8}	
U-234	0.0346(5)	0.0347(21)	0.0345(33)	0.025(20)	0.0362(24)	0.0358(9)
U-235	2.5136(14)	2.5121(14)	2.5119(30)	2.51(12)	2.562(34)	2.529(19)
U-236	0.451(22)	0.47(44)	0.47(86)	-	0.38(24)	0.474(24)
U-238	97.000(21)	96.9823(20)	96.9829(12)	97.47(12)	97.021(34)	96.961(20)

The methods developed for isotopic analysis by LA-ICP-MS were repeatedly cross–checked using the sample which originated from the HEU ITWG round robin exercise (2001) with excellent agreement as noted in Table 5 and 6.

TABLE 5. ISOTOPIC COMPOSITION OF THE ROUND ROBIN HEU SAMPLE BY HRGS AND LA–ICP–SFMS, IN COMPARISON WITH THE AVERAGE OF THE ROUND ROBIN INTERLABORATORY EXERCISE

	^{234}U	^{235}U	^{236}U	^{238}U
Average of RR	0.964±0.055	89.475±1.14	0.661±0.039	8.906±1.22
HRGS	0.98±0.07	89.8±0.7	0.6±0.1	8.6±0.5
LA-ICP-SFMS	0.965±0.05	90.032±1.30	0.681±0.040	8.322±0.25

Comparison of the results for the production date of the round robin HEU sample is seen in Table 6.

TABLE 6. PRODUCTION DATE OF THE ROUND ROBIN HEU SAMPLE BY HRGS AND ICP-SFMS, IN COMPARISON WITH THE AVERAGE OF THE ROUND ROBIN EXERCISE

Production date by HRGS	Production date by ICP-SFMS	Production date from RR
1978 (±3 *a*)	August, 1979 (±6 months)	February–July 1979

Uranium age determination

For determination of the production date of a uranium sample the ^{230}Th/^{234}U chronometer commonly used in geological applications was applied. Measurement of this parameter and the mentioned isotope ratios is possible with solution based ICP–MS and LA–ICP–MS techniques. The production date determination by ^{230}Th/^{234}U ratio is based on the decay of the relatively long-lived ^{234}U ($t_{1/2}$=245,250 years) to ^{230}Th ($t_{1/2}$=75,690 years) and the disequilibrium between these two radionuclides. After the last chemical separation of ^{234}U during the preparation of the nuclear material, the concentration of ^{230}Th daughter nuclide is continuously increasing in the uranium–oxide material with the age T, as represented by:

$$N_{Th230}(T) \approx N_{U234}\lambda_{U234}(0)T,$$

where N and λ are the respective number of atoms and the decay constant. Though the intensity ratio of ^{230}Th and ^{234}U can be directly obtained from the mass spectrum of the laser ablation measurements, correction has to be applied in order to convert it to ^{230}Th/^{234}U atom ratio, which is necessary for age determination. The direct determination of ^{230}Th/^{234}U atom ratio from the mass spectrum using laser ablation is hindered by the slightly different ablation and ionization efficiencies of ^{230}Th and ^{234}U. For the calculation of isotope ratios, the relative sensitivity factor (RSF) approach is commonly applied, which takes into account the differences in the ionization and ablation efficiencies of the elements at fixed conditions (constant laser energy, crater diameter and repetition rate).

Determination of production date of nuclear materials – destructive analysis involves:

- Digestion of the uranium–oxide sample
- Determination of ^{234}U–content by isotope dilution (ID)–ICP–MS analysis
- Determination of ^{230}Th–content by ID–ICP–MS analysis after extraction chromatographic separation
- Production of ^{229}Th tracer from ^{233}U–solution ("milking")

Further detail of the destructive analysis is provided in Table 7.

TABLE 7. DETERMINATION OF PRODUCTION DATE OF URANIUM

	^{230}Th Determination	^{234}U Determination
Sample Preparation		
Time of sample preparation	2 days	3 hours
Uranium separation factor	< 4.0×10^{9}	Not available
Recovery	53–86%	Not available
Detection Limit	0.20 picogram (0.15 mBq)	0.24 picogram (4.6×10^{-2} mBq)
Precision (*k*=2)	0.8–2.51%	0.67–1.2%
Analysis time	2 hours	2 hours
Uncertainty (*k*=2)	0.85–2.3%	0.49–0.73%

During our work ^{230}Th-content was determined by ID–ICP–MS analysis after extraction by a chromatographic separation. The ^{229}Th tracer used for the ^{230}Th determination was prepared by separating ("milking") the ^{229}Th from an 18–year–old ^{233}U standard solution (New Brunswick Laboratory, USA).

Concentrations of isotopes of interest measured by liquid sample introduction were calculated as a function of ^{230}Th/^{229}Th and ^{234}U/^{233}U ratios according to the isotope dilution method. All raw data was corrected taking into account instrumental mass bias using linear correction. The overall uncertainty was calculated taking into account the uncertainty of the weight measurements, tracer concentrations, measured intensities and half–lives according to ISO/BIPM guide. The calculations were carried out by the use of the GUM Workbench software.

The ^{230}Th/^{234}U ratio in the investigated uranium–oxide samples was also determined by LA–ICP–SFMS in chromatographic mode. The intensities of ^{230}Th and ^{234}U signals were recorded as a function of time.

The method developed for the analysis of uranium–oxide samples was applied for the production date determination of the confiscated nuclear materials and the round robin sample. The results obtained are summarized in Table 8.

TABLE 8. ANALYSIS RESULTS OF INVESTIGATED NUCLEAR MATERIALS BY THE DESTRUCTIVE ICP–MS METHOD

Sample	^{235}U/^{238}U ratio	Measured ^{230}Th/^{234}U atom ratio	Calculated age (a)	Production date
HU-DU	0.00258±0.00004	$(3.88\pm0.28)\times10^{-5}$	13.7±1.0	Aug 1993 (±12 months)
HU-NU	0.00713±0.00014	$(5.01\pm0.24)\times10^{-5}$	17.73±0.87	July 1989 (±10 months)
HU-LEU	0.0255±0.0013	$(4.38\pm0.28)\times10^{-5}$	15.51±0.98	Sep 1991 (±12 months)
RR-HEU	10.80±0.17	$(7.89\pm0.36)\times10^{-5}$	27.9±1.3	June 1979 (±16 months)

Trace element determination

Determination of impurities (non–radioactive elements) in nuclear materials may be used to identify nuclear forensic signatures imparted during material production or inherited from the ore body. Analysis of elemental composition was performed using ICP–MS. The given data (Figs. 9–12) are preliminary results from direct analysis from an acid digestion using diluted samples. More precise data requires extraction using a chromatographic separation.

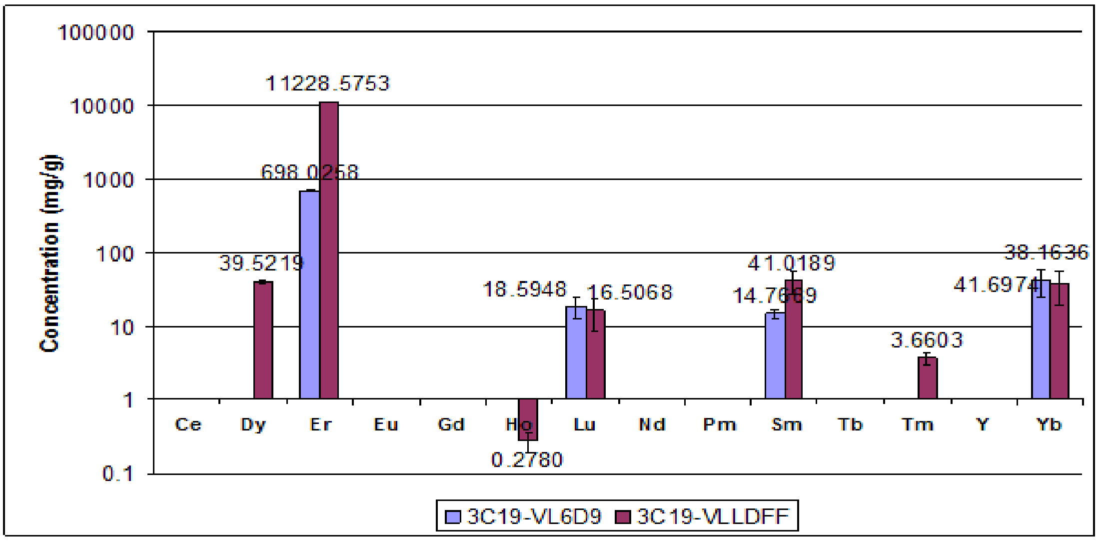

FIG 9. ICP–MS results: REE content in ITWG HEU round robin (2010) with sample A and B comparison.

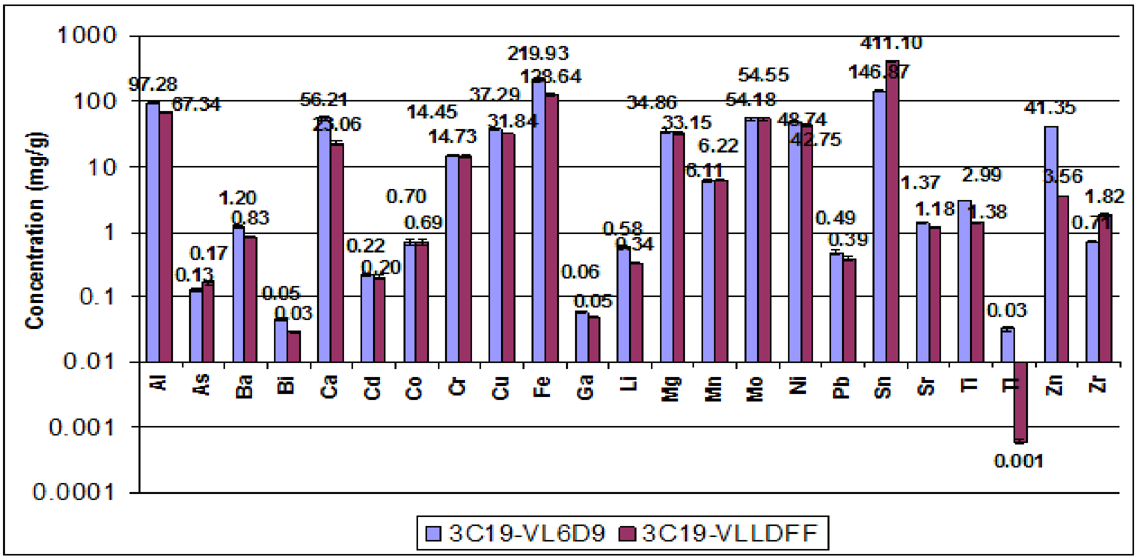

FIG 10. ICP–MS results: Impurities in ITWG HEU round robin (2010) with sample A and B comparison.

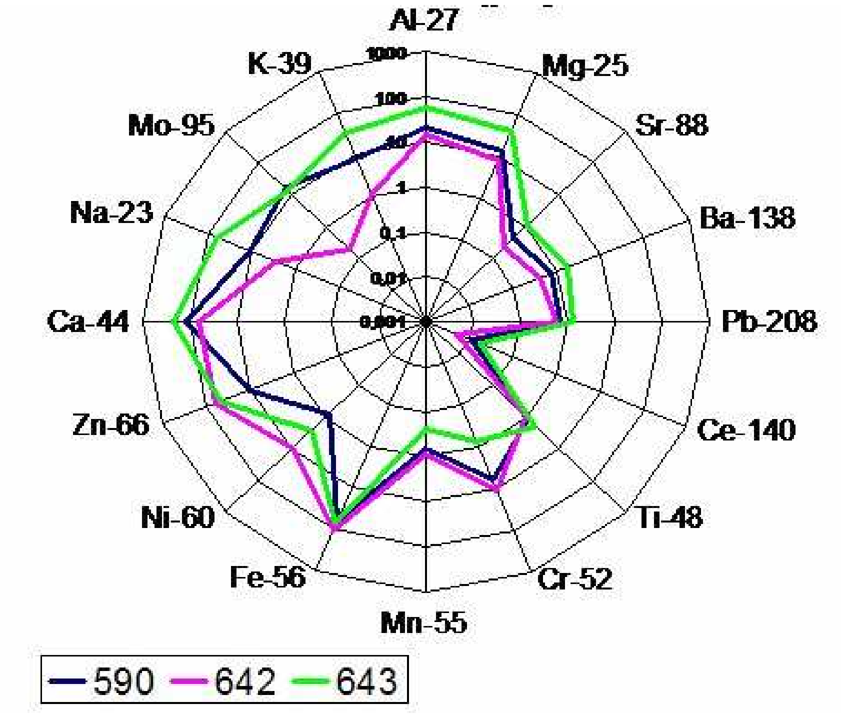

FIG. 11. *Trace element analysis in three confiscated U–oxide pellets. Samples: 590: NU; 642: LEU; 643: DU.*

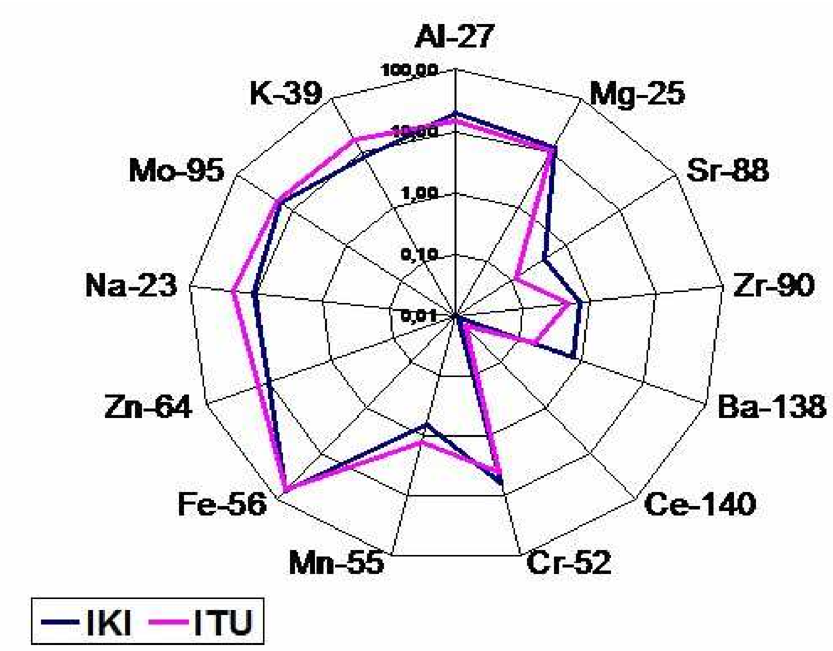

FIG. 12. *Analyses comparing results from the Institute of Isotopes (IKI) and the Institute for Transuranium Elements (ITU).*

Comparison of ICP–MS and HRGS results: isotopics

Table 9 displays the results for the isotopic composition of a ITWG round robin metal sample obtained by HRGS and ICP–MS are shown as an example. It can be seen that the accuracy of mass spectroscopy is better for the major U isotopes ^{234}U, ^{235}U, ^{236}U, ^{238}U. For ^{232}U and ^{237}Np HRGS data are only available. The results obtained by intrinsic calibration are in red italics, whereas those in black come from measurements done by the MGAU code. Gamma spectrometric data for ^{232}U, ^{236}U and ^{237}Np are available from the use of intrinsic calibration only.

TABLE 9: COMPARISON OF ICP–MS AND HRGS ISOTOPICS RESULTS:
ITWG RR 2010: SAMPLE A. (EXPANDED UNCERTAINTY: 2 STANDARD DEVIATIONS)

HRGS			**ICP–MS**		
Isotope	Abundance, %	Expanded uncertainty	Isotope	Abundance, %	Expanded uncertainty
U-232	*1.23E-8*	*± 0.15E-8*			
U-234	0.929 *0.996*	± 0.192 *± 0.03*	U-234	0.965	± 0.018
U-235	92.81 *92.6*	± 1.57 *± 1.0*	U-235	92.5	± 0.9
U-236	*0.30*	*± 0.03*	U-236	0.35	± 0.019
U-238	6.26 *6.1*	± 1.57 *± 1.0*	U-238	6.1	± 0.65
Np-237	*5.5E-4*	*± 0.6E-4*			

Comparison of ICP–MS and HRGS results: age dating

In Fig. 14 and Table 10 age data is shown as obtained for the two HEU ITWG round robin metal samples analyzed by HRGS, ID–ICP–MS and LA–ICP–MS. One can see that results are consistent and there is a systematic deviation between the ages of the two samples. According to supplier data, Sample B is younger by 7.5 months. Such a large difference is best approximated by HRGS data: 7.2 ± 1 month, while mass spectrometry data indicates the difference to be ~ 3 months. For comparison, supplier's data are 6.88 and 6.25 years. Although uncertainties are higher for HRGS, the values are nevertheless closer to the absolute ages.

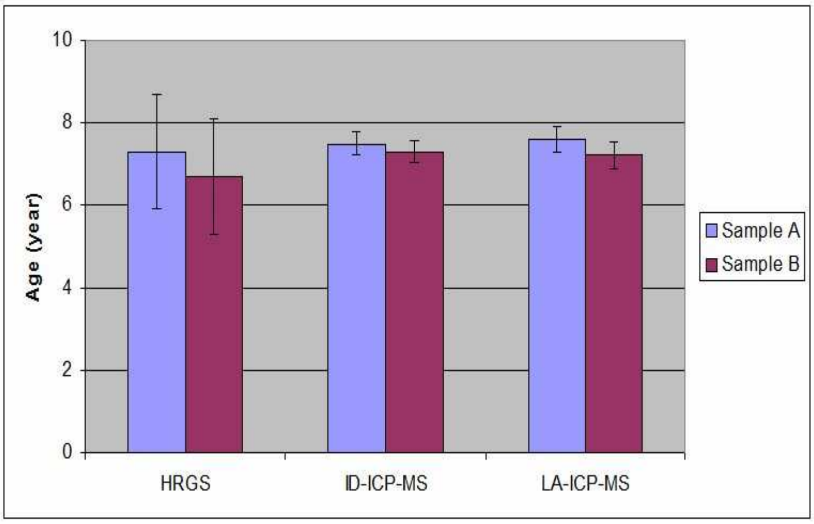

FIG. 14. Age dating:comparison of ICP–MS and HRGS results.

TABLE 10. COMPARISON OF AGE DATING RESULTS USING HIGH RESOLUTION GAMMA SPECTROSCOPY AND INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY

	Age (year) (*k*=2)		
	HRGS	ID–ICP–MS	LA–ICP–MS
RR 2010: Sample A	7.3 ± 1.4	7.59 ± 0.29	7.66 ± 0.34
RR 2010: Sample B	6.7 ± 1.4	7.35 ± 0.26	7.23 ± 0.32

ICP–MS is capable of dating NU older than 0.2 years; however, dating of even younger LEU and HEU materials is possible. LA–ICP–MS is not as sensitive as the methods involving dissolution and destructive analysis, but it may be the only applicable method in certain cases.

SCANNING ELECTRON MICROSCOPY / ENERGY DISPERSIVE SPECTROMETRY

Scanning electron microscopy (SEM) equipped with energy dispersive x–ray spectroscopy (EDS) is frequently used technique for surface study, elemental analysis and chemical characterization of samples. This technique is very useful for characterization of unknown materials (e.g. using element mapping of main components), but the sensitivity of the method does not allow for the investigation of trace elements. The lower detection limit of this x–ray technique is ~ 0.1% depending on the element measured: no isotopic information can be determined by SEM.

SEM/EDS method was also applied for the analysis of HEU samples. Surface and SEM/EDS spectra of the HEU samples were analysed.

X–RAY DIFFRACTION

X–ray diffraction (XRD) techniques are a family of NDA techniques which reveal information about the crystallographic structure, chemical composition, and physical properties of materials and thin films. These techniques are based on observing the diffracted intensity of an X–ray beam impinging a sample as a function of incident and scattered angle, polarization, and wavelength or energy. X–ray diffraction determines the geometry or structure of a molecule using X–rays.

The spectra of the samples indicated that the presence of α (alpha) uranium can be observed. Peaks of two uranium–oxide–hydrate ($UO_3 \cdot 2H_2O$) phases were observed in one of the samples; schoepite and metaschoepite.

CONCLUSIONS

Using the techniques mentioned here the following observations can be made:

High resolution gamma ray spectroscopy

High resolution gamma ray spectroscopy is an adequate tool for passive quantitative assay of isotopic composition of DU, NU, LEU and HEU samples of unknown origin. This technique can determine isotopics to an accuracy of a few percent with acquisition count times of 15–60 minutes. Accurate measurements can easily be made in the field.

A relative expanded uncertainty of $< 2\%$ was achieved for the measured value of the total U content. The matrix of the unknown sample can be characterized (if not a mixture) on the basis of the relative U–content. Two methods were developed that do not require reference material of known age: i) absolute: requires an efficiency–calibrated geometry and ii) relative efficiency calibration: applicable to an arbitrary measurement geometry and chemical form.

Results from HRGS analysis are in good agreement with mass spectrometric measurements. With an appropriate detector and by controlling the background, HRGS can be used for nondestructive age dating of LEU samples of arbitrary shape with age above ~10 years. The accuracy of age dating measurements by HRGS is close to that of age dating by of mass–spectrometry.

Low background gamma spectrometry was successfully used to show that a nuclear material was made from reprocessed uranium, by measuring the amount of ^{232}U in the sample. HRGS approximates the accuracy of mass spectrometry in determining major uranium isotopes (^{235}U, ^{238}U), to include age dating of HEU, and provides fast, non destructive analysis.

Inductively coupled plasma mass spectrometry

Destructive and non–destructive analytical methods as complementary techniques are applicable for characterization of unknown nuclear materials. ID–ICP–MS U isotopic composition results are in agreement with results from bulk and particle analysis by LA–ICP–MS. The production date can be also determined by direct LA–ICP–SFMS technique. It has the great advantage in not requiring the dissolution of the sample and only a small portion (approximately a few micrograms) is consumed for the analysis. This is essential for nuclear forensic applications where small amounts of radioactive materials constitute legal evidence in a court of law.

LA–ICP–MS is not as sensitive as the destructive method, but it may be the only applicable method in certain cases. Although the precision of LA-ICP–SFMS technique is inferior to that of the liquid sample introduction, the uncertainty of the measured production date is usually adequate for nuclear forensic purposes. Various ICP–MS techniques are applicable for precise isotopic measurements also in ultra–trace amount of analytes (e.g. Pu and minor U isotopes: ^{234}U and ^{236}U) can be analysed precisely using ICP–MS techniques. However, determination of ^{232}U isotope is only possible using HRGS. Although destructive analytical methods usually provide more sensitive analysis with lower detection limits, they cannot be used for the characterization of items which cannot be dismantled. A combination of different analytical techniques may increase confidence in the results and may help to further narrow down the set of possible origins and intended uses of the investigated materials. Measurements of trace elements in environmental materials are also possible using the ICP–MS instrument.

SEM/EDS and XRD techniques are applicable for the initial characterization of the samples; investigation of the surface and the structure and determination of the elemental composition.

PLANNED DEVELOPMENTS

The Centre for Energy Research will upgrade the low–background HRGS facilities for enhancing analytical sensitivity and accuracy. Research will focus on the correlation of ^{232}U – ^{236}U. The institute will also develop novel analytical methods for characterization of nuclear materials with unknown origin using solution based ICP–MS and LA–ICP–MS techniques.

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DETERMINATION OF THE ORIGIN OF UNKNOWN NUCLEAR MATERIAL THROUGH AN ISOTOPIC FINGERPRINTING METHOD

G. NIKOLAOU
School of Engineering
Democritos University of Thrace
Greece
Email: nicolaou@ee.duth.gr

INTRODUCTION

The determination of the provenance of nuclear material seized outside of regulatory control provides critical information to address and improve vulnerabilities in a State's nuclear security regime. The objective of this work is to demonstrate a procedure to differentiate different fuel-reactor combinations, and to identify the origin of spent nuclear fuel potentially encountered outside of regulatory control. The term "origin" refers to the type of the reactor in which the fuel was irradiated and its final burn up at the end of irradiation, as well as the type of the fuel introduced into the reactor.

PROCEDURE

The spent fuel composition is the consequence of, and reflects, the fresh composition of the fuel and its irradiation history, and carries information which may be uniquely related back to the provenance of the fuel. The identification procedure is based on the use of the isotopic composition in the spent fuel as measurable parameters (either the uranium and/or plutonium isotopic composition measured by mass spectrometry, and the identification of fission products). The isotopes considered are chosen on the basis that they can be easily measured by mass–spectrometry and high resolution gamma–spectrometry [1, 2, 3].

The procedure is based on the comparison of the isotopic compositions of uranium, plutonium or fission products, in the form of isotopic ratios, of an unknown material and known materials of well–defined origin. The isotopic ratios considered in the case of U and/or Pu are: ^{242}Pu/^{240}Pu, ^{238}Pu/^{xxx}Pu, ^{235}U/^{238}U, ^{240}Pu/^{239}Pu, ^{241}Pu/^{240}Pu, ^{239}Pu/^{235}U, ^{242}Pu/^{238}U, and for fission products: ^{134}Cs/^{137}Cs, ^{154}Eu/^{137}Cs, ^{154}Eu/^{134}Cs, ^{144}Ce/^{137}Cs, ^{106}Ru/^{144}Ce.

Isotopic ratios are then compared using the multivariate statistical technique of factor analysis [4]. Similar type fuels irradiated in the same type of reactor are plotted in 3 dimensions to visualize differences in isotopic composition. Such a graph may be used to compare unknown materials with a range of known reactor fuels, in order to interpret their origin. The origin of the unknown material is then identified in terms of the characteristics that are most similar in isotopic composition to the known material. Factor analysis is used for the comparison. The technique can use all information available in a single analysis reducing high dimensional data to three components which are then plotted in 3 dimensions. In this way, similarities between different materials may be identified by their isotopic composition. The method of comparison should be sensitive enough to avoid overlapping between the known materials, which may lead to erroneous interpretation of the origin. The known spent fuel/reactor compositions should ideally cover cases of all possible origins. Such information can be drawn from existing post irradiation examination (PIE) composition studies on spent fuels. In the absence of sufficient experimental data on U, Pu and fission product compositions in spent nuclear fuels, these were simulated for the reactor–commercial nuclear fuel combinations considered. The compositions were calculated using the zero–dimensional depletion computer code ORIGEN–2 [5].

Depletion calculations, required to simulate the composition of pressurized water nuclear reactor (PWR) and boiling water nuclear reactor (BWR) spent fuels from the SFCOMPO databank, were performed using the zero–dimensional isotope generation and depletion code ORIGEN–2. The nuclear data used in the depletion calculations were from the libraries ENDF/B-V [6]. The calculations were carried out with target burn up values given in Tables 1 and 2. Then the comparison between the experimental (E) and calculated (C) compositions could allow an assessment on the accuracy of the modeling. Considering that the chemical analysis was performed with mass spectroscopy techniques of the high accuracy (~0.1%) for the actinides, the experimental compositions could be used for the assessment of the modeling [7,8]. The accuracy for the measurement of the fission products of interest is ~2%.

MATERIALS CONSIDERED

The required known spent fuel/reactor compositions should ideally cover universe wide range of origins. Such information can be drawn from existing PIE composition studies on spent fuels. The measured compositions of the PWR and BWR nuclear fuels considered in the study are shown in Tables 1 and 2, respectively. Isotopic composition data at the end–of–irradiation (EOI) of these fuels are available in the SFCOMPO/OECD-NEA data bank.

The simulated compositions for spent nuclear fuels are shown in Table 3. The simulations performed were coupled with burn up–dependent cross–section libraries resembling, as closely as possible, the fuel type and neutron spectrum of irradiation.

TABLE 1. PWR UO_2 AND (UO_2–Gd)* FUELS CONSIDERED UNDER THE SFCOMPO DATA SET

REACTOR	FRESH FUEL (enrichment in ^{235}U)	BURN UP OF SAMPLES (GWd/tU)
Obrigheim	2.83%	24.22, 30.16, 35.64
Obrigheim	3.00%	25.81, 31.14, 36.26
Mihama-3	3.21%	21.2, 29.44, 32.3
Genkai-1	3.415%	38.1, 38.7
Calvert Cliffs-1	2.453%	31.4, 37, 46
Calvert Cliffs-1	2.72%	26, 33.17
Calvert Cliffs-1	3.038%	27, 37.12, 44,34
Takahama-3*	2.63%	28.2, 28.9
Takahama-3	4.11%	30.4, 35.42, 40.79, 47.03
Trino	3.897%	3.44, 7.61
H.B. Robinson	2.55%	24, 28

TABLE 2. BWR UO_2 AND (UO_2–Gd)* FUELS CONSIDERED UNDER THE SFCOMPO DATA SET

REACTOR	FRESH FUEL (enrichment in ^{235}U)	BURN UP OF SAMPLES (GWd/tU)
Monticello	1.45%	48, 55, 55.5, 57.3
Monticello	1.87%	44, 50, 53, 54.5
Monticello	2.14%	49, 51, 52.5, 54
Gundremmingen	2.53%	21, 23.5, 25
Monticello	2.87%	44, 44.4, 47, 48.2
Fukushima Daini-2	3.91%	25.6, 37, 42
Fukushima Daini-2*	3.41%	22, 32, 37
Monticello*	2.87%	39, 52

TABLE 3. SIMULATED REACTORS, FUELS AND TARGETED BURN UP VALUES

REACTOR	FRESH FUEL	BURN UP RANGE (GWd/tU)
PWR	2.5–5% ^{235}U	40–55
PWR	3.5% ^{235}U + Gd	40–55
	thermal MOX [95% U depleted, 5% Pu*]	35–50
	MTR high enriched (90% ^{235}U)	100–300
	MTR low enriched (25% ^{235}U)	50–100
BWR	UO_2, 2 – 3.5% ^{235}U	20–40
	2.75% ^{235}U + Gd	20–40
CANDU-N	natural U	4–10
CANDU-S	slightly enriched UO_2, 1–2% ^{235}U	7–30
LMFBR	MOX [74% U depleted, 25% Pu*]	60–100

*isotopic vector of plutonium 238:239:240:241:242 was 1.4:55:25.3:13.3:5.

RESULTS

The results were obtained using U and Pu isotopics as the characteristic signatures of spent fuels. However, similar conclusions are derived when only Pu or fission products are considered as the signatures.

Sensitivity analysis has been carried out on the PWR and BWR spent fuels from the SFCOMPO data bank given in Fig. 1 and Fig. 2, on the basis of their U and Pu compositions. The analysis has differentiated: (1) spent fuels of different ^{235}U enrichments from the same reactor; and (2) spent fuels of similar ^{235}U enrichments from different reactors.

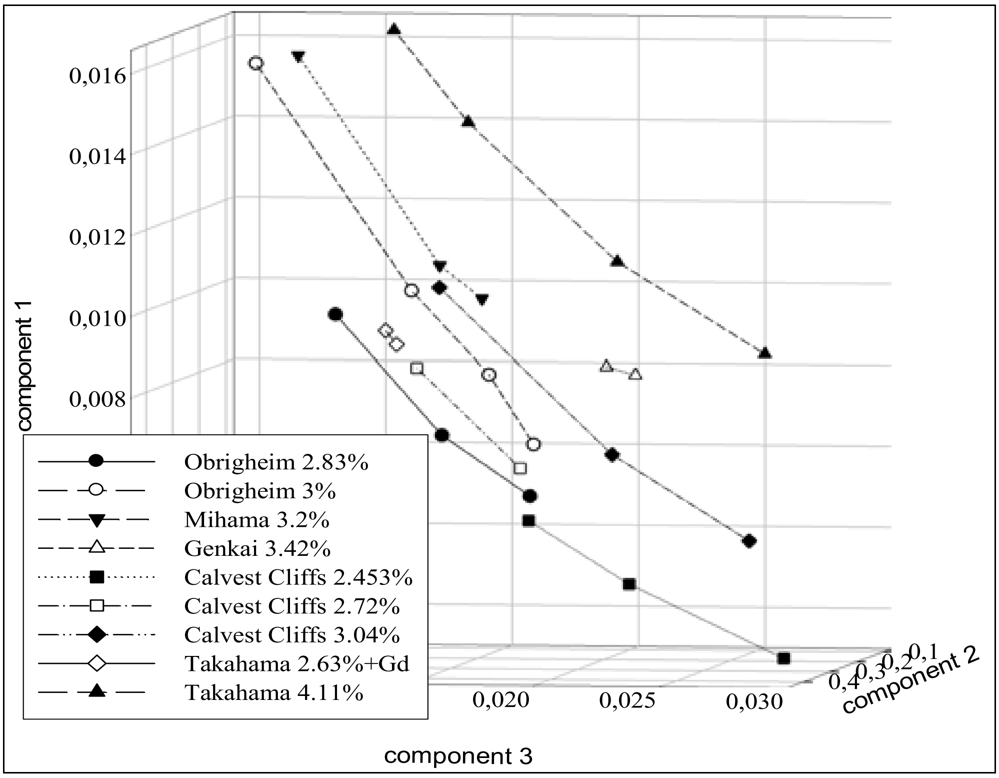

FIG. 1. Spent fuel PWR with different charge enrichments.

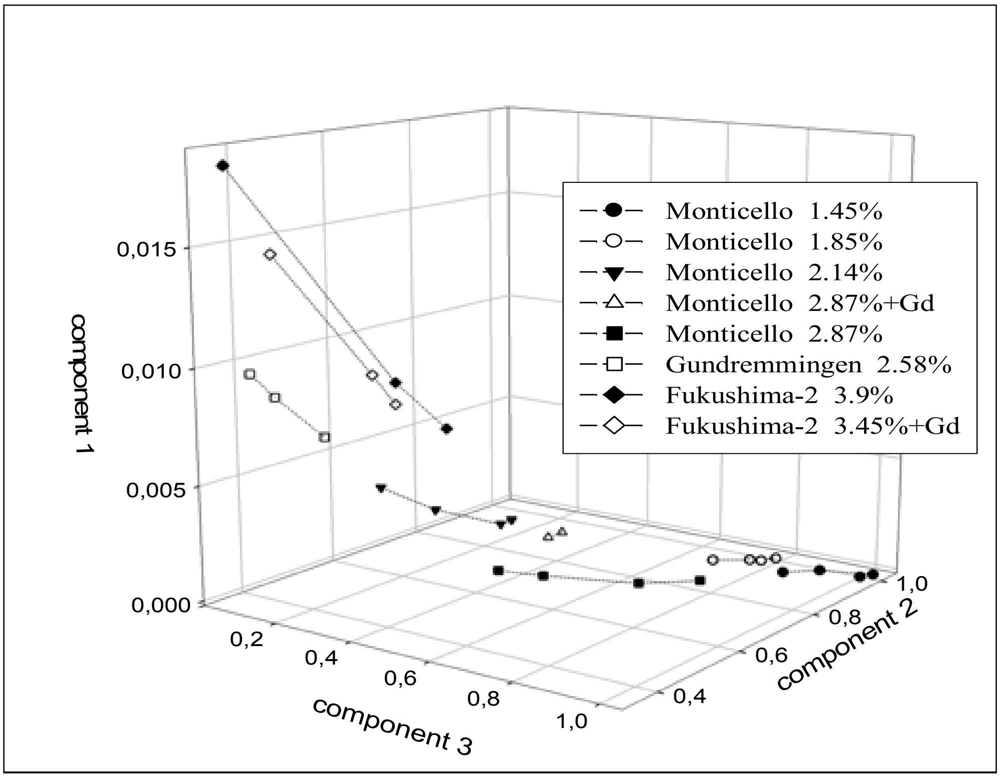

FIG 2. Spent fuel BWR with different charge enrichments.

In Fig. 3, the ten simulated fuels considered (Table 3), are clustered together on the basis of their similarities and differences with respect to the isotopic composition of each spent fuel, according to the charge fuel and the reactor type where the fuel was irradiated. There is clear differentiation between the fuels. In order to enhance the resolution of the graph, the MTR fuels are removed, revealing clearly the U and UGd fuels from the PWR and BWR reactors (Fig. 4).

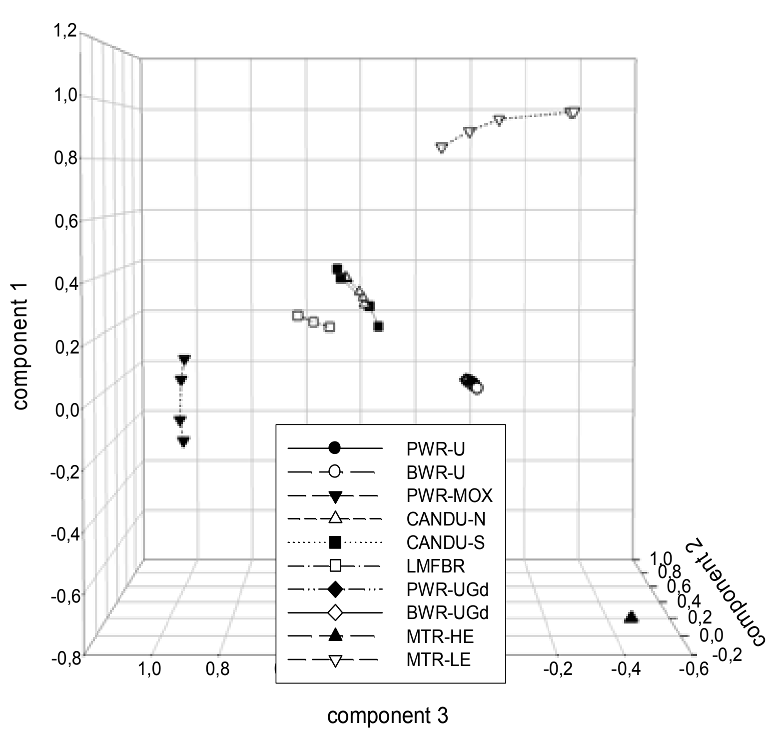

FIG. 3. The ten simulated nuclear fuels clustered on the basis of their isotopic composition.

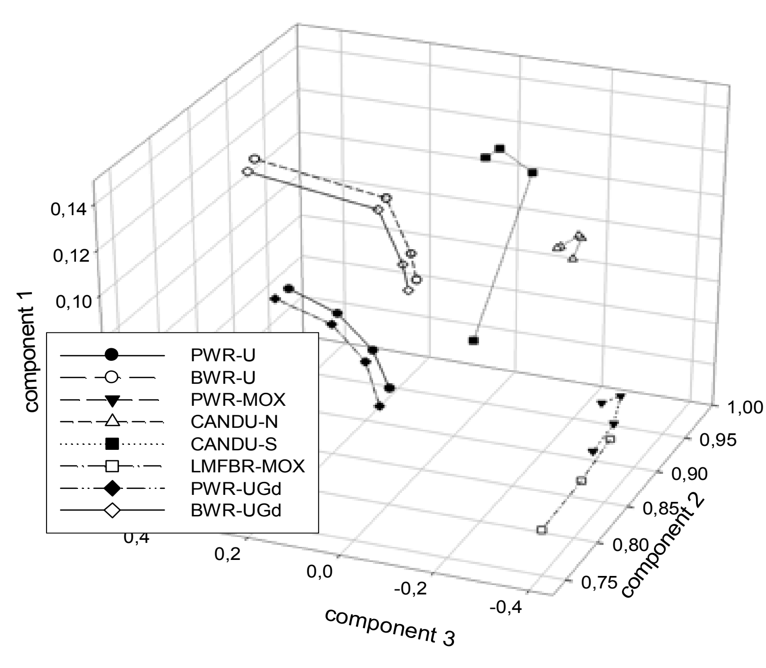

FIG 4. Enhanced resolution of Fig. 3 with removal of MTR fuels.

A sensitivity study has been carried out in order to investigate the potential of the procedure to differentiate: (1) different reactor types loaded with the same fuel type; and, (2) different enrichment of fuels within the same reactor. The study is concerned with PWR and BWR, reactors loaded with different enrichments of U fuels and UGd fuels, and the CANDU-S and –N reactors (Table 3).

On the basis of the U, Pu composition of the spent fuels, the PWR and BWR fuel cases in Table 3 are clustered in distinct families according to the reactor type where the fuels were irradiated: PWR–U and –UGd fuels, BWR–U and –UGd fuels, CANDU–N and CANDU–S (Fig. 5). The two latter cases are clearly resolved between them as well as from the light water nuclear reactor (LWR) families. Hence, different reactors with similar fuel types are well resolved. In order to improve the resolution of the graph, the PWR and BWR fuel cases are plotted separately in Figs. 6 and 7 respectively. Within each reactor type, the fuels considered are well resolved.

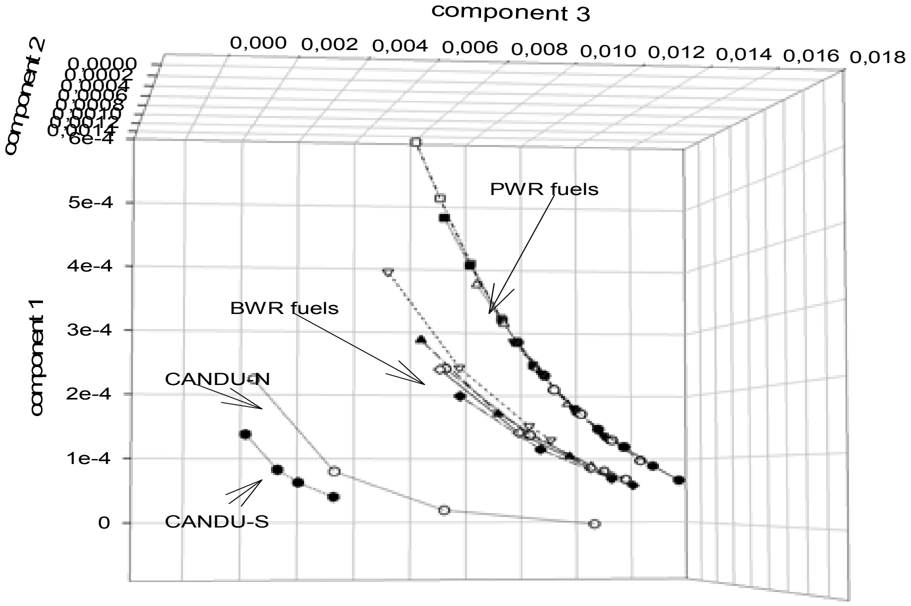

FIG. 5. Spent fuel CANDU, PWR and BWR cases with different charge enrichment.

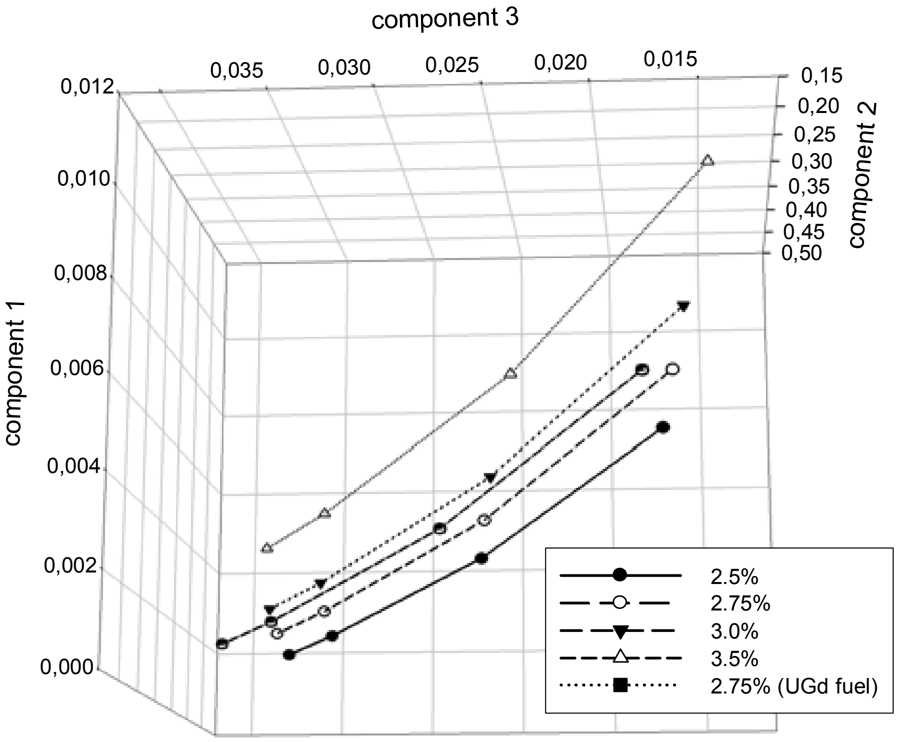

FIG. 6. Spent fuel PWR case with different charge enrichments.

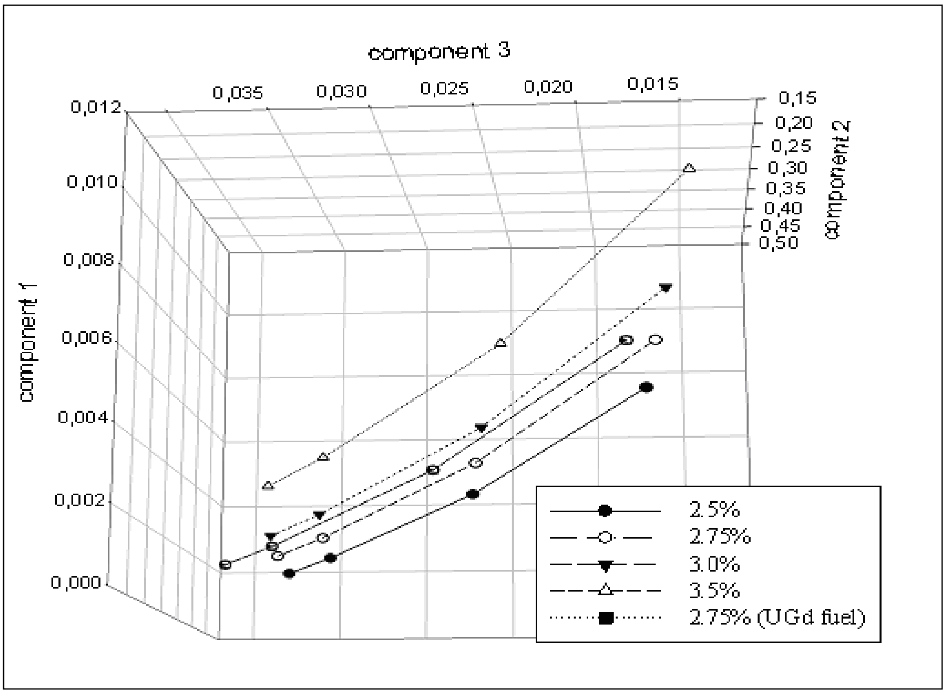

FIG. 7. Spent fuel BWR case with different charge enrichments.

Similar results are obtained when the identification procedure is based on the use of Pu isotopics or fission products as the characteristic signatures of the fuels. Fuels are clustered according to the reactor type and their charge composition.

The effect of the inclusion of ^{236}U at charge was investigated when simulating spent fuel compositions for the needs of the data bank. The purpose of these simulations was to assess if the inclusion of ^{236}U in the charge composition of the fresh fuels would affect the clustering of their corresponding spent fuels. Spent fuel compositions at EOI were simulated, using the code ORIGEN–2, for PWR and BWR uranium fuels with different charge compositions of ^{235}U (3%–5%) and ^{236}U (0 or 0.6%). The inclusion of ^{236}U at charge did not affect the clustering of the spent fuels in their corresponding distinct families and only shifted slightly the curves representing the fuels.

As expected, a great deal of variation in the calculated relative to experimental values is observed, ranging from one or very close to one (calculated / experimental), indicating a good model, to discrepancies of the calculated compositions from the experimental ones by more than 10%. This may be attributed to the nuclear data bank used. With respect to the former, studies have been carried out in order to correct discrepancies in neutron cross–sections within different nuclear data banks. The different level of accuracy observed between different reactor/fuel combinations may be the result of a different level in the accuracy of the neutron spectrum considered in the modelling and reflects how closely the modeling resembles the reactor/fuel combination considered.

CONCLUSIONS AND DISCUSSION

The method was sensitive enough to resolve fuels from different reactor types, including fuels from the same reactor but with different charge composition and, fuels of the same charge composition but from different reactors. The simulation of spent fuel composition when ^{236}U is included in fresh fuel may cause some overlap between different cases, particularly for very low burn up values.

The effect of cooling time on the sensitivity of the method to resolve spent fuels was tested by considering one of the simulated PWR spent fuels as the ‘unknown’. When using U and Pu as the characteristic signatures, the origin determination of the ‘unknown’ remained unaffected regardless of its cooling time; however, the burn up prediction is now within 20%. When using only Pu as the characteristic signatures, a discrepancy of one year on the cooling time yields an erroneous origin prediction. Furthermore, the prediction of the origin of the ‘unknown’ remained unaffected for an accuracy of less than ± 1% based on the measurement of the U and Pu composition which is easily attained with present day analytical techniques. In the case of radioactive fission products as the characteristic signatures, compositions at different cooling times have to be included in the procedure.

Outstanding issues associated with the isotopic compositions should be explored in order to assess the potential benefits and limitations of the procedure: the uncertainty of the ORIGEN simulations; the dependence of the composition on the sample axial or radial position within a fuel rod, the position of the rod within the core, and the position of the rod in the core relevant to any control rods which would affect the neutron spectrum; the accuracy of the nuclear data libraries used in the simulations; and the accuracy of the chemical analysis carried out to determine the composition of an ‘unknown’ material.

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ESTABLISHMENT OF PROCEDURES AND TECHNIQUES FOR NUCLEAR FORENSIC INVESTIGATIONS PART II – WORKSHOP ON NUCLEAR FORENSICS INSTITUTO DE PESQUISAS ENERGÉTICAS E NUCLEARES

J. DE SOUZA SARKIS, A. GODOY, D. ROSA, D. RODRIGUES, E. FRAJNDLIC, J. MARTINELLI, M. NASCIMENTO , M. TALHAVINE, M. COTRIN, N. LIMA, O. DE OLIVEIRA JUNIOR, O. NETO
Comissão Nacional De Energia Nuclear
Instituto de Pesquisas Energéticas e Nucleares
São Paulo, Brazil
Email: jesarkis@ipen.br

INTRODUCTION

The principal objective of this research contract was to implement a comprehensive program of nuclear forensic science in Brazil. The work plan had the following goals:

- Reviewing and upgrading procedures and methods for chemical, physical and isotopic characterization of nuclear materials;
- Demonstrating the reliability of the developed procedures by analyzing reference materials;
- Collecting and analyzing common nuclear material in Brazil; and
- Establishing a database for the nuclear forensic signatures of nuclear material in Brazil.

NETWORK OF LABORATORIES

The first achievement of the project was the creation of the Brazilian Network of Laboratories on Nuclear Forensic Science (BNLNFS) using the scientific expertise of Brazilian National Commission of Nuclear Energy (CNEN) nuclear scientists and existing infrastructure. The BNLNFS partners six laboratories located at Nuclear and Energy Research Institute, São Paulo, an additional laboratory in Poços de Caldas, Rio de Janeiro, and has the support of São Paulo State Police and Federal Police, Brasília. The associated laboratories implement techniques recommended by the IAEA to conduct an examination consistent with the nuclear forensics model action plan. In addition the BNLNFS have also the support of the Department of Radiological Protection, located at the Nuclear and Energy Research Institute, São Paulo.

Based on the IAEA recommendations governing the conduct of a nuclear forensic investigation (i.e., the nuclear forensics 'model action plan'), the procedures and techniques used in the nuclear forensic science were reviewed for their applicability. The action plan incorporates the main steps required during a nuclear security event including operational procedures on radiological crime scene management consistent with BNLNFS's operational plan and the sequence of laboratory analytical protocols.

The first case study was to establish the chemical fingerprint of thorianite, a radioactive mineral containing thorium, uranium, lead and rare earth elements (Fig. 1). During the last decades several tons of this mineral have been seized by Brazilian Federal Police in the north region of Amapa State, Brazil. The current work established nuclear forensic signatures of this mineral to assist in cases of international seizures of this mineral and confirm whether or not the mineral originates from Brazil.

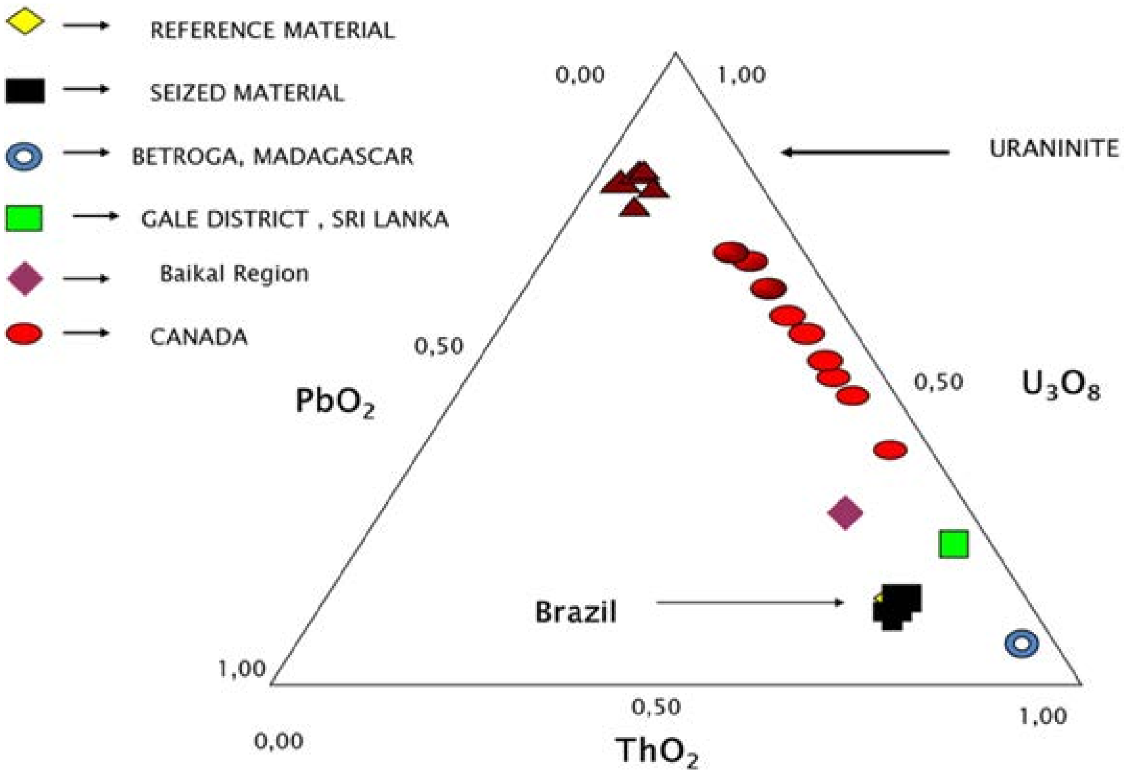

FIG 1: Nuclear forensic chemical fingerprint of thorianite.

The same strategy is now in progress involving other indigenous Brazilian radioactive minerals.

In order to initiate the establishment of a nuclear forensic signatures database, several naturally occurring nuclear materials were collected. Initially, uranium compounds and reference materials from different suppliers were analyzed. The initial results encompass the following data characteristics: visual appearance, density, dose rate, uranium content, particle size, x–ray diffraction patterns to determine mineral identification, and morphology. All of this data will be organized in an appropriate databank to facilitate nuclear forensic analysis and interpretation.

PARTICIPATION IN EXERCISES

The BNLNFS also participated in the Nuclear Forensics International Technical Working Group (ITWG) round robin #3 exercise than was conducted from from 2005 to 2010 and involved analysis of highly enriched uranium samples. Uranium isotopic analyses were performed using high resolution gamma spectrometry (HRGS) and metallic impurities determined by inductively coupled plasma atomic emission spectrometry technique (ICP–OES). All results were in good agreement with comparable analyses obtained by other participating laboratories.

A separate nuclear forensic exercise was planned and conducted involving all laboratories in the Brazilian network. The main objective was to verify the capability of the laboratories to categorize and characterize two samples according to the established action plan. The exercise assumed the existence of a national nuclear forensic library (NNFL) that allows States to make high confidence statements regarding the security of radioactive materials for which it has responsibility. Data —input to the library database — was obtained previously as part of the second objective of the contract.

The exercise simulated seizure of two suspected vials (Questioned Sample 1 and Questioned Sample 2) from a suitcase during a routine customs control in Guarulhos International Airport, São Paulo, Brazil. The exercise commenced with the identification of the presence of a nuclear material and the establishment of a chain of custody. Subsequently, the law enforcement officer at the scene initiated the action plan involving notification to the CNEN's nuclear security officials. Once the presence of nuclear material was confirmed, the site was secured, and conditions established for the safe and secure transport, the evidence was packed and sent for analysis following the network operational plan as well as the recommended analytical protocols of the IAEA and ITWG.

The data obtained was compared with the data reported in the database and used to tentatively identify the geographical origin. For this purpose, three different statistical techniques were employed utilizing the results analysis of rare earths elements: cluster analysis, principal component analysis and ternary graphs. The main conclusions of the investigation as reported to the authorities were:

- Questioned Sample 1 and Questioned Sample 2 are both U_3Si_2;
- The samples displayed a similar morphology;
- The samples displayed different particle size distributions;
- $^{235}U/^{238}U$ of Questioned Sample 1 was consistent with natural uranium;
- $^{235}U/^{238}U$ of Questioned Sample 2 was consistent with enriched uranium (19.95% ^{235}U);
- ^{236}U was not detected in either sample;
- The chemical signature based on rare earths elements is similar in both Questioned Sample 1 and Questioned Sample 2;
- The forensic evidence suggests the samples represent a similar origin but were produced during different periods;
- The chemistry of the seized samples, based on rare earth patterns, is not consistent with the characteristics represented by the samples reported in the national nuclear forensic library (see Fig. 2);
- The inconclusive results from comparisons with the national nuclear forensic library suggest further analyses are required to include additional elements;
- The geologic age dating of the questioned samples has yet to be determined.

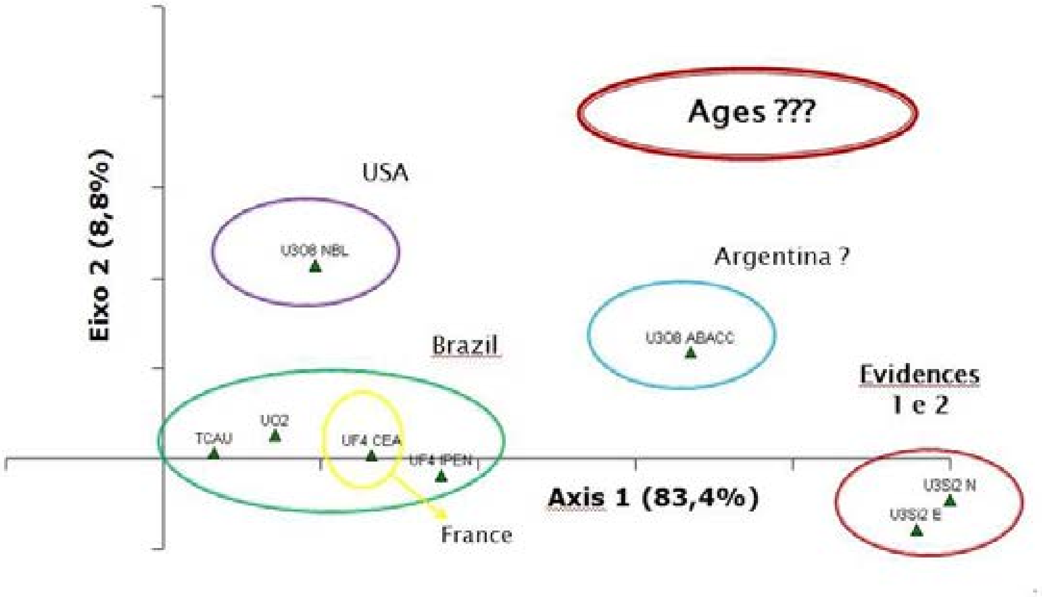

FIG 2. Principal component analysis (PCA) for rare earth elements measured in the Brazilian nuclear forensic exercise.

CONCLUSIONS

The results demonstrated that the laboratories belonging to the Brazilian network are capable of performing the full spectrum of analysis required in the conduct of a national investigation utilizing nuclear forensics. The establishment of an analytical plan facilitated the conduct of the exercise. However, lessons learned indicate improvements are necessary to the procedures for the distribution and reception of questioned samples, the standardization of documentation, and chain of custody associated with the nuclear forensic investigation. The determination of sample ages is paramount for the better interpretation of the results. The national nuclear forensic library is a very important tool for nuclear forensic interpretation. However, the library needs more comparative data to be reliable. Future exercises should seek to enhance the engagement of the law enforcement experts and better exploit classical forensic evidence.

LESSONS LEARNED AND FINAL COMMENTS

The principal objective of the contract was to establish a working group in Brazil dedicated to nuclear forensic science. The creation of the BNLNFS, the subsequent use of this infrastructure for analyzing several uranium compounds and reference materials, and the conduct of the first national nuclear forensic exercise is a direct result of this contract. The available infrastructure, as well as Instituto de Pesquisas Energéticas e Nucleares’s nuclear experts, demonstrated that the network was able to support a nuclear forensic investigation. A networked system is the most effective way to initiate a nuclear forensics program and avoids the high cost required to implement infrastructure dedicated to this activity. However, it is necessary to implement continuous training of all teams involved in the nuclear forensic analysis, establish a well–defined model for the nuclear forensic analytical plan, and standardize all the analytical documentation to include a chain of custody.

The experience of Brazil has emphasized the importance of IAEA training courses and workshops to promote nuclear forensic awareness and understanding, disseminate technical guidelines concerning analytical and interpretative methods, and promote the exchange of best practice among experts required to work together on real cases of nuclear or other radioactive material out of regulatory control.

Despite the progress in the program, further work remains to be done. The entire plan of action, as well as the analytical plan, needs to be documented in greater detail. Additional technical procedures have to be implemented while, for others, a better understanding of the analytical results is necessary. A comprehensive national nuclear forensic library must be developed for Brazil; as well, new chemical signatures of nuclear materials and minerals need to be studied prior to their inclusion in the library. In conclusion, the participation in this CRP research contract contributed to the improvement of the knowledge of the principles and procedures used in nuclear forensic science and, as a consequence, the improvement of the nuclear security infrastructure within Brazil.

INVITED PRESENTATIONS FROM EXPERTS AT THE THIRD RESEARCH COORDINATION MEETING, APRIL 2012

As part of the 3rd IAEA Research Coordination Meeting (RCM) in April 2012, experts from both the Russian Federation and the United States of America were invited to present their work related to nuclear forensics in the areas of improved techniques and procedures and the role of universities in advancing science that promotes nuclear forensic analysis and interpretation.

PRESENTATION FROM THE RUSSIAN FEDERATION

Dr. Vladimir Stebelkov of the Russian Federation presented results from work being performed in the Laboratory of Microparticle Analysis in Moscow applied to strengthening a States' nuclear security regime. The investigation of individual particles or micro–objects may be useful for cases where illicit trafficking of nuclear or radioactive material occurs but no bulk material is seized. In this case, traces of the radioactive material that can be detected on surfaces and may be useful in providing clues to the investigation. Another potential application involves the differentiation of products with similar gross characteristics that are manufactured in different facilities. In such cases, contamination present on questioned material or its packaging may indicate the origin of the material. The participants at the RCM noted that the use of α–autoradiography for detection of small amounts of alpha–emitting particles can be useful in nuclear forensic investigations.

PRESENTATION FROM THE UNITED STATES OF AMERICA

Professor Sue B. Clark of Washington State University USA addressed the role of the universities in the development of nuclear forensics. Basic research provides the foundation for innovation and discovery that may lead to the development of next generation analytical tools for nuclear forensics. In addition, university research programs in radiochemistry, radiation detection, and other disciplines that are traditionally exploited in nuclear forensics provide the opportunity to engage undergraduate and graduate students in science and engineering. These students represent the human capital 'pipeline' needed to develop the next generation of nuclear forensic scientists.

Professor Clark discussed recent developments in rapid radiochemical separations using capillary electrophoresis. This research, which is supported by the US Department of Homeland Security and US Defense Threat Reduction Agency, provides the foundation of basic chemistry associated with ligand–assisted capillary electrophoresis to expeditiously separate the lanthanide and actinide elements. From this foundational knowledge, microfluidic processes are under development that may be forward deployed, providing the possibility of rapid field triage of samples contaminated with the actinide and lanthanide elements. In addition, recent work on the development of electrochemical methods using mercury film and carbon paste electrodes for preconcentration of these elements holds promise for nuclear forensics. One objective of this work is to integrate the electrodes into the microfluidic devices to increase sensitivity. The substantive contributions of many students in radiochemistry and nuclear science ensured the success of this research programme.

CONCLUSIONS

THE COORDINATED RESEARCH PROJECT AS AN INTERNATIONAL TECHNICAL FORUM FOR NUCLEAR FORENSICS

The CRP provides a forum for sharing international technical experience in the field of nuclear forensics with a focus on improved procedures and techniques, optimization of nuclear forensic analysis, preservation of evidence, and provision of support to Member States. This project addresses both outstanding scientific needs to develop the discipline of nuclear forensics as well as to promote confidence in the application of analytical methods and understanding of the nuclear fuel cycle applied to nuclear forensics. The CRP also demonstrates that a common technical theme dedicated to promoting nuclear security capabilities for Member States can facilitate key international partnerships of nuclear forensics experts that can advance best practices in the discipline. The CRP focused on nuclear forensics applied in three areas: i) instrumentation and field work to include sample collection, ii) laboratory methods to include analytical procedures and techniques, and iii) predictive modeling and nuclear forensic interpretation.

SEQUENCING NUCLEAR FORENSICS ANALYSIS TO MEET DIVERSE REQUIREMENTS

The CRP covered a variety of measurement and interpretative tools relevant to nuclear forensics. These include non–destructive and destructive analytical methods, predictive tools, and information based technologies. The range of options reflects the requirements of using nuclear forensics to address the challenge of nuclear or other radioactive material out of regulatory control. States may encounter milligrams to kilograms of nuclear and other radioactive material out of regulatory control; these materials incorporate an array of isotopic, chemical, physical, and traditional forensic signatures that reflect their processing history and origin. Nuclear forensic methods must be poised to meet this broad range of requirements. In addition multiple nuclear forensic signatures are required to build confidence in conclusions. For this reason, analytical procedures must be identified and optimized relative to a State's existing measurement capabilities and organized for effective application.

Work conducted under the auspices of the CRP highlighted the need for nuclear forensic analysis to be staged to first return information to identify what the questioned material is, the immediate threat it poses to the public, responders or laboratory personnel, and to provide information necessary to guide the conduct of the ensuing comprehensive nuclear forensic analysis. The initial categorization stage utilizes non–destructive, in–situ analytical techniques that may include gamma ray spectroscopy. The ensuing nuclear forensic characterization involves destructive analyses where samples may be dissolved in strong acids, chemically separated using ion exchange resins, diluted in a carrier, and atoms analyzed by mass spectrometry. This sequenced arrangement also allows for conservation of small samples (i.e., < 5 grams). The development of the analytical plan should be chosen based upon the forensic examination plan and take into account the amount of sample available for analysis, prior information available on the sample, and the potential signatures (physical, chemical, elemental and isotopic) leading to precise interpretation. For this reason the forensic analytical plan must be developed and is specific to each investigation. The forensic examination plan also recognizes the need to provide information to investigators in a timely manner in the context of investigation. Suggested techniques using non destructive analysis will allow initial reporting 24 hours after receipt of samples, while subsequent reporting using destructive analysis by mass spectrometry or radiochemistry will occur between one week and two months after receipt of the sample by the laboratory [1].

THE ROLE OF PREDICTIVE NUCLEAR FORENSIC SIGNATURES

The results of the research program also underscored the need to employ both comparative and predictive nuclear forensic signatures. Questioned samples can be analyzed for isotopic, chemical, and physical signatures by nuclear forensic techniques and the data compared to like signature fields maintained in a national nuclear forensics library. It may also be the case that samples are not available for analysis. Using computation algorithms that simulate critical nuclear and chemical reactions characteristic of the nuclear fuel cycle, the relevant signatures of fuel cycle processes can be modeled. As noted by the work undertaken by the Democritos University of Thrace, the resulting predictive signatures allow data to be assembled where experimental data is otherwise unavailable (e.g., spent fuel, nuclear fuel reprocessing, or strategic facilities). As possible, predictive models need to be validated against primary measurement to ensure the validity of the simulations; this may include testing of model assumptions and/or performance.

NEED TO PRESERVE NUCLEAR FORENSIC EVIDENCE

The research further identified the important value of the preservation of evidence to nuclear forensics. Traditional forensic evidence includes, for example, hair, fibers, fingerprints, explosive residues, DNA, tool marks, and documents that can link people to places, materials and events. This is particularly important in supporting potential criminal prosecution of individuals or groups who would divert nuclear or other radioactive material out of regulatory control. The results reported by the Australian Nuclear Science and Technology Organisation indicates that while traditional forensic evidence can withstand exposure to elevated radiation fields, for it to be properly exploited when contaminated with radioactive material, either i) dedicated facilities with adequate controls need to be established that can receive and permit examination of evidence contaminated with radioactive material, or ii) the evidence needs to be decontaminated before it can be examined in a conventional forensic laboratory and is certified free from any residual radioactivity.

The Republic of Korea has identified the importance of evidence preservation as part of a nuclear forensics examination through the development of a national response plan. During meetings in South Korea with the IAEA in 2010, representatives of South Korean law enforcement, defense, emergency response, environmental monitoring, non–proliferation, nuclear safety, nuclear security gathered to address their ministry’s role and responsibilities in positioning nuclear forensics as part of a nuclear security infrastructure. Once the role and responsibilities were assigned, and assets and national capabilities fully identified, nuclear forensics is now applied to meet law enforcement and nuclear security requirements through a fully integrated program that best utilizes these distributed assets consistent with the IAEA’s Model Action Plan [1].

NUCLEAR FORENSICS SUPPORT TO A NUCLEAR SECURITY INFRASTRUCTURE

The objective of the research initiative is to provide technical assistance to Member States to elevate nuclear forensics as an effective tool in the detection and response to nuclear security events. The research program gathers scientists that can assist in this endeavor. Every State will have different requirements for nuclear forensics; it is essential to effectively articulate these requirements between the levels of national policy (e.g., law enforcement or the assessment of nuclear security vulnerabilities) and technical practitioners to ensure proper implementation. The science conducted by the researchers must reflect the needs of the national authorities consistent with the requirements for the nuclear forensics model action plan. In this regard, consideration must be given for the need to use technical guidelines, demonstrated competencies of personnel, and standards and reference materials to guide analysis. These needs must be balanced against the basic science necessary to identify priority nuclear forensic signatures at each stage of the nuclear fuel cycle, how these signatures become incorporated and persist in nuclear or other radioactive material and improved technical approaches to enable novel analysis and interpretation.

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RECOMMENDATIONS

In light of the outcomes of this CRP, it is recommended that the international nuclear forensics community continue to use this research platform to advance mutual interest in nuclear forensics. It is recognized that nuclear forensics is still an emerging science that can benefit from advances in measurement science as well as the considerable information collected by the related disciplines of geochemistry, radiochemistry, environmental monitoring, and nuclear engineering. Communication and sharing of information is critical, particularly for the development of best practices, to the broader nuclear forensics community. For this purpose, the investigators and experts were encouraged to disseminate information about the nuclear forensics CRP within their States and home institutions to encourage the development of new proposals and encourage young investigators. Specific recommendations from this CRP follow.

COMMON NOMENCLATURES AND TERMINOLOGIES FOR NUCLEAR FORENSICS

A key finding is the need to work on a common nomenclature of nuclear forensic terminologies. With the consistent application of common definitions, terminology and usage can be harmonized to ensure consistent understanding and application. This need is driven by requirements for clear communications between nuclear forensic practitioners, from different States, to include law enforcement officials, nuclear scientists, nuclear regulators and national level decision–makers. Consistent use of terminology in IAEA and related publications is essential.

THE ROLE OF PEER REVIEW IN NUCLEAR FORENSICS

The importance of basic science in nuclear forensics cannot be underestimated. Besides addressing fundamental questions concerning the need for reliable measurements of nuclear forensic signatures and their associated interpretation bearing on production processes across the nuclear fuel cycle, research also establishes the scientific validity of the methods applied to nuclear forensics. Through peer review of techniques and findings, for example, the use of nanometer scale secondary ion mass spectrometry to resolve variations in $^{18}O/^{16}O$ in UO_2 particles diagnostic of origin, or to exploit subtle variations in $^{235}U/^{238}U$ ratio in uranium ore concentrate using a magnetic sector inductively coupled plasma mass spectrometer indicative of different geological histories, can be subsequently applied to specific case studies involving materials returned from a nuclear security event.

ADVANCES IN THE DISCIPLINE

The research program further recommends that the results from its research program be incorporated within IAEA training curricula, as well as outreach and awareness in nuclear forensics. Important to the Agency's activities is conveying advancements in the discipline to improve nuclear forensic analysis and interpretation and aid in the development of best practice. This includes advancements undertaken by the IAEA Office of Nuclear Security as well as those of the IAEA Office of Safeguards Analytical Services pertinent to nuclear forensic analysis. Besides incorporating results from coordinated research activities, national research outputs and those from the Nuclear Forensics International Technical Working Group (ITWG) are essential to advancing the discipline. By enabling States to couple their own research and development base to nuclear forensics the national capability directly benefits. Regular dissemination of non–proprietary results through peer–reviewed publications, technical progress reports, presentations at international meetings and seminars, and through electronic internet portals is essential.

THE NEED FOR ADDITIONAL RESEARCH AND A NEW COORDINATED RESEARCH PROJECT

Due to the importance that the Office of Nuclear Security attaches to nuclear forensics as a key technical capability in a national nuclear security infrastructure, a viable research programme involving leading experts from the Member States is required. The CRP process furthers on–going and critical path IAEA nuclear forensic initiatives. For this reason a new CRP has been approved by the IAEA to provide data for the development of a national nuclear forensic library by the Member States. Nuclear forensics is a comparative discipline where a questioned sample is compared against data characteristics of known samples to ascertain whether there are similarities. A national nuclear forensic library enables States to make these comparisons quickly and with confidence. The new CRP (IAEA tracking number J02003) is entitled “Identification of High Confidence Nuclear Forensic Signatures for the Development of National Nuclear Forensic Library” and is focused on the identification of important isotopic, chemical and physical data characteristics (i.e., “signatures”) corresponding to stages of the nuclear fuel cycle (e.g., uranium ore mining, milling, conversion to UF_6, nuclear fuel manufacture, reactor operations, reprocessing, nuclear waste disposal, and radioactive source production) that facilitate comparative analysis of signatures between sample unknowns with known classes of the same for nuclear or other radioactive material using a national nuclear forensics library and associated material databases. The outputs will constitute guidance for States to include organization and measurements of key isotopic, chemical, and physical signatures to address nuclear fuel cycle process history. The IAEA anticipates involving leading nuclear forensic institutes from around the world in this next phase of research.

LIST OF PARTICIPANTS

Abedin-Zadeh, R.	International Atomic Energy Agency Wagramerstrasse 5 P.O. Box 100 A-1400 Vienna Austria e-mail:r.abedin.zadeh@iaea.org
Axelsson, A.	International Atomic Energy Agency Wagramerstrasse 5 P.O. Box 100 A-1400 Vienna Austria
Bartolomei, P.	Agenzia Nazionale per le Nuove Tecnologie l'energia e lo Sviluppo Economico Sostenibile (ENEA) Via Martiri de Monte Sole 4 40129 Bologna Italy e-mail: paolo.bartolomei@enea.it
Bashur, B.	International Atomic Energy Agency Wgramerstrasse 5 P.O. Box 100 A-1400 Vienna Austria e-mail:b.bashur@iaea.org
Baude, S.	Permanent Mission of France to the International Organizations in Vienna Schwarzenbergplatz 16 A-1010 Vienna Austria e-mail: stephane.baude@cea.fr
Clark, S.	Department of Chemistry Washington State University Pullman, Washington 99164-4630 USA e-mail: s_clark@wsu.edu
Colella, M.	Australian Nuclear Science and Technology Organization Locked Bag 2001 Kirrawee DC 2232 New South Wales Australia e-mail: mxc@ansto.gov.au
Colgan, P.	International Atomic Energy Agency Wagramerstrasse 5 P.O. Box 100 A-1400 Vienna Austria e-mail: p.colgan@iaea.org

Curry, M.
Office of Weapons of Mass Destruction Terrorism
US Department of State
2201 C. Street NW
Washington, DC 20520
USA
e-mail: currymr@state.gov

Davison, N.
The Royal Society
6-9 Carlton House Terrace
London
United Kingdom
e-mail: neil.davison@royalsociety.org

De Sousa Sarkis, J.
Comissão Nacional De Energia Nuclear
Instituto de Pesquisa Energéticas e Nucleares
Cidade Universitária, CEP
São Paulo
Brazil
e-mail: jesarkis@ipen.br

Dimayuga, F.
Atomic Energy of Canada Limited (AECL)
Chalk River Laboratories
Chalk River, Ontario K0J 1J0
Canada
e-mail: dimayugaf@aecl.ca

Doerfel, H.
IDEA System GmbH
Am Burgweg 4
Karlsruhe
Germany
e-mail: info@idea-system.com

Erastov, V.
State Atomic Energy Corp. (Rosatom)
ul. Bolshaya Ordinka 24/26
119017 Moscow
Russian Federation
e-mail: vverastov@rosatom.ru

Evans, T.
Australian Nuclear Science and Technology Organization
Locked Bag 2001
Kirrawee DC 2232
New South Wales
Australia
e-mail: tee@ansto.gov.au

Fischer, D.
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400
Vienna
Austria
e-mail: d.fischer@iaea.org

Garrett, B.
FBI Laboratory
2501 Investigation Parkway
Quantico, Virginia 22135
USA
e-mail: benjamin.garrett@ic.fbi.gov

Hinton, T.
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400
Vienna
Austria

Kim, S.C.
Korea Institute of Nuclear Safety
19 Gusung-Dong
PO Box 114
Daejeon 305-338
Republic of Korea
e-mail: k110ksc@kins.re.kr

Kristo, M.
Lawrence Livermore National Laboratory
7000 East Avenue
Livermore, California 94550
USA
e-mail: kristo2@llnl.gov

Lakosi, L.
Centre for Energy Research
1121 Budapest
Hungary
e-mail: lakosi.laszlo@energia.mta.hu

Larsson, C.
Defence R&D Canada – Ottawa
2701 Carling Avenue
Ottawa, Ontario
Canada K1A 0Z4
e-mail: carey.larsson@drdc-rddc.gc.ca

Mayer, K.
Joint Research Center
Institute for Transuranium Elements
European Commission
PO Box 2340
Karlsruhe
Germany
e-mail: klaus.mayer@ec.europa.eu

Nikolaou, G.
Democritos University of Thrace
School of Engineering
671 00 Xanthi, Prokat
Greece
e-mail: nicolaou@ee.duth.gr

Nogueira de Oliveira, C.
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400
Vienna
Austria
e-mail: c.nogueiradeoliveira@iaea.org

Ostropikov, V.
State Atomic Energy “ROSATOM”
International Cooperation
Ul. Bolshava Ordinka 24
119017 Moscow
Russian Federation
e-mail: VVOstropikov@rosatom.ru

Paillard, P.
CEA/DAM, Ile de France
B.P. 12 91680 Bruyères Le Châtel
France
e-mail: philippe.paillard@cea.fr

Peters, H.
FBI Laboratory
2501 Investigation Parkway
Quantico, Virginia 22135
USA

Ramebäck, H.
CBRN Defence and Security
Swedish Defence Research Agency
Cementvaegen 20
SE-901 82 Umea
Sweden
e-mail: henrik.ramebeck@foi.se

Samoilov, A.
Federal State Unitary Enterprise
A.A. Bochvar
5a Rogova Street
Moscow 123060
Russian Federation
e-mail: sam@bochvar.ru

Smith, D.K.
International Atomic Energy Agency
Wagramerstrasse 5
P.O. Box 100
A-1400
Vienna
Austria
e-mail:d.smith@iaea.org

Stebelkov, V.
Laboratory for Microparticle Analysis
Bolshaya Cheremushkinskaya st 25
117218 Moscow
Russian Federation
e-mail: stebelkov@mail.ru

Stefanka, Z.
Centre for Energy Research
1121 Budapest
Hungary
Email: stefanaka@iki.kfki.hu

Stoitcheva, G.
Overgas, Inc.
Philip Kutev Street 5
Sofia
Bulgaria
e-mail: stoitcheva@gmail.com

Szytel, L.
National Nuclear Security Administration
US Department of Energy
1000 Independence Avenue SW
Washington, DC 20585
USA
e-mail: lisa.szytel@nnsa.doe.gov

Wacker, J.
Pacific Northwest National Laboratory and
DOE IN-10
US Department of Energy
1000 Independence Avenue., SW
Washington, DC 20585
USA
e-mail: john.wacker@pnnl.gov

Wallenius, M.
Joint Research Center
Institute for Transuranium Elements
European Commission
PO Box 2340
Karlsruhe
Germany
e-mail: maria-s.wallenius@ec.europa.eu

Research Coordination Meetings

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LIST OF ABBREVIATIONS

AS	alpha spectrometry
AMS	accelerator mass spectrometry
AU	ammonium uranate
BNLNFS	Brazilian Network of Laboratories on Nuclear Forensic Science
BWR	boiling water nuclear reactor
BY40	basic yellow 40
C&C	command and control
CNEN	Comissão Nacional de Energia Nuclear (Brazil)
CRP	Coordinated Research Project
CRM	certified reference material
DA	destructive analysis
DFO	(1,8 – diazafluoren – 9 – one)
DNA	deoxyribonucleic acid
DU	depleted uranium
EC	European Commission
EDS	energy dispersive X-ray spectrometry
EOI	end-of-irradiation
FTIR	Fourier transform infrared spectroscopy
GDMS	glow discharge mass spectrometry
GICNT	Global Initiative to Combat Nuclear Terrorism
GIS	geographic information system
GPS	global positioning system
GUM	Guide to the expression of uncertainty in measurement
HEU	high enriched uranium
HPGe	high-purity germanium
HRGS	high resolution gamma spectrometry
ICP–MS	inductively coupled plasma mass spectrometry
ICP–OES	inductively coupled plasma optical emission spectrometry
IC	ion chromatography
ID	isotope dilution
IDA	isotope dilution analysis
ID–ICP–MS	isotope dilution inductively coupled plasma mass spectrometry
IDMS	isotope dilution mass spectrometry
IKI	Institute of Isotopes (Hungary)
ILC	identification, localisation and categorization
IND-Zn	indanedione-zinc
IR	infrared spectroscopy
ISO/BIPM	International Organization for Standardization/Bureau International des Poids et Mesures
ITDB	Incident and Trafficking Database
ITU	Institute for Transuranium Elements
ITWG	Nuclear Forensics International Technical Working Group
JRC/ITU	Joint Research Centre/Institute of Transuranium Elements
KED	K-edge densitometry
kGy	kilogray
KINS	Korea Institute of Nuclear Safety

LA–ICP–MS	laser ablation inductively coupled plasma mass spectrometry
LA–ICP–SFMS	inductively coupled plasma sector field mass spectrometry
LED	L-edge densitometry
LEU	low enriched uranium
LSC	liquid scintillation counting
LWR	light water nuclear reactor
MBq	megabecquerel
MC–ICP–MS	multi collector inductively coupled plasma mass spectrometry
MGA	Multi-Group Analysis
MSP	microspectrophotometry
MTR	Materials Test Reactor
NCC	neutron coincidence counting
NDA	non-destructive analysis
NIST	National Institute of Standards and Technology
NNFL	national nuclear forensics library
NU	natural uranium
PIE	post irradiation examination
ppm	parts per million
PWR	pressurized water nuclear reactor
RCM	Research Coordination Meeting
RDD	radiological dispersal device
REE	rare earth element
RR	round robin
R6G	Rhodamine 6G
RSD	relative standard deviation
RSF	relative sensitivity factor
SEM	scanning electron microscopy
SIMS	secondary ion mass spectrometry
SN–ICP–MS	solution nebulization inductively coupled plasma mass spectrometry
TEM	transmission electron microscopy
TIMS	thermal ionization mass spectrometry
U	uranium
UV	ultraviolet
UOC	uranium ore concentrate
WDS	wavelength dispersive X-ray spectrometry
XRD	X-ray diffraction
XRF	X-ray fluorescence

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