

Atomic Insights- Advancements In Nuclear Forensic

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Abstract

This review paper explores recent advancements in nuclear forensics, a multidisciplinary field that focuses on understanding nuclear security. It discusses quantitative morphological characterization techniques for carbide inclusions in uranium metal, the novel approach of using samarium isotope compositions in uranium ore concentrates, the determination of short-lived photo-fission product yields, isotope imaging, environmental monitoring, radio-analytical techniques, nuclear fuel irradiation test beds, image texture analysis, and micro-particle forensics. The paper emphasizes the importance of continuous advancements in nuclear forensics techniques and methodologies in enhancing global nuclear security efforts.

Keywords: Nuclear Forensic, Uranium, IAEA, multidisciplinary, nuclear security, radiological analysis, environmental monitoring.

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I. INTRODUCTION

As per (IAEA) the International Atomic Energy Agency, Nuclear Forensics is defined as “a discipline of forensic science involving the examination of nuclear and other radioactive material, or of other evidence that is contaminated with radionuclides, in the context of legal proceedings” (International Atomic Energy Agency, 2023). Nuclear Forensic is an emerging branch of forensic science which ascertains the history and the origin of nuclear and other radioactive elements by using different analytical techniques. Nuclear forensics is a multidisciplinary field that examines the chemical, isotopic, and physical properties of radioactive and nuclear materials, as well as related forensic evidence like tool marks, hair, fingerprints, DNA, and explosive residues. It does this by combining radiochemistry, nuclear physics, and other scientific disciplines.

In the 1990s, there were nuclear forensics emerged as a result of several high-profile incidents involving radioactive materials or other dangerous materials that escaped regulatory oversight.

II. INTERNATIONAL COOPERATION AND TREATIES

The **International Atomic Energy Agency (IAEA)** The International Atomic Energy Agency is the world's central intergovernmental forum for scientific and technical cooperation in the nuclear field. It works for the safe, secure, and peaceful uses of nuclear science and technology, contributing to international peace and security and the United Nations' Sustainable Development Goals. (International Atomic Energy Agency, 2023)

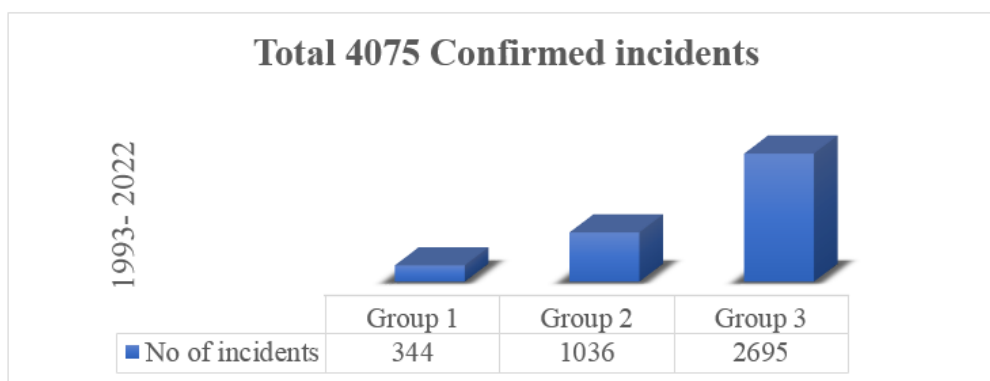
Incident and Trafficking Database (ITDB) is a part of IAEA information management systems which aids in the execution of the nuclear security plan of the IAEA. The authoritative data in the ITDB is voluntarily provided by the participating states by the officially nominated points of contact (PoC).

When the ITDB was first created, instances of the illegal trafficking of radioactive and nuclear materials were documented. Later, its purview was extended to encompass all instances in which radioactive materials, including nuclear material, are or were not under regulatory control.

III. DATA

In 2022, ITDB noticed a rise in reports with 146 new incidents logged by 31 states, which caused an increase by 26 incidents from previous years. These incidents encompassed a range of security concerns, including unauthorized activities, trafficking and potential misuse of radioactive and nuclear materials. ITDB categorized incidents into three groups:

- **Group 1-** includes incidents which are connected to malicious use or trafficking
- **Group 2-** includes the incidents of unclear intention.
- **Group 3-** includes the incident which are unrelated to any malicious use or trafficking. (*Incidents of Nuclear and Other Radioactive Material out of Regulatory Control*, n.d.; International Atomic Energy Agency, 2023)



Till 31st December 2022, 4075 confirmed incidents were registered since 1993.

- The majority of the reported incidents entails cases of theft or stolen materials of radioactive sources from industrial or medical applications. Potential thieves may find these sources appealing due to their perceived high resale or scrap metal value.
- It is reported that 52% of all thefts have happened during the transportation of these commodities since 1993 where 14% comprises nuclear material. Over the past decade, the figure has surged to nearly 62% underscoring the critical need to bolster transport security measures.
- 14% of all incidents entails nuclear material, while 59% comprises of radioactive material and 27% involves radioactively contaminated and other materials.
- In terms of reported thefts, 3.5% have been confirmed as linked to trafficking, with approximately 8.5% confirmed as unrelated to trafficking or malicious intent. The intention behind around 88% of thefts, whether related to malicious use or trafficking, remain undetermined. (*Incidents of Nuclear and Other Radioactive Material out of Regulatory Control*, n.d.)

The Global Initiative to Combat Nuclear Terrorism (GICNT) stands as a collaborative effort which comprises five international observer groups in addition to 88 states. Both the onlookers and the partners are united for a common goal of enhancing global capabilities against nuclear terrorism. Spearheaded by the United States and Russian Federation, this network operates with the shared leadership of nations like Finland, who coordinated efforts from 2017 to 2019. Within this framework, formal working groups, each chaired by a dedicated nation which focuses on the key aspects such as Argentina is in the forefront of response and mitigation, Canada is in command of forensics, and the UK is responsible for nuclear detection. Currently, GICNT is concentrated on planning meetings, seminars, workshops, and tabletop and field exercises that strengthen national capacities and promote international cooperation. (*GLOBAL INITIATIVE*, n.d.)

The Nuclear Forensics International Technical Working Group (ITWG) was founded by national authorities and bodies and consists of specialists from a range of disciplines, including science, law enforcement, and nuclear control. Their goal is to educate security experts as well as other interested parties about nuclear forensic analysis and its significance. They engage closely with global agencies such as INTERPOL and the IAEA. The ITWG's primary responsibilities are to determine the necessary resources for nuclear forensics, evaluate existing capacities, and recommend strategies for international cooperation in the fight against nuclear crimes. They also wish to guarantee that all nations are capable of securely handling nuclear materials and efficiently looking into crimes involving them. They stress the value of peer review in this discipline and accomplish these goals through conferences, activities, and publications. (Garrett et al., n.d.)

IV. RESEARCH AND DEVELOPMENT

Forensic characterization of Weapons- grade Plutonium by Destructive and Non-Destructive assay

The purpose of this work was to characterize signatures associated with nuclear forensics in weapons-grade Pu generated via thermal neutron irradiation of LEUO₂. The analysis used simulations of LEUO₂ neutron irradiation in a research reactor in addition to destructive and non-destructive techniques for signature characterization. The two primary components of the project were simulations and experiments, which helped validate and verify a MURR model using the code for simulating neutronics MCNP6.2. This work subjected LEUO₂ of milligram quantities to radiation in the MURR to simulate fuel irradiation from a PWR. The hope was that the LEUO₂ material would produce weapons-grade Pu. Light water is used for cooling and moderation in the annular core of the 10 MWt MURR reactor. Its eight symmetric parts, each with 24 fuel plates, make up its

core, which is powered by 93% enriched uranium-aluminide (UAlx) concentric plates. A water-filled flux trap located in the inner reactor section has a maximal $6 \times 10^{14} \text{ n cm}^{-2} \text{ s}^{-1}$ thermal neutron flux. Surrounded by an outer reflector made of graphite and a layer of beryllium reflector, outside the fuel zone, a neutron regulating blade and four boron neutron control blades are positioned radially. Two LEUO₂ samples were subjected to 602 hours of full power radiation in a research reactor as part of the study. It was discovered that the sample's burnup was rather near to the simulated amount. For several nuclides, the irradiated sample's gamma spectrometry closely matched the MCNP predictions, while for other nuclides, the discrepancies were more pronounced. Additionally, good agreement was found for certain nuclides but low agreement for others using mass spectrometry. The study showed that the time since uranium irradiation may be estimated using gamma spectrometry. The MCNP model of the research reactor was validated by experimental measurements, and the corresponding uncertainties and deviations were calculated. The findings from this study can be helpful in enhancing the analysis in the field of nuclear forensic. (Martinson et al., 2023)

Nobel metal radionuclides' impact on the environment and the subsequent utilization for Nuclear Forensics

Noble metal radionuclides were measured using mass spectrometry or radiometric methods after they were extracted, enriched, and isolated. The radionuclides were removed and isolated using different industrial and academic analytical methods, such as auto deposition and other selective resins. Later, the appropriate research equipment was used to separate the radionuclides which were exposed to determine the half-life, the radiation sensitivity for each type, and any interferences. The detection of radioactive silver isotopes (108 and 110 mg) in Japanese seafood samples contaminated by the Fukushima Daiichi NPP accident was another topic covered in the dissertation. For radio silver in the environment, even at high contamination levels as those experienced following a nuclear accident, the author established a separation process. Important details on reactor conditions, particularly those pertaining to control rods in pressurized water reactors (PWRs), may be revealed by this determination. The research also examined how radio cesium and radio silver are absorbed by and distributed among the elements in Shiitake mushrooms, as well as what happened to them in soil samples following a release scenario. The author created a previously unheard-of analytical technique for ICP-QQQ-MS measurements of long-lived ¹⁰⁷Pd. Using this method, ¹⁰⁷Pd with an activity level below mBq·kg⁻¹ was found in a cooling pond sediment sample from the Chernobyl nuclear power plant. (Weller, 2022)

To find out the Rare Earth Element in uranium-bearing material by ICP-MS for nuclear forensics.

The main goal of the study was to identify rare earth elements (REE) using ICP-MS in uranium-bearing material for nuclear forensics. ICP-MS analysis was performed with chromatographic separation of lanthanides employing selective extraction on TRUTM resin. Four UOC and two samples of uranium ore, which were collected from the Witwatersrand basin in South Africa, were analyzed. The overall REE (HREE) concentrations of the samples varied according to the study; UOC4 had the highest concentration. In comparison to UOC1, UOC2, and UOC3, UOC4 had the opposite pattern of LREE enrichment and HREE depletion when compared to the REE/Cl chondrites pattern. In U ore1 and U ore2, Ce anomalies were positive while Eu anomalies were negative. The difference in the mining sites was confirmed by a follow-up ANOVA of the REEs. These methods can accurately determine the concentrations of REEs in well-characterized UOC and uranium ore samples and the REE pattern may be used for source assignment and identification. The results might be applicable in forensic nuclear investigations as well as in fighting against illicit trafficking of nuclear materials. (Varga et al., 2010)

Comparative stage two examinations of nuclear materials on a micrometric scale: morphological, elemental, and chemical

Advantages of in-SEM The article talks about using Raman spectroscopy along with Scanning Electron Microscopy/Energy Dispersive Spectroscopy (SEM/EDS) for the examination of micrometric particles, particularly in the field of nuclear forensics. Combining all three analyses—morphology, elements, and chemistry on the same microscopic areas of particles is made possible by a specially designed linking device. In nuclear forensics, correlation analysis is crucial for differentiating between chemical phases that correspond to different morphologies or elemental compositions. The suggested procedures for conducting this research are outlined by the authors. These include determining which kind of support material is best for particle deposition, adjusting the power of the laser, and adding fiducial marks to help align images taken with different modalities. They emphasize reducing electron beam exposure before Raman analysis in order to preserve sample integrity. The method's effectiveness is demonstrated by examples drawn from international nuclear forensic exercises. The method separated plutonium dioxide from similar particles such as uranium dioxide and cerium dioxide and also detected more uranium compounds in one case (CMX-6). In a sample mostly made up of uranyl nitrate, a distinct exercise (CMX-7) identified a secondary phase of uranyl oxo hydroxide by its distinct shape and elemental

signature. Nuclear materials' length, storage conditions, and main and minor constituents, such as contaminants and aging byproducts like oxidation or hydration, can all be determined using this technique. The paper suggests that this analytical methodology has wider applicability to assess elemental homogeneity and demonstrate a link between morphological or phase changes and chemical heterogeneity. Characterizing microparticles in mixed oxide particles for reference materials is one instance of such an application. More study is urged in order to expand its uses and enhance understanding of material properties under different conditions. (Pointurier et al., 2024)

Utilizing Multi-modal Synchrotron Methods for Nuclear Forensic Applications

Nuclear materials can be analyzed non-destructively using synchrotron technologies, which can reveal intricate details about the chemical and structural properties of the materials without causing harm to the samples. This work focuses on the examination of particle materials in nuclear forensics utilizing synchrotron technology. Initially, surrogate particles composed of nickel oxide and molybdenum oxide were used in the study to evaluate) and X-ray Fluorescence (XRF) and synchrotron X-ray Diffraction (XRD) procedures. The researchers used specifically designed radioactive sample techniques to examine uranium standards including actinides after their initial experiments were effective. The samples were tested in 3D printed holders made to securely store powder samples with high concentrations. The goal of the project is to design a high-throughput, multi-modal analytical system to solve the difficulties associated with assessing particulate materials containing actinides. Detailed structural and chemical characterization at different length scales will be possible with this system. Among other things, it entails developing sophisticated sample containers for synchrotron facilities that handle hazardous materials like uranium and plutonium. The capacity to analyze powders in several modes at high throughput is essential. It enables the quick assessment of potentially questionable materials, environmental samples, or contamination traces on surfaces forms like soil and debris that are most likely to be found in the field. Rapid analysis capabilities like this cut down on sample collecting, transportation, preparation, and analysis time expenses considerably. The capacity to swiftly and precisely characterize particulate samples with trace impurities will also be improved by a deeper understanding of the particulate structure and chemical composition, which are influenced by variables such as impurity content and processing conditions (such as the uranium particulate's synthesis route, calcination temperature, and collection conditions). This development is essential to bolstering the arsenal of instruments available for quick forensic examination of nuclear materials. (Gill et al., n.d.)

Quantitative and Morphological Analysis of Uranium Carbide Inclusions

When uranium metal is being produced, interactions between the air and the equipment cause little structures known as uranium carbides (UCs) to develop. The aim of the study was determination of the structures evolved when the metal cools down. So, the researcher had melted uranium in a special machine and then cooled it at different speed. Next, the cooled metal was visualized using a powerful microscope. Very slow cooling, they found out, can also disrupt the geometry of UCs making them bigger with slower cooling. This implied that during the production of nuclear fuel one can manipulate the sizes as well as orientations of these structures. Additionally, such structures could provide insights into how metals are formed thereby giving a basis for more experiments. Knowing this would help in developing efficient fuels for new reactors and it may also help to identify where research materials made from metals come from. (Athon et al., 2022)

Samarium isotopes from uranium concentrates

This study might use better nuclear forensics capabilities to trace back confiscated uranium ore concentrate (UOC) and stop it from illegally going toward weapons. The team then took this technology and used it to measure the Sm isotope compositions of 32 UOCs obtained from various uranium mines around the world, after developing a new method for removing samarium (Sm) from U-rich samples. Around half of the UOCs had unique isotope compositions between 149 and 150 Sm, indicating that 149 Sm had snagged neutrons in the original ore body. Another interesting finding was that UOCs with strange Sm isotope compositions tended to be found in older, higher-grade ore bodies among other things such as neutron moderators possibly affecting this outcome. This study also represents a signature accomplishment in demonstrating the potential of Sm isotopes as an exceptional tool for nuclear forensics; it can help identify where detected UOCs came from while shedding light on geological conditions near the ore body itself. (Shollenberger et al., 2021)

Use of Bremsstrahlung 2 X-ray for experimental measurement of uranium and thorium Cumulative fission product yields

Cumulative fission product yields (CFPYs) for uranium and thorium isotopes have been measured experimentally using bremsstrahlung X-rays at 8, 14, and 20 MeV. The range of fission product half-lives covered by these measurements is 1.07 to 40.8 seconds. With a primary focus on nuclear models, the goal of this research is to fill the gap in the current photo fission product yield databases by providing more thorough data. Data on

short-lived fission products were gathered by researchers using a high-purity germanium detector and a pneumatic transfer system in the interim between accelerator irradiation and measurement cycles. The need for new isotope synthesis techniques in nuclear forensics and the improvement of nuclear data to aid in non-proliferation activities are what spurred this endeavor. (Foley & Yang, 2021)

Evaluation of isotope imaging and uranium inhomogeneity for nuclear forensics

In nuclear safety and forensics, laser ablation multi-collector inductively coupled plasma mass spectrometry (LA-MC-ICP-MS) is a potent instrument used to evaluate the isotopic homogeneity of solid uranium materials at the micron level. While the ^{235}U isotope abundances (around 1%) were similar, LA-MC-ICP-MS analysis of two UO_2 pellets from the Nuclear Forensics International Technical Working Group (ITWG) 5th Collaborative Materials Exercise (CMX-5) showed notable variations in isotopic heterogeneity. These pellets showed detectable differences at the micron level, despite coming from the same source materials but with different production paths. A new functionality enables software to detect the presence of a lyrical message and even analyze impact on human's mood. It is possible to change perception by amplifying/decreasing the intensity of certain frequencies and tones. This info concerning the constituent's setups, production routes and isotopic diversities care about nuclear forensic investigations which enable us to trace back and identify the cause of un-traced radioactive substances. (Varga et al., 2020).

Characterization of soil samples for uranium

The study focused on nuclear forensic examination of Uranium 6 soils obtained from the area of Botswana that is known as Serule gold mine. The parameters scrutinized here comprised: thorium 232 concentration activity levels, XRF mineralogy (X-ray fluorescence technique), ratios of Thorium 232 and Uranium 238 isotopes, Uranium 235 232 ratios radioactivity, impurities content rare earth elements amount coupled with ICP-MS (Inductively coupled plasma mass spectroscopy) techniques for measuring these factors. We found out that the concentration of isotopes and radioactivity within the uranium ore differed from one mine to another, the isotopic ratios in it were ^{238}U and ^{235}U as well as $^{232}\text{Th}/^{238}\text{U}$. As a matter of fact, one can say that there is significant variability between the concentration reactors that come out of this particular kind of ore depending upon where they have been mined from. Furthermore, deviations from the normal REE/chondrite ratios were observed during this study on the rocks from Serule area. These abnormal behaviors could be taken as indication of gold ore at Serule deposit with the distinctive characteristics. (Ntsohi et al., 2021)

Determination of "Age" by Radio Analytical Technique

The researchers have done an inquiring into the solid microscopic composition of Uranium nerve pellets used in a material balance exercise using the isotope ratio measurement technique. The capsules for which isotopic heterogeneity has been realized were made from similar feed materials through various processes. The two capsules that had different isotopic contents, however, had similar amounts of one of the uranium isotopes (^{235}U) during the scrutiny of the isotopic ratios within the various sites of the capsules, distinct constituent elements in each capsule and their respective isotopic proportions were identified by the scientists hence showing their internal formations differed. This data is crucial for nuclear studies because it can be used to trace unknown nuclear materials back to their origin as well as understand how they were synthesized. (Varga et al., 2020)

Nuclear Fuel Irradiation

For the purpose of addressing the ongoing need to measure and identify nuclear material signatures in the field of nuclear security, a distinctive test facility for nuclear fuel irradiation was developed. This novel test bed enables rapid diagnosis and evaluation of nuclear fuel samples aimed at several security uses in a cheap and effective manner. The experiment replica can contain 6 small samples of 'Micro Fuel' in hermetically-sealed container that may be bombarded for about 25 days in cycles in the High-Intensity Isotope Reactor (HIIR). The irradiation of up to nine targets can take place concurrently under alternate conditions like temperature and burnup levels. The main goal of post-irradiation tests is to make a track performance indicator and carry out detailed examination of the irradiated Fuel samples are simpler isotopic tracers for UO_2 have undergone this fast-track testing programme. (Wilson et al., 2023)

Colorimetry and Image Texture Analysis for Uranium ore

Uranium ore concentrates (UOCs) are significant in the context of nuclear security because they are traded in huge amounts, which raises the possibility of their usage "out of regulatory control." Understanding the origin and production process of a nuclear material is necessary once an illegal trade incident is detected. This means using analytical techniques capable of measuring the nuclear material's characteristic parameters, such as its physical, chemical, and isotopic properties, which are known as signatures in the field of nuclear forensics. The current work examines the possibility of using spectrophotometric colour determination along with picture texture

analysis, or the angle measure technique, to assess the origin of various UOCs. The classification of over 80 distinct samples into a few groups of UOC powders is made possible by the application of several complex statistical approaches, which makes this approach a prospective addition to the currently accepted nuclear forensics procedures. (Marchetti et al.)

Actinide detection and imaging of ^{242}mAm and ^{238}Pu in hot particles.

This creative technique examines micrometer-sized pollution particles using two potent instruments: resonant laser ionization and secondary ion mass spectrometry. Because those tiny elements could offer important details about their beginnings and possible impacts on people and the surroundings, they hold significant importance for scientists in different fields such as environmental studies, biology, astronomy, and criminal investigation. The advantage of this method is that it does not need changing the particle in any manner to give us complete information about the materials and isotopes contained in each unique particle. It is akin to closely analyzing every part without causing any damage to it. Furthermore, unlike other approaches, it can discriminate between various elements even when their masses are identical. Using a microscopic particle from the Chernobyl nuclear error, researchers tested this procedure and found tiny levels of a particular radioactive element called ^{242}mAm . This demonstrates the method's sensibility and accuracy, especially for uncommon elements. The most interesting aspect is that practically every element may be studied using this technique, giving scientists a wealth of new opportunities to examine particles in ways they were previously unable to. In general, it represents a significant advancement in our knowledge of the composition and effects of pollution particles in our environment. (Bosco et al.)

V. CONCLUSION

The papers being presented show a great diversity of methods and applications in the field of nuclear forensics. They cover a wide range of subjects, including the quantitative morphological characterization of carbide inclusions in uranium metal, the measurement of uranium inhomogeneity and isotope imaging for forensic applications, and the distinct nuclear forensic signature offered by samarium isotope compositions in uranium ore concentrates. They also look into the detection of short-lived photo fission product yields for nuclear forensic isotope production, the use of radio-analytical techniques to determine the age of nuclear materials, and the classification of uranium ore concentrate powders using colorimetry and image texture analysis. The topic also covers the identification of uranium in soil samples taken from prospective mining sites, the development of the emerging subject of microparticle forensics, which focuses on actinide imaging and the identification of specific isotopes in heated particles, and a test bed for nuclear fuel irradiation for security purposes. When taken as a whole, these papers significantly advance nuclear forensics by offering insights into analytical techniques, signature identification, and material characterization that are critical for enhancing nuclear security and non-proliferation efforts.

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