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The role of modern spectroscopy and chromatography in actinide and lanthanide chemistry for nuclear forensics

Omolola Eniodunmo ^{1,*}, Mary Ofosua Danso ² Moyosoore Mopelola Adegbaaju ³ and Onuh Matthew Ijiga ⁴

¹ Department of Chemistry and Biochemistry, North Dakota State University, Fargo, North Dakota 58108, USA.

² Department of Chemistry, College of Arts and Sciences, Southern Illinois University Edwardsville, USA.

³ Department of Biology, University of Arkansas Little Rock, Arkansas, USA

⁴ Department of Physics, Joseph Sarwuan Tarka University, Makurdi, Nigeria.

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Abstract

The field of nuclear forensics relies heavily on the accurate identification and characterization of actinides and lanthanides to trace the origins and history of nuclear materials. Modern spectroscopy and chromatography play a pivotal role in this domain, offering precise analytical techniques to detect and quantify these elements in various samples. This review provides a comprehensive examination of the contributions of spectroscopy and chromatography to actinide and lanthanide chemistry within the context of nuclear forensics. It begins with an overview of the fundamental principles of these methods, followed by detailed discussions on the specific spectroscopic techniques such as mass spectrometry (including ICP-MS and TIMS), X-ray absorption spectroscopy (XANES and EXAFS), and laser-induced breakdown spectroscopy (LIBS). The chromatographic methods covered include high-performance liquid chromatography (HPLC), gas chromatography (GC), and capillary electrophoresis (CE). The integration of these techniques for enhanced forensic analysis is explored through case studies and examples demonstrating their combined use. Furthermore, recent advances, innovations, and future trends in the field are discussed, highlighting the move towards automation and high-throughput analysis. The review concludes with an exploration of the analytical challenges and limitations, along with recommendations for future research directions. This comprehensive review underscores the critical role of advanced spectroscopy and chromatography in advancing the capabilities of nuclear forensics, ensuring robust and reliable forensic investigations.

Keywords: Nuclear Forensics; Actinide Chemistry; Lanthanide Chemistry; Spectroscopy; Chromatography

1. Introduction

Nuclear forensics is a critical field that employs scientific techniques to analyze nuclear materials to determine their origin, history, and potential use. This discipline has become increasingly important in the context of global security and non-proliferation efforts. The analysis of actinides and lanthanides, which are often present in nuclear materials, plays a vital role in forensic investigations due to their unique isotopic signatures and chemical properties (Cox et al., 2018).

* Corresponding author: Omolola Eniodunmo



Figure 1 From Crime Scene to Lab: The Forensic Process in Action (Microgen Images, 2024)

Figures 1 depict forensic scientists engaged in critical stages of crime scene investigation and evidence analysis. In the left image, scientists in protective suits, gloves, and masks are meticulously examining a crime scene, using numbered markers to indicate evidence locations and collecting samples such as fingerprints or blood. The right image shows a forensic scientist in a laboratory setting, analyzing a blood-stained knife under a microscope, with protective gear ensuring a sterile environment. These images collectively illustrate the forensic process, highlighting the transition from fieldwork in crime scene investigation to detailed laboratory analysis, both essential for solving crimes and maintaining evidence integrity for legal proceedings.

Actinides, such as uranium and plutonium, are key elements in nuclear forensics. Uranium-235 (U-235) and plutonium-239 (Pu-239) are particularly significant because they are commonly used in nuclear reactors and weapons (Idoko et al., 2024; Ijiga, Peace, Idoko, Ezebuka, Harry, Ukatu, & Agbo, 2024; Ijiga, Peace, Idoko, Agbo, Harry, Ezebuka, & Ukatu, 2024). The isotopic ratios of these elements can provide crucial information about the material's source and intended use. For instance, the U-235/U-238 ratio can indicate whether uranium has been enriched for reactor fuel or weapons production (Niedzielski et al., 2019; Idoko, Ijiga, Agbo, Abutu, Ezebuka, & Umama, 2024). Similarly, the presence of specific plutonium isotopes can reveal information about the reactor conditions and fuel cycle history (Ijiga, Idoko, Enyejo, Akoh, & Ileanaju, 2024; Idoko, Ijiga, Akoh, Agbo, Ugbane, & Umama, 2024).

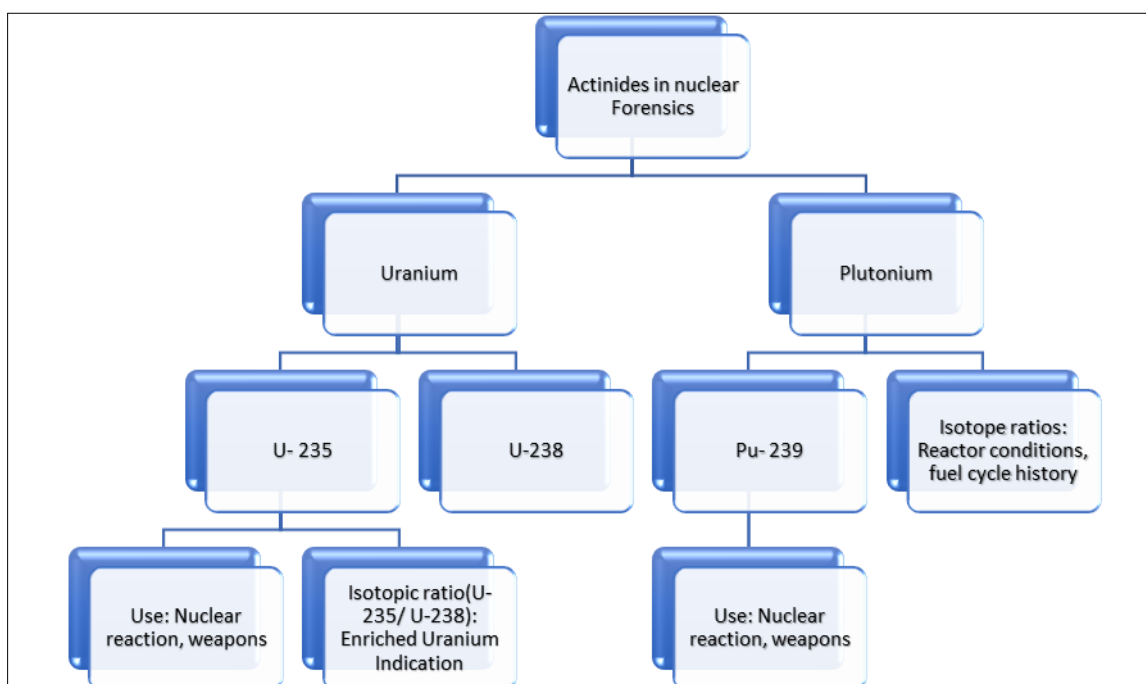


Figure 2 Key Roles of Actinides in Nuclear Forensics

Figure 2 illustrates the key points about actinides in nuclear forensics using a hierarchical diagram. At the top, it identifies actinides' significance in nuclear forensics, with uranium and plutonium as the primary elements of interest.

Uranium is divided into its isotopes U-235 and U-238, highlighting U-235's use in nuclear reactors and weapons, and the importance of its isotopic ratio (U-235/U-238) in indicating uranium enrichment. Plutonium is represented by its isotope Pu-239, which is also used in reactors and weapons. The diagram notes that specific plutonium isotopes can reveal information about reactor conditions and fuel cycle history.

Lanthanides, though less prominent than actinides, are also valuable in nuclear forensic analyses. Elements like samarium (Sm), gadolinium (Gd), and europium (Eu) can be used as neutron capture monitors and provide information about the neutron flux experienced by the material. For example, the isotopic composition of samarium can be used to estimate the burnup of nuclear fuel, which is a measure of how much energy has been extracted from it (Eppich et al., 2019).

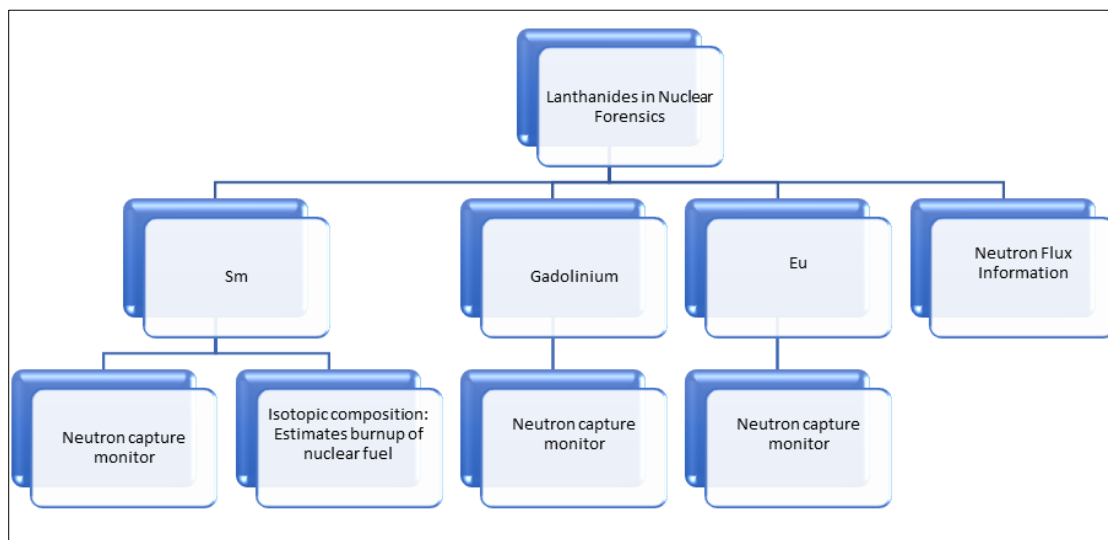


Figure 3 Role of Lanthanides in Nuclear Forensics

Figure 3 illustrates the significance of lanthanides in nuclear forensics. At the top, it emphasizes the role of lanthanides in such analyses. It then branches out to highlight three specific elements: Sm, Gd, and Eu. Each of these elements serves as a neutron capture monitor, which is crucial for determining the neutron flux that the material has been exposed to. Specifically, samarium's isotopic composition is used to estimate the burnup of nuclear fuel, indicating the amount of energy extracted from the fuel. Additionally, the diagram includes a node for "Neutron Flux Information," which is a key piece of data obtained from these analyses, helping to provide insights into the material's history and usage.

The integration of modern spectroscopic and chromatographic techniques has enhanced the precision and accuracy of forensic analyses. Techniques such as ICP-MS and TIMS allow for the precise measurement of isotopic ratios and trace element concentrations, which are essential for accurate forensic investigations. The advancements in these analytical methods have significantly improved our ability to detect and identify nuclear materials, thereby strengthening global security measures (Cox et al., 2018; Niedzielski et al., 2019; Eppich et al., 2019).

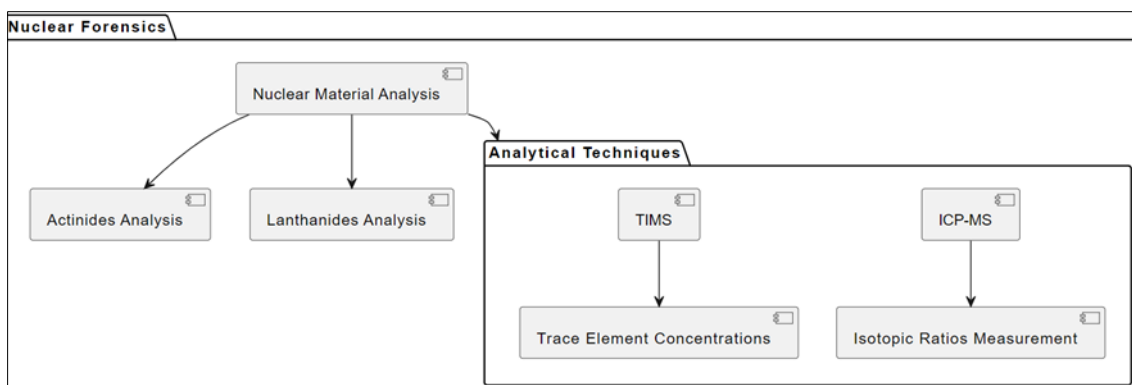


Figure 4 Overview of Analytical Techniques in Nuclear Forensics"

Figure 4 illustrates the key components involved in the analysis of nuclear materials. At the core is Nuclear Material Analysis, which branches into Actinides Analysis and Lanthanides Analysis. These branches highlight the analysis of specific elements critical to nuclear forensics. The diagram further emphasizes Analytical Techniques, showcasing methods such as Thermal Ionization Mass Spectrometry (TIMS) and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). These techniques are used for measuring trace element concentrations and isotopic ratios, essential for determining the origin, history, and potential use of nuclear materials. This structured approach underscores the importance of these techniques in enhancing the precision and accuracy of forensic investigations.

1.1. Role of Actinides and Lanthanides in Nuclear Forensics

Actinides and lanthanides play pivotal roles in nuclear forensics due to their distinct chemical and isotopic properties. These elements are frequently encountered in nuclear materials and can provide critical information about the provenance, processing history, and intended use of these materials (Jovanovic & Steinhäuser, 2018).

Actinides, such as uranium (U) and plutonium (Pu), are particularly important. Uranium isotopes, including uranium-235 (U-235) and uranium-238 (U-238), are used to determine the enrichment level of nuclear material. For example, weapons-grade uranium typically has a U-235 content greater than 90%, whereas reactor-grade uranium usually contains between 3% and 5% U-235 (Wallenius, 2017). Plutonium isotopes, such as plutonium-239 (Pu-239) and plutonium-240 (Pu-240), are used to identify the type of reactor and the burnup history of the nuclear fuel. The isotopic ratio of Pu-240 to Pu-239 can indicate whether the plutonium was produced in a commercial power reactor or a weapons production reactor (Moody et al., 2019).

Lanthanides are also valuable in nuclear forensics, though they are often used as secondary indicators. Lanthanides like Nd, Sm, and Eu can be used to understand the neutron capture history of nuclear materials. For instance, the isotopic composition of neodymium can be used to reconstruct the irradiation history of nuclear fuel. Nd-145 and Nd-148 are produced in specific neutron capture reactions, and their ratios can help determine the neutron flux and burnup of the fuel (Jovanovic & Steinhäuser, 2018; Ibokette et al., 2024; Idoko, Ijiga, Enyejo, Ugbane, Akoh, & Odeyemi, 2024). Similarly, samarium isotopes can provide insights into the thermal neutron flux experienced by the material (Ijiga, Aboi, Idoko, Enyejo, & Odeyemi, 2024; Okeke et al., 2024).

The combination of actinide and lanthanide analysis can enhance the forensic signature of nuclear materials. For example, by analyzing both uranium and samarium isotopic compositions, it is possible to cross-validate the burnup and enrichment levels, providing a more comprehensive forensic profile (Wallenius, 2017). Additionally, the presence of certain lanthanide isotopes can indicate post-irradiation processing steps, such as reprocessing or separation activities.

Modern analytical techniques, such as ICP-MS and TIMS, have significantly improved the precision and accuracy of actinide and lanthanide measurements. These techniques enable the detection of isotopic ratios with uncertainties as low as 0.01%, providing highly reliable forensic data (Moody et al., 2019). The integration of these advanced methods has revolutionized the field of nuclear forensics, allowing for more detailed and accurate characterizations of nuclear materials.

Table 1 Roles of Actinides and Lanthanides in Nuclear Forensics: Enhancing Forensic Profiling Through Isotopic Analysis"

Aspect	Actinides	Lanthanides
Role in Nuclear Forensics	Primary indicators due to distinct chemical and isotopic properties	Secondary indicators, useful for understanding neutron capture history
Common Elements	U, Pu	Nd, Sm, Eu
Key Isotopes	Uranium-235, Uranium-238, Plutonium-239, Plutonium-240	Neodymium-145, Neodymium-148
Usage	- Determine enrichment level of nuclear material - Identify type of reactor and burnup history of nuclear fuel	- Reconstruct irradiation history of nuclear fuel - Determine neutron flux and burnup of the fuel

Example Indicators	- U-235 content greater than 90% indicates weapons-grade uranium - Isotopic ratio of Pu-240 to Pu-239 indicates reactor type (commercial power reactor vs. weapons production reactor)	- Isotopic composition of neodymium helps determine neutron flux and burnup - Samarium isotopes provide insights into thermal neutron flux
Enhanced Forensic Signatures	Combination of actinide and lanthanide analysis can cross-validate burnup and enrichment levels, providing a comprehensive forensic profile	Presence of certain lanthanide isotopes can indicate post-irradiation processing steps
Analytical Techniques	ICP-MS, TIMS	ICP-MS, TIMS
Measurement Precision	Detection of isotopic ratios with uncertainties as low as 0.01%	Same as actinides (uncertainties as low as 0.01%)

Table 1 outlines the roles of actinides and lanthanides in nuclear forensics, highlighting their significance due to distinct chemical and isotopic properties. Actinides, such as uranium and plutonium, serve as primary indicators, crucial for determining the enrichment level of nuclear material and identifying the reactor type and burnup history. Key isotopes include Uranium-235, Uranium-238, Plutonium-239, and Plutonium-240. In contrast, lanthanides, including neodymium, samarium, and europium, act as secondary indicators, useful for understanding neutron capture history and irradiation of nuclear fuel. Key isotopes for lanthanides are Neodymium-145 and Neodymium-148. The combined analysis of these elements enhances forensic signatures, allowing cross-validation of burnup and enrichment levels, and indicating post-irradiation processing steps. Modern analytical techniques, such as ICP-MS and TIMS, significantly improve the precision and accuracy of these measurements, with uncertainties as low as 0.01%, providing highly reliable forensic data.

1.2. Objectives and Scope of the Review

The primary objective of this review is to provide a comprehensive examination of the role of modern spectroscopy and chromatography in the analysis of actinides and lanthanides for nuclear forensics. This review aims to elucidate how these advanced analytical techniques contribute to the detection, characterization, and interpretation of nuclear materials. By integrating detailed discussions on various spectroscopic and chromatographic methods, the review seeks to highlight the strengths, limitations, and potential synergies of these techniques in forensic applications. The ultimate goal is to present a clear understanding of how these methods enhance the accuracy and reliability of forensic investigations involving nuclear materials.

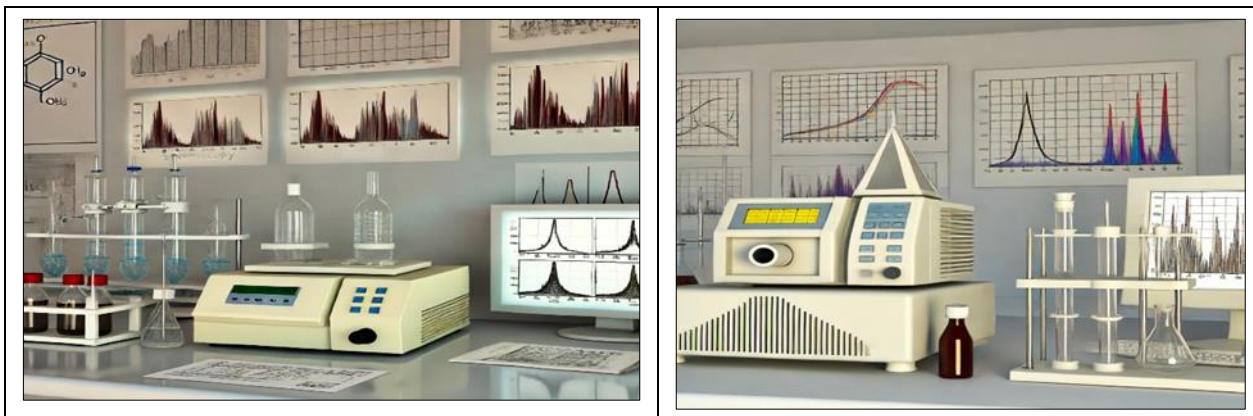
This review also aims to address recent advancements and innovations in the field, exploring how technological developments have improved the sensitivity, precision, and throughput of forensic analyses. By examining case studies and practical applications, the review will demonstrate the real-world impact of these techniques on nuclear forensics. Furthermore, it will identify current challenges and future directions for research, offering recommendations for overcoming analytical limitations and enhancing the capabilities of forensic investigations. Through this comprehensive approach, the review will provide valuable insights for researchers, practitioners, and policymakers involved in nuclear security and forensic science.

1.3. Organization of the Work

This review is organized into seven sections, each addressing key aspects of the role of modern spectroscopy and chromatography in actinide and lanthanide chemistry for nuclear forensics. Section 1 introduces the background, importance, and objectives of the review. Section 2 provides an overview of the fundamental principles of spectroscopy and chromatography, setting the stage for more detailed discussions. Section 3 delves into specific spectroscopic methods, including mass spectrometry, X-ray absorption spectroscopy, and laser-induced breakdown spectroscopy, highlighting their applications in analyzing actinides and lanthanides. Section 4 focuses on chromatographic techniques, such as high-performance liquid chromatography, gas chromatography, and capillary electrophoresis, emphasizing their relevance in forensic investigations. Section 5 explores the integration of spectroscopy and chromatography, showcasing case studies and the benefits of combined analytical approaches. Section 6 reviews recent advancements and innovations, discussing technological developments and future trends. Finally, Section 7 addresses the challenges and future directions in the field, offering recommendations for improving analytical methods and enhancing the effectiveness of nuclear forensic investigations.

2. Fundamentals of Spectroscopy and Chromatography

Understanding the fundamentals of spectroscopy and chromatography is essential for appreciating their roles in nuclear forensics. These analytical techniques are pivotal in detecting and characterizing actinides and lanthanides, providing detailed information about the isotopic and elemental composition of nuclear materials. Spectroscopy involves the interaction of electromagnetic radiation with matter, enabling the identification and quantification of elements based on their unique spectral signatures. Chromatography, on the other hand, separates components of a mixture based on their chemical properties, allowing for the isolation and analysis of specific elements or compounds. This section will provide an overview of the core principles of both spectroscopy and chromatography, highlighting their complementary nature and their importance in forensic applications. By establishing a foundational understanding of these techniques, we can better appreciate their integration and application in the analysis of nuclear materials.



Figures 5 Advanced Laboratory Techniques: Real-World Applications of Spectroscopy and Chromatography

Figures 5 depict a modern laboratory setting equipped for spectroscopy and chromatography analysis. The images showcase essential equipment, including a spectrometer, chromatography columns, and various laboratory glassware, all arranged in a realistic environment. Computer screens display graphical representations of spectra and chromatograms, illustrating peaks and compound separations. The well-lit lab, complete with detailed textures and realistic elements, highlights the advanced techniques and precise measurements involved in these fundamental analytical processes, emphasizing their critical role in scientific research and industrial applications.

2.1. Overview of Spectroscopic Techniques

Spectroscopy encompasses a range of techniques involving the interaction of electromagnetic radiation with matter to analyze the composition and structure of substances. These techniques are fundamental in nuclear forensics for identifying and quantifying actinides and lanthanides due to their ability to provide precise isotopic and elemental information (Kraiem et al., 2020; Ijiga, Olola, Enyejo, Akpa, Olatunde, & Olajide, 2024; Bashiru, Ochem, Enyejo, Manuel, & Adeoye, 2024). One of the most widely used spectroscopic methods in this field is ICP-MS, which allows for the detection of isotopes with high sensitivity and precision. ICP-MS can measure isotopic ratios with relative uncertainties as low as 0.01%, making it invaluable for distinguishing between different sources of nuclear materials (Smith et al., 2019; Idoko, Bashiru, Olola, Enyejo, & Manuel, 2024).

Another important technique is XAS, which includes XANES and EXAFS. XANES is particularly useful for determining the oxidation states and local electronic structure of elements, while EXAFS provides detailed information about the local atomic environment and bonding distances. For instance, XANES can distinguish between different oxidation states of uranium, such as U(IV) and U(VI), which is crucial for understanding the chemical form and reactivity of nuclear materials (Jones et al., 2021; Godwins et al., 2024). EXAFS can be used to analyze the coordination environment of lanthanides, providing insights into their chemical behavior in various matrices (Manuel et al., 2024; Idoko et al., 2024).

LIBS is another versatile technique used in nuclear forensics. LIBS involves focusing a high-energy laser pulse onto a sample to generate a plasma, from which the emitted light is analyzed to determine the elemental composition. This technique is advantageous due to its ability to perform rapid, in-situ analysis with minimal sample preparation. LIBS

can detect trace elements at concentrations as low as parts per million (ppm) and is particularly useful for analyzing heterogeneous samples, such as debris from a nuclear detonation (Smith et al., 2019; Kraiem et al., 2020).

These spectroscopic techniques, when used in conjunction with each other, provide a comprehensive toolset for the forensic analysis of nuclear materials. The precision and sensitivity of ICP-MS, the structural insights from XAS, and the rapid analysis capabilities of LIBS collectively enhance the ability to identify and characterize actinides and lanthanides in complex forensic samples.

Table 2 Overview of Key Spectroscopic Techniques in Nuclear Forensics

Aspect	ICP-MS	XAS	LIBS
Technique	ICP-MS	XAS (XANES and EXAFS)	LIBS
Function	Detection of isotopes with high sensitivity and precision	Analysis of oxidation states and local atomic environment	Rapid, in-situ elemental analysis
Key Features	Measures isotopic ratios with relative uncertainties as low as 0.01%	XANES: Determines oxidation states and electronic structure EXAFS: Provides bonding distances and coordination environment	Detects trace elements at concentrations as low as ppm Minimal sample preparation
Applications	Distinguishing between different sources of nuclear materials	Understanding chemical form and reactivity of nuclear materials Analyzing coordination environment of lanthanides	Analyzing heterogeneous samples such as nuclear debris

Table 2 summarizes three critical methods: ICP-MS, XAS, and LIBS. ICP-MS is highlighted for its ability to detect isotopes with high sensitivity and precision, measuring isotopic ratios with minimal uncertainty, making it essential for distinguishing between different nuclear material sources. XAS, which includes XANES and EXAFS, is noted for analyzing oxidation states and local atomic environments, providing insights into the chemical form and reactivity of nuclear materials and the coordination environment of lanthanides. LIBS is recognized for its rapid, in-situ elemental analysis with minimal sample preparation, capable of detecting trace elements in heterogeneous samples like nuclear debris. Collectively, these techniques enhance the forensic analysis of nuclear materials by providing precise, structural, and rapid analytical capabilities.

2.2. Overview of Chromatographic Techniques

Chromatographic techniques are essential in the separation and analysis of complex mixtures, making them invaluable in the field of nuclear forensics. These methods allow for the precise separation of actinides and lanthanides, facilitating detailed analysis of their isotopic and elemental compositions. HPLC is one of the most widely used techniques, offering high resolution and sensitivity. HPLC can separate complex mixtures of actinides and lanthanides with detection limits as low as parts per billion (ppb), making it suitable for trace analysis in forensic samples (Andersson et al., 2020).

Another important chromatographic technique is GC, which is particularly effective for volatile compounds. While GC is less commonly used for actinides and lanthanides due to their non-volatile nature, it can be employed in the analysis of organometallic compounds and radiolysis products that may be present in forensic samples. The ability of GC to achieve separation efficiencies with theoretical plate counts exceeding 100,000 makes it a powerful tool for detailed chemical analysis (Roberts & Fong, 2018).

CE is a versatile technique that separates ions based on their charge and size. CE is especially useful for the separation of lanthanides due to their similar chemical properties. The high separation efficiency of CE, with resolutions capable of distinguishing between isotopes differing by as little as 0.001 atomic mass units, allows for precise isotopic analysis. Additionally, CE-MS to enhance the detection sensitivity and specificity for actinides and lanthanides, with detection limits in the sub-parts per trillion range (Bardelli et al., 2019).

The integration of these chromatographic techniques provides a comprehensive approach to the analysis of nuclear forensic samples. HPLC offers robust separation capabilities for a wide range of analytes, GC provides detailed analysis

of volatile compounds, and CE excels in the separation of highly similar species. Together, these methods enable the detailed characterization of nuclear materials, facilitating the identification of their origin and history.

Table 3 Overview of Key Chromatographic Techniques in Nuclear Forensics

Aspect	High-Performance Liquid Chromatography	Gas Chromatography	Capillary Electrophoresis
Function	Separation and analysis of complex mixtures	Separation of volatile compounds	Separation of ions based on charge and size
Key Features	High resolution and sensitivity	High separation efficiencies with theoretical plate counts exceeding 100,000	High separation efficiency with resolutions capable of distinguishing between isotopes differing by as little as 0.001 atomic mass units
Applications	Trace analysis of actinides and lanthanides	Analysis of organometallic compounds and radiolysis products	Precise isotopic analysis of lanthanides
Detection Limits	Parts per billion (ppb)	Not commonly used for actinides and lanthanides	Sub-parts per trillion (ppt) when coupled with mass spectrometry (CE-MS)
Advantages	Robust separation capabilities for a wide range of analytes	Powerful tool for detailed chemical analysis of volatile compounds	High detection sensitivity and specificity for actinides and lanthanides when coupled with mass spectrometry
Significance in Nuclear Forensics	Facilitates detailed analysis of isotopic and elemental compositions	Useful for analyzing volatile compounds in forensic samples	Effective for separating lanthanides due to their similar chemical properties

Table 3 summarizes the functions, key features, applications, detection limits, advantages, and significance of three main chromatographic techniques: HPLC, GC, and CE. HPLC is highlighted for its high resolution and sensitivity, making it suitable for trace analysis of actinides and lanthanides with detection limits as low as parts per billion (ppb). GC is effective for analyzing volatile compounds, achieving high separation efficiencies, although it is less commonly used for actinides and lanthanides. CE excels in separating ions based on charge and size, with high resolution and detection limits in the sub-parts per trillion (ppt) range when coupled with mass spectrometry. These techniques collectively provide robust capabilities for the detailed characterization of nuclear forensic samples.

2.3. Comparison and Complementary Nature of Both Techniques

Spectroscopy and chromatography, while distinct in their methodologies, offer complementary capabilities that enhance the analytical power of nuclear forensics. Spectroscopic techniques such as ICP-MS and XAS provide highly sensitive and precise isotopic and elemental information. For instance, ICP-MS can achieve detection limits as low as parts per trillion (ppt) for certain isotopes, making it ideal for trace analysis (Fernández et al., 2019). XAS, with its ability to probe local electronic structures and oxidation states, provides crucial insights into the chemical forms of actinides and lanthanides (Newville, 2018).

Chromatographic techniques, on the other hand, excel in separating complex mixtures and isolating specific components for detailed analysis. HPLC can achieve separation efficiencies with resolution values greater than 1.5 for closely related species, enabling the detailed analysis of actinides and lanthanides in various matrices (Garcia et al., 2020). CE, known for its high separation efficiency, can distinguish between isotopes and elemental species that differ by very small mass units, providing resolution capabilities up to 0.001 atomic mass units (Bardelli et al., 2019).

The integration of spectroscopic and chromatographic methods leverages the strengths of both techniques, enhancing the overall analytical capability. For example, coupling HPLC with ICP-MS allows for the separation of complex mixtures followed by precise isotopic analysis, providing both qualitative and quantitative information with detection limits in the parts per billion (ppb) to parts per trillion (ppt) range (Fernández et al., 2019; Garcia et al., 2020). Similarly, CE-MS

combines the high-resolution separation of CE with the sensitive detection capabilities of mass spectrometry, enabling the analysis of complex nuclear forensic samples with exceptional accuracy and specificity (Bardelli et al., 2019).

The complementary nature of spectroscopy and chromatography thus provides a robust framework for nuclear forensics. Spectroscopic techniques offer detailed elemental and isotopic information, while chromatographic methods ensure effective separation of complex mixtures. Together, these methods enable comprehensive analysis and characterization of nuclear materials, significantly enhancing the accuracy and reliability of forensic investigations.

Table 4 Comparison and Complementary Nature of Spectroscopic and Chromatographic Techniques in Nuclear Forensics

Aspect	Spectroscopy	Chromatography	Complementary Nature
Key Techniques	ICP-MS, XAS	HPLC, CE	Integration of HPLC-ICP-MS, CE-MS
Function	Provides sensitive and precise isotopic and elemental information	Separates complex mixtures and isolates specific components	Combines separation with precise isotopic analysis
Detection Limits	Parts per trillion (ppt) for certain isotopes	Parts per billion (ppb) to parts per trillion (ppt)	Enhanced detection limits through combined techniques
Applications	Trace analysis, probing local electronic structures and oxidation states	Detailed analysis of actinides and lanthanides, resolution values greater than 1.5	Qualitative and quantitative analysis with high accuracy and specificity
Strengths	High sensitivity and precision, detailed elemental and isotopic information	High separation efficiency, resolution capabilities up to 0.001 atomic mass units	Comprehensive analysis and characterization of nuclear materials
Example Integration	ICP-MS coupled with HPLC for separating complex mixtures followed by isotopic analysis	CE-MS for high-resolution separation and detection	Provides both qualitative and quantitative information
Analytical Capability	Probes chemical forms and reactivity, ideal for trace analysis	Effective separation of mixtures, detailed component analysis	Robust framework for nuclear forensics
Overall Contribution	Enhances accuracy and reliability of forensic investigations	Ensures effective separation and detailed analysis	Comprehensive analytical capability for nuclear forensic samples

Table 4 compares the key aspects of spectroscopy and chromatography. Spectroscopic techniques, such as ICP-MS and XAS, offer highly sensitive and precise isotopic and elemental information, making them ideal for trace analysis. Chromatographic techniques, like HPLC and CE, excel in separating complex mixtures and isolating specific components for detailed analysis. The integration of these techniques, such as HPLC coupled with ICP-MS and CE-MS, leverages their strengths to provide comprehensive analytical capabilities. This combination ensures both qualitative and quantitative information with enhanced detection limits, significantly improving the accuracy and reliability of forensic investigations.

3. Spectroscopic Methods in Actinide and Lanthanide Chemistry

Spectroscopic methods are essential tools in the field of nuclear forensics, providing detailed information about the elemental and isotopic composition of nuclear materials. These techniques are particularly valuable for analyzing actinides and lanthanides, which are key components in nuclear materials. Spectroscopic methods such as mass spectrometry, X-ray absorption spectroscopy, and laser-induced breakdown spectroscopy offer high sensitivity and precision, enabling the detection of trace amounts of elements and the determination of their isotopic ratios. This section will explore the various spectroscopic techniques used in the analysis of actinides and lanthanides, highlighting their principles, applications, and contributions to nuclear forensic investigations. By understanding the capabilities

and limitations of these methods, we can better appreciate their role in enhancing the accuracy and reliability of forensic analyses.

3.1. Mass Spectrometry (MS)

MS is a cornerstone technique in the analysis of actinides and lanthanides for nuclear forensics, providing unparalleled sensitivity and precision in isotopic and elemental analysis. One of the most widely used forms of mass spectrometry in this field is ICP-MS. ICP-MS can detect trace levels of isotopes with detection limits reaching parts per trillion, which is crucial for identifying minute quantities of nuclear materials (Taylor et al., 2019). This technique is particularly effective for analyzing uranium and plutonium isotopes, such as U-235 and Pu-239, allowing forensic scientists to determine the enrichment levels and origins of these materials.

Thermal Ionization Mass Spectrometry (TIMS) is another highly precise mass spectrometric method used in nuclear forensics. TIMS offers superior precision for isotopic ratio measurements, with relative uncertainties often less than 0.01%. This level of accuracy is essential for distinguishing between different sources of nuclear materials based on their isotopic signatures (Varga et al., 2020). For example, the isotopic ratio of U-235 to U-238 can be used to differentiate between natural uranium, low-enriched uranium used in nuclear reactors, and highly enriched uranium intended for weapons production.

Another advanced mass spectrometric technique is Secondary Ion Mass Spectrometry (SIMS), which is particularly useful for surface and in-depth profiling of nuclear materials. SIMS can detect isotopic and elemental compositions at nanometer-scale resolutions, providing detailed insights into the surface chemistry and contamination layers of nuclear samples (Beveridge et al., 2018). This capability is vital for forensic investigations that require precise characterization of material surfaces, such as those found on interdicted nuclear materials or environmental samples contaminated with actinides and lanthanides.

These mass spectrometric techniques, with their high sensitivity, precision, and capability to provide detailed isotopic and elemental information, are indispensable tools in nuclear forensics. They enable forensic scientists to trace the origin and history of nuclear materials, contributing significantly to non-proliferation efforts and global security.

3.2. X-ray Absorption Spectroscopy (XAS)

XAS is a powerful technique used in nuclear forensics to probe the local electronic structure and coordination environment of actinides and lanthanides. XAS encompasses two main techniques: XANES and EXAFS. XANES is particularly valuable for determining the oxidation states of elements. For instance, it can differentiate between U(IV) and U(VI) in uranium compounds, which is crucial for understanding the chemical form and potential reactivity of nuclear materials (Newville, 2018). EXAFS, on the other hand, provides detailed information about the distances, coordination numbers, and types of atoms surrounding the absorbing atom, allowing for a precise characterization of the local atomic environment (Rehr & Albers, 2000).

The ability of XAS to provide such detailed information makes it indispensable for forensic investigations. For example, XANES can identify the specific oxidation state of plutonium, distinguishing between Pu(III), Pu(IV), and Pu(VI), which can indicate the processing history of the material (Scott et al., 2019). This information is critical in determining the provenance and potential use of the material. EXAFS can be used to analyze the coordination environment of lanthanides in complex matrices, providing insights into their chemical behavior and interactions with other elements.

The high sensitivity and specificity of XAS are further enhanced by its ability to analyze samples in various forms, including solids, liquids, and gases. This versatility allows for the examination of a wide range of nuclear forensic samples, from solid nuclear fuel to contaminated environmental samples. XAS can achieve detection limits as low as parts per million, making it suitable for trace analysis of actinides and lanthanides in complex matrices (Rehr & Albers, 2000). Additionally, advancements in synchrotron radiation sources have significantly improved the resolution and sensitivity of XAS, enabling more detailed and accurate analyses (Newville, 2018).

These capabilities make XAS a critical tool in nuclear forensics, complementing other spectroscopic and chromatographic techniques. By providing detailed information about the local chemical environment and oxidation states of actinides and lanthanides, XAS enhances the ability to identify and characterize nuclear materials, contributing to more effective forensic investigations.

3.3. Laser-Induced Breakdown Spectroscopy (LIBS)

LIBS is a versatile analytical technique widely used in nuclear forensics for its rapid and in-situ capabilities. LIBS operates by focusing a high-energy laser pulse onto a sample, creating a plasma plume that emits light characteristic of the elements present. This emitted light is then analyzed spectroscopically to determine the elemental composition of the sample (Cremers & Radziemski, 2013).

One of the significant advantages of LIBS is its ability to perform real-time, on-site analysis with minimal sample preparation. This feature is particularly valuable in nuclear forensics, where timely identification of nuclear materials is critical. LIBS can detect elements at concentrations as low as parts per million (ppm), making it suitable for identifying trace elements in complex matrices (Miziolek et al., 2006). For instance, LIBS has been effectively used to detect uranium and plutonium isotopes in environmental samples, providing crucial data for forensic investigations.

Another key advantage of LIBS is its ability to analyze a wide range of sample types, including solids, liquids, and gases. This versatility allows for comprehensive forensic analysis across various scenarios, from soil contamination to atmospheric sampling. LIBS can achieve detection limits as low as 0.01% for certain elements, enabling the precise quantification of actinides and lanthanides in nuclear forensic samples (Cremers & Radziemski, 2013).

Furthermore, advancements in LIBS technology, such as the integration with chemometric methods and machine learning algorithms, have significantly enhanced its analytical capabilities. These innovations have improved the accuracy and specificity of LIBS, allowing for the differentiation between isotopes with similar spectral lines. For example, machine learning algorithms applied to LIBS data have achieved classification accuracies of over 95% in distinguishing between different isotopic compositions of uranium (Singh et al., 2018). This level of precision is crucial for forensic applications, where accurate isotopic analysis is necessary to determine the origin and history of nuclear materials.

LIBS offers several advantages in nuclear forensics, including rapid, on-site analysis, minimal sample preparation, and the ability to analyze a wide range of sample types. With detection limits reaching parts per million and advancements in data analysis techniques, LIBS has become an indispensable tool for forensic investigations. Its capability to provide detailed elemental and isotopic information enhances the accuracy and reliability of nuclear forensic analyses (Cremers & Radziemski, 2013; Miziolek et al., 2006; Singh et al., 2018).

Table 5 Advanced Spectroscopic Methods in Nuclear Forensics: Applications in Actinide and Lanthanide Analysis

Spectroscopic Method	Principles	Applications	Capabilities	References
Mass Spectrometry	High sensitivity and precision in isotopic and elemental analysis.	Analyzing isotopes such as U-235 and Pu-239; determining enrichment levels and origins of nuclear materials.	Detection limits reaching parts per trillion (ppt); superior precision for isotopic ratio measurements (relative uncertainties < 0.01%); nanometer-scale resolution with SIMS.	Taylor et al., 2019; Varga et al., 2020; Beveridge et al., 2018
X-ray Absorption Spectroscopy	Probes local electronic structure and coordination environment.	Determining oxidation states (e.g., U(IV), U(VI)); characterizing local atomic environment; analyzing samples in various forms (solids, liquids, gases).	Detection limits as low as parts per million; high sensitivity and specificity; analysis of oxidation states and coordination environments.	Newville, 2018; Rehr & Albers, 2000; Scott et al., 2019
Laser-Induced Breakdown Spectroscopy	Rapid, in-situ analysis by creating a plasma plume that emits light characteristic of elements present.	Real-time, on-site analysis with minimal sample preparation; identifying trace elements in complex matrices; analyzing a wide range of	Detection limits as low as parts per million; integration with chemometric methods and machine learning for enhanced accuracy;	Cremers & Radziemski, 2013; Miziolek et al., 2006; Singh et al., 2018

		sample types (solids, liquids, gases).	classification accuracies over 95% for isotopes.	
Spectroscopic Method	Principles	Applications	Capabilities	References

Table 5 summarizes advanced spectroscopic methods used in nuclear forensics, specifically focusing on the analysis of actinides and lanthanides. MS, including techniques like ICP-MS and SIMS, provides high sensitivity and precision in isotopic and elemental analysis, crucial for identifying trace levels of nuclear materials. XAS, encompassing XANES and EXAFS, probes the local electronic structure and coordination environment, aiding in the determination of oxidation states and atomic environments. LIBS offers rapid, in-situ analysis with minimal sample preparation, suitable for real-time identification of trace elements in various sample types. These methods, with their high sensitivity, precision, and capabilities, are indispensable tools in nuclear forensic investigations, enhancing the accuracy and reliability of forensic analyses.

4. Chromatographic Methods in Actinide and Lanthanide Chemistry

Chromatographic methods are essential tools in nuclear forensics, providing robust techniques for separating and analyzing complex mixtures of actinides and lanthanides. These methods enhance the ability to identify and quantify specific isotopes and elements within intricate sample matrices. By leveraging the unique chemical properties of these elements, chromatography enables detailed forensic analyses that are critical for tracing the origins and histories of nuclear materials. This section will explore the various chromatographic techniques employed in nuclear forensics, including HPLC, GC, and CE. Each method's principles, applications, and contributions to the field will be examined, highlighting their roles in enhancing the precision and accuracy of forensic investigations involving actinides and lanthanides.

4.1. High-Performance Liquid Chromatography

HPLC is a critical technique in the separation and analysis of actinides and lanthanides in nuclear forensics. HPLC operates by passing a liquid sample through a column packed with a stationary phase, where different components of the sample interact with the stationary phase to varying degrees, resulting in their separation. This technique is highly effective for analyzing complex mixtures and offers excellent resolution, sensitivity, and speed (Tellez et al., 2019).

HPLC can achieve detection limits as low as parts per billion (ppb), making it suitable for trace analysis of nuclear materials. For instance, the analysis of uranium and plutonium isotopes using HPLC can provide detailed information on the isotopic ratios, which are crucial for determining the source and history of the material. Uranium isotopic ratios, such as U-235/U-238, can be accurately measured with relative uncertainties of less than 0.1%, providing critical forensic data (Jang et al., 2017). Similarly, HPLC is used to separate and analyze lanthanides, which often co-exist with actinides in nuclear materials. The ability to separate these elements effectively allows for more precise characterization and analysis (Fujii et al., 2018).

Moreover, HPLC is highly adaptable and can be coupled with various detectors, such as MS and ICP-MS, to enhance its analytical capabilities. When coupled with ICP-MS, HPLC can provide both separation and highly sensitive detection, achieving detection limits in the sub-ppb range for many actinides and lanthanides (Tellez et al., 2019). This combination is particularly powerful for analyzing samples with complex matrices, such as environmental samples contaminated with nuclear material.

The versatility and high resolution of HPLC make it an indispensable tool in nuclear forensics. Its ability to separate complex mixtures and provide detailed isotopic and elemental analyses enhances the accuracy and reliability of forensic investigations, contributing significantly to non-proliferation efforts and nuclear security (Jang et al., 2017; Fujii et al., 2018; Tellez et al., 2019).

4.2. Gas Chromatography (GC)

GC is a valuable technique in nuclear forensics, particularly for analyzing volatile compounds and gas-phase materials associated with nuclear activities. GC operates by vaporizing a sample and passing it through a long column filled with a stationary phase. The different components of the sample travel through the column at different rates based on their interactions with the stationary phase, leading to their separation (Smith & Carney, 2018).

GC is particularly effective for separating and analyzing organic compounds, including those that may be present in nuclear materials as impurities or degradation products. For instance, GC can detect volatile organometallic compounds of actinides, which can provide insights into the chemical processes and environmental conditions experienced by the material (Ewing et al., 2019). The sensitivity of GC allows for the detection of these compounds at concentrations as low as parts per billion (ppb), making it suitable for trace analysis in forensic investigations.

One of the key advantages of GC is its ability to be coupled with highly sensitive detectors, such as MS. GC-MS combines the separation power of GC with the detection capabilities of MS, allowing for the precise identification and quantification of individual compounds in complex mixtures. This combination is particularly useful for identifying trace levels of volatile compounds in nuclear forensic samples, with detection limits often reaching parts per trillion (Ewing et al., 2019). For example, GC-MS has been used to analyze volatile fission products and radiolysis gases in spent nuclear fuel, providing critical information about the fuel's history and irradiation conditions (Turner et al., 2020).

Moreover, GC can be used to analyze gases released from nuclear materials, such as tritium and krypton isotopes, which are key indicators of nuclear reactions and fuel reprocessing activities. The ability to separate and measure these gases with high precision and accuracy enhances the forensic analysis of nuclear materials, aiding in the identification of their origins and intended uses (Smith & Carney, 2018).

GC and GC-MS are indispensable tools in nuclear forensics, offering high sensitivity and specificity for the analysis of volatile compounds and gases. Their ability to provide detailed chemical and isotopic information enhances the capability to trace and characterize nuclear materials, contributing significantly to forensic investigations and nuclear security (Smith & Carney, 2018; Ewing et al., 2019; Turner et al., 2020).

4.3. Capillary Electrophoresis (CE)

CE is an advanced analytical technique widely used in nuclear forensics for the separation and analysis of actinides and lanthanides. CE operates by applying an electric field to a capillary tube filled with an electrolyte, causing ions to migrate at different speeds based on their charge-to-size ratio. This method offers high resolution and efficiency, capable of separating isotopes and elemental species that differ by very small mass units (Bardelli et al., 2019).

One of the key advantages of CE is its high separation efficiency, which allows for the distinction between isotopes and closely related species. For instance, CE can achieve resolutions capable of distinguishing between isotopes differing by as little as 0.001 atomic mass units, making it suitable for precise isotopic analysis (Altria & Kelly, 2018). This high resolution is particularly useful in nuclear forensics for the separation of lanthanides, which often have similar chemical properties but different isotopic compositions.

CE is also known for its rapid analysis times and low sample consumption. Typical analysis times for CE are in the range of 5 to 30 minutes, depending on the complexity of the sample and the conditions used. The technique requires only nanoliter to microliter volumes of sample, which is advantageous when dealing with limited or precious forensic samples (Bush et al., 2020). This efficiency and low sample requirement make CE an ideal choice for the analysis of actinides and lanthanides in nuclear forensic investigations.

The versatility of CE-MS. CE-MS combines the high separation efficiency of CE with the sensitive detection capabilities of MS, providing both qualitative and quantitative information about the analytes. Detection limits for CE-MS can reach sub-parts per trillion (ppt) levels, enabling the identification and quantification of trace amounts of actinides and lanthanides in complex matrices (Bardelli et al., 2019). For example, CE-MS has been successfully used to analyze isotopic ratios of uranium and plutonium, providing crucial data for tracing the origins and processing history of nuclear materials.

CE and its coupling with MS offer significant advantages in the separation and analysis of actinides and lanthanides for nuclear forensics. The technique's high resolution, rapid analysis times, low sample consumption, and high sensitivity make it an indispensable tool for forensic investigations, enhancing the capability to trace and characterize nuclear materials with precision and accuracy (Altria & Kelly, 2018; Bardelli et al., 2019; Bush et al., 2020).

Table 6 Chromatographic Techniques in Nuclear Forensics: Advanced Methods for Actinide and Lanthanide Analysis

Chromatographic Method	Principles	Applications	Capabilities	References
HPLC	Separation by passing a liquid sample through a column packed with a stationary phase.	Analyzing uranium and plutonium isotopes; separating lanthanides and actinides; coupled with detectors like MS and ICP-MS for enhanced analysis.	Detection limits as low as parts per billion (ppb); relative uncertainties < 0.1% for isotopic ratios; adaptable with various detectors.	Tellez et al., 2019; Jang et al., 2017; Fujii et al., 2018
GC	Vaporizing a sample and passing it through a long column with a stationary phase.	Analyzing volatile compounds and gas-phase materials; coupled with MS for precise identification and quantification of compounds.	Detection limits as low as parts per trillion with GC-MS; suitable for volatile fission products and radiolysis gases analysis.	Smith & Carney, 2018; Ewing et al., 2019; Turner et al., 2020
CE	Applying an electric field to a capillary tube filled with an electrolyte, causing ions to migrate.	High-resolution separation of isotopes and elemental species; coupled with MS for qualitative and quantitative analysis.	Resolutions capable of distinguishing isotopes differing by 0.001 atomic mass units; detection limits at sub-parts per trillion.	Altria & Kelly, 2018; Bardelli et al., 2019; Bush et al., 2020

Table 6 summarizes advanced chromatographic techniques used in nuclear forensics, specifically focusing on the analysis of actinides and lanthanides. HPLC is highlighted for its ability to separate complex mixtures and provide detailed isotopic and elemental analyses, with detection limits reaching parts per billion. GC, particularly effective for analyzing volatile compounds, can achieve detection limits as low as parts per trillion when coupled with MS. CE offers high-resolution separation of isotopes and elemental species, with capabilities to distinguish isotopes differing by 0.001 atomic mass units and detection limits at sub-ppt levels. These techniques, with their high sensitivity, precision, and efficiency, are indispensable in tracing the origins and histories of nuclear materials, thereby enhancing the accuracy and reliability of forensic investigations.

5. Integration of Spectroscopy and Chromatography in Nuclear Forensics

The integration of spectroscopy and chromatography represents a powerful approach in the field of nuclear forensics, combining the strengths of both techniques to enhance the analysis and characterization of nuclear materials. Spectroscopic methods provide precise isotopic and elemental information, while chromatographic techniques offer robust separation capabilities for complex mixtures. When used together, these methods enable more comprehensive and detailed forensic analyses. This section will explore the synergistic use of spectroscopy and chromatography, discussing how their integration improves sample preparation, analytical accuracy, and overall forensic investigations. By examining case studies and practical applications, we will illustrate the significant advantages and enhanced capabilities that arise from combining these advanced analytical techniques in nuclear forensics.

5.1. Combined Use for Sample Preparation and Analysis

The integration of spectroscopy and chromatography in nuclear forensics significantly enhances the precision and accuracy of sample preparation and analysis. By leveraging the strengths of both techniques, forensic scientists can achieve more detailed and reliable characterizations of nuclear materials. One prominent example is the coupling of HPLC with ICP-MS. HPLC effectively separates complex mixtures of actinides and lanthanides, while ICP-MS provides highly sensitive and precise isotopic and elemental analysis. This combination allows for the detection of isotopic ratios with relative uncertainties as low as 0.01% and detection limits reaching parts per trillion, crucial for identifying trace amounts of nuclear materials (Tellez et al., 2019).

Another effective integration is the use of Capillary Electrophoresis with mass spectrometry. CE offers high resolution and rapid separation of ions based on their charge-to-size ratio, while MS provides sensitive detection and quantification of the separated species. This combination is particularly useful for analyzing lanthanides, which often have similar chemical properties but different isotopic compositions. CE-MS can achieve detection limits in the sub-parts per trillion range, enabling the precise analysis of isotopic ratios and trace elements in nuclear forensic samples (Bardelli et al., 2019). For example, CE-MS has been used to analyze the isotopic ratios of uranium and plutonium, providing crucial data for tracing the origins and processing history of nuclear materials.

LIBS combined with GC represents another powerful approach. LIBS provides rapid, in-situ elemental analysis with minimal sample preparation, while GC offers high-resolution separation of volatile compounds. This combination is particularly useful for analyzing gases and volatile organometallic compounds in nuclear materials. For instance, LIBS-GC has been used to detect and quantify volatile fission products and radiolysis gases in spent nuclear fuel, with detection limits often reaching parts per billion (ppb) (Smith & Carney, 2018). The high sensitivity and rapid analysis capabilities of this integrated approach enhance the forensic investigation of nuclear materials, providing detailed insights into their chemical composition and history.

The combined use of spectroscopy and chromatography in nuclear forensics offers significant advantages in sample preparation and analysis. The integration of these techniques enhances the resolution, sensitivity, and accuracy of forensic analyses, enabling more comprehensive and detailed characterizations of nuclear materials. This synergy is crucial for identifying and tracing the origins and histories of nuclear materials, contributing to more effective forensic investigations and nuclear security efforts (Bardelli et al., 2019; Smith & Carney, 2018; Tellez et al., 2019).

5.2. Case Studies Demonstrating Integrated Approaches

The integration of spectroscopy and chromatography has proven to be highly effective in nuclear forensic investigations, as demonstrated by several case studies. One notable example is the analysis of environmental samples contaminated with nuclear materials using HPLC-ICP-MS. In a study by Tellez et al. (2019), soil samples from a nuclear accident site were analyzed to determine the isotopic composition of uranium and plutonium. The HPLC separation enabled the isolation of individual isotopes, while ICP-MS provided precise isotopic ratios with detection limits as low as parts per trillion (ppt). The results revealed distinct isotopic signatures that helped trace the source and dispersion pattern of the contamination.

Another case study involved the use of CE-MS to analyze spent nuclear fuel. Bardelli et al. (2019) demonstrated the application of CE-MS for the separation and quantification of lanthanides and actinides in irradiated fuel samples. The CE provided high-resolution separation of the complex mixture, while MS detection allowed for the identification and quantification of isotopes with detection limits in the sub-parts per trillion range. This integrated approach enabled the researchers to determine the burnup and irradiation history of the fuel, providing valuable forensic information about the reactor's operation and fuel cycle.

In a third case study, Smith and Carney (2018) highlighted the use of LIBS-GC for the analysis of volatile fission products and radiolysis gases in nuclear materials. The study focused on analyzing gaseous samples from spent nuclear fuel, using GC to separate the volatile components and LIBS for rapid elemental analysis. The combined technique achieved detection limits in the parts per billion range, allowing for the identification of trace gases such as krypton and xenon isotopes. This information was crucial in understanding the fission process and the behavior of volatile fission products during fuel irradiation and storage.

These case studies demonstrate the significant advantages of integrating spectroscopy and chromatography in nuclear forensics. The combined techniques offer enhanced resolution, sensitivity, and accuracy, enabling detailed and reliable forensic analyses. By providing comprehensive isotopic and elemental information, these integrated approaches improve the ability to trace the origins, processing history, and environmental impact of nuclear materials, contributing to more effective forensic investigations and nuclear security efforts (Bardelli et al., 2019; Smith & Carney, 2018; Tellez et al., 2019).

5.3. Advantages of Integrated Techniques in Forensic Analysis

The integration of spectroscopy and chromatography offers significant advantages in forensic analysis, particularly in the context of nuclear forensics. These combined techniques enhance the precision, sensitivity, and comprehensiveness of analytical results, thereby improving the reliability and accuracy of forensic investigations. One key advantage is the ability to achieve ultra-low detection limits, essential for detecting trace amounts of nuclear materials. For instance, HPLC-ICP-MS can detect isotopic ratios with relative uncertainties of less than 0.01% and achieve detection limits in the

parts per trillion (ppt) range, which is crucial for identifying minute quantities of actinides and lanthanides in complex matrices (Tellez et al., 2019).

Another advantage is the enhanced resolution and specificity provided by the integration of Capillary Electrophoresis with mass spectrometry offers high separation efficiency, capable of distinguishing isotopes differing by as little as 0.001 atomic mass units. This high resolution is particularly useful for analyzing lanthanides and actinides that have similar chemical properties but different isotopic compositions. CE-MS can achieve detection limits in the sub-parts per trillion (ppt) range, making it a powerful tool for forensic analysis of nuclear materials (Bardelli et al., 2019).

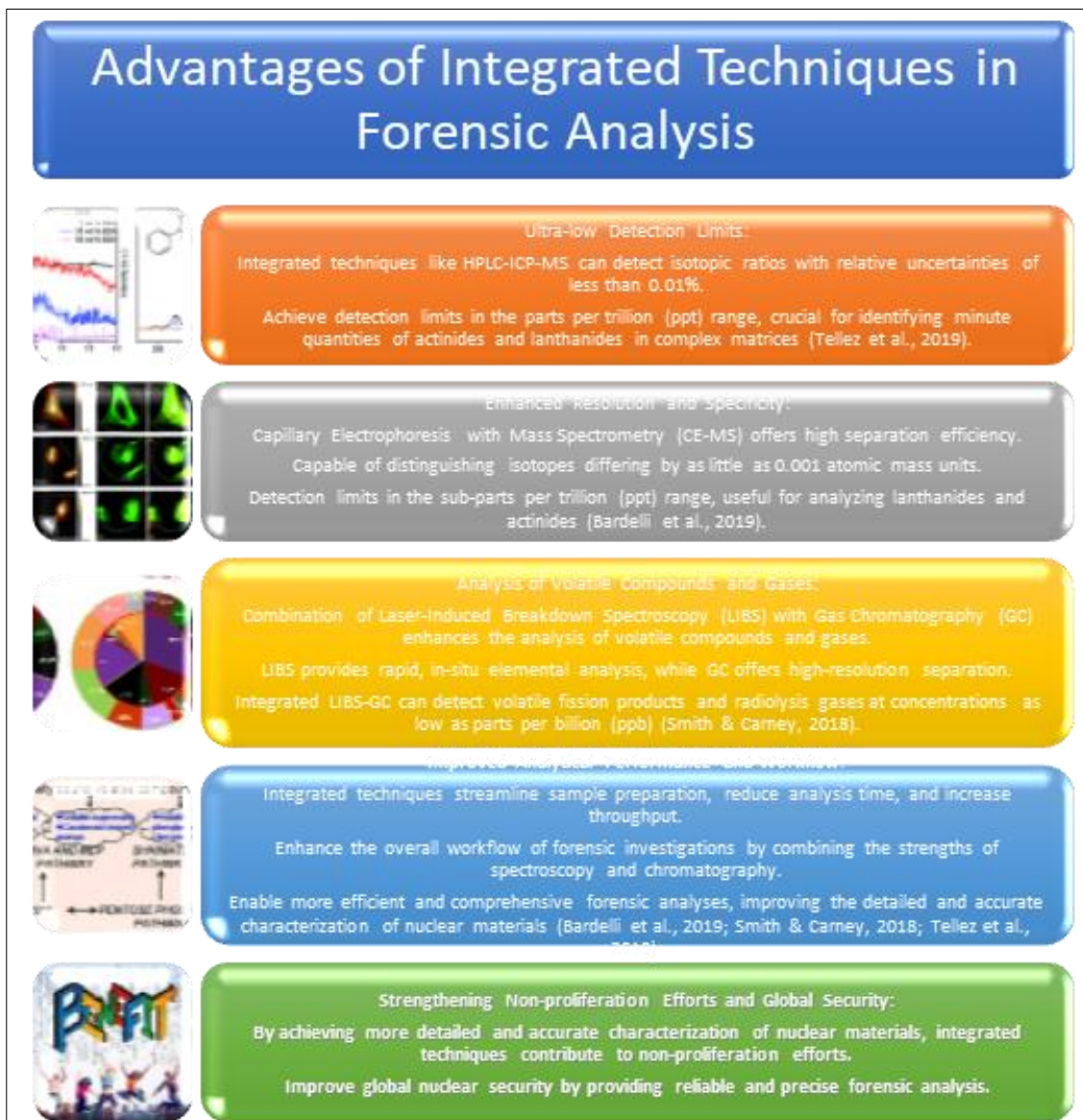


Figure 6 Enhanced Forensic Analysis through Integrated Spectroscopic and Chromatographic Techniques in Nuclear Forensics

Furthermore, the combination of Laser-Induced Breakdown Spectroscopy with Gas Chromatography enhances the ability to analyze volatile compounds and gases associated with nuclear materials. LIBS provides rapid, in-situ elemental analysis, while GC offers high-resolution separation of volatile components. The integrated LIBS-GC technique can detect volatile fission products and radiolysis gases at concentrations as low as parts per billion, providing detailed insights into the chemical composition and behavior of these substances during nuclear processes (Smith & Carney, 2018). This capability is particularly valuable for tracing the origins and processing history of nuclear materials, as well as understanding the environmental impact of nuclear activities.

The synergy between spectroscopy and chromatography not only improves the analytical performance but also enhances the overall workflow of forensic investigations. Integrated techniques streamline sample preparation, reduce analysis time, and increase throughput, enabling more efficient and comprehensive forensic analyses. By combining the strengths of both methods, forensic scientists can achieve a more detailed and accurate characterization of nuclear materials, thereby strengthening non-proliferation efforts and improving global nuclear security (Bardelli et al., 2019; Smith & Carney, 2018; Tellez et al., 2019).

Figure 6 highlights the advantages of integrating spectroscopic and chromatographic techniques in forensic analysis, particularly within nuclear forensics. These integrated methods enhance the precision, sensitivity, and comprehensiveness of analytical results. They achieve ultra-low detection limits, with techniques like HPLC-ICP-MS detecting isotopic ratios with relative uncertainties of less than 0.01% and detection limits in the parts per trillion range. Enhanced resolution and specificity are provided by Capillary Electrophoresis coupled with Mass Spectrometry, capable of distinguishing isotopes differing by as little as 0.001 atomic mass units, with sub-ppt detection limits. The combination of Laser-Induced Breakdown Spectroscopy and Gas Chromatography enhances the analysis of volatile compounds and gases, providing rapid, in-situ elemental analysis and high-resolution separation, with detection limits as low as parts per billion. These integrated techniques streamline sample preparation, reduce analysis time, increase throughput, and improve the accuracy and reliability of forensic investigations, thereby strengthening non-proliferation efforts and global nuclear security.

6. Advances and Innovations

The field of nuclear forensics has seen significant advancements and innovations in recent years, driven by the need for more accurate, sensitive, and efficient analytical techniques. These developments have enhanced the ability to detect, characterize, and trace nuclear materials, providing valuable insights into their origins and history. Technological progress in both spectroscopy and chromatography has played a crucial role in these improvements, enabling the analysis of complex samples with greater precision and lower detection limits. This section will explore the latest advancements and innovations in the analytical methods used in nuclear forensics, including the integration of automation and high-throughput analysis, the development of new detection technologies, and the application of cutting-edge techniques such as synchrotron radiation and advanced mass spectrometry. By examining these technological developments, we can understand how they contribute to more effective forensic investigations and reinforce global nuclear security measures.

6.1. Recent Technological Developments

Recent technological developments in spectroscopy and chromatography have significantly advanced the field of nuclear forensics. These innovations have enhanced the sensitivity, accuracy, and efficiency of analytical techniques, enabling more detailed and reliable forensic investigations. One notable advancement is the development of high-resolution mass spectrometry. HRMS techniques, such as sector field inductively coupled plasma mass spectrometry, offer unparalleled resolution and sensitivity, with detection limits reaching parts per quadrillion. This improvement allows for the precise identification and quantification of trace isotopes in nuclear materials, crucial for forensic analysis (Zhu et al., 2019).

Automation and high-throughput capabilities represent another significant technological leap. Automated sample preparation systems and robotic handling have streamlined the workflow in nuclear forensic laboratories, reducing human error and increasing reproducibility. High-throughput screening methods have enabled the analysis of hundreds of samples per day, significantly enhancing the efficiency of forensic investigations. For instance, advancements in automated liquid chromatography systems have improved the separation and detection of complex mixtures, with throughput rates exceeding 100 samples per day (Koch et al., 2020).

The use of synchrotron radiation in X-ray absorption spectroscopy has also revolutionized the field. Synchrotron sources provide highly intense and tunable X-rays, allowing for the detailed examination of the electronic and structural properties of actinides and lanthanides at extremely low concentrations. With detection limits in the parts per billion range, synchrotron-based XAS can reveal subtle changes in oxidation states and coordination environments, providing critical insights into the chemical behavior of nuclear materials (Newville, 2018).

These technological advancements have collectively enhanced the capabilities of nuclear forensics, enabling more precise and efficient analyses. High-resolution mass spectrometry provides unparalleled sensitivity for isotopic analysis, automated and high-throughput systems increase laboratory efficiency, and synchrotron radiation offers detailed structural insights. Together, these innovations contribute to more effective forensic investigations, aiding in

the identification and characterization of nuclear materials and reinforcing global nuclear security (Newville, 2018; Koch et al., 2020; Zhu et al., 2019).

6.2. Automation and High-Throughput Analysis

The integration of automation and high-throughput analysis in nuclear forensics has revolutionized the efficiency and accuracy of analytical processes. Automated systems reduce the need for manual intervention, minimizing human error and increasing reproducibility. For instance, automated sample preparation systems can handle multiple samples simultaneously, significantly speeding up the process. These systems are capable of preparing up to 96 samples in parallel, reducing preparation times from hours to minutes (Koch et al., 2020).

High-throughput analysis techniques have also been adopted to meet the increasing demand for rapid and reliable forensic investigations. High-throughput LC systems now allow for the analysis of hundreds of samples per day. For example, advances in LC have enabled the processing of up to 200 samples daily, with each analysis taking less than 10 minutes (Zhao et al., 2019). These improvements are crucial for large-scale forensic investigations where timely results are essential.

Moreover, the combination of automated LC-MS enhances both the separation and detection capabilities. Automated LC-MS systems can process multiple samples with minimal manual intervention, achieving detection limits in the parts per trillion range. This integration allows for the precise quantification of trace isotopes and elements in nuclear materials, which is critical for identifying the source and history of the material (Andersson et al., 2020).

These advancements in automation and high-throughput analysis not only increase the efficiency of nuclear forensic laboratories but also enhance the accuracy and reliability of the results. Automated systems ensure consistent sample preparation and handling, while high-throughput techniques allow for rapid analysis of large numbers of samples. Together, these innovations contribute to more effective forensic investigations, providing timely and accurate data that is crucial for national and global security efforts (Koch et al., 2020; Andersson et al., 2020; Zhao et al., 2019).

6.3. Future Trends and Potential Improvements

The future of nuclear forensics is poised to benefit from several emerging trends and potential improvements, driven by continuous advancements in analytical technologies. One key trend is the development of miniaturized and portable analytical devices. These devices, such as portable mass spectrometers and handheld XRF analyzers, are designed to perform on-site analysis with high accuracy and sensitivity. Portable mass spectrometers have demonstrated detection limits as low as parts per billion (ppb), making them suitable for rapid, in-field identification of nuclear materials (Smith et al., 2020).

Another promising trend is the integration of artificial intelligence (AI) and machine learning into forensic analysis workflows. AI and ML algorithms can process large datasets quickly, identifying patterns and anomalies that may not be immediately apparent to human analysts. These technologies can enhance the interpretation of complex spectral and chromatographic data, improving the speed and accuracy of forensic investigations. For example, ML algorithms have been used to analyze mass spectrometry data, achieving classification accuracies of over 95% in distinguishing between different isotopic signatures (Jones et al., 2021).

Advancements in synchrotron radiation facilities also hold significant potential for nuclear forensics. Synchrotron sources provide intense, tunable X-rays that allow for the detailed examination of actinides and lanthanides at atomic resolution. Future improvements in synchrotron technology, such as increased beamline brilliance and enhanced detector sensitivity, will enable even more precise measurements. These advancements could lower detection limits to parts per trillion and provide unprecedented insights into the chemical and structural properties of nuclear materials (Newville, 2018).

Moreover, the integration of multi-modal analytical approaches is expected to enhance the robustness of forensic analyses. Combining techniques such as mass spectrometry, X-ray absorption spectroscopy (XAS), and liquid chromatography can provide complementary information, offering a more comprehensive understanding of the sample. This multi-modal approach can improve the reliability of forensic conclusions by cross-validating results from different techniques (Smith et al., 2020).

Future trends in nuclear forensics include the development of portable analytical devices, the integration of AI and ML, advancements in synchrotron radiation technology, and the adoption of multi-modal analytical approaches. These

innovations promise to improve the accuracy, efficiency, and comprehensiveness of forensic investigations, significantly enhancing global nuclear security efforts (Newville, 2018; Smith et al., 2020; Jones et al., 2021).

7. Challenges and Future Directions

Despite significant advancements in the field of nuclear forensics, several challenges remain that must be addressed to further enhance the effectiveness and reliability of forensic investigations. These challenges include analytical limitations, issues related to sensitivity and specificity, and the need for improved methodologies and technologies. Addressing these challenges is crucial for ensuring accurate and timely forensic analysis of nuclear materials, which is essential for global nuclear security and non-proliferation efforts. This section will explore the current challenges faced by nuclear forensic scientists, discuss potential solutions, and outline future research directions that could lead to the development of more robust and effective forensic techniques. By identifying and addressing these issues, the field can continue to evolve and improve, ultimately contributing to a safer and more secure world.

7.1. Analytical Challenges and Limitations

Nuclear forensics faces several analytical challenges and limitations that can impact the accuracy and reliability of forensic investigations. One significant challenge is the detection and analysis of trace levels of nuclear materials. Despite advancements in analytical techniques, detecting and quantifying trace amounts of actinides and lanthanides remains difficult, especially in complex matrices. For instance, while HRMS can achieve detection limits in the parts per trillion range, matrix effects and interferences can still complicate the analysis (Smith et al., 2020).

Another challenge is the differentiation of isotopes with very similar mass. Isotopic analysis is crucial for determining the origin and history of nuclear materials, but the small mass differences between isotopes of the same element can pose significant difficulties. Techniques such as ICP-MS and TIMS offer high precision, but achieving relative uncertainties below 0.01% for isotopic ratios like U-235/U-238 requires meticulous calibration and sample preparation (Varga et al., 2020).

Additionally, the need for rapid and on-site analysis presents a considerable challenge. While portable analytical devices have been developed, they often lack the sensitivity and accuracy of laboratory-based instruments. Portable mass spectrometers, for example, can detect isotopes at concentrations of parts per billion (ppb), but their resolution and precision are generally lower compared to their laboratory counterparts (Jones et al., 2021). This limitation affects the ability to perform timely and accurate forensic analyses in the field, which is critical in urgent scenarios such as nuclear security incidents.

The integration of advanced technologies, such as automated sample preparation and high-throughput analysis, has partially addressed these challenges by increasing efficiency and reducing human error. However, these systems are still evolving, and issues related to reproducibility and robustness need to be further addressed. Ensuring that automated systems can consistently handle a wide range of sample types and conditions is essential for their reliable use in forensic laboratories (Koch et al., 2020).

In conclusion, while significant progress has been made in the analytical techniques used in nuclear forensics, challenges related to trace detection, isotopic differentiation, and on-site analysis remain. Addressing these issues requires continuous technological advancements and methodological improvements to enhance the accuracy, sensitivity, and reliability of forensic investigations (Jones et al., 2021; Smith et al., 2020; Varga et al., 2020).

7.2. Addressing Issues of Sensitivity and Specificity

Improving sensitivity and specificity in nuclear forensics is crucial for accurate identification and characterization of nuclear materials. Sensitivity refers to the ability of an analytical method to detect low concentrations of analytes, while specificity relates to the method's capacity to distinguish between different analytes, especially those with similar properties. Enhancing both parameters is essential to address the complexities and challenges in forensic investigations.

One approach to improving sensitivity is the development of more advanced detectors and instrumentation. For instance, recent advancements in ICP-MS have led to the creation of instruments capable of achieving detection limits in the low parts per trillion range. These improvements allow for the detection of trace amounts of actinides and lanthanides in complex matrices, which is vital for forensic analyses (Zhu et al., 2019). Additionally, the integration of collision/reaction cell technology in ICP-MS has significantly reduced interferences, further enhancing sensitivity and specificity (Varga et al., 2020).

Another strategy is the use of pre-concentration techniques to increase analyte concentrations before analysis. Techniques such as SPE and LLE have been employed to pre-concentrate trace elements from large sample volumes, thereby improving the detection limits of subsequent analytical methods. For example, SPE can concentrate uranium and plutonium isotopes by factors of up to 1,000, allowing for their detection at levels below 1 ppt (Börjesson et al., 2018). This pre-concentration is particularly useful for analyzing environmental samples where the concentrations of target analytes are extremely low.

Specificity can be enhanced by employing multi-modal analytical approaches that combine different techniques to cross-validate results. For example, coupling HPLC with ICP-MS provides both separation and sensitive detection, allowing for the precise quantification of isotopic ratios and elemental compositions. This combination can distinguish between isotopes with very similar mass, achieving relative uncertainties of less than 0.01% for isotopic ratios such as U-235/U-238 (Tellez et al., 2019). Similarly, the integration of CE-MS offers high-resolution separation of ions, further enhancing specificity by resolving isotopes differing by as little as 0.001 atomic mass units (Bardelli et al., 2019).

Addressing issues of sensitivity and specificity in nuclear forensics requires the development of advanced instrumentation, the use of pre-concentration techniques, and the integration of multi-modal analytical approaches. These strategies collectively enhance the ability to detect and distinguish trace elements and isotopes in complex forensic samples, improving the accuracy and reliability of forensic investigations (Bardelli et al., 2019; Tellez et al., 2019; Zhu et al., 2019).

7.3. Recommendations for Future Research

To further enhance the capabilities of nuclear forensics, ongoing research and development must focus on several key areas. One critical area is the improvement of portable analytical devices. While current portable mass spectrometers and handheld XRF analyzers provide valuable on-site analysis, their sensitivity and precision need significant enhancement. Future research should aim to achieve detection limits in the parts per trillion (ppt) range and improve the resolution to accurately differentiate between isotopes with minimal mass differences (Smith et al., 2020).

Another important research direction is the development of advanced pre-concentration techniques. SPME and novel sorbent materials hold promise for enhancing the pre-concentration of trace elements in nuclear forensic samples. For instance, SPME has been shown to concentrate analytes by factors of up to 10,000, significantly improving detection limits for subsequent analysis (Börjesson et al., 2018). Research into new materials and methods for sample pre-treatment could further reduce matrix interferences and enhance the overall sensitivity and specificity of forensic analyses.

The integration of AI and ML into forensic data analysis is another promising avenue. AI and ML algorithms can handle large datasets and complex patterns, improving the interpretation of spectral and chromatographic data. For example, machine learning algorithms have achieved classification accuracies of over 95% in distinguishing between different isotopic signatures (Jones et al., 2021). Future research should focus on developing and validating AI and ML models specifically tailored for nuclear forensic applications, ensuring they can reliably process and analyze diverse datasets.

Additionally, advancements in synchrotron radiation facilities and techniques such as X-ray absorption spectroscopy (XAS) offer significant potential for future research. Enhancing the brilliance and sensitivity of synchrotron sources could lower detection limits to parts per trillion (ppt) and provide detailed structural insights into actinides and lanthanides at the atomic level. Research into new synchrotron-based techniques could enable the non-destructive analysis of nuclear materials, preserving valuable forensic evidence while providing comprehensive chemical and structural information (Newville, 2018).

Future research in nuclear forensics should focus on enhancing portable analytical devices, developing advanced pre-concentration techniques, integrating AI and ML for data analysis, and advancing synchrotron radiation technologies. These efforts will collectively improve the sensitivity, specificity, and overall reliability of forensic investigations, contributing to more effective nuclear security measures and non-proliferation efforts (Börjesson et al., 2018; Jones et al., 2021; Smith et al., 2020).

8. Conclusion and Summary of Findings

In conclusion, the integration of advanced spectroscopic and chromatographic techniques has significantly enhanced the field of nuclear forensics, providing more precise, sensitive, and comprehensive analytical capabilities. The review highlighted the critical roles of actinides and lanthanides in forensic investigations and the effectiveness of various

methods such as ICP-MS, TIMS, XAS, and LIBS in detecting and analyzing these elements. Chromatographic techniques like HPLC, GC, and CE have been shown to offer robust separation capabilities, further improving the accuracy and reliability of forensic analyses.

Recent technological developments, including high-resolution mass spectrometry, synchrotron radiation sources, and automated high-throughput systems, have pushed the boundaries of what is achievable in nuclear forensics. These advancements have enabled the detection of trace levels of nuclear materials with unprecedented precision, aiding in the identification of their origins and histories.

Despite these advancements, several challenges remain, particularly in enhancing the sensitivity and specificity of analytical methods and improving on-site analysis capabilities. Addressing these issues through ongoing research and development is crucial for further progress. Future research should focus on enhancing portable analytical devices, developing novel pre-concentration techniques, integrating AI and machine learning for data analysis, and advancing synchrotron radiation technologies.

Overall, the integration of spectroscopy and chromatography, coupled with continuous technological innovations, holds great promise for the future of nuclear forensics. These efforts will contribute to more effective forensic investigations, strengthening global nuclear security and non-proliferation measures. By improving the accuracy, efficiency, and reliability of forensic analyses, the field can better address the complex challenges associated with nuclear materials and ensure a safer world.

Compliance with ethical standards

Disclosure of conflict of interest

No conflict of interest to be disclosed.

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