

Benefits and Drawbacks of Non-Destructive Assay (NDA) for Nuclear Safeguards - An Overview

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Abstract:

Nuclear safeguards are technical measures used to detect the diversion of nuclear material and technologies from their peaceful use. A key trait of safeguards is represented by measurements, which allow operators and inspectors to verify the nuclear activities and the amount of nuclear material declared by the States. In particular, an important class of measurements are Non-Destructive Assay (NDA) techniques, which allow one to characterize samples while preserving their physical integrity. NDA represents a rapid and effective method for measurements of nuclear materials in a variety of situations (from process lines to finished products, scrap and waste materials), with the possibility to perform on-site measurements, reducing radioactive waste production and offering wide margins for automatization and remote operation. However, there are situations where the use of NDA is not recommended or possible and one has to rely on Destructive Assay (DA) (i.e., techniques which alter the integrity of the item analysed). This happens, for example, in environmental sampling, when dealing with highly diluted nuclear materials, or when the radiation emitted by the sample is weakly penetrating. In conclusion, the integrated use of different NDA and DA techniques is the best possible solution for measurements in the frame of nuclear safeguards.

Keywords: Nuclear Safeguards, Nuclear Instrumentation, Nuclear Measurements, NDA techniques.

1. Introduction

Nuclear materials and technologies offer a powerful tool for the progress of society and industry across the world. However, the same instruments may also be used in a harmful way, like for the development of nuclear weapons. To detect the diversion of nuclear material from plants and facilities, preventing the proliferation of nuclear weapons and the illicit trafficking of nuclear and radiological materials, a series of international technical measures have been developed in the form of legally binding agreements. These measures are known as “nuclear safeguards”. Nuclear safeguards originated from the signing of the Non-Proliferation Treaty (NPT) in 1968, when non-nuclear weapon States officially declared that they would not develop nuclear weapons and agreed on establishing measures enabling the verifications of compliance to the NPT [1]. These verifications are performed by safeguards inspectorates: at an international level, the reference authority is represented by the International Atomic Energy Agency (IAEA) [2], while, for example, at the European Union level the reference authority is EURATOM [3], [4]. The European Commission collaborates actively with EURATOM in the frame of safeguards development via the DG ENER (EURATOM Safeguards) [5] and the Joint Research Centre (Nuclear Security Unit) [6], [7]. Another important contribution to European nuclear safeguards is given by the European Safeguards Research and Development Association (ESARDA), an association of European organizations established in 1969 to advance and harmonize R&D in the area of nuclear safeguards [8].

Initially, safeguards were only focused on the verification of activities and amounts of material declared by the States to the inspectors, but after the Iraqi crisis of 1991 [9], through the Additional Protocol (AP) (also known as INFCIRC/540) by IAEA [10], they became aimed at providing a more comprehensive picture of a State’s nuclear activities.

Through safeguards, a State can demonstrate to others that its nuclear material and technologies are being used for peaceful purposes. The IAEA safeguards were established about 60 years ago and nowadays it is clear that nuclear cooperation and the exchange of scientific and technological expertise among countries have benefited from these measures. At present, more than 180 States worldwide have put into force IAEA safeguards [11]. The continuous enhancement of safeguards efficiency is crucial to

improve the cost-effectiveness of safeguards mechanisms, to encourage international peaceful nuclear collaboration based on the exchange of nuclear technologies and materials, and to adapt to evolving/emerging technologies (e.g., Gen IV reactors, Small Modular Reactors, nuclear fusion, ...). Furthermore, by enhancing safeguards efficiency, the IAEA can maintain its credibility and effectiveness in implementing safeguards on a global scale.

To detect the diversion of nuclear material from declared activities to undeclared activities, it is necessary to account for the declared material via inspections and measurements. In particular, measurements of mass and/or volume and sample analysis play a key role in the control of nuclear material since they allow the characterization of the material both qualitatively (i.e., identifying its nature/components (isotopic composition)) and quantitatively. Measurement techniques can be divided into two main groups: Non-Destructive Assay (NDA) techniques and Destructive Assay (DA) techniques.

Non-Destructive Assay techniques allows the investigation of a sample preserving its physical integrity, as suggested by the name itself. NDA techniques have matured over the years and gained in precision, and nowadays these techniques allow for efficient and accurate measurement of nuclear material, including special nuclear material (SNM) like plutonium and enriched uranium [12], as well as other nuclear materials such as neptunium (^{237}Np) [13], which in turn allows authorities to draw elaborate safeguards for nuclear material accounting and control (NAMC), process control and perimeter monitoring [14].

In contrast to NDA techniques, DA techniques involve sampling from the material under analysis, affecting its physical integrity. Each sample taken from the original item must be representative of the bulk of the material from which it is taken. Moreover, these samples usually undergo a preparation prior to the measurement, which changes their physical/chemical form. For example, specific solutions or oxides may be required to carry out the measurement. Besides the analysis of samples of nuclear material for the verification of declared amounts of material, DA techniques are crucial for the detection of undeclared nuclear activities through environmental sample analysis. Furthermore, DA techniques are crucial to certify working standards used for the calibration of NDA techniques. The most common DA techniques for nuclear safeguards are based on precipitation/weighting of chemical analytes, analytical electro-/radio-chemistry and mass spectrometry.

Besides measurements, IAEA safeguards also involves containment and surveillance as key elements for drawing conclusions.

This paper describes the NDA techniques used for nuclear safeguards – focusing on the main techniques and providing some examples of advanced and novel methods – with

the aim of pointing out their benefits and drawbacks. Instead, a detailed discussion of DA techniques and of containment and surveillance, which is beyond the scope of the present work, can be found, for example, in [15].

2. Overview of NDA techniques

The main NDA techniques used for nuclear safeguards can be classified into three groups:

a) Gamma-ray spectrometry: this technique measures the gamma rays (and X-rays) emitted by radioactive nuclides in terms of energy and intensity. Because the photons are a “fingerprint” of the emitting nuclide, gamma-ray spectrometry allows a qualitative analysis of the sample (i.e., it allows one to recognize which nuclides it contains) besides the quantitative analysis linked to the abundance of isotopes in the sample. The reference tools for this NDA technique are gamma-spectrometers, which can include scintillators and semiconductor detectors. The detectors sense radiation based on the excitation (scintillators) and ionization (semiconductors) of the active material by the incoming γ rays [16]. In the context of nuclear safeguards, scintillators (like NaI(Tl) or LaBr_3), having a good efficiency but a relatively poor energy resolution, are commonly limited to uranium enrichment studies. On the contrary, semiconductors like high-purity germanium (HPGe), offering excellent energy resolution but a limited efficiency, are routinely used to obtain and unfold complex spectra such as the one of Pu isotopes. Figure 1 shows a comparison of spectra of U and Pu acquired with different instruments.

Nevertheless, it should be pointed out that gamma-ray spectrometry is not capable of measuring the isotope ^{242}Pu due to its very low specific gamma activity. Moreover, the drawback for the excellent resolution of HPGe is that the detector must be maintained at a low temperature (typically at 77 K), which makes the detector setup bulky and expensive. A good compromise between efficiency and resolution is represented by semiconductors like CdTe and CdZnTe, which can be operated at room temperature. In [16] a list of the state-of-the-art scintillators and semiconductor detectors is available, showing all their main features and discussing their performance.

In general, gamma-ray spectrometry is fundamental for the detection of divergences (“defect verification”) between the declared and measured values of spent fuel [18], [19]. As an example, semiconductors are routinely used for the detection of fission products in spent fuel.

Gamma-ray spectrometry is an NDA technique requiring sophisticated instrumentation and skilled operators: deep training [20] is crucial to master both the hardware and the software (including numerical codes) accompanying the spectrometers [21]. The relative error associated with gamma-spectrometry measurements varies with the isotope

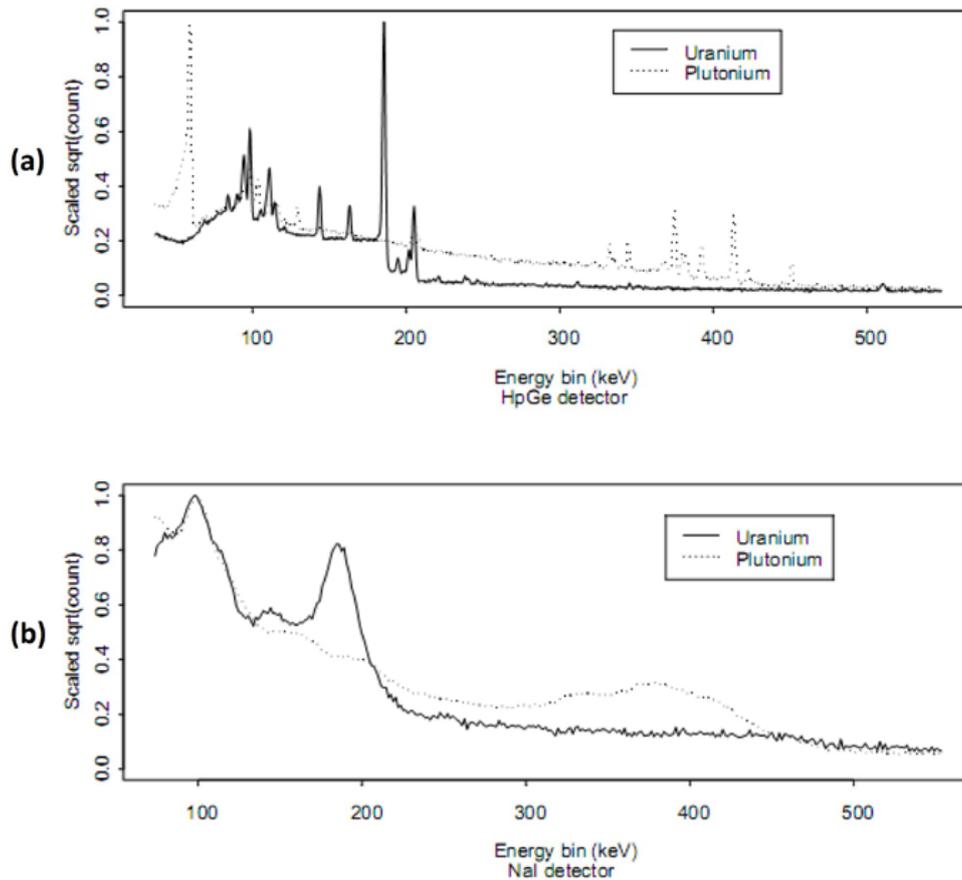
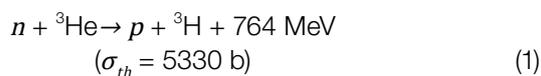


Figure 1. Examples of (a) high resolution (HPGe) and (b) low resolution (NaI) spectra for ^{235}U and ^{239}Pu . Image adapted from [17].

content (enrichment, fuel burnup, ...) and with the counting time: typical values are in the range of $\sim 0.1\%$ - 10% [22]. Generally, high-resolution gamma-spectrometry can detect directly a mass of $^{235}\text{U}/^{239}\text{Pu}$ greater than about 10 mg [23], [24].

It is worth mentioning that a novel technique based on gamma-ray detection such as partial defect verification: gamma-ray emission tomography [25].

b) Neutron assay: this technique is based on the detection of fission neutrons emitted by fissile and special nuclear materials either spontaneously or due to reactions induced by an investigation source. The detectors used for this assay are typically gas-filled detectors, mainly ^3He proportional counters which can detect thermal neutrons via the reaction:



The charged reaction products in Equation (1) interact with the filling gas of the detector (via ionization and excitation events), generating an electrical signal (charge pulse), from which the presence of neutrons is deduced. Since fission events generally release 2-3 neutrons [26], neutrons produced by fission can be discriminated from random

neutrons occurring from background natural/cosmic radiation and from the ones of interrogation sources by using electronics working in coincidence mode. The rationale for discrimination is the following: if two pulses are detected within a short time window ("gate") they are considered as temporally correlated events and are counted as a real fission event ("double"), on the contrary, if only one event is detected it is considered a random individual neutron. This is the basis of the neutron coincidence counting method (NCC), which is the most widespread NDA technique for the determination of the Pu mass in bulk samples. More complex algorithms, such as neutron multiplicity counting methods (NMC) have been developed to account for single, double and triple events detected within the gate, in order to extract quantitative information about the neutron multiplication effect within the sample [27]. If real coincident multiples are present, the distribution in time of events (detector pulses/counts) that follow an arbitrarily chosen starting event (pulse) is given by the Rossi-alpha distribution:

$$S(t) = A + R \cdot e^{-\frac{t}{\tau}} \quad (2)$$

where A is the accidental coincidence count rate, R is the real coincidence count rate and τ is the die-away time of the detector assembly [23]. The exponential form of the Rossi-alpha distribution comes from the fact that a population of coincident neutrons emitted at a time $t = 0$ in a

fission reaction will decay in time as $e^{-\frac{t}{\tau}}$. Figure 2 shows an example of a Rossi-alpha distribution: a prompt gate starting at $t = 0$ and ending when the distribution reaches a plateau collects real coincidences (“R”, green area) plus accidental events (“A”, red area), while a sufficiently delayed gate as wide as the first one collects only accidental events. Counting the events in the two gates and subtracting the events collected in the delayed gate from those collected in the prompt gate gives the real counts. This is done in practice using shift register electronics [15], [23].

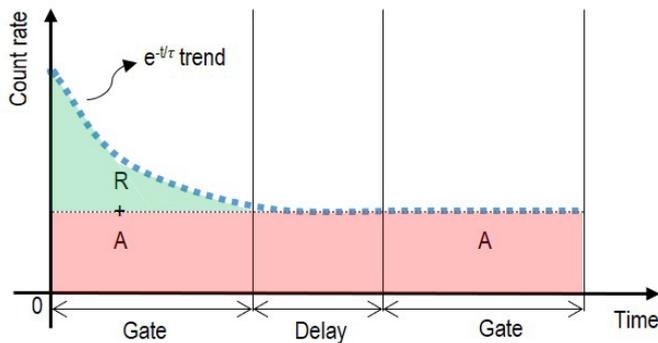


Figure 2. Rossi-alpha distribution (dashed line) of detection events. Key: R = real coincidence counts (green area), A = accidental coincidences (red area), dotted line: accidental coincidence background, τ = neutron die-away time.

The main advantage of passive NMC (i.e., NMC without an investigation source) is that it does not need calibration with a series of representative physical standards to determine the instrument response curve like NCC. Moreover, the additional precision of this technique makes it suitable for the analysis of heterogeneous Pu samples like scrap MOX material.

Since the Pu spontaneous fission yield (due to ^{238}Pu , ^{240}Pu and ^{242}Pu decays) is higher than that of U, Pu-containing samples can be investigated without the use of an external neutron source inducing fission, while for U measurements an investigation neutron source has generally to be employed. As ^{240}Pu is usually the major even isotope present in both low- and high-burnup plutonium, the plutonium mass measured with neutron assay is typically expressed as equivalent mass of ^{240}Pu ($m^{240\text{eff}}$), namely the mass of ^{240}Pu that would give the same coincidence response as that obtained from all the even isotopes in the actual sample [23]. For measurement of ^{235}U enrichment in uranium samples, an AmLi (α, n) source is typically used to induce fission in the sample because of the low average energy (around 0.5 MeV) of its neutrons, so that fast fission (e.g., on ^{238}U) is negligible. Note that an investigation source is also needed to measure ^{239}Pu mass [23].

Another option for performing neutron assay is by using liquid and plastic scintillators (or gaseous ones [28]), where fast neutrons are detected via the light nuclear recoils (mainly protons) they generate in the scintillator itself. As the

recoil energy is correlated to the initial neutron energy, this technique allows operators to perform a quantitative/spectrometric analysis of the fast neutron field. Moreover, if scintillators are equipped with thermal neutron absorber layers (containing B, Cd, Gd, Li, ...), they are able to detect (count) thermal neutrons too. The use of scintillators is more recent and less widespread than that of gaseous detectors [29].

Finally, note that fission chambers [16] are employed in cases where high neutron fluxes can overcharge the detectors, like in the measurements of spent nuclear fuel.

The most ubiquitous detectors for neutron assay based on ^3He are the neutron coincidence counter (NCC) – with or without an interrogation source – [23], the active well coincident counter (AWCC) [15], the EURATOM fast collar [30] (routinely used by the European Commission’s inspectors) and the underwater coincidence counter (UWCC) [31] (usually dedicated to Pu measurements in MOX fuel assemblies prior to irradiation). Instead, an example of detection system based on (liquid) scintillators is the fast neutron collar (FNC) [32], [33]. The instruments used for neutron counting contain a series of detectors which are typically arranged in compact geometries such as “wells” for relatively small samples or “collars” suitable for the analysis of bigger samples like PWR and BWR fuel pins/assemblies, etc. All these instruments offer compact and easy-to-use solutions for short-time on-site Pu and U measurements, relying on a large set of well-documented and internationally recognized procedures; however, they are bulky instruments often housing radioactive sources, so they are not usually customizable for measurement of “non-standard” samples. For instance, some counters are optimized only for PWR fuel assembly analysis and the presence of a radioactive source rises radiation protection issues. When dealing with massive/self-shielding samples, cadmium liners can be generally inserted in these instruments to improve the penetrability of the interrogating neutron flux in the sample: when cadmium liners are present, the instrument is said to operate in “fast mode” (e.g., the EURATOM fast collar operates in fast mode) [34].

Typically, the IAEA international target values (ITV) for the relative uncertainty of neutron assay techniques are of a few percent, but errors around 1% or less can be usually achieved by properly tuning the measurement time [22], [30], [35]. The minimum detectable mass of ^{235}U via neutron counting techniques lies commonly in the range 1 mg - 100 mg, while for plutonium the detection limit ranges roughly from 1 mg to 500 mg in terms of $m^{240\text{eff}}$, depending on both the method used and the material composition of the item analysed [23], [36], [37], [38].

c) Calorimetry: this technique is based on measuring the thermal power produced by the radioactive decay of samples placed inside containers, using calorimeters based on thermocouples and/or thermopiles. The main advantages

of the technique are that the assay is independent of the sample geometry and that it applies to a wide range of material forms. Calorimetry is mainly used to assess the total mass of plutonium inside a sample, but it requires knowledge of the plutonium isotopic mass ratios (usually given by gamma-ray spectrometry) [39], [40]. The Pu measurement via calorimetry is justified by the relatively high thermal power emitted by Pu isotopes and decay products, in particular ^{238}Pu and ^{241}Am [39], [40]. Calorimetry is the most accurate NDA technique available for plutonium mass measurements, and it has become a standard in the USA [39]. Nevertheless, it shall be noted that calorimetry measurements typically require a long time for thermal equilibrium to be reached and, moreover, a quite stable room temperature should be kept not to jeopardize the measurement performance, which makes calorimetry not suitable for industrial/workplace environments. The relative error associated with (the sole) thermal power measurement is typically below 1% [41]. It shall be noted that, due to the typical dimensions of the instrumentation used for calorimetry measurements, this technique is often restricted to small samples (< 80 L in volume). However, there exist systems for the calorimetry of samples with a volume of up to about 400 L [42]. The detection limit in terms of Pu mass generally ranges from a few tens of mg to about 10 g of Pu,

depending on the thermal power resolution of the calorimeter [42], [43].

NDA techniques are commonly divided into two categories, active and passive techniques: in the former case, one measures the radiation spontaneously emitted by the sample, in the latter case the radiation measured is induced in the sample employing external sources. Table 1 summarises the main NDA techniques discussed in this Section, while Figure 3 shows some examples of instrumentation typically employed to perform these assays.

The techniques discussed so far represent the most widespread and consolidated NDA methods used for nuclear safeguards (note that the IAEA inspectors use more than 100 different NDA systems [15]). More advanced and/or novel NDA techniques have been developed to increase the effectiveness and efficiency of nuclear safeguards. A few meaningful examples of these techniques are briefly outlined in the following lines, while a more detailed overview of these methods can be found in [15] and [34].

K-Edge Densitometry (KED) consists in irradiating a sample with highly collimated X-rays and measuring the abrupt change of the transmitted X-ray intensity in correspondence of the so-called “absorption edge” of the atomic

NDA	Category	Type	Instrument	Main applications	Advantages	Disadvantages
Gamma-ray spectrometry	Passive	Qualitative + quantitative	Spectrometers (scintillators and semiconductors)	U enrichment determination (LEU and HEU), Pu isotopic composition verification, fresh MOX fuel verification, spent fuel verification.	Possibility to precisely detect/identify nuclides, a wide range of software and codes for nuclide identification.	Complex instrumentation, need for deep operators' training, possible need to keep detectors at low temperatures, (sometimes) expensive instrumentation.
Neutron assay	Passive or active	Quantitative (and sometimes qualitative)	Gas-filled proportional counters, scintillators, fission chambers	Pu and U mass measurement at practically any enrichment and chemical/physical sample form.	Compact and easy-to-operate instrumentation, well-documented international procedures, high accuracy achievable, relatively short measurement time (< 1 h).	Bulky instrumentation, (possible) presence of a radioactive source (hence, dose to operators), (possible) need for reference field/sources for calibration, self-shielding effects in the sample can perturb measurements, fixed setup (sometimes).
Calorimetry	Passive	Quantitative	Calorimeters	Total Pu mass measurement.	Applicable to a wide range of material forms, very accurate, simple technology, relatively cheap instrumentation, no calibration source required.	Long time needed to reach thermal equilibrium and/or good statistics, need for near-constant ambient temperature, (often) adapt only for small samples, not suitable for U measurements.

Table 1. Summary of the key features of the main NDA technique used for nuclear safeguards.

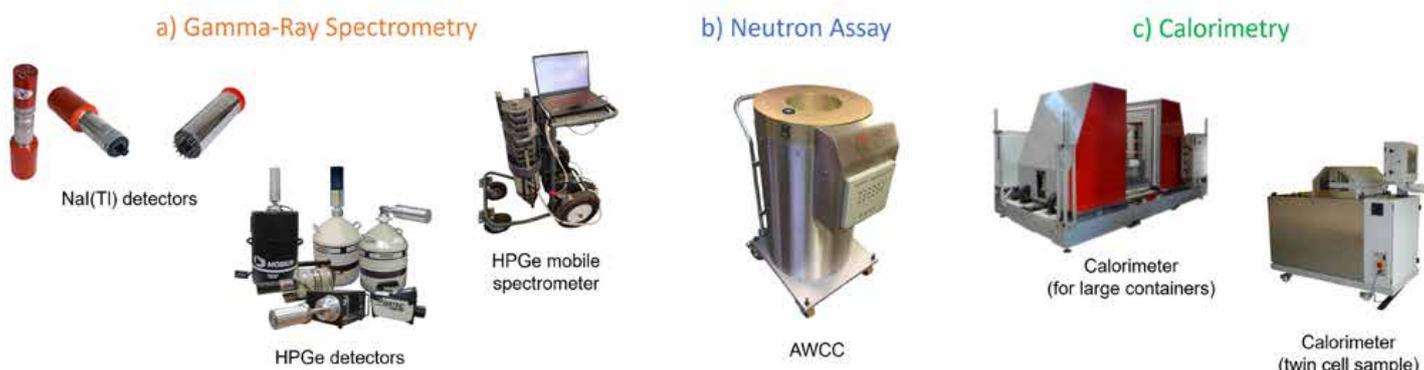


Figure 3. Examples of instrumentation used for the main types of NDA techniques (not to scale). Product Image courtesy of Mirion Technologies, Inc. (<https://www.mirion.com/>) for images a)-left. Image a)-middle courtesy of ORTEC® (<https://www.ortec-online.com/>). Images b) and c)-right courtesy of ANTECH Inc (www.antech-inc.com). Image c)-left courtesy of Setsafe (<https://setsafesolutions.com/>).

K-shell of a selected element to determine its concentration in the sample [44]. KED is generally applied to uranium or plutonium solutions with concentrations > 50 g/L, so that it is an ideal candidate for the analysis of reprocessing product solutions [22], [45]. The measurement uncertainty can be below 0.5% (depending on the counting time) [22].

Interferences can occur when elements having similar K absorption edges are present, so that corrections must be applied for samples containing both uranium and plutonium. Alternatively, when dealing with mixtures of uranium and plutonium, one can exploit Hybrid K-Edge Densitometry (HKED) [44], [45], which results from the combination of KED with X-ray fluorescence (XRF). HKED employs a single X-ray source for K-edge densitometry and fluorescence excitation. The concentration of the most abundant actinide is determined by KED, while the concentration of the other is derived from the XRF peaks. HKED is applicable to all U, Th, Pu, U-Th and U-Pu specimens containing at least 0.2 g/L of the main actinide, so that it allows the verification of reprocessing plants solutions having typical U concentrations of 150-250 g/L and Pu concentrations of 1-3 g/L [45]. An accuracy of about 0.20% for U and 0.60 % for Pu can be achieved in samples with a U/Pu ratio of about 100 [45]. Needing a fine X-ray spectrometry, both KED and HKED are normally performed with HPGe detectors [22]. Note that the accuracy of KED and HKED is better than that of high-resolution gamma spectrometry, leading to lower detection limits for the mass of nuclear materials. Another advantage is using an external, tunable investigating source instead of relying solely on the radiation emitted by the sample itself, which may be too weak to detect the source.

Moving to neutron measurements, an advanced NDA technique is the Neutron Resonance Transmission Analysis (NRTA) [46]. NRTA is based on the measurement of the intensity of neutrons that have traversed a sample as a function of the neutron energy. By so doing, it is possible to

correlate the reduction in the intensity of neutrons of a given energy to their absorption inside the sample due to the presence of specific isotopes, which are individuated and quantified. NRTA can determine the mass of all the isotopes of Am, Pu and U that are of interest for nuclear safeguards [46]. NRTA requires, upstream the sample to be investigated, a pulsed high-energy particle accelerator producing neutron bursts, and, downstream, a neutron detector (typically a ^6Li -glass scintillator coupled to a TOF (Time Of Flight) electronics for neutron spectrometry [47]). Each neutron burst comprises neutrons with a continuous energy distribution, but only neutrons with an energy in the 0.1-40 eV energy range are of interest for NRTA. NRTA has found a distinctive application in the analysis of spent fuel pin assemblies, which can be measured in parallel. Typical statistical uncertainties for NRTA are in the range 1%-4% [46]. In contrast to gamma spectroscopy, NRTA can be used to interrogate high-Z bulk material [47]. Moreover, differently from neutron counting techniques, which can detect only fissile and or fissionable nuclides, NRTA can characterize any isotope provided that it has strong and definite resonances in the energy region considered.

As a final example, a novel technique based on neutron measurement is the Differential Die-Away Self-Interrogation (DDSI) [15], [48], [49]. DDSI essentially consists in measuring fissile uranium and plutonium isotopes in spent fuel using spontaneous fission neutrons from ^{244}Cm that are present in the assembly as the interrogation source. The sensitivity of the fissile mass measurement is enhanced by measuring the sample with and without a cadmium liner between the sample and a surrounding moderator. In addition, the fertile mass can be assessed through the multiplicity analysis of the neutrons detected soon after the initial neutron pulse. Neutrons are detected using several ^3He detectors, which enhance the sensitivity of the measurement. Through DDSI, a statistical uncertainty of around 1% for an average spent fuel assembly (burnup around 40 GWd/tU

and 4% ^{235}U) can be achieved in few minutes [48]. DDSI is a powerful tool for detecting small quantities of fissile material, even if it is shielded or placed within a non-fissile matrix.

As described, NDA offers a powerful tool for supporting international safeguards; in the following section, the benefits and drawbacks of NDA for nuclear safeguards compared to other types of measurements and measures will be discussed.

As a final remark, it shall be highlighted that the border between NDA and DA becomes less clear when sampling is concerned: if an NDA technique is applied to a sample taken from an item, the same NDA technique can be considered non-destructive for the sample or destructive referring to the original item because its integrity has been compromised by taking the sample. For instance, this “destructive” sampling is needed when an item does not fit the detector dimensions and a portion of it is taken as sample and used for the selected NDA measurement. For example, HKED is classified as a non-destructive technique when it is assumed that it can be directly applied to the whole item under analysis (or when the sampling phase is neglected) [50], [51], while it is considered destructive when sampling is assumed to be required [45], and the same holds true for the other NDA techniques.

3. Discussion

In Section 2 the three main classes of NDA techniques were illustrated, discussing the specific advantages and disadvantages of each specific instrumentation type as they relate to nuclear safeguards. This section is dedicated to a discussion of the benefits and drawback of NDA in general.

Hereunder are listed the (main) benefits of NDA:

- 1) Preservation of the physical integrity of the material under analysis: non-destructive techniques offer the advantage of assessing nuclear material content without causing damage to the material itself. Hence, the physical (and chemical) state of the object investigated is not altered. This is crucial when dealing with valuable or sensitive materials, which can be used or further processed after the measurement.
- 2) Reduction of radioactive waste: NDA techniques do not generate of additional radioactive waste, for example with the preparation of a sample for analysis inside laboratories that, after the measurement becomes a waste. On the contrary, destructive techniques generally produce waste that needs to be managed and disposed of, involving radiation protection issues and, sometimes, safety/security issues.
- 3) Use at standoff distances: NDA methods are non-intrusive, namely they do not require physical contact with

the material being measured, mitigating the radiation hazard to personnel. This is particularly useful when dealing with items that are difficult to handle or manipulate directly. Moreover, this paves the way for automatization and remote controlling, which further reduces the exposure to operators.

- 4) Possibility to carry out on-site measurements: a lot of NDA techniques can be adapted for field measurements, allowing inspectors to assess the nuclear material content on-site, thus reducing the need for transportation of materials to dedicated measurement facilities, which in turn limits issues with radiation protection and/or safety and security. This way, issues related to the delay of international transfers of nuclear material which may be necessary for independent destructive analysis are avoided. In some cases, such as for the verification of spent nuclear fuel, on-site NDA is the only possible option.
- 5) Speed and cost: NDA techniques do not require sampling, which reduces the steps needed for a measurement. Not having to deal with the preparation and manipulation of a sample (generally in a dedicated laboratory) simplifies the measurement procedure and allows the user to get quickly to the result. This is particularly important in the context of nuclear safeguards, where it is crucial to timely verify the declared masses [13]. Furthermore, NDA instruments are typically cheaper than other laboratory instruments such as those used for DA [52].
- 6) Availability of methods and procedures: a great deal of well-documented, internationally recognized analysis procedures exist for NDA; modern software packages guide the user to perform the measurement from calibration, to data acquisition and analysis/interpretation (for example, the *MAESTRO*[®] software for gamma-ray spectrometry by ORTEC[®] [53]).
- 7) Possibility of cross-checks: as there are various NDA techniques available, one can use them for cross-validation on the very same sample (it is not modified by the non-destructive measurement), increasing the confidence in measurement results.

Despite the numerous advantages just presented, there are also a few specific drawbacks connected to the use of NDA, which are given below:

- 1) Limited precision: NDA methods often provide less sensitive, accurate and precise measurements (i.e., larger uncertainties) compared to destructive techniques, which can impact the accuracy of material accountancy and verification [23].
- 2) Difficulties with some sample geometry/setup: heterogeneous samples or large bulk samples can pose limits to

these techniques, for example, due to self-shielding techniques or due to a variable spatial distribution of burnable poisons. Similar problems arise with highly diluted nuclear materials. If NDA cannot guarantee to provide a measurement representative of the sample, other techniques have to be used; for instance, DA techniques are typically used for environmental sampling analysis for the detection of undeclared nuclear activities.

- 3) Sensitivity to environmental factors: NDA measurements can be performed on-site, but this makes them more dependent on environmental factors such as shielding, background radiation, and detector efficiency, which need to be carefully accounted for to achieve accurate measurements.
- 4) Need for specialized equipment and expertise: analytical methods used for nuclear safeguards, including NDA techniques, often requires sophisticated instruments (e.g., neutron sources) which may not be readily available or easily operated by non-experts. Additionally, the interpretation of data can be complex and requires skilled analysts to ensure accurate results [54].
- 5) Limited material identification: some NDAs can quantify the amount of nuclear material (quantitative techniques), but are not able to identify the specific isotopes present in the material. For this purpose, destructive techniques like mass spectrometry or radiochemical analysis can be employed. However, there are NDA techniques capable of directly quantifying specific isotopes in an assay, such as the traditional gamma-ray spectrometry already discussed in the previous section or more exotic techniques like nuclear resonance fluorescence (NRF) [55].
- 6) Relatively high detection limits: NDA techniques have detection limits that are generally higher than those of DA, meaning that they might not be suitable for measuring very low quantities of nuclear material or detecting highly diluted material or low-energy radiation. For the aforementioned cases, a destructive analysis is needed (for example, dissolving the sample into a liquid scintillator after a proper chemical treatment). For example, the limit of detection of NDA for fissile nuclides is typically in the range of micrograms to some milligrams (per gram of sample material), while the most advanced DA techniques like ion mass spectrometry can detect down to tens of picograms of the nuclide of interest [14], [34], [43], [56], [57], [58], [59].
- 7) Tampering risk: because NDA measurements are non-intrusive, there is a risk of tampering or attempts to deceive inspectors by altering the exterior of the material without affecting the nuclear content.

Pros	Cons
<ul style="list-style-type: none"> • Preservation of material integrity; • Reduction of radioactive waste and personnel's exposure; • Wide margins for automatization and remote control; • Possibility of on-site measurements; • Rapid and (relatively) cheap methods; • Wide international expertise available; • Possibility of cross-checks with different NDAs on the same sample. 	<ul style="list-style-type: none"> • Higher detection limits and uncertainty compared to DA; • Not applicable to weak radioactive sources (e.g., highly diluted radionuclides); • Sensitivity to environmental factors; • Need for specialized equipment and trained operators; • Limited material identifications with certain NDA (e.g., calorimetry); • Tampering risk.

Table 2. Summary of the main advantages and disadvantages of NDA.

4. Conclusion

NDA techniques are crucial to assess and verify the nuclear material content in various items, such as nuclear fuel, waste, or other materials, without altering the sample integrity.

NDA offers several advantages, but it also has a series of drawbacks. Despite NDA being generally less precise than DA, the precision of NDA methods has improved significantly over the years owing to advancements in detector technology, calibration techniques, and data analysis methods. Nevertheless, while NDA methods may have inherent limitations in achieving the same level of precision as destructive assays, they offer the main advantage of preserving the physical integrity of the materials investigated and of allowing on-site field measurements, which is crucial in many practical scenarios. Overall, NDA techniques stand out as valuable tools in nuclear safeguards, but their successful implementation depends on proper instrumentation, skilled analysts, rigorous calibration procedures, and careful consideration of potential uncertainties and errors in the measurements.

In conclusion, a combination of NDA and DA techniques, besides the use of complementary safeguard strategies like containment and surveillance, nuclear forensics and satellite imagery, is the best solution for nuclear safeguards purposes, to maximize the benefits and overcome the limitations of each approach allowing for a more comprehensive assessment of nuclear material content and safeguards effectiveness [60]. The fact that an NDA technique can be applied to a sample taken “destructively” from the item to be analysed is just another example of how non-destructive and destructive operations can be combined for the analysis of nuclear materials.

Table 2 offers a visual recap of what has been illustrated.

References

- [1] IAEA (1970). Treaty on the Non-Proliferation of Nuclear Weapons. Information Circular, INFCIRC/140, IAEA.
- [2] IAEA (2016). IAEA Safeguards – Delivering Effective Nuclear Verification for World Peace. Leaflet 16-17011.
- [3] Howlett, D. A. (1990). EURATOM and Nuclear Safeguards. London, UK: Palgrave Macmillan.
- [4] European Commission (2012). Consolidated version of the Treaty establishing the European Atomic Energy Community. EU Treaty, EUR-Lex 2012/C 327/01.
- [5] European Commission Directorate-General Energy (DG ENER) official website, <https://commission.europa.eu/about-european-commission/departments-and-executive-agencies/energy>
- [6] European Commission – Joint Research Centre (EC JRC) official website, <https://joint-research-centre.ec.europa.eu>
- [7] European Commission, Directorate-General for Research and Innovation (2012). Euratom research and training programme 2021-2025. EU Publications Office.
- [8] European Commission, ESARDA official webpage, <https://esarda.jrc.ec.europa.eu>
- [9] Wilson, R. (1991). Nuclear Proliferation and the Case of Iraq. *Journal of Palestine Studies*, 20(3), pp. 5-15.
- [10] IAEA (1997). Model Protocol Additional to the Agreement(s) between State(s) and the International Atomic Energy Agency for the Application of Safeguards. Information Circular, INFCIRC/540, IAEA (Vienna).
- [11] IAEA (2022). Safeguards. IAEA Bulletin Vol 63/3.
- [12] United States Nuclear Regulatory Commission (USNRC) (2023). USNRC webpage about Special Nuclear Material, <https://www.nrc.gov/materials/types/sp-nuc-materials.html>
- [13] M. Hua, Goddard, B., Lloyd, C., Leppink, E. Abraham, S. A., Noey, J., Clarke, S. D., Pozzi, S. A. (2019). Simulation of the Nondestructive Assay of ²³⁷Np Using Active Neutron Multiplicity Counting. *Nuclear Science and Engineering*, 194(2), pp. 154-162.
- [14] Gavron, A. (2001). Non destructive assay techniques applied to nuclear materials. ICONE 9: Ninth international conference on nuclear engineering, Nice Acropolis (France). France: Société Française d'Énergie Nucléaire.
- [15] IAEA (2011). International Nuclear Verification Series No. 1 (Rev. 2) - Safeguards Techniques and Equipment: 2011 edition. Vienna, Austria: IAEA.
- [16] Knoll, G. F. (2012). Radiation Detection and Measurement - 4th edition, USA: John Wiley and Sons.
- [17] Burr T., Hamada, M. (2009). Radio-Isotope Identification Algorithms for NaI γ Spectra. *Algorithms*, 2, pp. 339-360.
- [18] Mayorov, M., White, T. A., Lebrun, A., Brutscher, J., Keubler, J., Birnbaum, A., Ivanov, V., Honkamaa, T., Peura, P., Dahlberg, J. (2017). Gamma Emission Tomography for the Inspection of Spent Nuclear Fuel. IEEE Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC), Atlanta, Georgia (US). US: IEEE.
- [19] Jawerth, N. (2016). Revealing Facts Through Science for Nuclear Verification. Article on IAEA official website, <https://www.iaea.org/newscenter/news/auditors-lab-coats-revealing-facts-through-science-nuclear-verification>
- [20] European Commission, EU Science Hub – Training programmes webpage, https://joint-research-centre.ec.europa.eu/tools-and-laboratories/training-programmes_en
- [21] Vidmar, T., Aubineau-Laniece, I., Anagnostakis, M., Arnold, D., Brettner-Messler, R., Budjas, D., Capogni, M., Dias, M., De Geer, L., Fazio, A., Gasparro, J., Hult, M., Hurtado Bermudez, S., Jurado Vargas, M., Laubenstein, M., Lee, K., Lee, Y., Lepy, M., Maringer, F., Medina Peyres, V., Mille, M., Morales, M., Nour, S., Plenteda, R., Rubio Montero, M., Sima, O., Tomei, C., Vidmar, G. (2008). An intercomparison of Monte Carlo codes used in gamma-ray spectrometry. *Applied Radiation and Isotopes* 66(6-7), pp. 764-768.
- [22] IAEA, (2022). International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials, STR-368 (Rev 1.1). Vienna, Austria: IAEA.
- [23] Reilly, D., Ensslin, N., Smith, H. Jr., Kreiner, S. (1991). Passive nondestructive assay of nuclear materials. US: U.S. Nuclear Regulatory Commission.
- [24] Choi, H.-D., Kim, J. (2018). Basic characterization of uranium by high-resolution gamma spectroscopy. *Nuclear Engineering and Technology*, 50(6), pp. 929-936.
- [25] Jansson, P. (2002). Studies of Nuclear Fuel utilizing Nuclear Spectroscopic Methods. Uppsala, Sweden: Acta Universitatis Upsaliensis.
- [26] IAEA (2008). Handbook of Nuclear Data for Safeguards: Database Extensions. Vienna, Austria: IAEA.

- [27] Ensslin, N., Harker, W. C., Krick, M. S., Langner, D. C., Pickrell, M. M., Stewart, J. E. (1998). Application Guide to Neutron Multiplicity Counting. Los Alamos National Laboratory, Report LA-13422-M.
- [28] Lewis, J. M., Kelley, R. P., Murer, D., Jordan, K. A. (2014). Fission signal detection using helium-4 gas fast neutron scintillation detectors: theory and experiment, *Appl. Phys. Lett.*, 105.
- [29] Darby, F., Hua, M., Pakari, O., Clarke, S., Pozzi, S. (2023). Multiplicity counting using organic scintillators to distinguish neutron sources: An advanced teaching laboratory. *Am. J. Phys.* 91, pp. 936-945.
- [30] Clapham, M., Farese, N., Jaime, R., Lezzoli, L. Schillebeeckx, P. (1997). Performance comparison of different Active Neutron Interrogation Techniques for Safeguards Applications. Proceedings of the 19th ESARDA Symposium on Safeguards and Nuclear Material Management, Montpellier (France). Luxembourg: Publications Office of the EU.
- [31] Eccleston, G. W., Menlove, H. O., Abhold, M., Baker, M., Pecos, J. (1998). The underwater coincidence counter (UWCC) for plutonium measurements in mixed oxide fuels. Los Alamos National Laboratory, Report LA-UR-98-3303.
- [32] Swinhoe, M. T., Menlove, H.O., de Baere, P., Lodi, D., Schwalbach, P., Rael, C. D., Root, M., Tomanin, A., Favalli, A. (2021). A new generation of uranium coincidence fast neutron collars for assay of LWR fresh fuel assemblies. *Nucl. Instr. And Meth. A*, 1009.
- [33] Beaumont, J. S., Lee, T. H., Mayorov, M., Tintori, C., Rogo, F., Angelucci, B., Corbo, M. (2017). A fast-neutron coincidence collar using liquid scintillators for fresh fuel verification. *J. Rad. Nucl. Chem*, pp.803-812.
- [34] Abbas, K., Peerani, P., et al. (2023). Nuclear Safeguards and Non-Proliferation – ESARDA Course Syllabus. Luxembourg: Publications Office of the EU.
- [35] Tomanin, A., Bourva, L., Beaumont, J., De Baere, P., Terrasi, A., De pooter, V., Arias Arenas, M. (2019). Inter-Comparison Exercise for the Safeguards Verification of PWR Fresh Fuel Assemblies using Fast and Thermal Neutron Coincidence. Proc. 41st ESARDA 2019 conference, Stresa (Italy). Luxembourg: Publications Office of the EU.
- [36] Lousteau, A. L. (2017). Determining 235U Enrichment Using a Dual-Energy Approach for Delayed Neutron Measurements. PhD Diss., University of Tennessee (USA).
- [37] Parker, J. L., Beddingfield, D. H., Menlove, H.O. (1999). Low-Activity Solid Waste Measurements at Tokai Works. Los Alamos, Nex Mexico (US): Los Alamos National Laboratory.
- [38] De Felice, P., Bogucarska, T., Raiola, F., Pedersen, B. (2021). Good Practice Guide for Validation of a Waste Characterisation System for Very Low, Low and Intermediate Level Radioactive Waste. Luxembourg: Publications Office of the EU.
- [39] ASTM International (2009). ASTM-Standard C 1458-09: Standard Test Method for Nondestructive Assay of Plutonium, Tritium and ²⁴¹Am by Calorimetric Assay. West Conshohocken, Pennsylvania: ASTM International.
- [40] Bracken, D. S., Biddle, R. S., Carrillo, L. A., Hypes, P. A., Rudy, C. R., Schneider, C. M., Smith, M. K. (2002). Application Guide to Safeguards Calorimetry. Los Alamos National Laboratory, Report LA-13867-M.
- [41] IAEA, (2010). International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials, STR-368. Vienna, Austria: IAEA.
- [42] Setsafe – KEP Technologies. The Heat Check Line - Nuclear measurement. Data Sheet, 01/21, <https://set-safesolutions.com/app/uploads/sites/4/2021/03/HEAT-CHECK-LINE-ENG.pdf>
- [43] Tagziria, H., Bagi, J., Pedersen, B., Schillebeeckx, P. (2012). Absolute determination of small samples of Pu and Am by calorimetry. *Nucl. Instr. And Meth. A*, 691, pp. 90-96.
- [44] Ottmar, H., Eberle, H., Matussek, P., Michel-Piper, I. (1986). How to Simplify the Analytics for Input-Output Accountability Measurements in a Reprocessing Plant. Kernforschungszentrum Karlsruhe, Report 4012.
- [45] Balsley, S.D. (2010). Destructive nuclear material analysis for safeguards: Importance and future trends. IAEA symposium on international safeguards, Vienna, 2010. Vienna, Austria: IAEA.
- [46] Sterbentz, J. W., Chichester, D. L. (2010). Neutron Resonance Transmission Analysis (NRTA): A Nondestructive Assay Technique for the Next Generation Safeguards Initiative's Plutonium Assay Challenge. Idaho National Laboratory, Report INL/EXT-10-20620.
- [47] Klein, E. A., Naqvi, F., Bickus, J. E., Lee, H. Y., Danagoulian, A., Goldston, R. J. (2021). Neutron-Resonance Transmission Analysis with a Compact Deuterium-Tritium Neutron Generator. *Physical Review Applied*, 15.

- [48] Schear, M. A., Menlove, H. O., Tobin, S. J., Evans, L. G., Croft, S. (2011). Development of the Differential Die-Away Self-Interrogations Technique for Spent Fuel Characterization. Los Alamos National Laboratory, Report LA-UR-11-04130.
- [49] Trahan, A. C., McMath, G.E., Mendoza, P. M., Trelue, H. R., Backstrom, U., Pöder Balkeståhl, L., Grape, S., Henzl, V., Leyba, D., Root, M. A., Sjolund, A. (2020). Results of the Swedish spent fuel measurement field trials with the Differential Die-Away Self-Interrogation Instrument. Nucl. Instr. And Meth. A, 955.
- [50] Mirion Technologies, Inc. (2011). HKED™, Hybrid K-Edge Densitometry Software. Data Sheet C38779 – 04/2011.
- [51] Shizuma, T., Hajima, R., Hayakawa, T., Fujiwara, M., Sonoda, T., Seya, M. (2011). Proposal for an advanced hybrid K-edge/XRF densitometry (HKED) using a monochromatic photon beam from laser Compton scattering. Nucl. Instr. And Meth. A, 654(1), pp. 597-603.
- [52] Tsoulfanidis, N. (Editor) (2012). Nuclear Energy - Selected Entries from the Encyclopedia of Sustainability Science and Technology. New York (US): Springer.
- [53] ORTEC® (2012). MAESTRO® - Software User's Manual, Software Version 7.0, 2012.
- [54] Boulyga, S., Konegger-Kappel, S., Richter, S., Sangély, L. (2015). Mass spectrometric analysis for nuclear safeguards. Journal of Analytical Atomic Spectrometry, 30(7), pp. 1469-1489.
- [55] Ludewigt, B. A., Quiter, B. J., Ambers, S. D. (2011). Nuclear resonance fluorescence for safeguards applications. Lawrence Berkeley National Laboratory, Report LBNL-4350E.
- [56] Forbes, T. P., Szakal, C. (2018). Considerations for uranium isotope ratio analysis by atmospheric pressure ionization mass spectrometry. Analyst, 144(1), pp. 317-323.
- [57] Ramebäck, H., Vesterlund, A., Hedberg, M., Jonsson, S., Lagerkvist, P., Vidmar, T. (2023). Gamma spectrometric measurement of uranium isotopic composition and mass in sintered UO₂ pellets using the efficiency transfer method. Applied Radiation and Isotopes, 192.
- [58] Andrews, M.T., Goorley, J. T., Corcoran, E. C., Kelly, D. G. (2015). MCNP6 simulations of gamma line emissions from fission products and their comparisons to plutonium and uranium measurements. Progress in Nuclear Energy, 79, pp. 87-95.
- [59] European Commission - Joint Research Centre, European Safeguards Research and Development Association (2012). ESARDA Bulletin no. 48. Belgium: Publications Office of the EU.
- [60] Welsh, T. L., McRae, L. P., Delegard, C. H., Liebetrau, A. M., Johnson, W. C., Krick, M. S., Stewart, J. E., Theis, W., Lemaire, R. J., Xiao, J. (1995). Comparison of NDA and DA Measurement Techniques for Excess Pu Powders at the Hanford Site: Operator and IAEA Experience. Proceedings of the 36th annual meeting of the Institute for Nuclear Materials Management (INMM), Palm Desert, CA (US). US: INMM.