Nuclear and Atomic Data Needs for Safeguards

Chris A. Pickett[1, 2], **Christopher Ramos**[2], **Fredrik Tovesson**[2], **Karen Ventura**[2, 3]

[1] Oak Ridge National Laboratory
[2] National Nuclear Security Administration
[3]Pacific Northwest National Laboratory
pickettca@ornl.gov
Christopher.ramos@nnsa.doe.gov
Fredrik.Tovesson@nnsa.doe.gov
Karen.ventura@nnsa.doe.gov

Abstract

The International and Domestic Nuclear Safeguards programs are foundationally based on obtaining accurate and precise nuclear material measurements with quantifiable uncertainties. These programs employ several types of both non-destructive assay (NDA) and destructive analysis (DA) methods to measure material quantity, chemistry, and physical properties. Each measurement method has the goal of meeting the International Atomic Energy Agency (IAEA) established international target values (ITVs); which are based on the utilized measurement methods. Common NDA methods used for domestic and international Safeguards are; passive gamma spectroscopy, passive and active neutron correlation counting, nuclear calorimetry, and X-ray fluorescence techniques. The most common DA methods are mass spectrometry and Davis-Gray titration. All of the commonly used NDA and DA methods have dependence on atomic and nuclear data. These data typically exist in measurement codes (embedded software) that are either supplied by a commercial entity or government laboratory. However, it is not always clear what nuclear and atomic data sources are being utilized by these commercial and lab developed codes. Without this information or adequate calibration standards, accurate quantification of NDA and DA measurement uncertainties cannot be properly determined. In this paper, we highlight on-going efforts by the Defense Nuclear Nonproliferation Research and Development (DNN R&D) program that focus on determining nuclear and atomic data most pertinent for global Safeguards and other Nonproliferation missions. We also provide status on recent efforts to improve the quality of nuclear and atomic data used for NDA and DA measurements of special nuclear materials (SNMs).

Keywords: International Target Values, Safeguards, Nuclear Data, Atomic Data, NDA, DA

1. Introduction

Destructive and non-destructive measurement methods used to characterize and quantify special nuclear material (SNM) are foundational to all Domestic and International Safeguards programs. Additionally, they are utilized by many global nuclear safety and security programs. The primary safeguards objective for these measurements methods is to determine the completeness and correctness of a facility or state declared SNM inventory. The underlying codes (software) associated with these measurement methods are reliant on the quality of nuclear and atomic data being utilized to determine elemental and isotopic quantities. In many cases (for nuclear data being used today), the original source of the data and its associated uncertainties are unknown. Without this information, safeguards measurement methods are limited in their ability to accurately quantify measurement uncertainty; or in other words in their ability to effectively verify the completeness and correctness of a safeguards declaration.

Safeguards practices require excellence in the performance of **nuclear material accountancy** (NMA) and **containment/surveillance** (C/S), domestically referred to as material control. Material accountancy deals with measurements that determine the material type and the material amounts. Containment and Surveillance deals with **protecting the integrity of measured values** and works to maintain chain of custody of the SNM as it is processed, stored, and transported. Built on a foundation of effective NMA and C/S, these measures provide confidence and Continuity of Knowledge (CoK) that supports overall safeguards conclusions (see Figure1). Our ability to obtain quality measurements and maintain the integrity of these measured values is necessary for drawing meaningful safeguards conclusions!

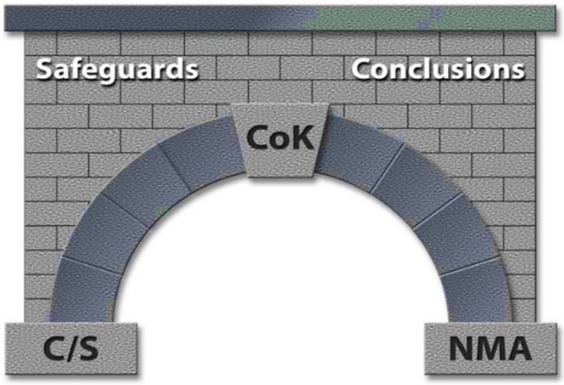

Figure 1: CoK is the Keystone for Drawing Safeguards Conclusions that relies on Effective Nuclear Material Measurements and Containment & surveillance

2. Deriving the Material Balance equation

Safeguards monitoring is summarized as a need to understand a particular process and identify key measurement points. Every process contains an input and an output (see Figure 2). In Safeguards, we must characterize both, taking into account that in some cases the understanding of the process to get from the input to the output is necessary. Consequently, the material balance equation becomes fundamental for determining and verifying nuclear material inventories.

A process in safeguards is anything that has inputs and outputs. We define Material Balance Areas or MBAs as distinct geographical areas where inventories can be performed (meaning where inputs and outputs can be measured and tracked). MBAs are typically actual physical processes, sub-processes, storage areas, shipments, etc. MBAs can be as large as an entire facility or as small as a source storage cabinet.

Figure 2: Simple Nuclear Material Process Model

Figure 2 illustrates a simple process. The material enters, leaves, and/or may remain in a process. If <u>all</u> nuclear material that enters the process leaves the process, then:

Inputs = Outputs (ideal case)

When some material remains in the process from previous processing, then the inventory at a specific point in time will be:

Inputs = Outputs + Ending Inventory (EI) (EI is the material left in the process)

Thus, for the very first inventory period (beginning inventory n = 0):

$$0 = Inputs - Outputs - EI$$

For a second inventory at a point in time, the ending inventory of the first period becomes the beginning inventory (BI) of the second period, i.e. $BI_2 = EI_1$:

$$0 = \text{Beginning Inventory}_2 + \text{Inputs}_2 - \text{Outputs}_2 - \text{Ending Inventory}_2$$

$$0 = BI_2 + I_2 - O_2 - EI_2$$

Which leads to:

$$0 = BI_n + I_n - O_n - EI_n$$

or

$$0 = EI_{n-1} + I_n - O_n - EI_n$$

Where n is the n^{th} inventory period.

However, in nearly all nuclear material processes, each term is subject to an unknown uncertainty. Therefore, we define the Inventory Difference (ID) or Material Unaccounted for (MUF) as:

$$ID = MUF = BI + I - O - EI \qquad \text{(Material Balance Equation)}$$

Note: Sometimes Inputs (I) and Outputs (O) are referred to as Additions (A) and Removals (R)

Unfortunately, most material balances at nuclear processing facilities are non-trivial. In these facilities, materials can change physical form and/or chemical composition as they are processed. These processes also result in process losses that cannot be recovered for measurement (see Figure 3).

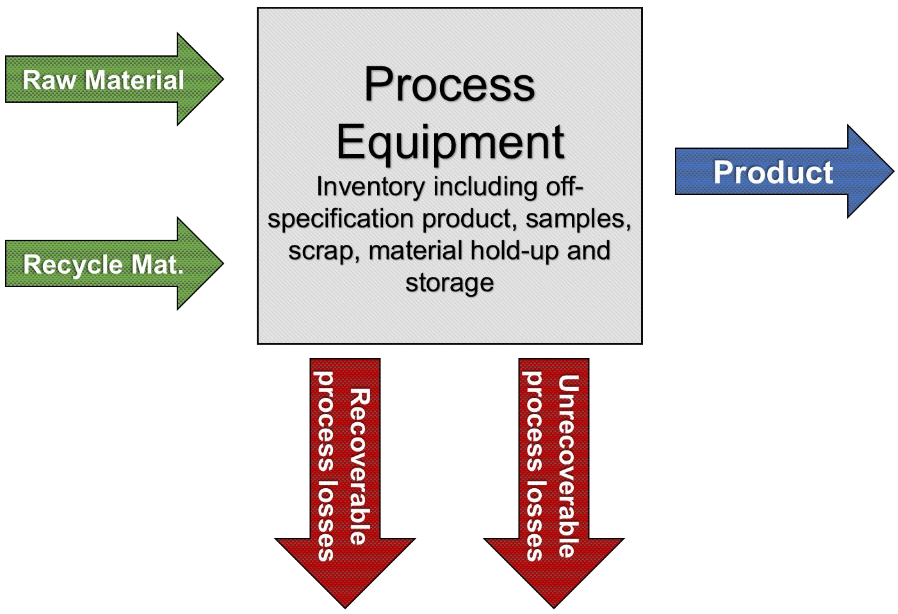

Figure 3: Representative Nuclear Material Process Model

Our ability to effectively determine IDs or MUFs is fundamental for obtaining meaningful safeguards conclusions. Examples of event that contribute to MUFs are:

- Errors in the inventory
- Errors in the inventory process
- Process upset
- Human errors

- Measurement uncertainty
- Incorrect adjustments
- Unmeasured losses
- Theft/Diversion.

Even when all sources of contribution to MUF are minimized, there still are factors that must be considered when drawing safeguards conclusions. These include:

- **Prior knowledge:** (What is the quality of past information? What do we know and what we not know?)
- **Technical capabilities:** (How good are the initial tools and methods versus the current tools and methods?)
- **Time** (When was the information acquired? Could the material have changed since it was last measured?)
- **Ability to monitor** (When and where can measurements be taken?)

Since there are many contributors to MUFs it is very important that we ensure that our measurement uncertainties can be sufficiently quantified. When MUFs start to equal significant quantities of nuclear material, the effectiveness of Safeguard measures for detecting material theft or diversion will be questioned. Some of the previously mentioned factors can be mitigated with an effective measurement control program, documentation of calibration methods, standards, and use of meta-data (i.e., operational records, etc.), and maintaining effective C/S.

Next, we will discuss the DA and NDA measurement methods used for safeguards and the underlying nuclear and atomic data that it relies on to accurately determine elemental and isotopic quantities of SNM.

3. Destructive Analysis Methods Used for Safeguards

Destructive analyses are methods that require obtaining a physical sample of the item for analyses. The obtained sample is typically “destroyed” as part of these analyses. The advantages of DA methods are high precision and accuracy. They are useful for the characterization of standards and allow for total analysis (providing information on other actinides of interest). On the other hand, the disadvantages of these techniques are the removal of material from the process, they are labour intensive, time consuming, subject to sampling errors, and they typically generate chemical, radiological and mixed wastes.

In 2010, the International Atomic Energy Agency (IAEA) STR-368 established International Target Values (ITVs) for DA methodologies used for Uranium and Plutonium [1]. The ITVs estimate the capability that could reasonably and realistically be expected from industrial-type laboratories on a routine basis.

Example of common DA techniques include:

- **Gravimetric Analysis**: Where uranium tetrafluoride (UF_4) is converted to uranium oxide (U_3O_8) using pyrohydrolysis in a furnace at 850 °C
- **Davies-Gray Uranium Titration**: Chemical titration where Uranium (U) is reduced to U^{IV} then titrated to U^{VI}
- **Coulometric Determination of Plutonium: Electrochemical** “titration” where Plutonium (Pu) is oxidized from Pu^{III} to Pu^{IV}
- **Mass Spectrometry**: Thermal ionization mass spectrometry (TIMS) and isotope dilution mass spectrometry (IDMS) (These measurements are not absolute. The measurements are relative to an external (standard bracketing) or internal (isotope dilution) standard)
- **X-ray Fluorescence**: Can be used to quantify U in materials; may be considered an NDA method as well depending on how the sample is prepared.

It is important to mention that the uncertainties for most of the DA methods are limited to the instrumental measurement uncertainty and availability of appropriate reference materials. In addition, DA methods depend on nuclear and atomic data, such as, atomic masses, half-lives, etc. One of the major safeguards needs in the area of destructive analysis is the ability to age date nuclear materials.

3.1 Nuclear Data Needs for Destructive Analyses

Atomic masses are used in a variety of techniques such as mass spectrometry calculations. The uncertainty of these values is so small that it is usually neglected. Most of the time these well-known values are treated as constants. However, there are ongoing discussions whether to continue to disregard these uncertainties.

Half-lives are very important when accounting for material. The short Half-lives of ^{241}Am, ^{238}Pu and ^{241}Pu have uncertainties significant enough to have non-trivial effects on accountancy. A near-term goal for the safeguards community is to have consistent values of half-lives among national laboratories. In addition, a literature review of the current state of use of such values could help us better understand the extent of the problem. It is also important to mention that ^{241}Pu has a string effect on measurements because it is used as a reference material. Some of the primary or most specific needs for improved nuclear data for destructive analyses methods involve:

- **Consensus/improved half-lives for ^{229}Th, ^{230}Th**. Improvements in these half-lives would benefit age dating of Uranium materials
- For Pu determinations, IDMS and TIMS isotopic methods can benefit from **consensus/improved half-lives for ^{238}Pu, ^{241}Pu, ^{241}Am**
- Age dating of Pu materials are dependent on how well we know the **half-lives of ^{241}Pu and ^{241}Am**

Some other important points to take into consideration when improving nuclear data to support DA methods are:
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- Improve data for ^{241}Am, ^{234}U and ^{230}Th. Special attention should be given to ^{229}Th, since discussion about the value of its half-life still exists and it is currently being used as an IDMS tracer for age dating of uranium materials.
- The half-life of ^{241}Pu has direct impact in accountancy when propagating a measurement through time.
- A study on burn-up effects on fission product is needed. The yield of these products can be used for quantifying the number of fissions in a sample

4. Nuclear and Atomic Data Needs for Nondestructive Data Analysis (NDA) Measurements and Metrology in Nuclear Safeguards

International Nuclear Safeguards and Nuclear Material Accounting and Control (NMAC) rely on accurate and quantifiable physical inventory measurements. For instance, the commercial nuclear fuel cycle requires the verification of material in a variety of forms under a diverse set of measurement conditions. To perform these measurements the nuclear safeguards inspectorates employ a wide suite of non-destructive assay techniques and instruments. NDA methods based on passive gamma spectroscopy, passive and active neutron correlation counting, nuclear calorimetry, and x-ray fluorescence techniques are the most widely established, but a number of techniques are under development to meet emerging difficult to measure items and material flows.

A common feature for all NDA techniques is their dependence from implementation to analysis and interpretation on atomic and nuclear data. A physical model supported by data may be used to justify technique selection. Scientific design and optimization of measurement systems using forward prediction models rest on the quality of physical data. Basic data is often needed to support the characterization and calibration of instruments. During NDA analyses, correction factors and interference corrections typically require basic data as does the inversion of the measurement data collected into quantitative assay results and the interpretation of the nuclear material source term. Historically, nuclear safeguards measurement applications have relied on atomic and nuclear data that was evaluated for purposes other than safeguards. The uncertainties in this nuclear and atomic data are often the limiting factors in the overall uncertainties achievable with an NDA technique. For accurate uncertainty quantification (UQ), it is also important to evaluate co-variance data, but unfortunately this is rarely done.

Minimizing systematic uncertainties due to nuclear and atomic data would improve the accuracies that are achieved by the NDA instruments. Efforts in this area, will drive the revision of ITVs [see e.g. STR-368 (2010), ESARDA Bulletin 48 (2012)], resulting in better measurements. The ITVs reflect the current state of practice, given the knowledge of the uncertainties. We are obligated to develop and utilize the best metrological practices.

The following sections provide details on the current status of different types of nuclear data that is utilized by common NDA measurement methods [2].

4.1 Status of Existing Nuclear Data Uncertainties: Fission yields

- Fission Yields: Accurate estimation of neutron absorbing fission products is vital.
  - Build-up of neutron absorbing fission products reduces the net neutron population inside and escaping from the source and, therefore, the count rate measured by an NDA instrument.
  - ORIGEN estimations of fission products such as ^{133}Cs, ^{143}Nd, ^{149}Sm, ^{154}Eu are within a few % of experimental values.
  - Absorption cross sections of some of the fission products (^{155}Gd) have relatively large uncertainties (~5.3%).
  - Calculated/Experimental ratios for ^{109}Ag, ^{106}Rh, and ^{125}Sb: 170%, 67%, and 100%, respectively.
  - Inconsistencies have been observed with respect to quoted uncertainties on legacy nuclear fission yield data on noble gas fission products; (e.g. ^{85}Kr)
  - ^{244}Cm is the dominant source of spontaneous fission neutrons as well as delayed neutrons from spent fuel: the nuclear data uncertainties are relatively high (8%).

4.2 Status of Nuclear Data Uncertainties – Actinide Reaction Cross-sections

- High-fidelity covariance matrices for evaluated ENDF/B-VII files are available for three major actinides, $^{235,\,238}$U and ^{239}Pu [3]
- Covariance matrix evaluations for all major reaction cross sections are available- total, capture, fission, elastic, total inelastic, and (n,xn). [3]
  - Need: Angular distribution, uncertainties for discrete inelastic reaction cross sections
- Fission cross sections: Important for source term definition and interpretation of the response of active and passive neutron NDA system measurements for safeguards (e.g. Active Well Coincidence Counter, Neutron Coincidence Collar).
- Neutron-induced fission cross section of ^{235}U was evaluated by the IAEA Standards Group [4],
  - ENDF/B-VII.0 evaluation incorporates their findings without modification, **including the associated covariance matrix for this reaction**
  - UQ for the neutron-induced fission cross-section of ^{235}U is of major importance as most other actinide fission cross-section uncertainties are driven by it.
- Evaluation by the IAEA Standards Group is the result of major efforts from experts in the domain.

5. The Type of Measurements Needed for (α, n) Reaction Data for Safeguards Science

Neutron emissions include spontaneous fission, induced fission, and (α, n) reactions. Nuclear data related to neutron emission are vital in accurate definition of the source term and the detector response. UF_6 is the most abundant material in the fuel cycle, and one way that the material production is verified is by neutron counting highlighting the importance of the (α, n) reactions. A 10% uncertainty in (α,n) cross sections measurements can represent several significant quantities (SQs) of uncertainty in the MUF at the quantities of material handled in industrial facilities.

5.1 Sealed sources

Sealed neutron source measurements are convenient, since sources can be transferred to other centres for independent measurement, as part of inter-comparison exercises, or to use and calibrate different kinds of instruments. Once source measurements are taken they can be used for several years (working life in excess of 20 years, although the radiolysis of high activity sources and helium gas build up must be considered). They are readily accessible for laboratory experiments as routine quality control items. These source measurements can be set up to validate new spectroscopy and yield measurement methods that might be developed. They can be certified for absolute neutron emission using the $MnSO_4$-bath technique and provide an important link to national standards. Sources of various types can be used to validate the energy dependence of detectors to realistic (α,n) spectra. By developing an (α,n) yield and spectrum measurement program, sealed sources can play an essential role as well as providing important benchmark/integral/normalization data in their own right.

5.2 Accelerator measurements

Thick target (α-particles stop in the material) integrated-over-angle (TT IOA) yield measurements in steady state using flat a well characterized energy 4π are needed for the elements mentioned. A variety of target materials should be measured to check consistency and scaling rules. Using the same instrumentation is recommended to avoid bias. Target degradation under bombardment should be included as part of the experimental evaluation

6. Nuclear Data Needs for NDA Methods for Safeguards Applications

Accurate knowledge of gamma ray energies, half-life, and gamma ray yields are extremely important for identifying and quantifying radionuclides of interest for non-proliferation applications. NDA instruments based on gamma spectrometry, and analysis software depend on gamma ray related nuclear data and their associated uncertainties. Nuclear data related to gamma ray emission are used in libraries for nuclide identification in gamma ray analysis, and activity or mass determination.

Branching ratios of gamma-rays emitted by uranium, plutonium, and other actinide isotopes are needed with greater accuracies so that the uncertainties in the isotopic analyses can be driven down. Similarly, atomic data such as mass attenuation coefficients of actinide elements and X-ray yield data have large uncertainties. These limit the accuracy of U and Pu elemental concentration results that are of importance to nuclear safeguards.

A Workshop for Applied Nuclear Data Activities (WANDA) was held at the Elliot School of International Affairs at George Washington University in January 2019 [5]. The purpose of WANDA was to bring subject matter experts from the national laboratories, universities and industry together with government program managers and their advisors to develop collaborative plans of action (e.g., roadmaps) to address outstanding issues in nuclear data that affect applications in nuclear non-proliferation. During a brainstorming session focussed on NDA measurements used in Safeguards, the following comprehensive list of nuclear data needs were obtained:

- Knowledge of nuclear and atomic data can become the limiting factors in design and calibration of NDA systems and physics-based modelling of responses from NDA systems used in safeguards applications.
- The neutron yields from (α, n) reaction on low Z nuclides (for example the F(α, n) reaction, important in the safeguards of the enrichment of uranium) are not well known. [6]
- Uncertainty quantification, taking into account co-variances, is needed for cross-section (fission and other reactions) data in the evaluated nuclear data libraries.
- Relative abundances of delayed neutron groups available in the literature have large uncertainties.
- Branching ratios of gamma-rays emitted by uranium, plutonium, and other actinide isotopes are needed with greater accuracies so that the uncertainties in the isotopic analyses can be improved.
- Atomic data such as interaction cross sections and X-ray yield data have large uncertainties, which limit the accuracy of U and Pu elemental concentration results that are of importance to nuclear safeguards.

- It is a high priority to have improved (α,n) cross sections on low-Z material (e.g. Fluorine, Oxygen, Nitrogen) for uranium enrichment measurements.
  - Improved stopping power measurements are needed for alphas in these materials for alpha energies of ~10 MeV down to threshold of relevant (α,n) reactions.
- ^{13}C (α,n) is important for calibration neutron detectors used in safeguards.
  - This information is also relevant to molten salt reactor studies such as low-Z isotopes found in F, Li, Be in other salts.
- A study on burn-up effects of fission products is needed. The yield of these products can be used for quantifying the number of fissions in a sample.
- Intensity of gamma-ray emissions and branching ratios in the decay of ^{234m}Pa can have immediate impact on safeguards applications.
- Delayed gamma-ray spectra and models thereof are affected strongly by fission yields. These calculations of fission yields are important for assaying nuclear materials.
  - For fissile materials and a variety of energy-group/energy-differential irradiations. ^{235}U, ^{238}U, ^{239}Pu, ^{241}Pu
  - For fast neutron spectra more fissionable isotopes become relevant
  - Carefully selected energy groups can be useful to different applications because they can be selected/applied in an intuitive fashion.

- There are a number of active projects to measure fast neutron fission. [7] However, low-energy neutron irradiations would likely require additional efforts.
- High-energy gamma intensities for fission products.
- A list of high priority fission products is needed, especially those with short half-lives (< 10 min) and high energy gamma emission lines over 2.5 MeV
- Eventually it would be ideal to obtain fission yields for minor actinides as these are also found in spent fuel.
- There are about 156 fission products. Not all of them contribute to the fission spectra. We are looking for high energy gammas. (^{142}La, Cm that is in spent fuel has a high cross section that causes some interference, ^{237}Np, ^{233}Pu)
- New high-precision measurements of ^{252}Cf (nu-bar), including the quantification of the delayed neutron component are needed. Moments and distributions are desired.
- Evaluations of distributions need to be updated for applications like MCNP [8].
- Improved measurements of low energy X- and gamma-ray line intensities of U and Pu.) and their daughters

A summary comment made by several workshop attendees indicated that just like there are benchmarks for criticality safety, benchmarks are needed for safeguards measurements; however there first should be an effort to define a safeguards benchmark. If appropriate safeguards benchmarks can be established, then we can truly perform measurements of excellence!

7. Concluding Remarks

The entire global safeguards community needs to achieve a better understanding of the underlying nuclear and atomic data used by safeguards measurement systems. It is very important that the source(s) of nuclear and atomic data utilized by these measurement methods can be referenced and the uncertainties with this data are well understood. This will improve our ability to quantify measurement uncertainty and move the global safeguards community toward measurements of improved quality.

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