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Measurement Techniques for Warhead Authentication with Attributes: Advantages and Limitations

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A survey of different measurement techniques applicable in the context of the attribute approach for warhead authentication is presented to demonstrate current capabilities and gaps. Therefore, their technical advantages and limitations will be assessed, in particular in terms of correctly characterizing an item whose configuration is not entirely known to an inspecting party. Passive and active neutron and gamma measurements assessing the presence of fissile material, isotopic composition and fissile mass of plutonium and uranium are considered.

Currently, verification of nuclear arms control refers to the verification of delivery vehicles. This is the case for instance in the New START Treaty between Russia and the United States.[1] Warheads are counted indirectly via the delivery vehicles they are associated with. Regarding the longer-term future of nuclear arms control and disarmament, there seems to be fairly broad agreement that verification should become more intrusive and that direct verification of warheads, deployed as well as non-deployed, could play a vital role in this approach. Scenarios where warhead (and warhead component)[2] verification might be required are warhead and component inventory declarations or during a dismantlement process where warheads are disassembled into their components; verification related to both could be based on the Nonproliferation Treaty (disarmament obligations under Article VI) or other bilateral or multilateral arms control or disarmament regimes, examples could include a future United States/Russian regime or regional regimes, forinstance in the Middle East.

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Part of such a verification regime could be warhead authentication. In this context authentication is the process during an on-site inspection through which it is assessed by measurements whether a specific item is a nuclear warhead (or component). States declaring their nuclear arsenals will very likely not allow inspections that reveal warhead design information, neither if the inspector belonged to a nuclear-armed state, nor if he belonged to a non-nuclear weapon state, be it for proliferation or national security concerns or other reasons. Assuming a likely case where verification measures are decided upon in a cooperative manner, mutual agreement on warhead authentication activities can only be reached in the case where nuclear-armed states are confident that their warhead design information is not at stake. The inspecting party, whose interest is to gain maximum confidence on the true nature of a declared warhead, would likely prefer rather intrusive and comprehensive authentication measures. The goal is to create a warhead authentication system which is in a position to build confidence while preventing unacceptable levels of intrusion that could leak information the inspected state is unwilling to share. This intuitively appears to be somewhat contradictory.

A solution would be to take potentially intrusive measurements containing sensitive information, but to automatically process the measurement information via an algorithm so that the only output visible to the inspector would be of non-sensitive nature (e.g., a green or red light indicating "specified warhead" or "not specified warhead," other output options could be possible). Preventing the leakage of sensitive information would be the task of a so-called information barrier. Main requirements of such an authentication system are (a) that automatic measurement and analysis function properly so that the best possible indication of authenticity is given and (b) that no sensitive information is released. (a) is in the interest of both host and inspector since an honest host would like to get credit for having dismantled a true warhead and prevent false alarms, while the inspector would like to ensure that he is not cheated upon. Therefore, ideally, the system should accept all specified warheads and reject all other items, showing low probabilities of false positives and false negatives. (b) is certainly in the host's, but also in the inspector's interest since all parties must adhere to the Nonproliferation Treaty's Articles I and II.[3] This puts restrictions on the possible output options and perhaps also on allowable measurement and analysis methods. Therefore authentication systems must be designed in a way that both host and inspector can ensure that the host has not built in a capacity to manipulate measurement results, and that the inspector has not built in a capacity which could enable assess to sensitive information. Some plausible concepts how the information could be processed can be thought of and shall be acknowledged, in particular the template approach,[4] however this article will address the attribute approach: Under this concept, the inspected party would declare attributes that characterize the warhead that will be authenticated and that do not contain sensitive information. For

robust authentication, the attributes should be chosen in a way to minimize the possibility of cheating, so that other items that are not warheads or warhead components will not fulfill the attributes, but that the authenticated warheads will. Attributes might be of qualitative (e.g., the presence of fissile material) or quantitative nature (e.g., minimum mass thresholds).

The purpose of this article will be to offer a survey of different measurement techniques applicable in the context of the attribute approach and assess the technical advantages and limitations they offer, in particular in terms of correctly characterizing an item whose configuration is not entirely known to an inspecting party. It is based on a review of existing publications, complemented by some research results by the authors. These nuclear measurement techniques would be well suited for verifying signatures of the fissile warhead component. A range of non-nuclear measurement methods could be applicable for attribute determination as discussed by Kouzes and Geelhood;[5] this article will however discuss neutron and gamma measurements. Furthermore, neutron imaging techniques are currently developed and may play a role, but are not included here.[6]

Technical Boundary Considerations

In most situations where radioactive samples are characterized, certain knowledge exists prior to the measurements. The geometry and other properties might be known, at least roughly. Quantitative analysis methods can rely on detector calibration with representative standards. In warhead (component) authentication this is not the case. Especially in the case of an inspector from a non-nuclear weapon state, weapon or component design information will not be available. This raises the problem that the designed system must authenticate an item whose configuration such as geometry remains largely unknown. Therefore the authors propose that the optimum measurement system should be chosen in such a way that it is not dependent on calibration with materials representative of the warhead or component and that a minimum amount of assumptions have to be made regarding the nature of the item.

Part of this issue is possibly unknown shielding. In the case of fully assembled warheads, shielding might arise from material surrounding the fissile component or self-absorption of radiation within the fissile material itself. Furthermore, most nuclear warheads are stored in containers for safety reasons that could act as further shielding. Warhead components would also be stored in containers which could act as shielding. A range of such containers exists. The AT-400 and 9975 containers are two examples that could be used for components.[7,8] The AT-400 container mainly consists of two stainless steel shells (0.1 cm and 0.2 cm) separated by a polyurethane layer (7.0 cm). It hosts a containment vessel, a 0.6 cm thick stainless steel cylinder.[9] The 9975 container

consists of a 0.1 cm stainless steel outer shell, then an 11.7 cm thick Celotex® shell, then a lead shield of 1.6 cm.[10]

Such issues could have an impact on the measurement, and therefore on the output, of the attribute analysis. It shall be recognized that it is in the interest of the inspected country to ensure a positive outcome of the authentication activity. If the impact of the configuration is so strong that it could give inconclusive results, it would be in the interest of the inspected country to offer an alternative method or to declare the shielding to the inspecting country. Still, it is advisable (and one might find it essential) to use measurement methods where the inspecting party has confidence in its functionality apart from assurances by the inspected party.

To the knowledge of the authors, most technical research to date has focused on developing measurement methods that do not release warhead design information when applied with an information barrier. More research is found wanting to investigate proposed methods in terms of possible requirements of the inspecting party.

Notes on Attributes

A range of attributes has been suggested by the United States, often in cooperation with Russia. This list has changed and developed over time. While plutonium, technically a much easier authentication task, was exclusively dealt with in the beginning, more research is conducted on uranium attribute measurements. A list of the proposed attributes is given in Table 1.[11–14] All the attributes are chosen to yield a binary yes/no answer; some are of qualitative nature while others set quantitative thresholds. Thresholds provide the benefit that it is not necessary to declare the actual values which might be sensitive. However, threshold values should not differ from the actual values to the point that they are not suited to exclusively describe warheads, otherwise the attribute authentication activity loses relevance. Attributes need not necessarily be limited to thresholds and binary outputs, though all of the proposed attributes have been to date.

While the choice of some of the attributes is intuitive, one might come up with different attributes nevertheless. Little is published on why specific attributes were chosen, but one could think that the attributes are chosen in a way that they can be successfully measured (at least theoretically) given the geometries and shielding properties. To state one theoretical example, the plutonium mass threshold in at least one initiative was considered to be 500 g.[15] This value could have been chosen in such a way that a measurement would yield a mass higher than 500 g for the warheads to be authenticated, given possible undeclared shielding which would make the measurement result lower than the actual fissile mass present. Reducing the dependency of measurements on geometry and shielding could allow more meaningful

Table 1: Attributes proposed by various initiatives. AVNG refers to the Attribute Verification with Neutrons and Gamma Rays, FMTTD refers to the Fissile Material Transparency Technology Demonstration, 3G-AMS refers to the Third Generation Attributes Measurement System.

Presence of plutonium	Trilateral Initiative, AVNG, FMTTD, 3G-AMS
Ratio of plutonium-240 and plutonium-239	Trilateral Initiative, AVNG, FMTTD, 3G-AMS
Plutonium mass threshold	Trilateral Initiative, AVNG, FMTTD, 3G-AMS
Plutonium age	FMTTD
Absence of oxide	FMTTD
Symmetry	FMTTD
Presence of uranium-235	3G-AMS
Uranium enrichment	3G-AMS
Uranium-235 mass	3G-AMS
High explosive mass	3G-AMS

attribute thresholds as close as possible to the real values so that the level of confidence gained from such a system can be greater.

NUCLEAR MEASUREMENT TECHNIQUES

While a range of different attributes have been considered (Table 1), nuclear measurement techniques relevant for assessing six attributes are addressed here, namely the presence, isotopic composition, and fissile mass of plutonium and uranium. One might consider these attributes as rather essential for successful authentication. The major nuclear measurement techniques discussed to date will be presented without necessarily achieving completeness. An overview of the analysis is given in Table 2; the details are found in this section. Some novel techniques that are in an earlier stage of research and might reveal future potential are also included.

Whether the item to be authenticated is a fully assembled warhead or a warhead component, the attribute assessment refers to the properties of the fission component (or "primary" in the case of thermonuclear warheads). Only a choice of attributes that can be assessed by a certain technique will be presented. Further attributes that could be assessed by the same technique will not be discussed in detail when it is found that the attribute can be assessed more easily with a different technique.

Passive Gamma Measurements

Gamma measurements could be used to determine the presence and isotopic composition of plutonium warheads and only in the case of minimal shielding, possibly also for uranium warheads. In the case of plutonium, it may be suited for a minimum mass estimate.

Table 2: Non-exhaustive selection of measurement techniques possibly suitable for attribute determination discussed in the article. Advantages and limitations of specific techniques are listed.

Attribute	Measurement method	Advantages	Limitations
Plutonium presence	Gamma spectrometry	Not vulnerable to little external shielding	Vulnerable to significant external shielding Requires homogeneous isotopic composition
Plutonium isotopic composition	Gamma spectrometry	Not vulnerable to little external shielding	Vulnerable to significant external shielding Requires homogeneous isotopic composition
Plutonium mass	Passive neutron multiplicity counting when isotopic composition known	Calibration without representative standards	Somewhat vulnerable to external shielding
	Scintillator cross-correlation measurements when isotopic composition known		Somewhat vulnerable to external shielding Calibration requires representative standards
	Gamma spectrometry (minimum mass estimate)		Physically limited through self-absorption
Uranium presence	Gamma spectrometry		Little shielding hinders analysis
	Active Neutron Multiplicity Counting	Not vulnerable to moderate external shielding	
	Active scintillator cross-correlation and time-of-flight measurements	Not vulnerable to moderate external shielding	

(Continued on next page)

Table 2: Non-exhaustive selection of measurement techniques possibly suitable for attribute determination discussed in the article. Advantages and limitations of specific techniques are listed. *(Continued)*

Attribute	Measurement method	Advantages	Limitations
Uranium enrichment	Gamma spectrometry		Little shielding hinders analysis Protactinium-234m low activity
	Active neutron multiplicity counting	Little shielding does not prohibit analysis	Somewhat vulnerable to external shielding Calibration with representative standards may be required due to source-sample coupling Triples precision limited
	Active scintillator cross-correlation and time-of-flight measurements	Little shielding does not prohibit analysis	Somewhat vulnerable to external shielding Calibration requires representative standards due to source-sample coupling
Uranium mass	Active neutron multiplicity counting		Somewhat vulnerable to external shielding Calibration with representative standards may be required due to source – sample coupling Triples precision limited
	Active scintillator cross-correlation and time-of-flight measurements		Somewhat vulnerable to external shielding Calibration requires representative standards due to source-sample Coupling

Plutonium Presence and Isotopic Composition

Plutonium-239 has a wealth of gamma lines, but plutonium-240 has significantly fewer. The significant lines are at 104.2 keV, 160.3 keV and 642.4 keV.[16] In all of these cases, lines of plutonium-239 are in proximity (for example 103.0 keV; 160.2 keV, 161.5 keV; 640.0 keV, 645.9 keV). This is helpful for the analysis as the energy-dependent efficiencies and self-attenuation are not an issue, but in practice even with the use of high purity germanium (HPGe) detectors is the unfolding of the interfering peaks required. Due to resolution requirements, HPGe detectors are assumed in the following discussion. In case of aged plutonium spectrum evaluation the ingrowth of americium-241 resulting from the decay of plutonium-241 must also be considered, for example the peak at 641.5 keV.

Attribute Determination. The presence of plutonium (in particular plutonium-239) can be determined given the amount of plutonium peaks.

The isotopic composition can be determined from analyzing the 640 keV peak region where the triplet of plutonium-239, plutonium-240 and americium-241 can be solved. This energy region can be seen in Figure 1 showing a measurement performed at the PERLA laboratory of the Joint Research Centre in Ispra with a weapon-grade plutonium metal source na,med "PM1" (12.5 g plutonium mass) behind 2 mm lead shielding.[17] At the measurement date major isotopes present were plutonium-239 (95.4 percent), plutonium-240 (4.5 percent), uranium-235 (0.1 percent) and americium-241 (0.02 percent). The triplet and the other peaks required for the analysis were visible, even though count rates were low due to the small sample mass.

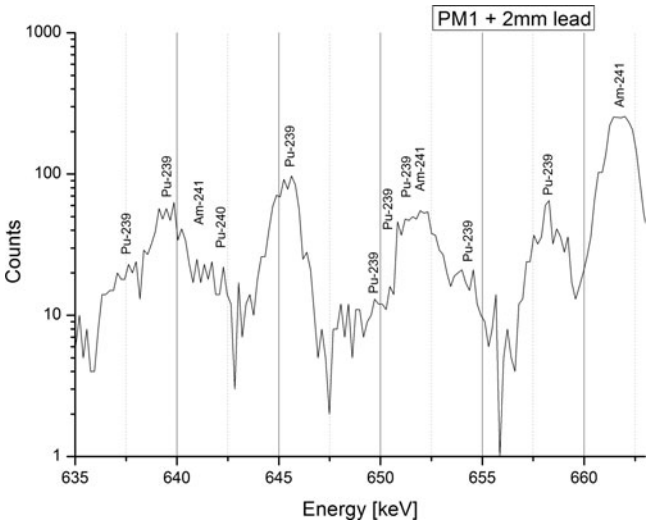

Figure 1: Gamma ray spectrum of PM1 sample. The measurement was performed with 2 mm lead shielding between sample and detector. Plutonium and americium peaks should theoretically be present and are indicated, though only some of them are visible.

Several codes have been developed that analyze this region according to the procedure explained above. The code Pu600 was developed in the United States as part of a collaboration on an attribute measurement project between Russia and the United States.[18] The method was implemented in further initiatives, for example in the Fissile Material Transparency Technology Demonstration (FMTTD),[19] as part of a project called Next Generation Attribute-Measurement System (NG-AMS) and is considered for the Third Generation Attributes Measurement System (3G-AMS).[20,21] Most recently, this analysis method was implemented and tested in an information barrier developed in the United Kingdom and Norway. Both the United Kingdom–Norway code as well as Pu600 yielded rather satisfactory results, though some bias was observed.[22]

Discussion. Gamma shielding can be an issue, especially at energies below 200 keV as can be seen from the measurements at the Joint Research Centre (see Figure 2). Table 3 shows the very short lead thickness required to strongly attenuate photons at 150 keV. While peaks related to lead could be easily identified (and one americium-241 peak was present), neither plutonium-239 nor plutonium-240 peaks could be evaluated easily in the region up to 170 keV. Without the lead shielding, these peaks became visible. Figure 2 also shows a measurement with the AT-400 container. For this measurement, the detector was further away from the sample than for the previous two. The measurement times for the previous two measurements were 2000 s, and for the AT-400

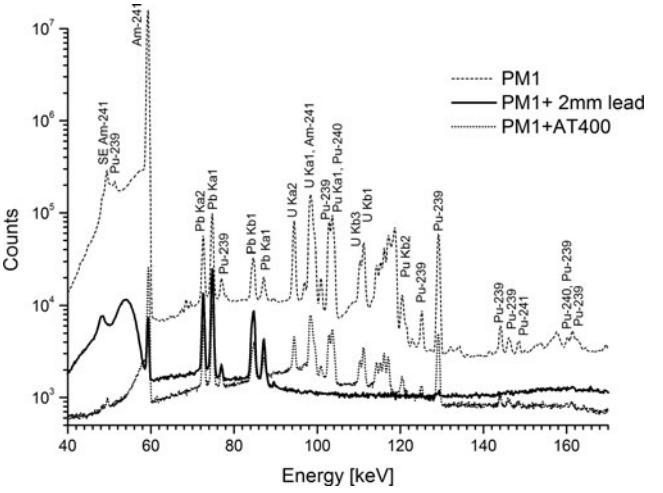

Figure 2: Gamma ray spectra of plutonium metal sample source "PM1." One measurement was performed without shielding, in another a 2 mm lead plate was placed between sample and detector, in the third measurement the sample was placed inside the AT-400 container. Indicated are characteristic x-ray peaks (K_α and K_β denoted as Ka and Kb) from lead placed around the detector; uranium and plutonium, a peak from X-ray fluorescence in the detector crystal (SE americium-241) and the characteristic gamma radiation.

Table 3: Half-value layers (where 50 percent of radiation is absorbed) of lead, uranium and plutonium metals for photons of different energies.

	Lead	Uranium	Plutonium
150 keV	0.3 *mm*	0.1 *mm*	0.1 *mm*
600 keV	5.2 *mm*	2.6 *mm*	2.4 *mm*
4 MeV	14.7 *mm*	8.3 *mm*	7.9 *mm*

measurement it was 12000 s. Some of the plutonium peaks could be identified rather easily in the AT-400 measurement. As this container is suited to store weapon components, it could be possible to determine the isotopic composition in this lower energy region in a verification scenario. However, minimal additional shielding could still make such an analysis difficult. It shall also be noted that the measurement was done without a containment vessel in place which would have constituted additional shielding.

The lead peaks are the result of photon interaction processes with the lead. These peaks can be used to establish the presence of lead as shielding material. To the author's knowledge, no research has been conducted to establish the presence of lead or quantify it in combination with the analysis of attributes. For example, the presence of significant amounts of lead with no plutonium lines above the detection threshold may both mean that plutonium is absent or that plutonium is present but that its peaks are shielded.

Shielding has much less influence above 600 keV, see Table 3.[23] The measurement of the PM1 source without lead shielding yielded similar results as in Figure 1, though the lead decreased the count rate. The same should hold for the AT-400 container. Therefore measurements in this region are preferred since assessments would be less dependent on external factors such as shielding that the inspecting party might not know about.

A potential issue is that gamma radiation from the inside of the fissile material is attenuated to a large extent before reaching the surface so that an isotopic composition assessment does not give the average isotopic composition of the volume, but rather at the volume's surface. This is problematic when the isotopic composition varies over the fissile material volume, for example when both plutonium and uranium are present.

Overall, assessing the presence and isotopic composition through gamma spectrometry at 640 keV is not very vulnerable against limited external shielding such as the AT-400 container and should give good results as long as the isotopic composition in the fissile material is homogeneously distributed. Therefore, this method is considered robust. Moreover, if shielding is not excessive, its presence may be deduced from the measurement.

Plutonium Fissile Mass

Attribute Determination. Self-absorption of gamma radiation by plutonium would result in only measuring a portion of the total plutonium mass. The influence can be seen in Table 3, which shows for example that the gamma ray intensity is reduced to 50 percent after passing through 2.4 mm of plutonium at 600 keV. The ratio of the measured minimum and actual mass would be strongly dependent on the plutonium configuration.

The "Trusted Radiation Attribute Demonstration System", developed by Sandia National Laboratory, analyzes gamma radiation to obtain such a minimum mass estimate.[24] It assumes that self-absorption does not take place and that shielding is uniform, recognizing that evaluating a spectrum based on these two assumptions will yield a minimum mass. Taking into account gamma ray intensity at different peak energies as well as the intensities of scattered photons with energies approaching the incident gamma peaks, shielding properties are calculated. The measured spectrum is fitted accordingly to obtain the minimum mass estimate, which also requires an approximate energy calibration. A measurement of a solid ball containing 2230 g plutonium-239 yielded an estimate of 290 g.[25]

Discussion. An argument for this technique is that the mass attribute is usually considered a minimum mass threshold and is therefore not required to measure the exact mass. It should be taken into account though that for large plutonium masses, the underestimation is severe so that the attribute would need to be much lower than the actual value.

Uranium Presence, Isotopic Composition and Fissile Mass

While uranium-235 has many low intensity gamma peaks, there are four peaks at higher intensities. A peak at 185.7 keV has a 57.2 percent intensity, three further peaks with intensities of 5–11 percent are present at 143.8 keV, 163.4 keV and 205.3 keV (intensities range from 5–11 percent). Uranium-238 has two low energy peaks with very low intensities, however uranium-238 could be identified via gamma spectroscopy by identifying peaks of protactinium-234m (half-life 70.2 s), which is the daughter nuclide of thorium-234 (half-life 24 days) which uranium-238 decays into. Protactinium-234m inter alia has three peaks between 740 and 790 keV and the strongest one at 1001.0 keV.

Attribute Determination. In the absence of shielding, the presence of uranium-235 could be determined by identification of the peak at 185.7 keV. The presence of uranium-238 could be determined through the 1001.0 keV protactinium-234m peak.

The isotopic composition could be determined from a comparison of the two above-mentioned peaks. If the 185.7 keV uranium-235 peak and protactinium-234m peaks are visible, an issue is that the main peaks of protactinium-234m

are in a different energy region than uranium-235 which is problematic because of the energy-dependent detection efficiency due to the energy-dependent attenuation. Therefore, an intrinsic self-calibration technique would need to be performed. Relative efficiencies can be obtained at energies showing different peaks of the same isotopes by analyzing the ratios of these peaks. Such a technique is implemented in the FRAM code.[26]

In order to assess uranium mass, a similar approach as the minimum mass estimate for plutonium could be considered. To the authors' knowledge, no authentication system to date was based on such a technique. Here, all the issues pertaining to uranium measurements stated above add to the issue of self-absorption which makes such an analysis path unattractive to pursue.

Discussion. The relevant peaks of uranium-235 are all in the low energy region and can be easily shielded (Table 3). This severely limits a successful determination of the presence and isotopic composition for uranium warheads. Identifying and quantifying uranium-238 through protactinium-234m could be a challenge as well because the activity of protactinium-234m would likely be low due to the limited mass of uranium-238 in weapons-grade fissile material and its long half-life.

Assessing the isotopic composition using protactinium-234m requires the assumption that protactinium-234m is in equilibrium with uranium-238. This assumption is true since the activity of thorium-234 will reach its maximum in 867 days which is the equilibrium criterion. The fissile material used in nuclear weapons is much older, at least in the United States and Russia.

This technique is implemented with some degree of success in configurations with very limited shielding: A measurement campaign at Los Alamos National Laboratory deducing uranium enrichment via FRAM was done with 200 g and 1 kg uranium sources shielded by 22 cm polyethylene, which is much less effective in gamma shielding than high-Z elements (it attenuated 94 percent of the 185.7 keV gamma ray and 76 percent of the 1001.0 keV gamma ray).[27] Most of the actual enrichments were within 3σ standard deviation of the measurements, some within 1σ; samples of high mass and high enrichments yielded better results than lower mass and lower enrichment samples. Certainly, under configurations of stronger shielding, this measurement method would be much less robust.

Overall, due to the low energy of the principal uranium-235 gamma line, this technique does not seem to be robust in the presence of shielding from storage containers or the warheads themselves.

Passive Neutron Measurements

Passive neutron measurements can be used to determine the fissile mass of plutonium when the isotopic composition is known.

Plutonium Mass Through Multiplicity Counting

Plutonium-240 emits neutrons through spontaneous fission at a rate of $1.02 \times 10^3 n\,{}^*s^{-1}\,{}^*g^{-1}$. The multiplicity distribution (i.e., number of neutrons emitted per fission) has a mean of 2.16 for spontaneous fission; the emitted neutrons follow a Watt spectrum with an average energy of 1.96 MeV.[28] The mean multiplicity is 2.88 for thermal neutron induced fission in plutonium-239 and 3.16 for 2 MeV neutrons.[29]

Neutrons can react via elastic or inelastic scattering as well as neutron-induced nuclear reactions. Elastic scattering slows them down and changes their direction. The average energy loss due to elastic scattering is given as $2E \times A/(A+1)^2$, resulting in an effective thermalization by light materials only.[30] The probability of many inelastic neutron-induced reactions drops off rapidly with increasing neutron energy and is usually high for thermal and epithermal neutrons (<1 eV) only, though also here the resonance integrals are considerable. Self-absorption of neutrons by plutonium cannot be neglected, especially in the resonance energy region, but is less pronounced compared to gamma absorption since fission neutrons are fast neutrons.

Attribute Determination. In order to deduce the plutonium mass through passive neutron counting, the spontaneous fission rate would need to be measured from which the plutonium-240 mass could be deduced. Then, the total plutonium mass can be calculated when the isotopic composition is known, e.g. through gamma spectroscopy.

The neutron flux emitted by a fissile sample is affected by a number of possibly unknown properties:

- Total spontaneous fission rate which depends on the fissile mass
- Neutron multiplication across the sample, in particular through induced fission by plutonium-239
- (α, n) reactions if oxides are present

Passive neutron multiplicity counting measures the multiplicity distribution (i.e., the distribution of the number of neutrons detected for gates of a defined length) which can be expressed in terms of its factorial moments which can be related to the factorial moments of the emitted distribution which includes spontaneous and induced fission as well as the (α,n) reactions, called the singles, doubles and triples rates.[31] The emitted distribution is based on the "superfission model" which assumes that all induced fission neutrons are emitted simultaneously with the original spontaneous fission or (α,n) reaction and can therefore be combined in one distribution for which an analytical form can be given.[32] One can solve the three unknowns through the three parameters that are measured.

Typically, helium-3 detectors are used for multiplicity analyses. Since the capture cross-section for neutrons in helium-3 decreases with the energy, fast neutrons are moderated by polyethylene surrounding the helium-3 tubes. This requires a certain time, so that coincidence gates must remain open for a sufficient time (usually 32–64 μs). Coincidence gates are triggered by neutron pulse so that correlated neutrons will fall into the gate. However, for such gate lengths, uncorrelated events will also be detected. This also requires randomly triggered gates with uncorrelated events to subtract those from the events in the coincidence gate to obtain the real coincidences.

This method has been implemented in some verification projects' authentication systems, including the Trilateral Initiative, the FMTTD, the NG-AMS, and 3G-AMS.[33–36]

In principal, other detector types could also be used for performing multiplicity analyses. It has been proposed to use the Nuclear Materials Identification System (NMIS) at Oak Ridge National Laboratory, which uses scintillators, to conduct multiplicity measurements.[37,38] In scintillation detectors, fast neutrons are detected via scattering without needing prior neutron moderation; this detector type enables much better time resolution and thus significantly lower background of non-correlated neutrons,[39] but shows lower neutron detection efficiencies than helium-3 detectors. A major issue is that already detected neutrons can exit the scintillator and re-enter after scattering or enter another detector, which can lead to multiple detection of a single neutron and therefore false coincidences. While deducing mass analytically by the described multiplicity analysis has not yet succeeded using scintillation detectors, development is under way.[40] If, however, calibration curves (plutonium-240 mass vs. doubles) are available, it is possible to estimate the fissile mass.[41]

Discussion. Neutron multiplication and the (α,n) reaction rate highly depend on the fissile material configuration. Neutron multiplicity counting separates these effects from the plutonium-240 spontaneous fission rate. Further simplifications that may introduce a bias depending on the configuration include the assumption that all neutrons have the same energy. The probability of inducing fission (uniform multiplication) is assumed to be homogeneous over the sample volume as well, as the detection efficiency of neutrons does not depend on their emission location ("point model"). The "superfission model" is usually considered reasonable given the small time scale of the fission process compared to the detection process, the validity of the "point model" depends on the detector design and the sample.[42] Some studies on limitations of these assumptions have been performed and modifications suggested, e.g. the weighted point model which takes into account non-homogeneous multiplication, but these continue to depend on the sample configuration.[43,44] Additional research is needed for quantifying these effects given the potential large range of sample and detector configurations.

Initial determination of the detector efficiency can be done with a californium-252 source. Multiplicity counting based on this calibration can be slightly biased because of a detector's different efficiencies between californium-252 and plutonium fission neutrons. This is a result of the slightly faster neutron energy spectrum of californium-252 compared to plutonium-239 and plutonium-240. Due to the similarity of the distributions though, the efficiency difference is small (in an MCNP[45] simulation the efficiency difference between a 2 kg plutonium metal source and californium-252 was 3.3 percent). The specific sample and detector configuration is described below. It is therefore well-suited for warhead authentication where representative calibration standards (warhead components) are not available and where the configuration of the fissile material remains largely unknown to the inspector, requiring approaches independent of sample configuration. The small bias could be minimized by establishing correction factors between the californium-252 and plutonium efficiencies for specific detectors.

Multiplicity counting however, is dependent on the configuration (shielding) between fissile material and detector. From the results presented below it appears that component storage containers could have an effect on the quantitative assessment of mass, but not even when some reasonable amount of neutron absorber is present do they prevent the sufficient detection of neutrons.

MCNP simulations were performed for a 2 kg plutonium sample (solid sphere, 95 percent plutonium-239, 5 percent plutonium-240) situated within the 9975 container. A detector consisting of 8 slabs, each filled with polyethylene and 6 helium-3 tubes, aligned around the container was used. The neutron surface current around helium-3 tubes is shown in Figure 3.[46] Simulation results with the 9975 container were compared to a simulation without the container and a simulation where the Celotex® from the 9975 container was replaced by 5 percent borated Celotex®. The result was that the count rate was increased by 33 percent with the 9975 container present and by 22 percent with the borated Celotex® configuration compared to the measurement of the bare sample. Three processes lead to this result: Firstly, Celotex® consists of low-Z materials that effectively reduce the neutron energies due to elastic scattering. This effect was in addition to elastic scattering in the detector polyethylene, so it contributed to slowing down the neutrons to increase their detection probability as shown in Figure 3. This effect is detector-specific and in this case means that the detector was not ideally moderated, since neutron moderation through elastic scattering competes with neutron capture (mainly in hydrogen with 1/v dependence). Thus, when increasing the amount of moderating material the neutron detection rate could be reduced due to increased capture rate of thermal neutrons in the moderator. Multiplicity counters are usually designed to minimize the dependence of the count rate on neutron energy,[47] but this is difficult for large detectors as required for this purpose (i.e.,

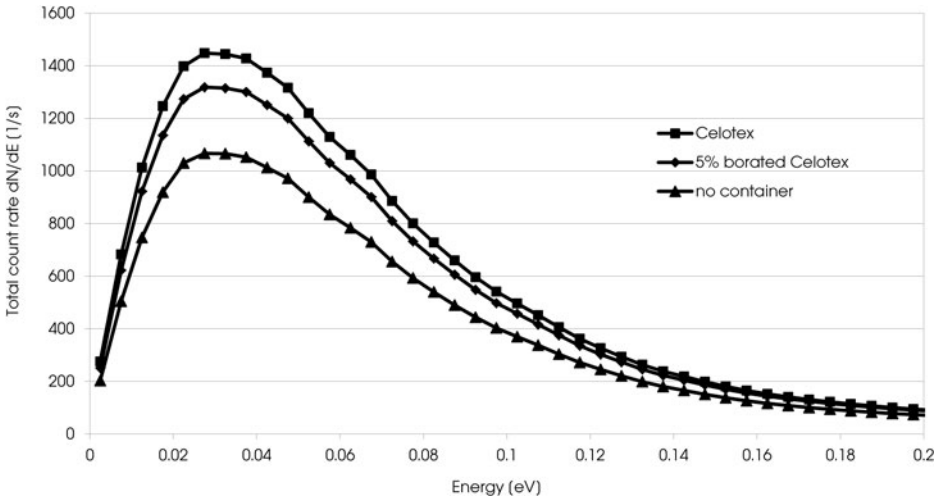

Figure 3: Energy spectrum of low energy neutrons passing through helium-3 tube surfaces for a 2 kg weapon grade metal plutonium source with a 9975 container (energy bin size 0.005 eV).

those where appropriate storage containers fit), as can be seen from the count rate results of the detector simulated above. Secondly, in the presence of boron (or another neutron absorber) epithermal and thermal neutrons are captured (the (n,α) cross-section of boron-10 is 1933 b at 0.1 eV). Celotex® moderates neutrons to epithermal and thermal energies, where the boron capture cross-section becomes large. However, Figure 3 shows that with 5 percent boron content, the count rate was decreased but did not prevent sufficient detection. Neutron capture rates in other materials such as stainless steel present in the container are much smaller. Thirdly, neutrons are scattered back into the plutonium source and induce further fission reactions, therefore increasing the count rate through increased multiplication.

In summary, this technique is considered little dependent from the fissile material configuration, though this should be investigated for specific configurations. Furthermore, the influence of external shielding on a mass assessment must be considered, as the presented simulation results suggest. The multiplicity analysis with scintillation detectors is successful when calibration standards are available, but it is unclear how large deviations become when sample configurations differ from the calibrated samples used for the mass analysis.

Plutonium Mass through Scintillator Cross-Correlation Measurements

Attribute Determination. In a distribution showing the detection of two correlated neutrons in different scintillation detectors as a function of time difference of detecting these (Figure 4), the width of the distribution is a function

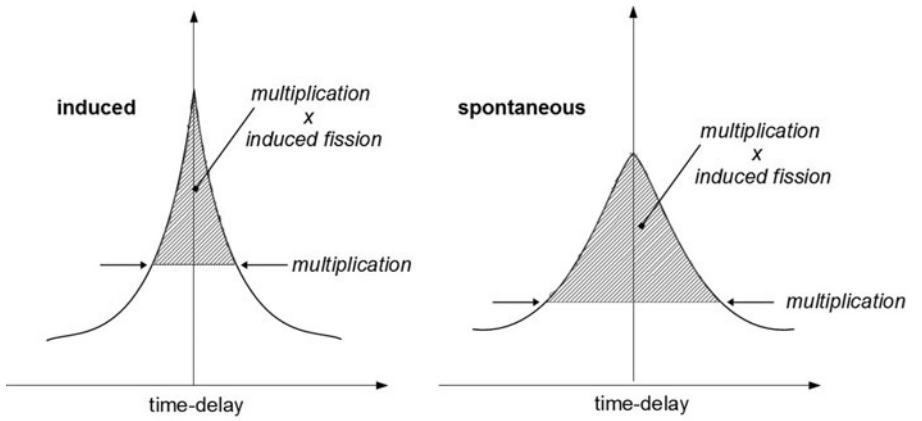

Figure 4: Scintillator coincidence time distribution as obtained from NMIS.

of the self-multiplication in the fissile sample.[48] The integral of the distribution is proportional to the product of the multiplication and the fissile mass. The NMIS uses the described approach to assess plutonium mass.[49] In principle, it is also possible to measure higher order cross-correlations between more than two detectors.[50]

Discussion. Determining the relationship between distribution width and multiplication relies on calibration with materials representative of the samples to be authenticated in terms of geometry, composition, density, and others.[51,52] With appropriate calibration curves, a plutonium mass analysis is feasible.[53] If these are not available to an inspecting party, this technique could be applied for mass determination only if a procedure will be developed for calculating the multiplication from the distribution width. The authors are not aware of studies investigating whether higher order cross-correlations could be helpful.

External shielding may have an effect on a quantitative analysis as discussed above in the context of multiplicity counting.

Uranium Analysis

For uranium-235, the spontaneous fission yield is $2.99 \times 10^{-4} n \cdot s^{-1} \cdot g^{-1}$, for uranium-238 it is $1.36 \times 10^{-2} n \cdot s^{-1} \cdot g^{-1}$.[54] This is about 5 orders of magnitudes less than for plutonium-240. For such low count rates, passive neutron multiplicity counting is not feasible.

Neutron-Induced Neutron Measurements

Active neutron measurements are relevant for determining uranium presence, enrichment and mass.

Uranium Presence, Enrichment and Fissile Mass Through Multiplicity Counting

Uranium isotopes can undergo neutron-induced fission from an external source. The (n,f) cross-sections for uranium-235 (uranium-238) are 665 b (1.9×10^{-5} b) at 20 meV, 1.3 b (0.5 b) at 2.5 MeV and 2.1 b (1.1 b) at 14 MeV.

Typical neutron sources used are, for example, americium-lithium (average neutron energy 300 keV) or 14 MeV neutrons emitted from a D-T generator.

Assessing the uranium presence, enrichment and mass with helium-3 detectors has been suggested for 3G-AMS.[55] Research for 3G-AMS appears to be on-going, the concepts are presented below.

Attribute Determination. The presence of pure uranium can be determined through combining (a) a passive measurement yielding no significant count rate and (b) an active measurement yielding a count rate signaling induced fission. Depending on the energies of the incident neutrons, both uranium-235 (at all energies) and uranium-238 (at high neutron energies) will be fissioned. Assuming that both uranium and plutonium are present in a warhead with a composite fissile core, the plutonium mass and isotopic vector would be determined passively, then an active multiplicity counting measurement would be conducted to evaluate whether the response would be higher than that expected for plutonium (since also plutonium isotopes could undergo induced fission).[56] Then, uranium enrichment and fissile mass could be determined through active neutron multiplicity measurements with neutron sources at two different energies. First, the mass of uranium-235 could be determined by multiplicity measurements using a neutron source with energies below the fission energy threshold of uranium-238, e.g., americium-lithium, and then the mass of uranium-238 by multiplicity measurements using a neutron source above the energy threshold of uranium-238, e.g., from a D-T generator by evaluating the neutron response above that expected from the already characterized isotopes.[57] The equations translating the measurement data to the multiplication and fissile mass are fairly similar as in the passive case.[58]

Discussion. A significant issue is that the coupling between neutron source and sample introduces another unknown quantity. Epithermal neutrons (<1eV), for example, do not penetrate far through the sample volume since they have a short mean free path due to the high induced fission cross-section of uranium-235. This would result in fission that is not distributed homogeneously throughout the sample which introduces a dependency of mass assessments on geometry, isotopic composition etc. Various approaches to determine the coupling have been developed, but they usually rely on calibration with representative samples.[59] Only in the case of homogeneous fission (when only fast neutrons reach the sample), calibration with representative samples is not required since the induced fission rate per mass only depends on the

incoming neutron flux and the induced fission cross-section for the energy of the source neutrons.[60]

Unfortunately, in warhead (component) verification the spectrum of the source neutrons arriving at the fissile material could include epithermal neutrons as they undergo scattering prior to their absorption. In a warhead storage container, they need to penetrate through Celotex® or polyurethane (in the case of the 9975 container and the AT-400 container, respectively), in a fully assembled warhead, they need to penetrate through the warhead's high explosive.

For the most efficient neutron multiplicity counter (the Epithermal Neutron Multiplicity Counter) and a 1000 s measurement time, the relative standard deviation is less than 1 percent only for assessing a 1000 g uranium-235 mass. For the more widely existing Active Well Coincidence Counter, a 1 percent precision is only achieved for a 4 kg uranium-235 sample.[61] However, a weapon or component would require a different detector (due to the requirements to the cavity size to fit an appropriate container) likely having a lower efficiency. Therefore, statistics might well become an issue even for large masses. Therefore the efficiency will be an important criterion when evaluating different detector designs.

In summary, moderate external shielding does not inhibit determining uranium presence. For quantitative analyses (enrichment and mass), however, the dependence on the sample configuration may be strong. While shielding has an influence on the estimates as discussed for passive multiplicity measurements and because of coupling as explained above, little shielding does not prohibit the measurement altogether as a reasonable number of neutrons would be detected. However, requirements for a sufficient precision constitute a challenge. Certainly, establishing techniques to reliably assess uranium attributes is in an early stage of development.

Uranium Presence, Enrichment and Fissile Mass Through Scintillator Cross-Correlation and Time-Of-Flight Measurements

The NMIS uses a D-T generator or a californium-252 source and could possibly be considered to determine uranium presence, enrichment, and mass through coincidence counting as explained below.[62]

Attribute Determination. The presence would be determined via the absence of spontaneous fission but the presence of induced fission.[63] It is proposed to determine the enrichment via an active cross-correlation measurement with 14 MeV neutrons to determine the mass of uranium-235 through induced fission and a neutron transmission measurement to determine the sum of uranium-235 and uranium-238 mass as both uranium-235 and uranium-238 would attenuate the transmission of the source neutrons through inelastic scattering and absorption.[64] The publication does not consider that uranium-238 also undergoes induced fission for 14 MeV neutrons.

The mass of uranium-235 would be determined through assessing the rate of induced fission neutrons from the coincidence distribution integral. Another approach of NMIS to determine mass and enrichment compares moments of the time correlation distribution of detected neutrons using a californium-252 source to calibration values yielding mass and enrichment as a function of the moments, but this technique requires calibration with representative materials.[65,66]

In addition, NMIS analyses the temporal decay (die-away profile, time-of-flight) of neutron emissions after pulsed neutron irradiation which is an indicator of uranium-235 mass. A slow neutron decay after pulsed interrogation with thermal neutrons indicates a longer fission chain (multiplication) which could correspond to highly enriched uranium, while a faster decay signaling less or no multiplication could correspond to depleted uranium.[67] Shortly after a pulse, detected events are primarily interrogation neutrons, while later, the contribution from induced fission becomes dominant. Accordingly, dividing the temporal count distribution into two segments, the ratio of the integral of the later part to the early part shortly after the pulse has been demonstrated to reliably estimate the isotopic composition when representative calibrations are available.[68]

It is also possible to combine the time-of-flight with the cross-correlation approach. By establishing two equations (by measuring calibration curves with representative samples) linking the die-away integral and the cross-correlation integral to mass and enrichment, both can be determined.[69]

Discussion. There are no apparent issues prohibiting qualitatively determining the presence of uranium. For determining enrichment and mass, similar considerations apply as discussed for active multiplicity measurements, i.e., heterogeneous coupling as well as external shielding. While shielding has an influence on the estimates, little shielding does not prohibit the measurement altogether as a reasonable number of neutrons would be detected. Calibration with representative materials would be required not only due to the source-sample coupling, but also due to the analysis method via the time distribution and the dependence between width and multiplication. A rough estimate is possible without representative calibrations (e.g. distinguishing between highly enriched uranium with high and depleted uranium with almost no multiplication).[70] The issue that the cross-correlation depends on both multiplication and mass could be solved by additionally taking into account the die-away profile. If representative materials are available or if the configuration of the item is known (so that Monte-Carlo simulations can be used), the combination of die-away profile and cross-correlation yields results with low uncertainties for mass and enrichment.[71]

The authors are not aware of studies testing the approaches in cases where the configuration of the item is not known. Quantifying the effect of sample geometry on the calibration curves should be given priority.

Novel Active Technologies

In addition to the technologies presented above which have reached a certain level of sophistication, emerging active technologies are discussed here that might in the future be helpful for measuring uranium and plutonium isotopic composition or mass. At the current stage of their development, their potential for attribute determination is difficult to assess. Many of the novel technologies are developed for nuclear security purposes such as border control where the focus is the general detection of possibly shielded fissile material in short measurement times, whereas a precise assessment of isotopic composition or fissile mass is secondary. For some of the following techniques, even apart from the challenge of implementing theoretically sound concepts in practice, the point of departure must be the enhancement of available nuclear data in order to consider implementation for attribute determination. A summary of the state-of-the-art is provided by Runkle et al.[72]

Interrogation sources can either be gammas or neutrons. Gamma radiation undergoes Compton scattering, pair production, and absorption in atoms. Radiation of some MeV (usually 1.5 to 8 MeV sources are used)[73] can excite nuclei and lead to the emission of gamma radiation at characteristic energies which depend on the individual isotope's nuclear structure. This phenomenon is called nuclear resonance fluorescence. At gamma energies between 10 and 15 MeV, the photofission cross-sections become large.[74] Furthermore, interrogating gammas can undergo (γ,n) or (γ,2n) reactions in fissile and non-fissile material, though the reaction threshold is higher than the photofission reaction threshold for most materials.[75] Neutron radiation can induce fission which competes with (n,γ) and other absorption reactions in fissile and non-fissile material.

Most research in the last decade has focused on β-delayed neutron and gamma radiation after fission induced by a pulsed source. It has the main advantage that no background from the interrogating beam exists. The "nuclear car wash" project interrogates fissile material with fast neutrons to observe the delayed gamma radiation with gamma energies $<$7 MeV which effectively penetrates both low- and high-Z shielding.[76] Furthermore, most (non-fissile) induced activation products emit photons of comparably low energies so that high energy radiation above 3 MeV indicates the presence of fissile material.[77] Observing delayed gamma radiation after interrogation with gammas, Reedy et al. successfully distinguish fissile from non-fissile sources when detecting radiation 13 ms after the interrogating pulse.[78] They further propose to use this method to determine the isotopic composition of the fissile material. This can theoretically be achieved by identifying the fission fragments from their specific high energy gamma disintegrations and by determining the fissioning isotopes from the fission fragment distribution. However, experimental tests have not been performed yet.

Kinlaw and Hunt introduce a technique to assess the isotopic composition from the delayed neutron signature measured with helium-3 detectors: They observe different decay rates of the delayed neutrons depending on the fissile isotopes during the first 130 ms after irradiation with a 20 MeV photon source.[79] Specific issues of this technique include that decay rates of relevant isotopes (e.g. uranium-235 and plutonium-239) are similar,[80] and that delayed neutrons are emitted only after 0.3 to 5 percent of the fission reactions.

Less experience exists with detecting prompt radiation, apart from neutron coincidence and multiplicity measurements with neutron interrogation as described above. The advantage of prompt radiation measurements is mainly that the intensity of prompt emissions can be orders of magnitudes higher than delayed emissions. On the other hand, however, the fraction of emitted radiation is usually small compared to the source intensity and prompt background effects (e.g., absorption reactions) from all materials.[81,82]

A promising prompt photon spectroscopy technique measures nuclear resonance fluorescence. Since the emitted gamma rays are isotope-specific, this technique in principal allows for the determination of the isotopic composition. Both the photon interrogation and emission energies are in the order of a few MeV and can therefore penetrate high-Z shielding. Nuclear resonance fluorescence may also be a candidate for fissile mass determination.[83] Today, development is still limited to identifying resonances of interest and conducting proof-of-principle measurements.[84] General experimental issues concerning "fast" spectroscopy are the high resolution requirements likely requiring HPGe detectors, which, however, pose the problem of small detection efficiencies, in particular for photons >1 MeV.

Given the early proof-of-principle stage of most of the techniques discussed in this section, any quantitative evaluation would be premature. However, limited effects of the geometry of the configuration measured and of eventual shielding will be required. For mass assessments, induced reactions in the fissile material should be as isotropic as possible. Last, the emitted radiation must also sufficiently penetrate eventual shielding between material and detector. As these novel technologies may prove useful to meet the required criteria, further research should be promoted.

CONCLUSIONS

There has been a range of research on measurement methods relevant for attribute determination which is discussed in this article. A summary of these methods addressing advantages and limitations is found in Table 2. The presented novel active technologies are not included due to their early state of research. Some limitations might be solved through further research. Advantages are listed when the specific measurement technique is not or is less

vulnerable to an issue than is another measurement technique. If no limitations are listed, the measurement technique fulfills the requirements addressed in this article. The main differences between the individual techniques in terms of correctly characterizing an item whose configuration is not entirely known to an inspecting party are the dependence on shielding external to the fissile material and the dependence on the fissile material configuration (i.e., whether detector calibration must be performed using representative fissile material standards). A technique should be as independent as possible of these issues in order for the inspecting party to be assured that it functions properly.

Generally, measurement techniques for plutonium attributes are much further in their development than for uranium. It appears that the presence and isotopic composition of plutonium can be determined using gamma spectrometry when there is no significant external shielding. Determining plutonium mass through passive neutron multiplicity counting is a technique that is usually assumed to be little dependent on the plutonium configuration (though this should be re-evaluated for specific cases) but is influenced by external shielding which could have an effect on the mass assessment. Determining uranium presence and enrichment through gamma spectrometry would only be possible when no external shielding is present. Likely there will be too much shielding. Neutron multiplicity measurements can determine uranium presence. However, for assessing enrichment and uranium mass through active neutron multiplicity counting, much further research is necessary to demonstrate its robustness and reliability. Besides having the same vulnerability towards external shielding as passive multiplicity counting, the largest issue is the unknown coupling between neutron source and fissile material which introduces a strong dependence on the configuration. Novel active technologies may also be helpful for uranium characterization, but are in an early state of research in regard to warhead authentication making it currently difficult to assess their future potential.

It may make a difference whether a fully assembled warhead would need to be authenticated or a warhead component. A warhead would contain materials that shield the radiation emitted from the fissile material and might also be placed in a container. For active measurements, interrogating particles would need to penetrate through these components before reaching the fissile volume which could modify both their flux density and energy distribution. Authentication of warhead components might have similar problems (though possibly less sincere) since they would be placed in storage containers that may act as shielding. In the case of warhead component authentication, one approach may be to share the container design or elements of it with the inspecting party and/or include the storage container in the calibration of the detector. For fully assembled warheads, the issues will be much more difficult to resolve due to the sensitivity. It would be beneficial if it were possible for the host to share

some information on the expected shielding without breaking nonproliferation obligations. Without sufficient shielding information, none of the quantitative attribute assessments are robust. In this light, even those measurement techniques usually considered robust require further assessment.

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