IPNDV Working Group 3: Technical Challenges and Solutions
Nuclear Materials (4) Technology Data Sheet

October 27, 2016

Nuclear Materials (NM) Technology Name: Passive Neutron Counting

Physical Principle/Methodology of Technology:

Contemporary nuclear explosive devices (NED) or nuclear weapons primarily containing plutonium (Pu) with ≥ 90 percent ^{239}Pu and/or highly enriched uranium (HEU) with ≥ 20 percent ^{235}U (for which active interrogation is more suitable). In addition, the so-called "primary" (or pit) may be made of Pu and/or HEU metal and as part of a "secondary" may also include extra fissile materials (in general HEU). It is expected that a nuclear weapon would contain about 8 kg of Pu (or ^{233}U) or 25 kg of ^{235}U in HEU. Passive neutron counting makes use of the neutrons emitted by spontaneous and induced fission processes in Pu and U to measure the amount of nuclear material present. The very penetrating nature of neutrons facilitates this use by making it possible to measure neutrons from the entire item.

The neutrons from nuclear material are created by three process:

- Spontaneous fission where the nucleus randomly separates into two fragments that then emit a distribution of neutrons and gamma rays.
- Induced fission where a fission event is driven by an incoming neutron interacting with the nucleus that then fissions.
- (α , n) reactions where lighter elements (e.g., ^{18}O or ^{19}F) react with a decay α

For well characterized material, the total or gross neutron counting rate is proportional to the mass of nuclear material present. The characterization requirement is substantial, however, and this approach is rarely used to quantify the mass of nuclear material as variations in the material composition and shape strongly affect the neutron rate. Neutron coincidence counting systems (both passive and active) have over many decades been successfully designed, adapted and used in safeguards for the accurate Non-Destructive Assay (NDA) of Pu and U containing items.

In essence, by measuring the correlated spontaneous fission rates of the item of interest, a passive neutron coincidence counter (two neutrons correlated are counted) or a multiplicity counter (more than two neutrons in coincidence) once calibrated leads to the determination of the mass of Pu provided the isotopic composition of the item is known by (e.g., gamma-ray spectrometry) and that the other competing reactions and conditions are taken into account. For neutrons from either spontaneous fission (^{238,240,242}Pu isotopes) or induced fission (^{239}Pu and ^{235}U) are emitted almost simultaneously (in coincidence) and detected within a gate width in the range of 40 to 80 µs.

The train of electronic pulses produced by the neutron detector is recorded and its distribution in time is determined.

Neutrons from background and from (α , n) reactions that must be accounted for are fortunately either uncorrelated or arrive randomly in time. Standard coincidence electronics such as the shift-registers or pulse train analyzers (or recorders) exploit this fact so that the detectors are insensitive to those unwanted neutrons.

In a traditional NCC the total neutron rates (Totals) and the time-correlated rates (Reals or Doubles) are measured within a gate width of typically 64 µs (depending on counter) following a few µs pre-delay. By subtracting the accidental rates counted within the same gate width but 1,000 µs later (i.e., once the fission neutron has died away) the pure correlated pulses are measured, which leads to the determination of the Pu mass and neutron multiplication (M) in the sample, provided the ratio of random to coincidence neutrons (α) is known. The latter is not always easily known especially for impure samples or items for which information is restricted and/or whose chemical composition is unknown. This problem of more unknowns (^{240}Pu mass, neutron multiplication M and α) than equations is solved by using multiplicity counters with high efficiency, which allow to measure the third order terms of the multiplicity distribution, i.e., the triplets rates (three correlated fission neutrons) and subsequently extract the mass of ^{240}Pu equivalent (or effective).

Table 1: For Indication, Decay Modes and Spontaneous Fission (sf) of Pu and U isotopes

Isotope	Main **Decay Mode**	**Half-Life (years)**	SF Neutrons **(1/(g·s))**	**Neutrons per SF**
^{238}Pu	α to ^{234}U, SF	87.74	2,600	2.07
^{239}Pu	α to ^{235}U	24,100	0.022	2.16
^{240}Pu	α to ^{236}U, SF	6,560	910	2.21
^{241}Pu	β^- to ^{241}Am	14.4	0.049	2.25
^{242}Pu	α to ^{238}U, SF	3.76×10^5	1,700	2.14
^{235}U	α to ^{234}Th	7.04×10^8	$3.0\text{x}10^{-4}$	1.86
^{238}U	α to ^{234}Th ... β^- to ^{234}U	4.47×10^9	0.0136	2.07

Note: NEDs typically contain no more than 7 percent ^{240}Pu due to its high spontaneous fission rate.

Potential Monitoring Use Cases (pre-dismantlement, dismantlement, post-dismantlement, storage stage):

The simple form of passive neutron counting—total neutron counting—can be used to verify the presence or absence of nuclear material. Although there are other neutron emitters, in conjunction with other information, a non-zero neutron count rate is an indicator of fission activity.

Considering the simple scenario set by the IPNDV and given boundary conditions to be ensured by strong information barriers, the verification of attributes in nuclear disarmament for a Pu -based device would ultimately seek to:

(1) Confirm the presence of Pu,
(2) Measure the ^{240}Pu to ^{239}Pu ratio (typically ≤ 0.1 for NEDs),
(3) Measure the mass of ^{240}Pu, and
(4) Consequently extract the mass of Pu from (2) and (3) in order to verify whether the mass exceeds an agreed threshold.

If a mass attribute is used, passive neutron coincidence or multiplicity counting could measure that quantity with sufficient accuracy and within a reasonable time (approximately an hour or less).

Count rates depend on the geometry of the fissile material due to strong neutron self-multiplication. However, for spherical geometries at least, approximate correction factors can be calculated.[1]

Used to measure U, Pu, or U and Pu:

[1] M. Göttsche and G. Kirchner, "Improving Neutron Multiplicity Counting for the Spatial Dependence of Multiplication: Results for Spherical Plutonium Samples," *Nuclear Instruments and Methods in Physics Research A* 798 (2015): 99–106.

Most suitable for Pu (in passive mode) but U can be assayed if used in active mode. Active mode is very similar to the discussion above where the addition of a neutron source (e.g., AmLi because of its low energy spectrum, thermal to 1.5 MeV) induces fission in the nuclear material creating a signature of the amount of material present.

For detection technologies, what does the method determine/measure (e.g., presence of nuclear material, isotopics, mass, etc.)?

Presence of nuclear material and mass of Pu if isotopic composition is known.

Physical Description of Technology (e.g., approximate size, weight):

The cavity dimension is determined by the dimension of the item of interest, which in turn determines the weight, size, and footprint of the neutron coincidence counters (NCC). NCC, the most commonly used of which is the High-Level NCC (HLNCC), developed at the Los Alamos National Laboratory, are generally made of ^{3}He detector tubes, which are highly efficient, robust, gamma-insensitive, and reliable. There are 18 tubes in the HLNCC, which has 17.8 percent efficiency, whereas high-efficiency multiplicity counters such as the High-Efficiency Passive Counter (HEPC) or Plutonium Scrap Multiplicity Counter (PSMC, 80 ^{3}He tubes) may contain many tens of tubes (efficiency can be as high as 45 to 65 percent) and a drum monitor containing 130 tubes. The detector sensors are all embedded in a few centimeters of HD Polyethylene to make use of the high capture cross-section of $^{3}He(n,p)T$ reaction for thermal neutrons. Other alternatives to ^{3}He and other inexpensive technologies are now available. Figure 1 below shows indications.

(1) (2) (3) (4) (5)

Figure 1: (figures not to scale) (1) the HLNCC-II cavity diameter 17.5 cm, outside diameter 34 cm, active length 50.8 cm, 43 kg eff. 17.8 percent; (2) JSR-12 Neutron Coincidence Analyzer; (3) JSR14 Neutron Analyzer Shift Register (multiplicity plus traditional coincidence counting); (4) the PSMC 80 kg, sample cavity 20 × 40 cm (up to vol. 10 L, eff. 50 percent for ^{240}Pu SF neutrons); (5) A drum monitor at JRC for 200 gal., cavity 1.2 m × 80 cm, 3–4 tons including shielding, 146 ^{3}He tubes, eff.> 60 percent.

If the application is not limited to existing commercial systems, the use of neutron slab detectors (walls of polyethylene with embedded ^{3}He detector tubes or alternatives to ^{3}He based technologies e.g., LiZnS, B10 etc.) linked together can be used to do low efficiency coincidence counting or, if sufficient in number, can be used to essentially build a multiplicity counter around an item. For large items that may be encountered in this application, this may be the only option short of a new, custom-built well counter. Design can be developed and optimized using Monte Carlo simulation.[2]

Time Constraints (e.g., measurement times including distance from object, time to install the equipment):

Measurement time to measure 500 g of Pu (0.1 for ^{240}Pu/^{239}Pu) or 500 g of ^{235}U at 1 m from the surface of the container (order of magnitude: seconds, minutes, hours, days):

Distance N/A. The preference is that the item be fully enclosed within a cavity whose dimension is to be determined by the item of interest. The general principle is that as much of the solid angle around the item be covered as close as possible to maximize the detector efficiency. Typically a few minutes measurements in a standard NCC.

Will this method work in the presence of shielding? If so, what is the maximum amount of shielding that will still allow the method to work?

Neutrons are very penetrating and many tens of centimeters in low Z materials (PE, C, H_2O etc.) will substantially thermalize and suppress neutrons. Some thermalization is desirable as the neutron detectors used in this application are most sensitive to thermal neutrons. Also, cadmium and boron are strong neutron poisons and will be very effective shielding against thermalized neutrons.

Technology Complexity (e.g., hardware, software, and ease of use by personnel):

Moderate but information barrier necessary for gamma-ray spectrometer and NCC.

Infrastructure Requirements (e.g. electrical, liquid nitrogen, etc.):

Voltage power supply.

Well-counters tend to be fairly large and heavy.

Technology Limitations/Variations (e.g., detection limits for nuclear material, operational temperature range, differences in technology detector materials):

- Knowledge of isotopic composition is required.
- Temperature range: 0° to 45° for ^{3}He-based counters are very insensitive to temperature variations and have good neutron-gamma discrimination. Organic scintillators are more sensitive to temperature variations.
- Due to the shortage of ^{3}He, alternative technologies have been developed for nuclear security and safeguards, which make use of ^{10}B in various forms (B-lined, BF3 gas etc.), Organic scintillators, LiZnS-based etc. Monte Carlo modeling has been a key tool for the design, optimization, calibration, and characterization of most counter-developments in recent years and will continue to be as important, especially where one seeks to adapt counter-designs to new and often stringent boundary conditions and geometries, especially cavity size and performance requirements such as efficiency, neutron die-away time in moderator, sensitivity to matrix materials, etc.[3] This is expected to be the case in disarmament verification.

[2] M. Looman, P. Peerani, and H. Tagziria, "Monte Carlo Simulation of Neutron Counters for Safeguards Applications," *Nuclear Instruments and Methods in Physics Research A* 598 (2009): 542–50.
[3] Ibid.

Information Collected by the Technology (used to help determine if an information barrier is required for use):

Presence of Pu, mass of Pu provided isotopic composition is known. Information barrier(s) required for both gamma-ray spectrometer and neutron NCC separately or/and together. See Attribute Verification with Neutrons and Gamma-rays below.

Safety, Security, Deployment Concerns:

It is a passive technique with generally no major concerns especially in systems using ^{3}He, ^{10}B lined, or plastic scintillators. BF3 based alternatives may have some safety concerns due to toxicity. New liquid scintillators used in nuclear safeguards and security have high Flash Point (FP), low vapor pressure, low chemical toxicity (e.g., EJ-309, FP=144°C).

Technology Development Stage (Technology Readiness Level, TRL):

Both the HLNCC and PSMC and equivalent counters are commercially available.

Counters can be ordered to user requirements (efficiency, footprint, cavity size, etc.) including alternatives to ^{3}He based systems. Information barrier is system dependent and needs development. Systems such as those given as examples below are in advanced TRL (6+).

Additional System Functionality (e.g., outside the monitoring use case):

None.

Where/How the Technology Is Currently Used (e.g., international safeguards, border protection):

International safeguards, nuclear security (in radiation portal monitors), disarmament verification (see below).

Examples of Equipment:

(1) Commercially available NCCs such as HLNCC-II (JCC 31 of Canberra), HEPC, PSMC etc.
- http://www.canberra.com/products/waste_safeguard_systems/neutron-safeguards-systems.asp
- http://www.antech-inc.com/product-categories/neutron/

(2) AVNG (Attribute Verification with Neutrons and Gamma-rays) with information barrier built by RFHC in VNIIEF Russia with support from LANL and LLNL (USA) within the Trilateral Initiative (IAEA, Russia, USA 1996-2002): High Purity Germanium (HPGe) gamma-ray spectrometer and Multiplicity Neutron Counter: 164 ^{3}He tubes, 30 percent efficiency inside AT-400R container (491 mm diam. and 503 mm height); demonstrated in 2009 in VNIIF to US observers.
- D. Budnovic et al., INMM paper LA-UR-05-4144 (July 2005)
- J. Thron et al., LA-UR-0-03606 (authentication features) INMM 51

(3) The Fissile Material Transparency Technology Demonstration (FMTTD; 2000) at LANL with LLLNL and PNNL able to measure six attributes (Pu presence, presence of weapon grade Pu, Pu mass and age, absence of Pu oxide and symmetry of Pu source) using a multiplicity counter and two gamma-ray detectors
- http://www.lanl.gov/orgs/n/n1/FMTTD/index_main.htm

(4) The third generation Attributes Measurement System (3G-AMS), which aims to measure:

- Pu presence, ^{239}Pu and ^{240}Pu mass, Pu (time since last separation), Pu density, ^{240}Pu to ^{239}Pu ratio, ^{235}U presence, U enrichment, ^{235}U mass, U density, HE presence, HE mass, HE thickness
- https://www.nti.org/media/pdfs/Third_Generation_Attribute_Measuring_System_3G-AMS_.pdf?_=1438892653

(5) The Institute for Nuclear Physics and Chemistry (INPC, in China) developed a system similar to the AMS with information barrier able to measure six Pu attributes.[4]
- http://www.caep.ac.cn/kxjsnew/wlx/13392.shtml

References:

N. Ensslin, "Principles of Neutron Coincidence Counting," in *Passive NDA of Nuclear Materials* (D. Relly et al., eds.) Nureg/CR-5550 LA-UR-90-732 (1991): 457–91.

H.O. Menlove, "Neutron Coincidence Instruments and Applications," in *Passive NDA of Nuclear Materials* (D. Relly et al., eds.) Nureg/CR-5550 LA-UR-90-732 (1991): 493–527.

W. Hage and D.M. Cifarelli, "Correlation Analysis with Neutron Count Distribution for a Paralysing Dead-Time Counter for the Assay of Spontaneous Fissioning Material," *Nuclear Science and Engineering* 112, no. 2 (1992): 136–38.

[4] See also Jie Yan and A. Glaser, "Nuclear Warhead Verification: A Review of Attribute and Template Systems," Science & Global Security 23, no. 3 (2015): 157–70.