Neutron Techniques in Safeguards

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Abstract. An essential part of Safeguards is the ability to quantitatively and nondestructively assay those materials with special neutron interactive properties involved in nuclear industrial or military technology. Neutron techniques have furnished most of the important ways of assaying such materials, which is no surprise since the neutronic properties are what characterizes them. The techniques employed rely on a wide selection of the many methods of neutron generation, detection, and data analysis that have been developed for neutron physics and nuclear science in general.

The term 'Safeguards'[1] refers to a continuing and developing effort by national and international organizations to prevent by certain control measures the diversion to illicit use of 'special nuclear material' ('SNM'). Usually SNM refers to various fertile or fissile nuclides, especially U or Pu, but is also interpreted to include substances such as D_2O or graphite useful for production of such nuclides. The control measures being developed are an integrated triad of physical security, classical accounting, and the actual assay of the flow of SNM in its various forms into, out of, and within nuclear plants and installations. The goal of assay systems is a material balance whose uncertainty is less than a 'strategic quantity'. No law of nature guarantees that this goal is attainable; indeed, more often it may not be. Thus the importance of considering Safeguards as being composed of the three mutually supporting factors, each insufficient by itself.

The radioactivity of typical SNM makes non-destructive assay ('NDA') attractive since it can be accomplished without breaching containment, and nuclear NDA methods usually the obvious choice. Most of the nuclear NDA involves neutrons in a significant way, not surprising since it is the unusual manner in which neutrons react with them that defines SNM. Though this paper discusses only neutron techniques it should be noted that other techniques, nuclear or not, are often needed to provide auxiliary information needed to interpret neutron technique derived data. For example, interpreting a spontaneous fission rate would require isotopic ratios obtained through gamma or mass spectrometry.

In the ~15 years of Safeguards instrumentation development it seems that virtually every trick known to neutron, nuclear, or reactor physics, and in various combinations, has been tried as a way of assaying the various forms in which SNM appears in the nuclear economy: feed stock or raw material, scrap, waste, finished product (nuclear fuel rods, plates, or assemblies), spent fuel, and processing or recovery solutions. The most manageable classification of the myriad of schemes seems to be in terms

of the incoming or interrogation radiation, its energy spectrum and means of production: accelerator, reactor, isotopic or none (relying on spontaneous fission), and the outgoing radiation, and its manner of detection, including the electronic processing of the detector pulse train. We elect here to first consider three techniques of widest applicability and importance as primary examples of neutron safeguards techniques.

(i) Neutron correlation[2] was originally developed to take advantage of the multiplicity of neutrons emitted in the spontaneous fission (s.f.) of the 'even' ('fertile') nuclides such as ^{238}U and $^{238,240,242}Pu$, expressed as the probability P_ν that ν prompt neutrons are emitted in a time $\sim 10^{-15}$s. The point of using the number of fissions rather than just the number of neutrons as an assay 'signature' is that these nuclides are emitters and therefore much or even most of the neutrons from the sample may be due to (α,n) reactions with light elements present perhaps incidentally, e.g. F, O, Al, etc. These reactions will depend on the chemical and physical composition, rather than only on the SNM. In fact, the problem is to separate out the bursts of 'correlated' neutrons, from fissions that are randomly distributed in time, from the 'uncorrelated' (α,n) neutrons (and those from background), which occur singly and also at random. (However when chemical and physical form are rigidly controlled, and background minimal, 'gross n' counting can be satisfactory.)

The basic neutron correlation detector, the 'well counter', is a hollow cylinder of moderator with (gamma insensitive) $^{10}BF_3$ or ^3He proportional counters embedded. The well is used to hold the sample. The purpose of moderator is reducing the energy of the neutrons to where they react efficiently with the ^{10}B or ^3He. The fast neutrons will slow down in a few μs, followed by diffusion in the counter until they either escape, are captured by the moderator, or react with the ^{10}B or ^3He, giving rise to pulses. The time τ_o for diffusion, $\sim$(15-150)μs depending on the size, material, and geometry of the detector assembly, is the lifetime of neutrons introduced into the well. Thus, in a trade-off, to obtain efficient detection, the original $\sim 10^{-15}$s distribution has been broadened by the stochastic slowing down-process to a distribution of the form $\tau_o^{-1}\exp(-t/\tau_o)$. A kind of correlation performed on a pulse train formed from the added outputs of all the detectors now becomes preferable to coincidence, since τ_o is much greater than the pulse width.

An incoming pulse causes the electronics to interrogate what is in effect a delay line of length τ (chosen to be of the order of τ_o) as to how many pulses preceded it in that time. A tally of the number of such preceding pulses is kept and compared with a similar tally of the pulses in an identical delay line which is separated from the first by many times τ_o. Pulses in the first delay can have two origins. Either they originated in the same fission as the incoming pulse (are correlated) or did not and are uncorrelated. Pulses in the second delay however must be uncorrelated. The difference or 'net' count N then depends on the correlated count alone. A proportionality between N and the number of fissions can be derived from first principles in terms of the P_ν and half-life for s.f. of the nuclides, the detection efficiency ε and neutron lifetime τ_o, τ and other parameters of the electronics which can be measured independently. The theory starts to break down for two different reasons as sample size increases. For large enough samples there is an appreciable probability that neutrons from a fission will induce further fission, so that the number of fissions is no longer in one to one correspondence with the amount of SNM. However,[3] often the region where the linear cali-

bration holds can be extended by introducing a 'multiplication' factor M such that the neutron emission multiplicity ν is replaced by $M\nu$; otherwise the calibration must be done empirically. Analogous to the way in which a radioactive source emitting coincident radiation can be calibrated absolutely without knowledge of the detector efficiency, it can be shown that G^2/N, where G, the 'gross' count, is also independent of detector efficiency to a high degree and so this can be used to reduce geometry and matrix effects. The other flaw is that for very high count rates particularly if largely uncorrelated, even if the electronics performs as designed, the relative statistical precision in N suffers. This is affected by ε and τ_o, and there are natural limits to further improvement.

Neutron correlation has been extended to 'odd' ('fissile') nuclides principally ^{235}U by using an external isotopic neutron source to induce fission ('active' mode) as well as the 'passive' mode described above, and the basic well counter has been proliferated into many specialized forms for particular applications such as fuel rods, assemblies, raw material, scrap, and waste of both Pu and U.

(ii) Fast coincidence techniques[4] employing plastic scintillator-photomultiplier detectors offer an interesting contrast with the preceding as a way of getting a signature for fission. The detectors are sensitive to fast neutrons and gammas and so take advantage of the fact that $\sim$2.5n and 7.5γ are produced within 10^{-15}s of each fission. The coincidence resolution time is however limited to (10-50)ns or more by the transit time of the radiation in the sample volume and detectors, whose dimensions are typically in the range (0.2-2.0)m. Since fission gammas tend to be less penetrating than neutrons, some instruments try to rely solely on neutrons by interposing Pb shielding to lessen sensitivity to 'matrix'. This does however limit count rate, hence the statistical precision for a given sample size and assay time, and, since only two-fold coincidence can be used, increases sensitivity to background. The calibrations tend to be more linear and less matrix dependent than instruments that also accept gammas. With the latter three-and even four-fold coincidences can be used and still obtain useable count rates.

Usually these instruments are used in the active mode, typically with a 'subthreashold' isotopic source such as ^{241}Am-Li(α,n), $\bar{E}_n \sim .4$MeV, or ^{124}Sb-Be(γ,n), $\bar{E}_n \sim .025$ MeV, although 'superthreshold' ^{252}Cf(s.f.,n), $\bar{E}_n \sim 2.3$MeV, is also used. In principle, by using these alternately, information on both the even and odd nuclides would result, since the superthreshold source would induce fission in both.

In contrast to correlation, where all counters form one superdetector, in coincidence the total detector volume is divided among several channels, with a resulting possible trade off between signal (true coincidences) to background (accidentals) ratio, and statistical precision. Also there is no simple theory or calculation of response. The calibration is purely empirical, using functions of the form $Am(1+Bm)^{-1}$ or $A(1-e^{-Bm})$ with m the mass of SNM, and A and B determined from standards and strongly matrix, geometry, and 'interrogation' source dependent. Large samples cause problems because of multiplication, and also from absorption by the sample of ingoing and outgoing radiation; this latter leads to a lack of sensitivity (decreased slope or 'saturation') in the calibration. An interesting recent development, analagous to using G^2/N in correlation, is to use as a signature various algebraic combinations of the various multiplicities of true coincidences available in a several-fold coinci-

dence circuit to derive quantities proportional to the amount of fission but independent of detector efficiency, alleviating geometry and some matrix effects. Thus, e.g. for a 3-fold arrangement, instead of using just N_{123}, quantities such as $(N_{12}N_{23}N_{31}/N_{123}^2)$ and $(N_1N_2N_3/N_{123})^{1/2}$ are used as indicators.

(iii) Bombardment with neutrons from an isotopic source followed by observation of the delayed gammas from the induced fission is an important technique with nearly all fuel rods assayed this way. The rods are moved at a controlled speed past a collimated neutron source, ^{252}Cf or ^{238}Pu-Li(α,n) is favored, so that only a small length, say $\sim$1 cm, is irradiated. Down stream a collimated gamma sensitive detector picks up the induced activity. The method is sensitive enough to detect individual pellets of incorrect enrichment, as well as assay the total content in minutes per rod, and is an outstanding example of how the manufacturer's interest in quality control often coincides with Safeguards.

The above methods implemented in various ways make up the bulk of applications, but by no means all.[5] In the following several others will be described briefly for general interest because of the physical principles entailed, but with relatively less claim for universality or importance:

(i) Accelerators, and accelerator and isotopic pulsed sources. Pulsed source techniques have the virtue that the detectors can be operated to sense radiation emitted from the sample when the interrogation source is not operating, in principle enhancing the signal to background ratio. Certain accelerators are readily operated in a suitable pulsed mode. 'Sealed beam' devices, miniaturized Cockroft-Walton generators of the type used for oil well logging, have been used to provide $\bar{E}_n \sim 14$ MeV via the $T(d,n)^3He$ reaction. Electron linacs developed for industrial or medical radiography have been used in several ways. Electron energies of $\gtrsim$5MeV can be used to make Bremmsstrahlung gammas capable of inducing photofission directly in the SNM sample, or produce from a ^{238}U target neutrons then used to bombard the sample; lesser electron energies can be used to make photoneutrons from D or Be targets. Van de Graaff accelerators are particularly flexible in the wide range of energies of monoenergetic neutron yielding reactions, e.g., $T(p,n)^3He$, $^7Li(p,n)^7Be$, $D(d,n)^3He$, $T(d,n)^4He$, etc. that can be used with them. Isotopic sources can also be pulsed. In the 'Shuffler' arrangement, an intense ($\sim 10^9$ n/s) ^{252}Cf(s.f.,n) source briefly irradiates the sample and is then rapidly withdrawn into a neutron shield by a flexible cable. Another method uses a gamma source passing through a hollow Be (or D containing) cylinder. Most used is ^{124}Sb yielding $\sim$0.025 MeV neutrons with Be. The detectors are of moderator pierced by BF_3 or ^3He proportional counters surrounding the volume where the sample is introduced.

Whatever the pulsing method, the delayed fission neutrons (and or gammas) can be used as a signature. Since the delayed neutrons are only $\sim$1% of the prompt, the advantage of the interrogation neutrons not being present is only partly realized. In fact, 'room return' and the 'on/off' ratio of neutrons from the source become important considerations. Typically, irradiation is for $\sim$1s, followed by a several second observation of the delayed neutron decay. The initial slopes of the properly normalized decay curves is found to be different for ^{238}U compared to ^{235}U or ^{239}Pu.

It is also possible to use the prompt neutrons as a signature, but record them after the interrogating pulse of neutrons. The 'slab detec-

tor', consisting of a flat array of BF_3 or ^3He counters in moderating material, has a characteristic neutron lifetime. The prompt neutrons entering the slab during the irradiation time therefore will give rise to an exponentially decreasing count rate, conveniently viewed on a few hundred μs time scale. This takes advantage of the relative intensity of the prompt group and the fact that the neutron source is off during the counting period, but does require more sophisticated electronics, e.g., to desensitize the counters during the massive initial pulse, so that they will recover soon enough after the pulse to take advantage of the maximum signal-to-background ratio available then.

Up to now, accelerators have not been very successful in industrial settings because of insufficient reliability and lack of qualified operators, while isotopic sources have reliable and predictable behavior and are comparatively inexpensive, and so have been favored. However, recent availability of better medical linacs, and significantly improved sealed beam sources, combined with a growing need to assay large containers (210 ℓ drums or boxes 1-1.5m on a side) which requires the greater penetrability of a more energetic neutrons source, may change this. Van de Graaff accelerators do not seem as amenable to industrial use and so their greater versatility may be confined to laboratories.

(ii) The lead 'slowing-down' spectrometer has been used to assay fuel 'rods'. A pulsed 14 MeV neutron beam from a sealed beam source enters a block of lead ~2m on a side. The neutrons tend to slow down as a group in such a medium with their average energy a function of time. Fissions induced in the rods inserted into the lead block are detected with proton recoil counters sensitive primarily to the high energy fission neutrons. Cross sections for induced fission are energy dependent, with differences among the nuclides. Thus, the fission neutron production will be a function of time, which, normalized with respect to the slowing-down neutron flux (measured with a BF_3 proportional counter) provides the total fissile content and isotopic ratios.

(iii) An interesting illustration of neutron methods in Safeguards is furnished by recent work to develop NDA techniques for measuring D_2O/H_2O concentration of mixtures in 210 ℓ drums without opening them. Several neutron based nuclear methods were tried and all proved basically workable: (a) transmission of neutrons from an ^{241}Am-Li source through the drum, taking advantage of differences in the scattering cross section of D vs. H, (b) the detection of 2.226 MeV gammas from the $H(n,\gamma)D$ reaction, which clearly indicates the presence of H, again using an isotopic source for the neutrons, (c) neutron detection from $D(\gamma,n)H$ reactions using 2.61 MeV gammas from ^{208}Tl to bombard the drum (d) measurement of the lifetime of a neutron bunch injected into the drum from a pulsed accelerator, this lifetime being a delicate function of the respective absorption cross sections, transport mean free paths, etc., of D and H.

It is difficult to quote typical accuracies (which involve random and systematic errors) or precisions (stability or reproducibility), or the ultimate sensitivity, of a particular technique. This is because they are usually extremely dependent on the nature of the sample including total amount, concentration, matrix, isotopics, physical or chemical form, possible interferences, and the availability of suitable standards. Establishing such accuracies and precisions is in fact a major part of the Safeguards instrumentation effort, with obvious implications as to how well the system can work. At their best, these methods can achieve

accuracies and precisions of a few tenths of a percent, more typically (1-10)%, at their worst, say (20-50)%. Nature is however often kind in that the poorer results are usually in those cases where the material is very diffuse and poorly characterized, therefore of low monetary and or strategic value, and the quantities need not be so well known. Ultimate sensitivities to SNM are often measured in mg to dg, and the largest assayable quantities in a few kg, though the best accuracy doesn't go with such extremes. The minimal assayable amounts depend on the allowable assay time and instrument environment, and the development of specialized versions of the basic instruments will aid assay of both minimum and maximum quantities.

This report covers work done at the U.S. DOE laboratories (principally LANL, but also BNL, ANL, LLNL, and Mound), the members of the ESARDA consortium (principally JRC ISPRA, KfK, and Harwell), under the auspices of the US NRC, and certain U.S. Companies: SAI, IRT, and NNC.[7]

Notes and References

1. As in other fields, a mostly self-explanatory jargon has developed; such words will be introduced with quotation marks ' '.

2. Selected references on neutron correlation counting by M. Zucker, et al. are (a) "Neutron Correlation Counting", Proc. ANS, May 15-17, 1978; (b) "Assay of Low-Enriched Uranium.....", Proc. of the 2nd Annual ESARDA Conf., p. 313, May 1981; (c) "Apparatus Characterizations as a Standard for Neutron Correlation Counting" Proc. of the 4th An. ESARDA Conf., p. A1-A10, April 27-29, 1982; (d) R. Sher, "...Characteristics of Neutron Well Counters...." BNL 50332, 1972.

3. Selected references for multiplication are (a) N. Ensslin, et al., "Self-Multiplication Corrections Factors for Neutron Coincidence Counting", Nuclear Materials Management, Vol. VIII, no. 2, p. 60, 1979; (b) M.S. Krick, "Neutron Multiplication Corrections, etc." LANL Report LA-8460-MS, 1980; (c) see also ref. 2(b).

4. The best overall single reference for this section and the following (which contains exhaustive specific references to work at LANL, IRT, etc.) is T. Gozani, "Active Nondestructive Assay of Nuclear Materials" U.S. NRC, Jan. 1981 (NUREG/CR-0602, SAI-MLM-2585). We regard this also as an excellent review of the basic physics and technology involved in Safeguards. See also Sher and Untermyer, "The Detection of Fissionable Materials....", ANS monograph, 1980.

5. The wide range and scope of safeguards methods as revealed by available instrumentation, both neutron based NDA and other methods as well, is illustrated by Fishbone and Keisch "Safeguards Instrumentation A Computer-Based Catalog", BNL Report BNL 51450, August 1981.

6. Fainberg, Zucker, et al. "Assay of Heavy Water..." 3rd ESARDA Symposium May 1981. Though technically and aesthetically satisfying, the nuclear methods proved inferior to a non-nuclear technique (measurement of the acoustic velocity) as far as field applications go, showing the danger in riding one's hobby horse too hard!

7. In preparing this paper we appreciated the efforts and consultation of T. Gozani (SAI), H. Menlove (LANL), and L. Kelly (BNL).