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NUCLEAR DATA FOR SAFEGUARDS

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Abstract: A review is presented of nuclear data needs for safeguards. Several areas where new measurements are required are identified.

(review, nuclear data, safeguards application)

Introduction

The Treaty for the Non Proliferation of Nuclear Weapons places a number of obligations on signatory states. Non Nuclear Weapons States (NNWS) agree to forego the development of nuclear weapons or other nuclear explosive devices and those states with nuclear weapons (NWS) agree not to transfer the technology to NNWS which would allow them to develop such weapons or devices. To verify fulfilment of their obligation under the terms of the treaty, Article III requires each NNWS to accept safeguards as set forth in negotiated agreements with the International Atomic Energy Agency. Effectively two regimes of safeguards have been established - international carried out by the IAEA and national programs carried out by appropriate national bodies. The cornerstone for safeguards is the concept of materials accounting whereby all materials entering and leaving a specific location or crossing a "material balance area" boundary, are recorded in a materials ledger. In support of materials accounting records there is a need for materials verification measurements whose accuracy, frequency and timeliness are determined according to safeguards criteria. These measurements require special instrumentation. The development of such instrumentation for use by national authorities is of course the responsibility of that particular state. For international safeguards for which the IAEA is responsible, the IAEA does not develop its own instrumentation but relies upon voluntary contributions from member states to carry out its responsibilities under the NPT.

Classification of Safeguards Measurements

Safeguards measurements can be conveniently divided into two classes of techniques - Destructive Assay Techniques (DAT) and Non Destructive Assay Techniques (NDAT). The former are generally applied at major analytical laboratories, e.g. Siebersdorf Analytical Laboratory (SAL) to verify measurements made in the field essentially by NDAT. The advantage of DAT is their relatively higher accuracy. However from the safeguards viewpoint their principal liability is their lack of timeliness. Typical techniques include isotope dilution mass spectrometry, x-ray fluorescence, alpha counting and gamma ray and neutron resonance absorption. Typically the quality of the existing nuclear data is adequate for such measurements and the accuracy is generally superior to the accuracies inherent in the specific technique, e.g. radio chemical techniques, source preparation.

Non Destructive Assay techniques are performed in the field by IAEA inspectors generally to verify data supplied by operators of the specific installation. The instruments used must generally be small and readily transportable and extremely user friendly. Since the inspector may be obliged to apply measurement techniques to almost an infinite variety of samples the instruments must be fairly sophisticated despite these other properties. It is in the area of NDAT that the nuclear data needs are the most pressing. Figure 1 taken from a relatively old review by Dragnev[1] illustrates the range of NDAT employed. Table 1 from the review paper by Lammer[2] presents a summary of the safeguards methods that use nuclear data.

For the present review a comprehensive listing of nuclear data requirements for safeguards will not be given. This question is currently being addressed by the Nuclear Data Section of the IAEA who are preparing a handbook of Nuclear Data for Safeguards. This review will concentrate on several specific examples of NDAT where the data requirements are the most pressing. The cases quoted involve almost exclusively the NDA of batch samples which are extremely important as the majority of nuclear material is in this form. Not surprisingly these techniques generally involve neutron interactions.

Neutron Coincidence Counting

Neutron coincidence counting is the technique that is generally employed in the non destructive assay of bulk quantities of nuclear material and in particular, quantities of plutonium. Figure 2 taken from the review article by Menlove[3] illustrates the range of applications of the technique. Effectively the family tree of neutron coincidence counters can be subdivided into two classifications, (a) active in which external neutron sources are required, and (b) passive in which spontaneous fission within the sample itself is employed.

The technique makes use of the multiplicity of neutrons emitted in the fission process whether from spontaneous fission in passive systems or from neutron induced fission in active systems. The sample is surrounded by a moderating medium incorporating neutron sensitive detectors. The neutrons are emitted in the fission process within 10^{-16}s of fission, however, the detection process generally requires moderation

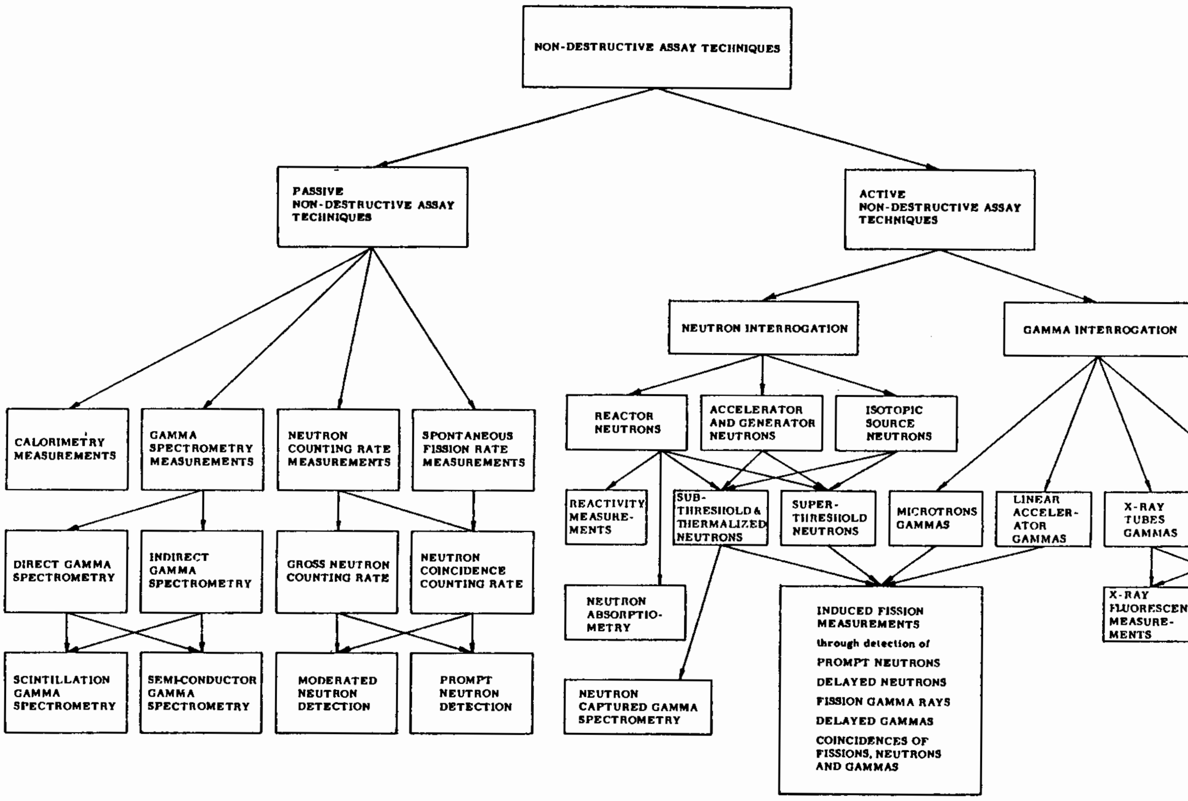

Fig.1. The structure of non destructive assay measurements (from Ref.1).

of the neutrons and their detection in time following the source fission event is determined principally by the lifetime of the neutron in the system comprising the detector and sample.

A great variety of instrumentation based on neutron coincidence counting has been developed as is clear from Figure 2. Most of these systems have employed polyethelyne moderators incorporating ^{3}He ionisation chambers for neutron detection. BF_3 tubes for neutron detection have generally not been used because of their poorer efficiency. (The Europeans prefer BF_3 because it is cheaper in Europe and because it is a little less gamma-ray sensitive). Other systems studied include fast neutron recoil detectors plus liquid and gas scintillators. Because of the very high count rates that typify neutron coincidence counting, considerable attention has been given to the development of specialised counting systems of which the shift register[4] warrants special mention.

There are three principal problems in neucron coincidence counting, namely matrix effects, (α,n) backgrounds and multiplication caused by both fission neutrons from the primary event and by the (α,n) background. These problems are handled at present by the development of sophisticated correction procedures and by the provision of an extensive inventory of reference standards which are intended to cover most measurement scenarios. Several more sophisticated detector systems are being studied in which it is hoped that sufficient additional information is generated to allow direct corrections for these three effects. The new detector systems include the high efficiency multiplicity counter by Krick et al.[5] the Fatima detector by Prosdocimi et al.[6] the Euratom Time Correlation Analyser by Bondar[7] and the liquid scintillator and hybrid neutron coincidence counter[8] at our laboratory.

For all of these detection systems there is a number of data problems where significantly higher accuracy is required than is presently available. The more important data problems and an assessment of their priority are discussed below .

The assay of plutonium in bulk material proceeds as follows. The isotopic composition of the plutonium is determined by gamma ray spectroscopy and the amount of ^{240}Pu is determined by the application of neutron coincidence counting.

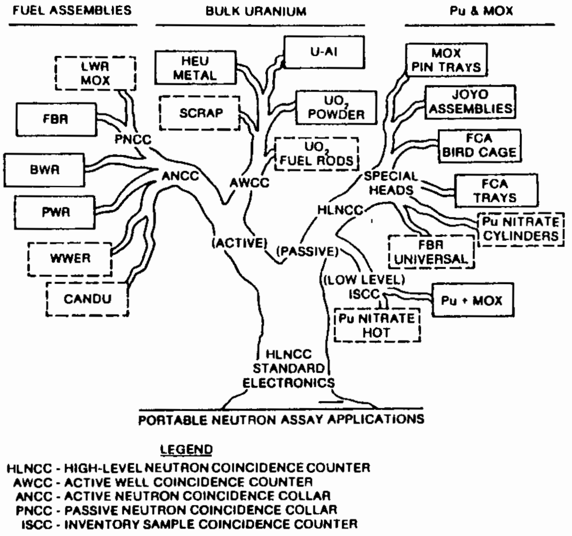

Fig.2. *Family-tree* diagram of active and passive neutron coincidence systems and applications based on the standard shift-register electronics package developed for the HLNCC. (from Ref.3).

Table 1. Safeguards Methods that use Nuclear Data (from Ref.2).

method	nuclear data	purpose
1)fresh fuel assay		
gamma spectrometry of recycled U fuel	half-life of U-232, γ-ray energies and intensities of its daughter products	correction for interference of U-232 daughters with U-235 γ-rays
gamma spectrometry of Pu containing fuel	half-lives of Pu isotopes and Am-241, γ-ray energies and intensities of their α-decay daughters	quantitative analysis of the γ-ray spectrum of Pu containing fuel
active neutron iterrogation (standards)	library of yields, half-lives, γ-ray energies and intensities of FP's	investigation of activation build-up in calibration standards
coincident counting techniques	prompt γ and prompt neutron multiplicity distributions from fission of U-235, Pu-239, (Pu-241) and spontaneous fission of Pu-238,240,242. Possibly delayed neutron yields as a function of time (induced fission)	optimization of coincident counting instrumentation layout
X-ray fluorescence	X-ray energies and intensities of Th, U, Pu	spectrum analysis
2) spent fuel assay		
FP γ-ray spectroscopy	thermal fission yields of Zr-95, Ru-106, Cs-133, Cs-137, Ba-140, Ce-144, Eu-153 from U-233, U-235, Pu-239 (Pu-241). Half-lives and γ-ray intensities of Zr-Nb-95, Ru-Rh-106, Cs-134, Cs-137, Ba-La-140, Ce-Pr-144, Eu-154; capture cross-sections of Cs-133, Eu-153.	γ-spectrum analysis, interpretation of measured activities and their ratios
passive neutron assay	Pu-238, 239, 240, 242, Am-241, Cm-242, 244: α-decay and spontaneous fission half-lives, $\bar{\nu}$ for spontaneous fission; fission and capture cross sections also for U-238, Pu-241, Am-242, Cm-243 and half-lives for the last 3 nuclides; O^{18} (α,n) cross section.	calculation of neutron emission from irradiated fuel for a better understanding of the method and interpretation of the results
3) dissolved fuel (reprocessing plant)		
isotope correlations	fission and capture cross-sections of U-234, 235, 238,Pu-238 to 242; cumulative fission yields from U-235, Pu-239, Pu-241; capture cross sections for: Kr-82 to 84, 86, Xe-131 to 136, Nd-143 to 146; half lives of Xe-133, 135	help to resolve discrepancies between measured and calculated correlations.

Clearly therefore, the spontaneous fission half life of ^{240}Pu is needed for all assays when standards are not used and the accuracy of $T_{\frac{1}{2}}$ for ^{240}Pu appears directly in the accuracy of the assay. Table 2 lists measurements of this parameter. There has been a great deal of scatter in the data, which is clearly unsatisfactory for this parameter. A new measurement in progress from Dytlewski et al.[9] aims at better than 1 percent which is the required precision. Spontaneous fission half lives are also required for the transuranic elements for this application, specifically data for ^{238}Pu and ^{242}Pu. Currently the data are adequate and are listed by Zucker and Holden[10].

Table 2. Measurement of the Spontaneous Fission Half Life for ^{240}Pu (from Ref.10).

Experiment		Value (10^{11} years)
Budtz-Jorgensen et al.	(1980)	1.15 ± 0.03
Fieldhouse et al.	(1967)	1.170 ± 0.025
White	(1967)	1.27 ± 0.05
Malkin et al.	(1963)	1.45 ± 0.02
Watt et al.	(1962)	1.34 ± 0.015
Mikheev et al.	(1959)	1.20
Chamberlain et al.	(1954)	1.20
Barclay et al.	(1954)	1.225 ± 0.030
Kindermann	(1953)	1.314 ± 0.026

The shape of the fission neutron spectra are also important in neutron coincidence counting. The neutron detectors that are employed are limited in size because of the need for tranportability. Consequently, their efficiencies vary fairly rapidly with source neutron energy. The understanding of the shape of fission neutron spectra has improved considerably in the last few years with consensus achieved for the shape of the reference standard, the spontaneous fission of ^{252}Cf[11], and the development of a number of theoretical descriptions such as the Madland Nix Model[12] and the Complex Evaporation Model[13]. In fact for most applications, theory provides a sufficiently accurate description for safeguards applications. However, in the specific case of the spontaneous fission of ^{240}Pu the situation is unsatisfactory. Current practice is to use a calculated spectrum from Madland Nix in which the average energy is given as 1.98 MeV. There have been only two experimental determinations of the spectrum. Average energies (Maxwellian) of 1.86 ± 0.045 and 1.80 ± 0.045 were obtained by Aleksandrov et al.[14] and Bonner et al.[15] respectively. Clearly there is a need for new measurements.

The $\bar{\nu}$ and P_ν distributions are of paramount importance in neutron coincidence counting. Both classes of data are generally in excellent shape following recent measurements by Gwin et al.[16], revision and extension of earlier measurements by Boldeman and Hines[17] and several comprehensive reviews of the experimental data by Zucker and Holden[18]. However, there remain two problem areas. Multiplication in neutron coincidence counting arises from secondary fission (either from fission neutrons or (α,n) neutrons) of the fissile elements in the sample. Because of the design of neutron coincidence counters the majority of the secondary fission is induced by neutrons of energy considerably higher than thermal energies. Measurements of P_ν for such fission reactions are extremely difficult to make. In fact, there are virtually no data in the literature. Zucker and Holden from the systematics of the little data[19] that exist have been unable to derive satisfactory values and strongly recommend additional measurements. A second area of concern is the $\bar{\nu}_p$ and P_ν data for spontaneous fission of ^{238}Pu for which a correction is necessary. The correction is significant for high-burnup reactor-grade plutonium which the IAEA must measure, and for $^{238}PuO_2$ heat sources that are used to calibrate calorimeters, which in turn are used to calibrate neutron coincidence counters.

Another area which has caused concern is the data for the (α,n) cross section for ^{19}F which is a major source of background neutrons in typical samples for neutron coincidence counting. The neutron emission spectra for this reaction are also required with improved precision.

Finally with the development of increasingly sophisticated neutron coincidence counting systems, there has been a need for a better understanding of the fission process itself. Important matters are the complete systematics of the multiplicity and total energy of the gamma ray emission and the variation of neutron parameters (e.g. energy) with multiplicity.

Decay Data Problems

Generally, the accuracy of the half lives for the isotopes typically measured in safeguards inspections are better than the errors inherent in either the measurement technique on the interpretation based on experimental data. Several minor problems are known. For Pu isotopic measurements, there is a problem with the 152.7 keV gamma ray from the decay of ^{238}Pu. Current practice in safeguards measurements is to use a value 2.5 percent lower than the published value. In addition there are several other biases in the experimental data that need consideration[19].

Conclusion

It may be concluded from present review, that the accuracy of nuclear data employed in safeguards measurements is generally satisfactory except for a few problem areas referred to above. However, the development of safeguards instrumentation is continuing and as more sophisticated techniques are developed greater demands will be placed on the accuracy of the appropriate data. This review was hampered to some extent by the lack of a current file of nuclear data problems in safeguards. It is recommended that the IAEA give higher priority to the preparation of a data handbook for safeguards applications.

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