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## Editorial

In 2006, ESARDA introduced in issue 34 of its bulletin the idea of special issues aimed at addressing in the same journal topical subjects, thematic areas and detailed studies such as benchmarking and inter-comparison exercises. In this issue 48, we are pleased to continue this tradition, presenting the results of two collaborative projects in the field of nuclear material measurement. Both documents have been extensively worked and peer reviewed by a large number of experts.

*International Target Values (ITV) 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials* provides the latest update to a document that originated within the ESARDA DA WG and has since been adopted by safeguards authorities and operators. This latest edition is reproduced from an original Safeguards Technical Report of

the IAEA, prepared in collaboration with a number of parties with expertise in nuclear material measurement including the DA and NDA Working Groups of ESARDA.

*Performance Values for Non-Destructive Assay (NDA) Techniques Applied to Wastes* offers an important extension in consideration of performance values for nuclear material measurements and reflects a range of work evaluated within the ESARDA NDA WG. This original ESARDA report provides the first dedicated review of performance values for waste assay since publication of the first NDA performance values in 1993. As such, we hope that this document too will prove to be a valuable reference for safeguards practitioners.

**The Editor**  
**Hamid Tagziria**



# **International Target Values 2010 for Measurement Uncertainties in Safeguarding Nuclear Materials**

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**November 2010**

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## Abstract

This issue of the International Target Values (ITVs) represents the sixth revision, following the first release of such tables issued in 1979 by the ESARDA/WGDA. The ITVs are uncertainties to be considered in judging the reliability of analytical techniques applied to industrial nuclear and fissile material, which are subject to safeguards verification. The tabulated values represent estimates of the 'state of the practice' which should be achievable under routine measurement conditions. The most recent standard conventions in representing uncertainty have been

considered, while maintaining a format that allows comparison with the previous releases of the ITVs. The present report explains why target values are needed, how the concept evolved and how they relate to the operator's and inspector's measurement systems. The ITVs-2010 are intended to be used by plant operators and safeguards organizations, as a reference of the quality of measurements achievable in nuclear material accountancy, and for planning purposes. The report suggests that the use of ITVs can be beneficial for statistical inferences regarding the significance of operator-inspector differences whenever valid performance values are not available.

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## List of Acronyms

<b>ABACC</b>	Agencia Brasileiro-Argentina de Contabilidade e Controle de Materiais Nucleares
<b>ANSI</b>	American National Standards Institute, USA
<b>BIPM</b>	International Bureau on Weights and Measures
<b>CGM</b>	Consultants' Group Meeting
<b>D</b>	MUF difference estimate
<b>DA</b>	Destructive Analysis
<b>EQRAIN</b>	Programme d'Evaluation de la Qualité des Résultats d'Analyse dans l'Industrie Nucléaire, CETAMA, France
<b>ESARDA</b>	European Safeguards Research and Development Association
<b>ESARDA/WGDA</b>	ESARDA Working Group on Techniques and Standards for Destructive Analysis
<b>ESARDA/WGNDA</b>	ESARDA Working Group on NDA methods
<b>GS</b>	Gamma spectrometry
<b>GUM</b>	Guide to the Expression of Uncertainty in Measurement
<b>HEU</b>	High enriched uranium ( $^{235}\text{U}$ abundance > 20%)
<b>IAEA</b>	International Atomic Energy Agency
<b>IEC</b>	International Electrochemical Commission
<b>IFCC</b>	International Federation of Clinical Chemistry and Laboratory Medicine
<b>INMM</b>	Institute of Nuclear Materials Management, USA
<b>ISO</b>	International Organization for Standardization, Geneva, Switzerland
<b>ISO/TC85/SC5</b>	ISO/TC85 Subcommittee 5 on Nuclear Measurements
<b>ISO/TC85/SC5/WG1</b>	Working Group on Analytical Methodology in the Nuclear Fuel Cycle
<b>ITVs</b>	International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials
<b>IUPAP</b>	International Union of Pure and Applied Physics
<b>JCGM</b>	Joint Committee for Guides in Metrology
<b>LEU</b>	Low enriched uranium (0.7 % < $^{235}\text{U}$ abundance < 20 %)
<b>LSD</b>	Large Size Dry Spike for Isotope Dilution Mass Spectrometry
<b>LWR</b>	Light Water Nuclear Reactor
<b>MOX</b>	Mixed Uranium/Plutonium Oxide Nuclear Fuel Material
<b>MUF</b>	Material Unaccounted For
<b>NDA</b>	Non-Destructive Analysis
<b>NRTA</b>	Near Real Time Accountancy
<b>OIML</b>	International Organization of Legal Metrology
<b>QA</b>	Quality Assurance
<b>QC</b>	Quality Control
<b>SAGSI</b>	IAEA Standing Advisory Group on Safeguards Implementation
<b>SI</b>	International System of Measurement Units
<b>VIM</b>	International Vocabulary of Metrology
<b>XRFA</b>	X-ray fluorescence analysis

## 1. Introduction

Safeguarding nuclear material involves a quantitative verification of the accountancy of fissile materials by independent measurements. The effectiveness of these verifications depends to a great extent upon the quality of the accountancy measurements achieved by both the facility operator and the safeguards inspectorate. For this reason a typical safeguards agreement based on INFCIRC/153<sup>1</sup> stipulates that:

*The Agreement should provide that the system of measurements on which the records used for the preparation of reports are based shall either conform to the latest international standards or be equivalent in quality to such standards.*

Although the above requirement is directed to the facility operators, it indeed applies equally well to the safeguards inspectorates.

The International Atomic Energy Agency (IAEA) had defined in the 1970s a set of international standards of nuclear material accountancy<sup>2</sup>, which lists the 'values of measurement uncertainty expected for closing a material balance' for five different types of nuclear facilities. In the absence of relevant international standards of measurements, safeguards evaluators, as well as plant measurement specialists, need references regarding the performance capabilities of measurement methods used for the determination of the volume or mass of a material, for its sampling, and for its elemental and isotopic assays. Such information is needed for the various nuclear materials encountered in the nuclear fuel cycle.

The Working Group on Techniques and Standards for Destructive Analysis (WGDA) of the European Safeguards Research and Development Association (ESARDA) pioneered the way in 1979 by presenting a list of 'Target Values' for the uncertainty components in destructive analytical methods<sup>3</sup> to the safeguards authorities of Euratom and of the IAEA. Revised estimates were prepared in collaboration and published as the 1983 Target Values<sup>4</sup> after four years of extensive discussion and consultation with and within operators' laboratories and safeguards organizations. The

international acceptance of the concept grew further with the next review, which involved, besides the ESARDA/WGDA and IAEA, the active participation of the members of two specialized committees of the Institute of Nuclear Materials Management (INMM). The 1987 Target Values, published as a result of this review<sup>5</sup>, defined, as in the previous editions, the values of 'random' and 'systematic' error parameters to be aimed for in elemental and isotopic analyses of the most significant types of materials using common destructive analytical methods. The same groups took a new step in the 1988 edition<sup>6</sup> when they agreed to define the values of the random error parameter to be met in the elemental assays as a result of sampling.

Following a 1988 recommendation of the IAEA Standing Advisory Group on Safeguards Implementation (SAGSI), the IAEA convened a Consultants Group Meeting in June 1991 to provide expert advice on international standards of measurements applicable to safeguards data. A concept of International Target Values (ITVs) was proposed on the model of the 1988 ESARDA Target Values and included estimates of the 'random and systematic error' uncertainties originating from the measurements of volumes or masses of nuclear materials. The scope of ITVs was also extended beyond destructive analysis (DA) methods to include non-destructive assay (NDA) methods, which had won acceptance as accountancy verification tools.

Specialists from four continents took part in the discussion of the proposed concept. The result was the publication of an IAEA Safeguards Technical Report in March 1993, entitled '1993 International Target Values for Uncertainty Components in Fissile Isotope and Element Accountancy for the Effective Safeguarding of Nuclear Materials'<sup>7</sup>. Articles in the ESARDA Bulletin<sup>8</sup> and in the Journal of the INMM<sup>9</sup> widely publicized the IAEA technical report. The report itself was translated into Japanese<sup>10</sup>.

<sup>1</sup> INTERNATIONAL ATOMIC ENERGY AGENCY, The Structure and Content of Agreements between the Agency and States Required in Connection with the Treaty on the Non-Proliferation of Nuclear Weapons. INFCIRC/153 (corrected), para. 55 Vienna (1972).

<sup>2</sup> INTERNATIONAL ATOMIC ENERGY AGENCY, IAEA Safeguards Glossary, 2001 edition, para. 6.35, International Nuclear Verification Series No. 3, Vienna (2002).

<sup>3</sup> DE BIEVRE, P., DE REGGE, P., "1979 Target Values for uncertainty components of destructive analysis methods", Minutes of the Meeting of the ESARDA/WGDA in Brussels, April 23-24, 1979, SCK, Mol, Belgium (1979).

<sup>4</sup> DE BIEVRE, P., et al., 1983 Target Values for uncertainty components in fissile element and isotope assay, ESARDA Bulletin 6 (1983) 1.

<sup>5</sup> DE BIEVRE, P., et al., "1987 Target Values for uncertainty components in fissile isotope and element assay", (IAEA Proc. Symp. Nucl. Safeguards Technology Vienna, 1986), IAEA, Vienna (1987) 649-662.

<sup>6</sup> DE BIEVRE, P., et al., "Random uncertainties in sampling and element assay of nuclear materials; target values 1988", ESARDA Bulletin 13 (1987) 8.

<sup>7</sup> KUHN, E., et al., "1993 International Target Values for Uncertainty Components in Fissile Isotope and Element Accountancy for the Effective Safeguarding of Nuclear Materials", IAEA STR-294, IAEA, Vienna (1994).

<sup>8</sup> DERON, S., et al., "1993 International Target Values for Uncertainty Components in Measurements of Nuclear Material for Safeguards Purposes", ESARDA Bulletin 23, JRC, Ispra (1994).

<sup>9</sup> DERON, S., et al., "1993 International Target Values for Uncertainty Components in Fissile Isotope and Element Accountancy for the Effective Safeguarding of Nuclear Materials", Journal of Nuclear Materials Management, Volume XXII, Number II, January 1997.

<sup>10</sup> "1993 International Target Values for Uncertainty Components in Fissile Isotope and Element Accountancy for the Effective Safeguarding of Nuclear Materials", Japanese Translation, Science and Technology Agency, Japan, (1993).

In 2000, international experts reviewed the experience gained with the use of the 1993 ITVs and the progress made in accountancy and safeguards verification measurements. Subsequently, 'International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Materials' was published as an IAEA Safeguards Technical Report in April 2001<sup>11</sup>, in the ESARDA Bulletin<sup>12</sup> and by INMM<sup>13</sup>.

Each ITV bears a date, reflecting a recognition that the quality of measurements may change and that new methods and instruments may be developed and implemented. The ITVs also reflect the current understanding of the structure of the uncertainty components in nuclear material accountancy measurements which may change in the future as this understanding improves or varies.

In preparation for the ITVs-2010 the IAEA conducted 'Verification Measurement Performance Evaluations', using data reported by facility operators and the results of independent measurements performed on the same material by the inspectors<sup>14</sup>. These historical operator-inspector paired data, accumulated from over 20 years, represent the most relevant and complete set of information. Based on these performance evaluations and the IAEA's experience from using the ITVs-2000, a set of draft ITVs-2010 tables were prepared, which included some changes in the target values, the deletion and addition of analytical techniques or methods, and changes in the format of the tables.

As in the earlier formulation and revision of ITVs, the IAEA counted on the expertise available in the Working Groups for DA and NDA of ESARDA, the ANSI/INMM 5.1 Analytical Chemistry Laboratory Measurement Control Committee, the Working Group 1 on Analytical Methodology in the Nuclear Fuel Cycle of the ISO TC85/SC5 Subcommittee, the Japanese ITV-2010 Expert Group, and the inspectorates of Euratom and ABACC. The above panels and organizations were asked to review the draft document and provide comments. In addition they were asked to report on measurement quality experience, as derived from QC/QA and inter-laboratory programmes, instrument qualification, or from verification activities. Representatives of the above

groups and organizations participated in a Consultants Group Meeting, convened at the IAEA in March 2010. Their comments and recommendations are reflected in this document.

As with the previous lists, the ITVs-2010 should be achievable henceforth under the conditions normally encountered in typical industrial laboratories or during actual safeguards inspections. They do not represent the measurement uncertainties, which would only be achieved under exceptional or ideal laboratory conditions, or with most recently developed methods, which have not yet found wide use for daily and routine measurements. It is expected that the ITVs-2010 will continue to be a motivating goal for beginner laboratories and be used as an independent reference for experienced laboratories and safeguards evaluators. With the growing acceptance of modern quality assurance concepts it is suggested that the ITVs-2010 can also constitute a good reference against which analytical laboratories would validate their measurement systems.

## 2. Safeguards Accountancy and Verification Measurements

Facility accountancy and safeguards verification measurement data are the major source of information on which the ITVs are based. Conversely, the derived ITVs are primarily used to evaluate the accountancy and verification results in safeguards. Therefore, a description of the safeguards measurement system in connection with the sources of uncertainties at each step of the measurement process is important in order to understand how the derived ITVs should be interpreted.

Figure 1 describes the basic measurement scheme followed in safeguards verification measurements. For each inspection,  $j$ , the inspector selects, in accordance with a random sampling plan, the items or batches of nuclear materials to be verified by an independent measurement. The inspector then compares the result of the verification measurement,  $Y_{ij}$ , to the result,  $X_{ij}$ , which the operator has obtained on the same batch or item,  $i$ , and which the operator has declared to the inspectorate. The ability of the inspector to detect whether the difference  $d_{ij}$  is significantly different from zero depends upon the overall uncertainties in the results  $X_{ij}$  and  $Y_{ij}$ . Figure 1 identifies the major steps of the measurement process where uncertainties can arise, although not all steps may be relevant for every method (e.g., several of the steps may be omitted or combined under a single step for NDA methods).

Step 1 corresponds to the measurement of the volume or mass of the item or batch of material. This so-called 'bulk' measurement, when needed, takes place in the plant area and involves a calibration procedure.

<sup>11</sup> AIGNER, H., et. al., "International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Material", IAEA, STR-327, IAEA, Vienna (2001).

<sup>12</sup> AIGNER, H., et. al., "International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Material", ESARDA Bulletin 31, JRC Ispra (2002).

<sup>13</sup> AIGNER, H., et. al., "International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Material", Journal of Nuclear Materials Management, Vol. XXX, No. 2 ([www.inmm.org/itvs/1698.htm](http://www.inmm.org/itvs/1698.htm)).

<sup>14</sup> L. Bevaart, W. Fuhr, K. Zhao, and E. Kuhn, 'International Target Values for Measurement Uncertainties in Safeguarding Nuclear Materials: preparing for the 2010 Update', 31st ESARDA Annual Meeting, Vilnius, May 25-28, 2009.

Step 2, the 'sampling', involves removing, for the purpose of the analytical measurement, a representative portion of the material from the batch or item to be analyzed. This may be a complete item in the case of an NDA measurement.

Step 3 concerns the precautions which must be taken in the way the sample is 'conditioned' and packaged at the sampling station so that all characteristics to be measured are preserved during its transport to the location or laboratory where the characteristics will be measured. For instance, the 'conditioning' of powder or solution samples must include a weight measurement to facilitate a correction for weight changes between sampling and analysis.

Step 4, the 'shipment', is the transport of the sample to the location where it can be measured.

Step 5, the 'treatment', is intended to bring the sample into the most appropriate geometrical, physical and/or chemical form for the measurement. The treatment of a sample taken for destructive analysis may involve a sequence of individual steps, such as sub-sampling, dissolution, dilution, spiking, chemical treatment or chemical separation, etc.

Step 6 represents the 'measurement' itself. In general terms, a measurement is based on a calibration from which the parameters linking the observed signal and the measurand are determined. Typical examples are HLNC calibration curves for Pu mass determination, calibrations of gamma spectrometers for  $^{235}\text{U}$  abundance determination, or the determination of the mass-discrimination correction factor for a mass-spectrometer. The standardization of a titrant solution is another example of a calibration, although it is frequently not recognized as such. Calibration functions may be as simple as a single calibration factor (actually representing a straight line through the origin), or may be complex and represented by an empirically determined calibration curve. Calibrations based on recognized references, such as certified reference materials or well-known physical constants, establish the traceability chain between the measurement result and the International System of Units (SI). Calibrations may be valid and used without modifications for a long time, repeated on a daily basis or even performed with each individual measurement. Sometimes, elaborate calibration exercises for determining the fundamental characteristics of the calibration function are combined with more frequently repeated 'normalization' measurements to correct for short-term effects or minor deviations from the overall calibration function at the specific working range.

Step 7, the 'calculation', consists in transforming the results of the physical or chemical measurement obtained in the preceding step into an estimate of the amount of fissile element or isotope in item or batch  $i$ . Particularly when the operator and inspector use DA, this involves estimating the total element content, by combining the result of the bulk measurement  $w_{ij}$  with the elemental concentration  $c_{ij}$ . In the case of Uranium materials, this is combined with the isotope abundances  $f_{ij}$  of the fissile isotope ( $^{235}\text{U}$  or  $^{233}\text{U}$ ) to yield a measure of the amount of fissile isotope in item or batch  $i$ , according to equations (1) and (2), respectively.

$$X_{ij} = w(O)_{ij} \cdot c(O)_{ij} \cdot f(O)_{ij} \quad (1)$$

$$Y_{ij} = w(I)_{ij} \cdot c(I)_{ij} \cdot f(I)_{ij} \quad (2)$$

Every stage of the process, starting with bulk measurements, must be performed under well-controlled conditions. Hence, quality control measures are imperative at every step of the process. Quality control on sampling can be done by taking replicate samples after different mixing times or taking samples from a number of items of the same batch of bulk materials. Quality control materials or samples can be introduced at specific steps to monitor the quality of the whole process or any part of it, including the conditioning and shipment steps. Figure 1 shows an example where control materials are used independently by the operator and the inspector to check the quality of the processes following the sampling. Quality control measures should be performed in the frame of a documented quality system.

The uncertainties in the measurements of element concentrations and isotope abundances in the ITVs refer to the combined effects of the uncertainties in Steps 3 to 6 occurring after the taking of the sample in Step 2.

Step 8, the 'reporting' of the results, is purely clerical but unfortunately it can be a source of errors. Uncertainties arising from such errors are not considered in the ITVs proposed in this document. Yet it is essential that appropriate quality assurance measures be taken to avoid the occurrence of clerical errors.

When NDA is used, attention is mostly focused on the measurement (Step 6), as the preceding steps have usually less impact or may even be omitted. For example, bulk measurements and sampling are not needed if the NDA method allows direct measurement of the total amount of fissile element or isotope contained in a whole item or batch of nuclear material, as with various neutron counters.

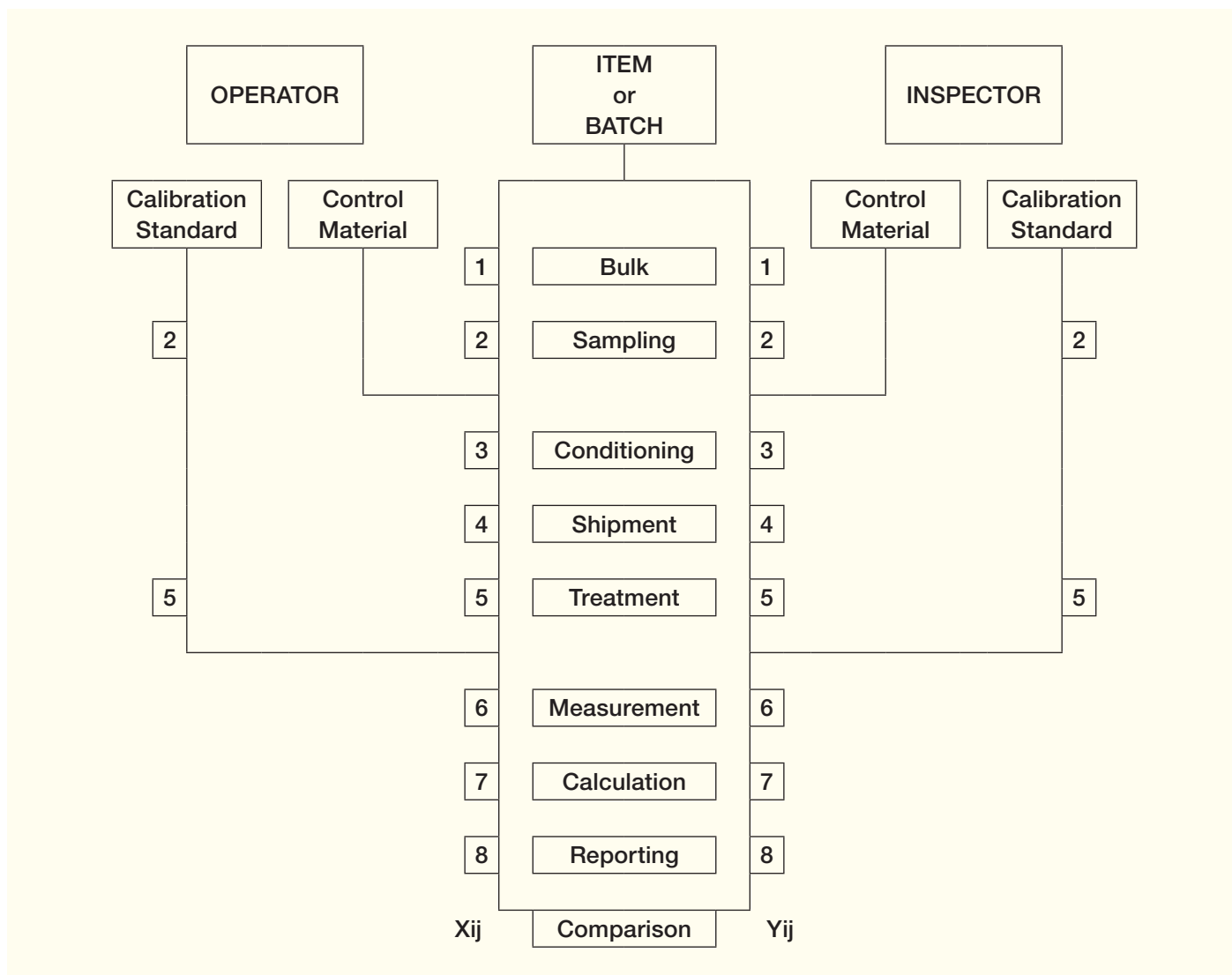


Figure 1: Accountancy and Verification Measurement Scheme for Item i during Inspection j

### 3. Evaluation of Safeguards Accountancy Verification Measurements

Nuclear material accountancy involves measurement systems for the determination of particular quantities of interest (**measurands**) such as the net weight or volume of an item (bulk measurement), the elemental and/or isotopic concentration for sampled material, and, in the case of NDA, the mass of nuclear material in an item.

Measurement errors are inherent to all the accountancy measurement results for both operators' declarations and inspectors' verification. The **measurement error** of a given result, defined mathematically as the result minus the true value, often represents the combined effect of many sources of errors in the measurement system, which might include statistical sampling error, bulk measurement error, material sampling error, analytical error, and other errors.

With regard to the different behaviours of measurement results on nuclear material accountancy, three broad

categories of errors related to safeguards applications can be identified as follows<sup>15, 16</sup>:

- **random error**, varies in an unpredictable way under **repeatability conditions**, i.e., conditions where independent results are obtained with the same method on identical items using the same equipment by the same operator within short intervals of time. Counting statistics or the repeatability of measurements under constant conditions are typical examples of random errors. The effects of random errors can be reduced by repeated measurements, but it is not possible to correct random errors.

<sup>15</sup> ISO 3534-1:1993, "Statistics - Vocabulary and Symbols - Part 1: Probability and general statistical terms", ISO Standards Handbook, Statistical Methods for Quality Control, Vol. 1, Fourth Edition, ISO, Geneva, (1995).

<sup>16</sup> BIPM, IEC, IFCC, ISO, IUPAC, IUPAP, OIML, International Vocabulary of Basic and General Terms in Metrology, Second Edition, ISO, Geneva, (1993).



- **bias**, remains constant under **reproducibility conditions**, i.e., conditions where results are obtained using the same method on identical items when using different equipment operated by different operators.
- **short-term systematic error**, remains constant for a short term when measurement conditions or settings, typically expressed as parameters such as calibration curves, normalization parameters, subtracted background, etc., are not altered while varying in an unpredictable way on a long-term perspective due to the random changes of these conditions or settings.

**Uncertainty** is a statistical parameter, associated with the result of a measurement, to characterize the dispersion of measurement values that could reasonably be attributed to a measurand. This parameter may be a standard deviation, in which case the uncertainty is called standard uncertainty, or the width of a confidence level. In the context of expressing uncertainties in safeguards accountancy measurements, **random uncertainty component,  $u(r)$** , and **systematic uncertainty component,  $u(s)$** , are simply defined as the standard deviations of the random errors and short-term systematic errors, respectively.

The operator-inspector paired differences are examined to determine whether they are commensurate with the characteristic uncertainties of the operators' and inspectors' measurement systems. For this purpose, the inspectors need to quantify the major uncertainties of the actual data collected during their verification activities.

A basic assumption for the estimation of the uncertainty components is that  $u(r)$  and  $u(s)$  are characteristics of the type of material, its chemical and physical form and of the method of measurement. A further assumption is that  $u(s)$  is constant for a given inspection period, but that it varies in a random manner from one inspection to another, for both the operator and the inspector.

Consequently, the inspectors group the data pairs, the operator's value  $X$  and the inspector's value  $Y$ , originating from one inspection period,  $j$ , by material balance areas (MBA), by strata of materials of similar characteristics and by measurement methods<sup>17</sup>. For a given MBA and stratum, the operator-inspector relative difference,  $d_{ij}$ , for item  $i$  in inspection  $j$ , is given by:

$$d_{ij} = (X_{ij} - Y_{ij}) / X_{ij} \quad (3)$$

where

- $i = 1, 2, \dots, m_j$
- $j = 1, 2, \dots, K$
- $K$  = the number of inspections
- $m_j$  = the number of items verified during inspection  $j$

The assumed error model is given by:

$$d_{ij} = d + \Delta_j + \varepsilon_{ij} \quad (4)$$

where

- $d$  = the mean difference over the  $K$  inspections,
- $\Delta_j$  = the systematic error of the operator-inspector difference during inspection  $j$ , and
- $\varepsilon_{ij}$  = the random error of the operator-inspector difference for item  $i$  during inspection  $j$ .

In addition, the expected values of  $\Delta$  and  $e$  are assumed to be zero. An analysis of variance of the operator-inspector differences,  $d_{ij}$ , according to this model equation, gives the estimates of the variance  $s^2(\varepsilon)$  of the random component and of the variance  $s^2(\Delta)$  of the systematic component within the given inspection period<sup>18</sup>. In performing this analysis of variance, it is generally assumed that  $\Delta_j$  and  $\varepsilon_{ij}$  are normally distributed and that the variances of the random error are the same for all inspections. The set of results are therefore screened for outliers prior to performing this evaluation.

Paired comparisons of this type are done separately for bulk measurements, element concentrations and isotope abundances, as well as for the masses of fissile elements and isotopes. Under the assumption that the errors of the operator and of the inspector are independent from each other, one obtains, for each type of measurement, an estimate of the combination of the actual uncertainty components for the operator's and inspector's measurement systems:

$$s^2(\varepsilon) = u^2(r,O) + u^2(r,I) \quad (5)$$

$$s^2(\Delta) = u^2(s,O) + u^2(s,I) \quad (6)$$

where  $u(r,O)$  and  $u(r,I)$  are the standard random uncertainty components, and  $u(s,O)$  and  $u(s,I)$  are the standard systematic uncertainty components for the operator and the inspector respectively.

In one simplified situation where the operator's and inspector's uncertainty parameters can be assumed to be equal as a result of using similar measurement methods,  $u(r)$  and  $u(s)$  can be determined based on  $s^2(\varepsilon)/2$  and  $s^2(\Delta)/2$ , respectively, for both the operator and the inspector. In another simplified situation where the operator's DA results may be compared with much less precise and/or accurate inspector's results obtained for example by some NDA methods,  $u(r,I)$  and  $u(s,I)$  can be estimated based on  $s^2(\varepsilon)$  and  $s^2(\Delta)$ , respectively, whereas  $u(r,O)$  and  $u(s,O)$  must be derived from a comparison with inspector's measurements also obtained by DA.

<sup>17</sup> IAEA, "Statistical Concepts and Techniques for IAEA Safeguards", Fifth Edition, IAEA/SG/SCT/5, IAEA, Vienna (1998)

<sup>18</sup> JAECH, J.L., "Statistical analysis of measurement errors", Wiley, New York (1985).

In the IAEA data analysis, various statistical techniques<sup>17</sup> applicable to more general cases are used to derive separate estimates of the operator's and inspector's uncertainty parameters based on the collection of historical operator-inspector differences. The results of these evaluations are 'Performance Values' obtained for each MBA/stratum/measurement method combination. These Performance Values, which are generally updated once a year as more historical data becomes available for DA and NDA, are used in planning inspections<sup>19</sup> and in drawing inferences based on the operator's declared values and the inspector's verification measurement values. There are, however, situations where insufficient historical data is available to derive Performance Values. In these instances ITVs are used until sufficient measurement data are accumulated.

The most recent 'Performance Values' are the basis for updating the two columns of random and systematic uncertainty components,  $u(r)$  and  $u(s)$ , in this ITV-2010 document for each measurement method. New to the ITV-2010 document is a column labelled ITV, which reflects an internationally-adopted standard approach to measurement uncertainty evaluation (*Guide to the Expression of Uncertainty in Measurement* – GUM). This column complements the use of ITVs in the evaluation of operator-inspector data and is provided as a reference for the laboratories.

#### 4. GUM and the use of ITVs by measurement laboratories

The current *Guide to the Expression of Uncertainty in Measurement* (GUM) was published in 2008 by the Joint Committee for Guides in Metrology (JCGM) in the name of the JCGM member organisations (BIPM, IEC, IFCC, ILAC, ISO, IUPAC, IUPAP and OIML) as JCGM 100:2008<sup>20</sup>.

The goal of the GUM is to provide measurement laboratories with a standardized, methodical approach to determining a quantitative statement of the measurement uncertainty associated with a measurement result. This standardized approach helps to ensure inter-comparability of results between methods and laboratories, ensures transparency (and traceability) in calculation, and by design adds some additional assurance that laboratories are identifying significant contributors to their measurement's uncertainties. This approach has been adopted by many safeguards laboratories and provides important information to laboratory operators and internal and external evaluators. The GUM is not intended to replace quality control systems or other data verification/validation schemes, but

to provide laboratory staff, measurement data users, and regulators with useful, comparable information regarding the performance of particular measurement methods on particular sample types.

The GUM approach consists of modelling the measurement process using mathematical relationships between a measurand and its input quantities, and determining the combined standard uncertainty for a measurement result using the standard procedure of error propagation from the standard uncertainties of the input quantities based on expert knowledge, QC data or other means, and reporting the result with the inclusion of the expanded uncertainty (a coverage factor multiplied by the combined standard uncertainty). In addition, it is recommended that the uncertainty budget or a statement indicating the significant contributors to the uncertainty, and the model equation(s) used be provided.

Typically, a laboratory will establish GUM uncertainties for each of its measurement methods during method validation. Once a GUM evaluation has been completed, the laboratory may choose to monitor the performance of the method and the constraints on those input quantities which affect the uncertainty (e.g. temperature) over a specified period of time or number of measurements and either re-calculate the uncertainty for each measurement using a template, or report a conservative 'stock' uncertainty that is validated by comparing the latest set of results with the previously validated results to ensure the measurement uncertainty remains unchanged. Thus, users do not have to perform a complete GUM uncertainty evaluation every time they perform a routine measurement. Non-routine measurements or significant changes in the sample, calibration standards, or measurement system would require a re-evaluation of measurement uncertainty.

The use of GUM uncertainties in inter-laboratory comparisons is particularly useful for participants and regulators, as it allows all users to immediately compare performance between facilities for the variety of methods and samples in use. The GUM uncertainties in this case may serve as good representations of typical measurement uncertainties achievable by comparable laboratories with real-world samples.

**According to GUM, uncertainty** is a statistical parameter associated with the result of a measurement to characterize the dispersion of measurement values that could reasonably be attributed to a measurand. This ITV-2010 document seeks to link the GUM approach essential for the measurement laboratories with the needs of the evaluators. The GUM does not have the concept of a random and systematic uncertainty component<sup>20</sup>, as described earlier.

For this reason, new to the ITV-2010 document, the column labelled 'ITV' provides safeguards laboratories with

<sup>19</sup> JAECH, J., Russell, M., Algorithms to Calculate Sample Sizes for Inspection Sampling Plans, STR-261, Rev. 1, IAEA, Vienna (1991).

<sup>20</sup> JCGM 100:2008, Evaluation of measurement data – Guide to the expression of uncertainty in measurement, Joint Committee for Guides in Metrology.

additional useful information, reflecting the combination of the  $u(r)$  and  $u(s)$  parameter. This is provided as a useful reference for the laboratories, and reflects a state-of-the-practice reasonable expanded uncertainty under routine measurement conditions.

## 5. The International Target Values 2010

The International Target Values 2010 for Measurement Uncertainties (ITVs-2010) are values for uncertainties associated with a single determination result, e.g., this may be the result reported by one laboratory on one sample (independent of the analytical scheme applied internally in the laboratory), or the result of an NDA measurement performed on a single item. The ITVs-2010 take into account actual practical experiences and should be achievable today under the conditions normally encountered in typical industrial laboratories or during safeguards inspections.

In preparation for the update of the ITVs-2000, the IAEA conducted 'Verification Measurement Performance Evaluations', using data reported by facility operators and the results of independent verification measurements<sup>14</sup>. The IAEA's database, consisting of operators' declared and inspectors' verified data, most likely represents the largest data set of results obtained for the various types of industrial materials and by the most commonly used measurement techniques. Based on these performance evaluations and the IAEA's experience in using the ITVs, a set of draft ITVs-2010 tables were prepared and distributed to the Working Groups for DA and NDA of ESARDA, the ANSI/INMM 5.1 Analytical Chemistry Laboratory Measurement Control Committee, the Working Group 1 on Analytical Methodology in the Nuclear Fuel Cycle of the ISO TC85/SC5 Subcommittee, the Japanese ITV-2010 Expert Group, and the inspectorates of Euratom and ABACC for review and discussion.

In addition the above panels and organizations were asked to report on measurement quality experience, as derived from QC/QA and inter-laboratory programmes<sup>21, 22, 23</sup>, instrument qualification, or from verification activities. Representatives of the above groups and organizations participated in a Consultants' Group Meeting (CGM), convened at the IAEA in March 2010.

The ITVs-2010 were thus selected by the CGM on the basis of a critical discussion of the inspectorates' performance evaluations of actual historical data and their comparison with the ITVs-2000. They were also chosen to be consistent with uncertainty assessments available from

experimental validation of measurement methods and instrumentation, from inter-laboratory measurement evaluation programmes, or from individual laboratories.

The ITVs-2010 are applicable to the accountancy data collected by the inspectorates. They do not represent the ultimately achievable performance of a measurement system, which would be obtained under exceptional or ideal laboratory conditions. However, they reflect reasonably well the progress observed during the past several years in the routine performance of measurements carried out for the purpose of material accountancy and verification.

The ITVs-2010 also intend to take into account all sources of measurement uncertainties, including sources which may not be apparent in Performance Values resulting from paired comparisons of operators' and inspectors' measurements.

## 6. Structure and Content of the ITVs-2010

The presentation of the 2000 ITVs involved seven different tables. Based on the experience in using them, a different format was chosen for the presentation of the ITVs-2010, which now include the following 10 tables:

<b>Table 1</b>	Measurement Method/Instrument Codes
<b>Table 2</b>	Bulk and Density Measurements
<b>Table 3</b>	Sampling Uncertainties for Element Concentration and <sup>235</sup> U Abundance
<b>Table 4a</b>	Uranium Element Concentration Measurements (DA)
<b>Table 4b</b>	Plutonium Element Concentration Measurements (DA)
<b>Table 5a</b>	<sup>235</sup> U Abundance Measurements (DA)
<b>Table 5b</b>	<sup>235</sup> U Abundance Measurements (NDA)
<b>Table 6</b>	Plutonium Isotope Assay of Pu and U/Pu materials
<b>Table 7a</b>	Total Mass of <sup>235</sup> U (direct NDA)
<b>Table 7b</b>	Total Mass of Pu (direct NDA)

- Each of the tables 2 and 4a to 7b identifies separate ITVs according to the type of material and measurement method, as appropriate.
- Table 3 lists the  $u(r)$  values for the step of sampling nuclear materials. It has not yet been possible to propose  $u(s)$  values applicable to sampling, except for a few cases where this parameter was found to be measurable. It should also be noted that random sampling errors were frequently not assessed on the basis of experimental data and are based on expert opinion and facility experience.
- In the tables the two parameters,  **$u(r)$**  and  **$u(s)$** , characterize the quality, which should be aimed for in a specific measurement of a given material using a specified

<sup>21</sup> Safeguards Measurement Evaluation Program, SMEP ([www.nbl.doe.gov](http://www.nbl.doe.gov)).

<sup>22</sup> Regular European Interlaboratory Measurement Evaluation Programme, REIMEP (<http://irmm.jrc.ec.europa.eu>).

<sup>23</sup> Évaluation de la Qualité des Résultats d'Analyses dans l'Industrie Nucléaire, EQRAIN ([www.cetama.cea.fr](http://www.cetama.cea.fr)).



method at a single laboratory;  $u(r)$  and  $u(s)$  are specific subsets of the combined standard uncertainty comprising all uncertainties arising from random effects and systematic effects, respectively, according to the description in chapter 3.

- New to the ITV-2010 tables is a column labelled **ITV**, which reflects the combination of the  $u(r)$  and  $u(s)$  parameters.

$$u_c(ITV) = [u(r)^2 + u(s)^2]^{1/2} \quad (7)$$

- The values in the **ITV** column are provided as a reference for the laboratories to allow them to easily compare their measurement uncertainties, estimated by the GUM approach, against the ITVs-2010. They are expressed as relative combined standard uncertainties.

**Table 1:** Measurement Method/Instrument Codes

Method/Instrument Code	Technique
ALPH	Alpha Spectrometry
AWCC	Active Well Coincidence Counter
CALO	Calorimetry
CMPU	Combined Product Uranium Concentration and Enrichment Assay (COMPUCEA)
COUL	Coulometry
EBAL	Electronic Balance
ELTM	Electromanometer
ENMC	Epithermal Neutron Multiplicity Counter
FRSC	Fuel Rod Scanner
GBAS	Glove Box Assay System
GRAV	Gravimetry
GSMS	Gas Source Mass Spectrometry
HEPC	High Efficiency Passive Counter
HKED	Hybrid K-Edge/K-XRF Densitometer
HLNC	High Level Neutron Coincidence Counter
HMMS	Hulls Monitor and Measurement System
HRGS	High Resolution Gamma Spectrometry
IDMS	Isotope Dilution Mass Spectrometry
IMCG	Inspector Multichannel Analyzer with Ge detector
IMCN	Inspector Multichannel Analyzer with NaI detector
INVS	Inventory Sample Coincidence Counter
KEDG	K-Edge Densitometer
LCBS	Load-cell Based Weighing System
LMCA	Laboratory Multichannel Analyzer (HRGS)
LMCN	Laboratory Multichannel Analyzer (NaI detector)
MCICP	Multi-Collector Inductively Coupled Plasma Mass Spectrometry
MMCG	Mini-Multichannel Analyzer with Ge Detector
MMCN	Mini-Multichannel Analyzer with NaI Detector
PCAS	Plutonium Canister Assay System
PNCL	Plutonium Neutron Coincidence Collar
POLA	Polarography
PSMC	Plutonium Scrap Multiplicity Counter
PUSP	Pu-VI Spectrophotometry
TIMS	Thermal Ionization Mass Spectrometry
TITR	Titration
UBVS	Uranium Bottle Verification System
UNCL	Uranium Neutron Coincidence Collar
USP	Uranium Spectrophotometry
VTDM	Vibrating Tube Density Meter
WDAS	Waste Drum Assay System
XRF	X-ray Fluorescence

**Table 2:** Bulk and Density Measurements

Measurement	Instrument	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
Mass	LCBS	0.05	0.05	0.07	
	EBAL	0.05	0.05	0.07	
Volume <sup>1/</sup>	ELTM (Accountability tanks)	0.05	0.1	0.12	2/
	ELTM (Process tanks; high concentration)	0.2	0.2	0.28	3/
	ELTM (Process tanks; low concentration)	1	1	1.4	4/
	ELTM (Accountability tanks)	0.3	0.2	0.36	5/
Density	ELTM (Accountability tanks)	0.05	0.05	0.07	6/
	ELTM (Process tanks; high concentration)	0.1	0.1	0.14	
	ELTM (Process tanks; low concentration)	0.7	0.7	1	
	VTDM	0.05	0.05	0.07	

**1/** Volume determinations are made on the basis of level pressure, density and temperature measurements. The volume measurement uncertainties are highly dependent on the homogeneity of the liquid, the quality of the density measurements and of the calibration equation determined in the calibration process. The volume measurements may also involve an absolute error component which has to be taken into consideration when determining the overall uncertainty of volume measurements.

**2/** For accountability tanks in newly built large-throughput facilities, uncertainties of 0.05% for u(r) and 0.1% for u(s) at full volume are achievable if: i.) A carefully designed calibration procedure has been implemented under well-controlled environmental and stable temperature conditions; and ii.) Measurements, using high precision electro-manometers, are performed on a well-characterized and homogenized liquid.

**3/** Process tanks for high Pu concentration solutions are generally also equipped with high precision electro-manometers, however, the calibration effort and tank design specifications may be lower.

**4/** Equipped with standard electro-manometers, lower calibration effort.

**5/** The values apply to older facilities where the tank design was not driven by optimized ELTM volume measurement capabilities.

**6/** The same comments as given for the volume measurements apply; one additional important calibration parameter is the determination of the probe (dip tube) separation.

**Table 3:** Sampling Uncertainties for Element Concentration and <sup>235</sup>U Abundance

Material	Uncertainty Component (% rel.)				Minimum Sample Size <sup>1/</sup>
	Concentration		<sup>235</sup> U Abundance		
	u(r)	u(s)	u(r)	u(s)	
DUF6	0.1	*	1 <sup>2/</sup>	1 <sup>2/</sup>	5-10 g
NUF6, LEUF6	0.05	*	0.1	*	5-10 g
U-oxide Powder	0.2	*	0.05	*	10-20 g
U-oxide Pellets	0.05	0.05	0.05	0.05	1 pellet
U-oxides (with Gd)	0.2	*	0.05	*	1 pellet
U Scrap (clean) <sup>3/</sup>	1	*	1	*	30 g
U Scrap (dirty) <sup>4/</sup>	10	*	10	*	2x30 g
Reprocessing Input Solution	0.3	0.2 <sup>5/</sup>	0.05	*	2x1 mL
High Active Liquid Waste	5	5	*	*	2x1 mL
U Nitrate Solution	0.1	*	0.05	*	10 mL
Pu, U/Pu Nitrate Solution	0.2	*	0.05	*	10 mL
Pu-oxide	0.1	*			2x1 g
FBR MOX	0.2 (Pu) 0.1 (U)	* *	0.1	*	2x1 pellet, or 2x2 g

Material	Uncertainty Component (% rel.)				Minimum Sample Size <sup>1/</sup>
	Concentration		<sup>235</sup> U Abundance		
	u(r)	u(s)	u(r)	u(s)	
LWR MOX	0.7 (Pu) 0.1 (U)	* *	0.1	*	2x1 pellet, or 2x5 g
MOX Scrap (clean)	1	*	1	*	2x5 g
MOX Scrap (dirty)	10	*	10	*	2x10 g
HEU Metal and Alloys	0.2	*	0.05	*	1 - 5 g

\* Values have not yet been defined.

1/ According to STR-69 (Destructive Analysis and Evaluation Services for Nuclear Material Accountancy Verification, STR-69, Rev. 5, IAEA, Vienna, 2004).

2/ Additional sampling uncertainties are expected for  $DUF_6$  when samples are taken from the gas phase of a non-homogenized cylinder.

3/ Scrap with low impurity content and suitable for recycling.

4/ Sampling uncertainties can vary widely depending on material heterogeneity and sample size.

5/ Sampling uncertainties of systematic nature, resulting from airlift effects (evaporation).

**Table 4a:** Uranium Element Concentration Measurements (DA)

Concentration measurements on powders and solutions require weight change correction because of sample instability.

Method	Material	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
GRAV	U (pure compounds)	0.05	0.05	0.07	1/
	U (with Gd)	0.1	0.1	0.14	
TITR <sup>2/</sup>	U (pure compounds)	0.1	0.1	0.14	
	U Alloys	0.2	0.2	0.28	
	Mixed U/Pu	0.1	0.1	0.14	
POLA	U (with Gd)	0.05	0.05	0.07	
IDMS	Hot Cell Conditions	0.2	0.2	0.28	3/ 4/
		0.3	0.3	0.42	3/ 5/
	Glove Box Conditions	0.15	0.1	0.18	3/ 4/
		0.2	0.2	0.28	3/ 5/
KEDG	U and U/Pu Solution	0.2	0.2	0.28	6/
HKED	Spent Fuel Solution	0.2	0.2	0.28	6/
XRF	U Solution (low conc.)	2	2	2.8	7/
CMPU	U Compounds	0.2	0.2	0.28	8/
USP	U Solution (low conc.)	2	2	2.8	7/ 9/

1/ Materials containing non-volatile impurities < 1000 ppm.

2/ Davies & Gray Method.

3/ For all materials typically encountered in the nuclear fuel cycle.

4/ Under conditions of sufficiently different isotopic compositions of spike and sample and near optimum sample to spike ratio, using large size spikes (such as LSD).

5/ Under conditions of sufficiently different isotopic compositions of spike and sample and near optimum sample to spike ratio, using small size spikes.

6/ For samples in solution with > 50 g/L U and measurement time of 3 x 1000 sec.

7/ 1 to 50 g/L U.

8/ 200 g/L U in dissolved samples.

9/ For process analysis under hot cell conditions.

**Table 4b: Plutonium Element Concentration Measurements (DA)**

Concentration measurements on powders and solutions require weight change correction because of sample instability

Method	Material	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
GRAV	Pu Oxide	0.05	0.05	0.07	1/
TITR	Pu Oxide and Nitrate	0.15	0.15	0.21	2/
	U/Pu Oxide and Nitrate	0.2	0.2	0.28	
COUL	Pu (pure compounds)	0.1	0.1	0.14	3/
IDMS	Hot Cell Conditions	0.2	0.2	0.28	4/ 5/
		0.3	0.3	0.42	4/ 6/
	Glove Box Conditions	0.15	0.1	0.18	4/ 5/
		0.2	0.2	0.28	4/ 6/
KEDG	Pu and U/Pu Solution	0.3	0.3	0.42	7/
HKED	Spent Fuel Solution	0.8	0.5	0.94	8/
XRF	Pu Solution (low conc.)	2	2	2.8	9/
PUSP	Process Solutions	2	2	2.8	10/
ALPH	Waste Solution (low conc.)	7	7	10	
INVS	Pu, U/Pu Oxides	2	1	2.2	11/ 12/
ENMC	U/Pu Oxides	1	1	1.4	12/
CALO	Pu, U/Pu Materials	0.4	0.4	0.56	12/ 13/

1/ Materials containing non-volatile impurities &lt; 1000 ppm.

2/ Techniques such as the MacDonald and Savage and the AgO methods.

3/ For samples containing &gt; 25 µg Fe/g Pu correction or chemical separation is required.

4/ For all materials typically encountered in the nuclear fuel cycle.

5/ Under conditions of sufficiently different isotopic compositions of spike and sample and near optimum sample to spike ratio, using large size spikes (such as LSD).

6/ Under conditions of sufficiently different isotopic compositions of spike and sample and near optimum sample to spike ratio, using small size spikes.

7/ For samples in solution with &gt; 40 g/L Pu and measurement time of 3 x 1000 sec.

8/ Typically 150 to 250 g/L U with a U/Pu ratio of 80 to 150 and measurement time of 3 x 1000 sec.

9/ Pu concentration between 1 and 50 g/L.

10/ &gt;0.1 g/L Pu.

11/ Counting time 600 sec.

12/ Pu isotopic composition determination by mass spectrometry.

13/ Lower uncertainties are achievable for materials containing low burn-up Pu.

**Table 5a: <sup>235</sup>U Abundance Measurements (DA)**

Method	Material	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
GSMS	DUF <sub>6</sub> & NUF <sub>6</sub>	0.1	0.1	0.14	1/
	LEUF <sub>6</sub>	0.05	0.05	0.07	
TIMS, MCICP	DU (<0.3% <sup>235</sup> U)	0.5	0.5	0.70	
	U (0.3% < <sup>235</sup> U <1%)	0.2	0.2	0.28	
	LEU (1% < <sup>235</sup> U <20%)	0.1	0.1	0.14	
	HEU (>20% <sup>235</sup> U)	0.05	0.05	0.07	
LMCN	LEU oxides	0.3	0.2	0.36	2/
	HEU oxides	0.2	0.2	0.28	
CMPU	LEU oxides	0.4	0.2	0.45	3/

1/ Sampling uncertainty may be dominating if cylinder contents are not homogenized.

2/ For materials not containing reprocessed uranium.

3/ Measurement time 1000 sec.; adjusted for age of source when necessary.

**Table 5b:**  $^{235}\text{U}$  Abundance Measurements (NDA)

Method	Material	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
IMCN, MMCN <sup>1/2/</sup>	DUF <sub>6</sub>	20	8	22	3/ 4/ 5/
	NUF <sub>6</sub>	10	3	10	
	LEUF <sub>6</sub>	5	3	5.8	
	NU (pure materials) <sup>6/</sup>	5	3	5.8	4/ 5/
	LEU (pure materials) <sup>6/</sup>	3	2	3.6	
	NU, LEU Scrap (dirty) <sup>7/</sup>	15	5	16	
	LEU Rods	3	1	3.2	
	HEU Metal, Alloys	3	1	3.2	
IMCG, MMCG <sup>2/</sup>	DUF <sub>6</sub>	20	8	22	3/ 4/
	NUF <sub>6</sub>	10	2	10	
	LEUF <sub>6</sub>	5	2	5.4	
	LEU Oxides <sup>6/</sup>	3	2	3.6	4/ 8/
	HEU Metal and Alloys	3	1	3.2	
UBVS	UO <sub>3</sub> (reprocessed)	5	3	5.8	

1/ For materials not containing reprocessed uranium.

2/ Measurement time 300 sec.

3/ Includes uncertainty associated with ultrasonic thickness gauge measurement of the UF<sub>6</sub> cylinder.

4/ Spectrum analysis: infinite thickness method (enrichment meter principle).

5/ Similar uncertainties should be achievable for peak fitting based spectrum analysis methods.

6/ Includes scrap with low impurity content and suitable for recycling.

7/ Uncertainties for dirty scrap can vary widely due to matrix heterogeneity.

8/ Similar uncertainties are expected for intrinsic calibration based spectrum analysis methods (e.g., MGAU).

**Table 6:** Plutonium Isotope Assay of Pu and U/Pu Materials

Material Type	Isotope Ratio	Typical Value for Ratio (*100)	Method					
			TIMS, MCICP <sup>1/</sup>		IMCG <sup>2/</sup>		LMCA <sup>3/</sup>	
			u(r)	u(s)	u(r)	u(s)	u(r)	u(s)
High-Burnup Pu	$^{238}\text{Pu}/^{239}\text{Pu}$	1.7	1.5	1	1	2	1	1
	$^{240}\text{Pu}/^{239}\text{Pu}$	43	0.1	0.05	1	1	0.7	0.7
	$^{241}\text{Pu}/^{239}\text{Pu}$	13	0.2	0.2	1	1	0.7	0.7
	$^{242}\text{Pu}/^{239}\text{Pu}$	8	0.2	0.3				
Low-Burnup Pu	$^{238}\text{Pu}/^{239}\text{Pu}$	0.02	10	10	5	10	5	5
	$^{240}\text{Pu}/^{239}\text{Pu}$	6	0.15	0.1	1.5	2	1.5	1.5
	$^{241}\text{Pu}/^{239}\text{Pu}$	0.2	1	1	1.5	2	1	1
	$^{242}\text{Pu}/^{239}\text{Pu}$	0.05	2	2				

1/  $^{238}\text{Pu}/^{239}\text{Pu}$  by alpha spectrometry/mass spectrometry combination.

2/ Measurement time 1000 sec.

3/ Measurement time 3 x 1000 sec.; 0.5 g Pu.

**Table 7a:** Total Mass of <sup>235</sup>U (direct NDA)

Method	Material	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
<b>AWCC</b>	HEU Metal, Alloys	5	3	5.8	1/
	HEU Fuel Elements	3	3	4.2	
<b>FRSC</b>	LEU Fuel Rods	1	1	1.4	
<b>UNCL</b>	LEU Assemblies	4	2	4.5	2/
	HEU Assemblies	1	1	1.4	
<b>HEPC</b>	LEU Items	3	1	3.2	

1/ Measurement time 600 sec.; fast mode operation.

2/ These values are valid for LWR fresh fuel without Gd or with a Gd content, not exceeding the calibration range. In the presence of higher Gd content, u(s) can increase up to 10.

**Table 7b:** Total Mass of Pu (direct NDA)

Method	Material	Uncertainty Component (%rel.)		ITV (%rel.)	Notes
		u(r)	u(s)		
<b>HLNC</b>	Pu Oxide	1	0.5	1.1	1/ 2/
	MOX (> 10% Pu) <sup>3/</sup>	2	0.5	2.1	
	MOX (< 10% Pu) <sup>3/</sup>	4	1.5	4.3	
	MOX (clean scrap) <sup>3/ 4/</sup>	5	2	5.4	
	MOX Rods	2	1	2.2	
	FBR MOX Assemblies	2	1	2.2	
<b>PNCL</b>	LWR MOX Assemblies	1	3	3.2	
<b>PSMC</b>	Pu Oxide	1	0.5	1.1	
	MOX (clean scrap) <sup>4/</sup>	4	1	4.1	
	MOX (dirty scrap) <sup>4/</sup>	5	1	5.1	
<b>GBAS</b> <sup>5/</sup>	Glove Box Inventory	10	5	11	
<b>WDAS</b> <sup>6/</sup>	Pu Waste Drums	10	5	11	
<b>HMMS</b>	Hulls Drums	10	10	14	
<b>VCAS</b>	Vitrified Waste Canisters	10	10	14	

1/ Measurement time 300 sec.

2/ Isotopic determination by mass spectrometry and alpha spectrometry.

3/ Including HLNC based canister counters (e.g., PCAS).

4/ Uncertainties for scrap represent average performance observed on historical data; material heterogeneity in the Pu isotopic composition and presence of absorbers or moderators in the matrix can cause much higher uncertainties.

5/ Including other facility-specific measurement systems for Pu inventory in glove boxes.

6/ Including other facility-specific measurement systems for low Pu content solid waste items.

## 7. Use of ITVs

ITVs are considered to be achievable in routine measurements involved in the determination of the amount of nuclear material for material accountancy and safeguards verification purposes. They are intended to be used as a reference by plant operators, State safeguards systems, and international safeguards organizations. However, they should not be normally used in place of performance uncertainties determined from actual measurement results, when assessing the statistical significance of operator-inspector differences or MUF. Analytical laboratories can find it useful to determine experimentally the actual uncertainties of their measurements, and to compare them with the corresponding values in ITVs-2010.

Safeguards authorities regularly compare the performance values with the current ITVs. In cases where the performance values are significantly higher than the ITVs, and too high to allow the IAEA to meet its detection goals, the safeguards authorities, along with the laboratories, will examine means of improving performance. When reliable performance values are not available, ITVs may be used instead to calculate sampling plans, to set rejection limits, and to calculate estimates of the combined uncertainties of inventories, throughputs, MUF and Ds.

Such applications of the ITVs require having a good insight of the measurement and verification systems. It is in particular important to recognize that, because of practical constraints, some measurement steps may be common to the operator and the inspector. It should also not be forgotten that the operator-inspector differences can carry errors which are not related to measurement uncertainties.

The following three examples illustrate how the tabulated ITVs can be used to calculate ITVs for combined uncertainties applicable to practical situations.

### Example 1

#### *Target Values for the Determination of the Total Mass of Fissile Element on Independent Samples*

Consider a situation where the operator and the inspector determine fully independently the total amount of plutonium in a batch of LWR MOX pellets. The operator measures the plutonium concentration by titration on ten randomly

selected pellets, and the inspector measures by IDMS on an independently selected single pellet.

The Target Values for the combined relative standard uncertainties applicable to the determination of the total mass of plutonium by the operator are derived from the following equations, respectively for the random uncertainty ( $u_c(r,O)$ ), the uncertainty of systematic character ( $u_c(s,O)$ ), and their combination  $u_c(O)$ :

$$u_c(r,O) = \sqrt{\left[ \sum_i u_i^2(r,O)/n_i(O) \right]} \quad (8)$$

$$u_c(s,O) = \sqrt{\left[ \sum_i u_i^2(s,O) \right]} \quad (9)$$

$$u_c(O) = \sqrt{u_c^2(r,O) + u_c^2(s,O)} = \sqrt{(0.0555 + 0.0425)} = 0.31\% \quad (10)$$

where  $i$  is the index to the  $i^{\text{th}}$  item in the batch of  $n_i(O)$  pellets. The above values would be used in the calculation of Target Values for the relative standard uncertainties to be expected in the inventory, throughput and MUF declared by the operator.

Similar equations are used to calculate the corresponding values applicable to inspector's measurements,  $u_c(r,I)$ ,  $u_c(s,I)$  and  $u_c(I)$ . The Target Value for the combined uncertainties on the total Pu mass measured by the inspector is equal to:

$$u_c(I) = \sqrt{u_c^2(r,I) + u_c^2(s,I)} = \sqrt{(0.5150 + 0.0125)} = 0.73\% \quad (11)$$

Its magnitude is dominated essentially by the random sampling uncertainty component. This is also true for the Target Value applicable to the Operator-Inspector difference:

$$u_d = \sqrt{u_d^2(r) + u_d^2(s)} = \sqrt{(0.5705 + 0.0550)} = 0.79\% \quad (12)$$

Assuming that the values of Target Values ( $u_c$  values given in Table 8) are effectively achieved, the 95% confidence intervals of the final results of the operator, of the inspector and of their difference, would be respectively equal to:

$$CL(O) = k u_c(O) = 2 \times 0.31 = 0.62\% \quad (13)$$

$$CL(I) = k u_c(I) = 2 \times 0.73 = 1.46\% \quad (14)$$

$$CL(d) = k u_c(d) = 2 \times 0.79 = 1.58\% \quad (15)$$

where the coverage factor  $k$  is 2.

**Table 8:** Target Values for Total Pu Mass  
with Independent Samples and DA (Example 1)

	Step		Method	$n_i$	ITV (% rel. Std. Dev.)			Variance Component	
			Instr.		$u_i(r)$	$u_i(s)$	Table	$u_i^2(r)/n_i$	$u_i^2(s)$
O P E R A T O R	1-	Bulk	EBAL	1	0.05	0.05	2	0.0025	0.0025
	2-	Sampling Pu-Conc.		10	0.70	nd	3	0.0490	
	6-	Pu-Conc.	TITR	10	0.20	0.20	4b	0.0040	0.0400
	Sum of variance components							0.0555	0.0425
	Combined Std. Uncertainties, $u_c(r,O)$ and $u_c(s,O)$ , (% rel.)							0.24	0.21
I N S P E C T O R	1-	Bulk	EBAL	1	0.05	0.05	2	0.0025	0.0025
	2-	Sampling Pu-Conc.		1	0.70	nd	3	0.4900	
	6-	Pu-Conc.	IDMS	1	0.15	0.10	4b	0.0225	0.0100
	Sum of variance components							0.5150	0.0125
	Combined Std. Uncertainties, $u_c(r,I)$ and $u_c(s,I)$ , (% rel.)							0.72	0.11
D I F F	Variance of Rel. Operator-Inspector Difference							0.5705	0.0550
	Standard Uncertainties of Rel. Diff., $u_d(r)$ and $u_d(s)$ , (% rel.)							0.76	0.23

**Example 2***Target Values for the Determination of the Total Mass of  
Fissile Element on a Common Sample*

In situations where the inspector analyzes a sub-sample of a homogeneous operator's sample, the sampling errors no longer contribute to the uncertainty of the operator-inspector difference. An example of this situation could be a co-operative effort to identify the existence of biases in the chemical analysis.

Applying these conditions to the first example as shown in Table 9, the Target Value for the operator-inspector difference and its 95% confidence interval will be:

$$u_d = \sqrt{(0.0675 + 0.0550)} = 0.35\% \quad (16)$$

$$CL(d) = k u_d = 2 \times 0.35 = 0.70\% \quad (17)$$



**Table 9:** Target Values for Operator-Inspector Difference on Total Pu Mass, with Common Sample and DA (Example 2)

	Step		Method	$n_i$	ITV (% rel. Std. Dev.)			Variance Component	
			Instr.		$u_i(r)$	$u_i(s)$	Table	$u_i^2(r)/n_i$	$u_i^2(s)$
O P E R A T O R	1-	Bulk	EBAL	1	0.05	0.05	2	0.0025	0.0025
	2-	Sampling Pu-Conc.		1	0.70	nd	3		
	6-	Pu-Conc.	TITR	1	0.20	0.20	4b	0.0400	0.0400
	Sum of variance components							0.0425	0.0425
	Combined Std. Uncertainties, $u_c(r,O)$ and $u_c(s,O)$ , (% rel.)							0.21	0.21
I N S P E C T O R	1-	Bulk	EBAL	1	0.05	0.05	2	0.0025	0.0025
	2-	Sampling Pu-Conc.		1	0.70	nd	3		
	6-	Pu-Conc.	IDMS	1	0.15	0.10	4b	0.0225	0.0100
	Sum of variance components							0.0250	0.0125
	Combined Std. Uncertainties, $u_c(r,I)$ and $u_c(s,I)$ , (% rel.)							0.16	0.11
D I F F	Variance components of relative difference							0.0675	0.0550
	Standard Uncertainties of Rel. Diff., $u_d(r)$ and $u_d(s)$ , (in %)							0.26	0.23

**Example 3***Estimation of the Uncertainty of Operator-Inspector Differences for NDA Sampling Plan Calculations*

Consider a situation where an inspector must calculate the sample size for verifying the content of LEU  $UF_6$  containers using MMCN. The operator's declarations for the material are based on DA measurements of  $^{235}\text{U}$  abundance and the stoichiometric value for U-concentration in  $UF_6$ . No historical inspector measurement data is available. Therefore ITVs need to be used to provide an estimate of the uncertainty which may be associated with the operator-inspector difference.

The variance components calculated from the ITVs-2010 are given in Table 10. The standard combined uncertainty associated with the operator-inspector difference in this example is equal to:

$$u_d = \sqrt{[u_d^2(r) + u_d^2(s)]} = \sqrt{(25.0175 + 9.0075)} = 5.83\% \quad (18)$$

In the absence of an uncertainty estimate based on historical measurement data, the inspector would thus use the above value calculated from the ITVs for performing sample size calculations and establishing rejection limits. In this example, the relatively large uncertainty associated with the NDA measurement almost entirely dominates the overall uncertainty of the operator-inspector difference.

**Table 10:** Target Values for Operator-Inspector Difference (Example 3)

	Step		Method/Instr.	ITV (% rel. Std. Dev.)			Variance Component	
				$u_i(r)$	$u_i(s)$	Table	$u_i^2(r)$	$u_i^2(s)$
O P E R A T O R	1-	Bulk	EBAL	0.05	0.05	2	0.0025	0.0025
	2-	Sampling $^{235}\text{U}$ wt. %		0.1		3	0.0100	
	6-	U-Conc.	Stoichiom. Val.					
	6-	$^{235}\text{U}$ wt. %	GSMS	0.05	0.05	5a	0.0025	0.0025
	Sum of variance components						0.0150	0.0050
	Combined Std. Uncertainties, $u_c(r,O)$ and $u_c(s,O)$ , (% rel.)						0.12	0.07
I N S P E C T O R	1-	Bulk	EBAL	0.05	0.05	2	0.0025	0.0025
	6-	U-Conc.	Stoichiom. Val.					
	6-	$^{235}\text{U}$ wt. %	MMCN	5	3	5b	25.0000	9.0000
	Sum of variance components						25.0025	9.0025
	Combined Std. Uncertainties, $u_c(r,I)$ and $u_c(s,I)$ , (% rel.)						5.00	3.00
D I F F	Variance components of relative difference						25.0175	9.0075
	Standard Uncertainties of Rel. Diff., $u_d(r)$ and $u_d(s)$ , (in %)						5.00	3.00

## 8. Organizations Involved

The following groups of experts and organizations participated in the review of the draft ITVs-2010 tables and nominated their representatives to participate in a Consultants Group Meeting (CGM) at the IAEA. In addition to these consultants several observers attended the discussions which led to the definition of the ITVs-2010.

The following internal experts from the IAEA also participated in the review process and/or the CGM:

- Division of Analytical Services (S. Balsley, S. Buerger, G. Duhamel, A. Fajgelj, F. Tadjer)
- Division of Information Management (J. Baute, C. Norman, K. Zhao)
- Divisions of Operations (R. Binner)

**Table 11:** Consultants and Observers (CGM March 2010)

Organization	Consultants' Group Meeting	
	Consultant	Observer
ESARDA Working Group on DA	K. Mayer	J. Tushingham
ESARDA Working Group on NDA	P. Peerani	
ANSI/INMM 5.1 Analytical Chemistry Laboratory Measurement Control Committee	C. Pietri	P. Mason
ISO TC 85/SC5 Subcommittee on Analytical Methodology in the Nuclear Fuel Cycle	S. Tapodi	T. Morris
Japanese ITV-2010 Expert Group	Y. Tsutaki	S. Hara
EURATOM Safeguards Directorate	M. Boella	O. Alique
Brazilian-Argentine Agency for Accounting and Control of Nuclear Materials (ABACC)	G. Renha Jr.	F. Cordeiro Dias

# **Performance Values for Non-Destructive Assay (NDA) Technique Applied to Wastes: Evaluation by the ESARDA NDA Working Group**

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## Abstract

The first evaluation of NDA performance values was undertaken by the ESARDA Working Group for Standards and Non Destructive Assay Techniques and was published in 1993. Almost ten years later in 2002 the Working Group reviewed those values and reported on improvements in performance values and new measurement techniques that had emerged since the original assessment. The 2002 evaluation of NDA performance values did not include waste measurements (although these had been incorporated into the 1993 exercise), because although the same measurement techniques are generally applied, the performance is significantly different compared to the assay of conventional Safeguarded special nuclear material. It was therefore considered more appropriate to perform a separate evaluation of performance values for waste assay.

Waste assay is becoming increasingly important within the Safeguards community, particularly since the implementation of the Additional Protocol, which calls for declaration of plutonium and HEU bearing waste in addition to information on existing declared material or facilities. Improvements in the measurement performance in recent years, in

particular the accuracy, mean that special nuclear materials can now be accounted for in wastes with greater certainty.

This paper presents an evaluation of performance values for the NDA techniques in common usage for the assay of waste containing special nuclear material. The main topics covered by the document are:

- Techniques for plutonium bearing solid wastes
- Techniques for uranium bearing solid wastes
- Techniques for assay of fissile material in spent fuel wastes

Originally it was intended to include performance values for measurements of uranium and plutonium in liquid wastes; however, as no performance data for liquid waste measurements was obtained it was decided to exclude liquid wastes from this report.

This issue of the performance values for waste assay has been evaluated and discussed by the ESARDA NDA Working Group, and uses data from international round-robin exercises and in-the-field measurements.

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## Glossary

<b>AAS</b>	Add-A-Source
<b>AGR</b>	Advanced Gas-cooled Reactor
<b>ANCC</b>	Active Neutron Coincidence Counting
<b>ANMC</b>	Active Neutron Multiplicity Counting
<b>BEGe</b>	Broad Energy Germanium
<b>BGO</b>	Bismuth Germanate
<b>BWR</b>	Boiling Water Reactor
<b>CHILW</b>	Contact Handled Intermediate Level Waste
<b>CsI</b>	Caesium Iodide
<b>CZT</b>	Cadmium Zinc Telluride
<b>DA</b>	Destructive Analysis
<b>DDA</b>	Differential Die-Away
<b>DPA</b>	Differential Peak Attenuation/Absorption
<b>EQRAIN</b>	Evaluation de la Qualité des Resultats d'Analyse dans les Installation Nucleaire
<b>ESARDA</b>	European Safeguards Research and Development Association
<b>FISPIN</b>	A computer code for nuclide inventory calculations
<b>FRAM</b>	Fixed-energy Response-function Analysis with Multiple efficiency
<b>FWHM</b>	Full Width Half Maximum
<b>HPGe</b>	High Purity Germanium
<b>HRGS</b>	High Resolution Gamma Spectrometry
<b>IGA</b>	Isotopie Gamma des Actinides
<b>LOD</b>	Limit of Detection
<b>LRGS</b>	Low Resolution Gamma Spectrometry
<b>MBA</b>	Material Balance Area
<b>MDA</b>	Minimum Detectable Activity
<b>MGA</b>	Multi-Group Analysis
<b>MOX</b>	Mixed Oxide
<b>NDA</b>	Non-Destructive Assay
<b>ORIGEN</b>	A computer code for calculating the build-up, decay and processing of radioactive materials
<b>PCM</b>	Plutonium Contaminated Material
<b>PNCC</b>	Passive Neutron Coincidence Counting
<b>PNMC</b>	Passive Neutron Multiplicity Counting
<b>Pu</b>	Plutonium
<b>PVC</b>	Polyvinyl Chloride
<b>PWR</b>	Pressurised Water Reactor
<b>RHILW</b>	Remote Handled Intermediate Level Waste
<b>ROI</b>	Region of Interest
<b>RTR</b>	Real Time Radiography
<b>SGS</b>	Segmented Gamma Scanner
<b>SLB</b>	Standard Large Box
<b>SNM</b>	Special Nuclear Material
<b>SWB</b>	Standard Waste Box
<b>TGS</b>	Tomographic Gamma Scanner
<b>TMU</b>	Total Measurement Uncertainty
<b>TNC</b>	Totals Neutron Counting



## 1. Introduction

In 2002, the ESARDA Non-Destructive Assay (NDA) Working Group published a performance values report for Non-Destructive Assay techniques applied to Safeguards (Reference [1]). The 2002 report did not include performance values for waste measurements, although these had been included in the inaugural 1993 ESARDA performance values evaluation (Reference [2]). It was decided that the waste performance values should be part of a separate evaluation, because although the NDA techniques involved are predominantly the same, the application is very different as are the performance characteristics of the assay instrumentation.

This report therefore provides information on the performance of different NDA techniques applied to the measurement of waste materials containing Special Nuclear Material (SNM) and spent fuel wastes. It is intended that this report is used alongside the 2003 Safeguards Performance Values report to provide a complete coverage of the NDA measurements of interest to Safeguards. To this end, the format of the Waste Performance Values report has been structured to closely match that of the 2003 report, to make it as user-friendly as possible.

The aim of the ESARDA Waste Performance Values Report is to provide the Safeguards inspector with reference performance values for different measurement techniques that can be applied to determine the SNM content of waste. This data is intended to represent achievable performance using proven readily available current techniques. The performance value of interest for nuclear materials accountancy is the measurement accuracy, expressed in terms of measurement uncertainty (random and systematic).

It is noted that the spent fuel wastes that are considered in this report are waste streams in which fragments of spent fuel are present as opposed to waste streams comprising whole spent fuel elements or rods. It is considered that intact spent fuel elements or rods would be treated as bulk materials rather than waste, and because different measurement techniques may be applicable, the performance values are considered in the earlier ESARDA Performance Values report (Reference [2]).

Originally it was intended to include performance values for measurements of uranium and plutonium in liquid wastes; however, as no performance data for liquid waste measurements was obtained it was decided to exclude liquid wastes from this report. It is noted that many of the radiometric techniques discussed within this report can also be applied to liquid wastes; however, there are additional techniques that are generally only applicable to solutions containing plutonium or uranium, such as absorption-edge densitometry and X-Ray fluorescence, which have not been considered due to the absence of performance information.

The most obvious difference between waste items and SNM products (i.e. product cans, fuel assemblies) normally measured by Safeguards is that a wide range of other materials may be present in the waste container in addition to the SNM. These so-called matrix materials can range from soft waste (i.e. plastics / PVC) to hard waste (i.e. metals / concrete), and there may be other radioactive materials or chemicals present that interfere with the measurement. The waste items are also packaged in a wide range of different containers, ranging from small bags or cans (volume  $\approx 2$  litres) to large crates (volume  $\approx 1\text{--}3\text{ m}^3$ ). The SNM present could be in the form of residues (e.g. sweepings from glovebox operations) in significant quantities (i.e. 10's to 100's of grams) or be secondary waste items (e.g. protective clothing, tools) that have negligible or very low levels of SNM contamination (e.g. milligram levels). Obviously, it is the higher end of the mass range which is of interest for Safeguards, but in many cases the same measurement technique and instrument system will be required to assay a wide range of SNM masses, from milligrams up to hundreds of grams. Clearly the materials present in the waste, the packaging of the waste and the amounts of SNM present can have a significant affect on the measurement performance. These factors must therefore be taken into consideration when assessing the performance of waste measurement techniques.

Another factor to consider is that waste has not traditionally been subject to Safeguards, and as a consequence the assay equipment may not have been designed with a Safeguards measurement in mind. For example, the primary purpose of the measurement may be criticality safety in many cases (or other applications such as nuclear material transport, waste characterisation and / or segregation), and the assay system must demonstrate that there is less than a certain amount of SNM within a waste container to comply with transport or storage criticality safety limits. In such cases the instrument calibration may be biased high to ensure that unsafe underestimation of the SNM content does not occur, and the measurement is therefore not as accurate as might otherwise be achievable (that is, it does not represent the best estimate SNM inventory).

The purpose of this document is to define the performance that is achievable using existing waste systems performing measurements for nuclear material accountancy purposes. The design intent of the system has therefore been taken into consideration when interpreting performance data from existing systems, to ensure that the performance values represent those achievable for Safeguards measurements. However, it should be noted that many in-the-field systems may not be able to meet the performance values if their measurement purpose is different.

The assessment of the waste measurement performance values has been made by the ESARDA Non-Destructive Assay (NDA) Working Group. The performance information

presented has been obtained, reviewed and endorsed by the NDA Working Group.

A more detailed examination of the issues that are prevalent to waste assay is given in Section 1.1.

### 1.1 Definition, Scope and Use of NDA Waste Performance Values

The aim of the ESARDA Waste Performance Values Report is to provide the Safeguards inspector with reference performance values for different measurement techniques that can be applied to determine the SNM content of waste. This data is intended to represent achievable performance using proven readily available current techniques.

The NDA performance values are intended to be used for the following purposes:

- for planning of inspections by Safeguards Authorities;
- to provide guidance to users in judging the quality of their NDA measurements;
- to decide if, under fixed settings, repeated measurements, repeated sampling or reported inspections are an appropriate tool to reduce the overall uncertainty;
- to analyse operator-inspector differences in Safeguards verification and accountancy;
- to define the required accuracy level of NDA standards;
- to serve as guidance for input into accountancy error propagation models for Material Balance Areas (MBAs) or entire plants;
- to compare the updated performance values with earlier editions, in order to get an impression of improvements in measurement quality (change to lower values) or improvements in realistic perception (change to higher or lower values);
- to provide orientation for national or international regulators for the definition of basic parameters (detection limits, goal quantities, anomaly definition).

This information is also of more general use, since it facilitates intercomparison between different techniques, aiding the selection of the most appropriate assay technique for a given type of waste (in this regard, other national / international standards and “good practice guides” may also assist users in selection of appropriate techniques).

As mentioned in the Introduction, there are a number of issues that affect the performance of waste assay measurement systems, which need to be considered further and defined in more detail when determining and interpreting the performance values. These issues have been grouped into the following categories, and each is examined in more detail in the sub-sections that follow:

- Type of SNM present / waste stream characteristics.
- Packaging / containment of the waste.

- Matrix characteristics.
- Calibration philosophy and assumptions.
- Instrument system design and application regime.
- Expert analysis.

#### 1.1.1 Type of SNM Present / Waste Stream Characteristics

The categorisation of types of SNM present in waste that was used in the previous evaluations of performance values by the NDA Working Group (Reference [1], [2]) is also appropriate for waste assay, and will therefore be utilised for consistency. The following categories have therefore been defined:

- Solid Plutonium-bearing wastes.
- Solid Uranium-bearing wastes.
- Assay of fissile material in Spent Fuel wastes.

Originally the intention was to include performance values for the measurement of uranium and plutonium in liquid wastes. However, as no performance data was obtained for this type of waste it has been excluded from the report. If performance data for liquid waste measurements becomes available in the future, then this report can be reissued to include this information. It is noted that liquid wastes are less relevant to NDA and more relevant to sampling and Destructive Analysis (DA).

In most cases the plant or process from which the waste was generated will dictate which of the above categories are appropriate. However, in some cases, for example historical wastes where the information on the origin of the waste is unavailable or unreliable, it may not be possible to define in advance the categorisation in terms of type of SNM. However, in such cases it is normal for preliminary investigative measurements to be performed, for example using High Resolution Gamma Spectrometry (HRGS), to determine the type of SNM present before more quantitative assay is applied.

Within these overall categories of SNM type, the characteristics of the SNM will vary according to the plant or process from which the waste was generated. For example, if the waste originates from a MOX fuel manufacturing plant, the SNM could be in a variety of different physical or chemical forms such as plutonium oxide powder, plutonium fluoride powder, uranium oxide powder, mixed oxide powder, metals, fuel pellet fragments, contaminated equipment and also combinations of uranium and plutonium.

The mass of SNM present will also affect the performance of the assay technique, in particular the random uncertainty component. Certain systematic uncertainty effects may also only be significant in certain mass ranges, for example neutron self-multiplication which although dependent on a number of factors (including the neutron

energy spectrum, sample composition, sample density, sample geometry and neutron reflection) is only significant at larger plutonium masses, i.e. gram levels and higher. Another example is thermal neutron self-shielding, which is again dependent on several factors (including sample geometry, enrichment, density, chemical composition and neutron energy spectrum) but can be considered negligible when there are no significant lumps of material present. The performance for different masses of SNM has therefore been taken into consideration in the performance values tables.

In addition to variations in the physical / chemical form of the SNM, there could also be other radionuclides, chemical contaminants or items present depending on where exactly the waste originated from. These may interfere with the measurement technique and affect the performance. For example, waste comprising spent fuel residues is likely to have a very high gamma emission (from fission and activation products) and neutron emission (from curium, i.e. Cm242 and Cm244). The high neutron count rate from curium makes it difficult to use PNCC to determine the Pu240 equivalent mass. Similarly, for spent fuel the high gamma background means that it is unlikely that HRGS can be used to measure the plutonium or U235 masses. Similarly the presence of certain chemical compounds, such as neutron poisons, may significantly affect the performance of certain NDA measurement techniques. Another example is the presence of materials that when intimately mixed with the SNM generate a high (alpha, n) neutron emission, such as fluorine or beryllium, which will affect the performance of neutron measurement techniques.

The different types of waste are often referred to as waste streams (a term used by plant operators / waste consignors), although it should be noted that the waste stream definition often refers to a building rather than a particular part of the process, and can therefore encompass several

different physical / chemical forms of the SNM. Clearly the characteristics of the waste stream can affect the measurement performance as discussed above.

These factors are difficult to take into consideration in the performance values, and in cases where the performance of a particular technique may be affected by the physical / chemical form of the SNM or the presence of other contaminants then notes have been added to indicate the magnitude of the effect on the performance.

### 1.1.2 Packaging / Containment of the Waste

The following container types have been considered as representative of those normally used for the containment of waste arisings in the nuclear industry. Note that these are the commonly encountered waste containers, and several unusual or bespoke designs may also be in use, but these have been excluded to avoid over-complication. In cases where a bespoke design is encountered, it should be possible to find a similar container within the list of standard containers below to provide an indication of the measurement performance.

In order to avoid having a large number of container types, containers with similar design have been grouped together. Each container type therefore represents a range of containers having similar designs, and there are likely to be minor differences in the volumes, overall dimensions, wall thicknesses and construction materials used (e.g. mild or stainless steel), within each particular type. These differences are not considered important in the context of this performance values document. The parameters reported in Table 1 are nominal values that best represent each container type.

Note that the first column of Table 1 defines the name of each generic container type that may be used elsewhere in this report.

**Table 1:** Waste Containers for Performance Values

Container Name	Detailed Description	Waste Volume, litres	Typical Dimensions, mm	Additional Information
Waste Sorting Tray	Tray used to inspect and sort waste before filling another container	Variable	1m x 50cm	
Waste Bag / Piece	PVC waste bag or metal plant item (piece)	2 – 100	Various	Usually soft waste, but can include metal plant items (pieces).
Small Can	6 inch diameter can, various heights	3 – 12	150 Ø x 150 h, 460 h, 660 h	Product can or similar
Polythene bin	Polyethylene drum	40	300 Ø x 500 h	e.g. Lacalhe containers
100l Drum	100 litre steel drum	100-120	460 Ø x 610 h	Typically 1.1-1.5mm wall thickness
200l Drum	200 litre steel drum	200-220	610 Ø x 870 h	

Container Name	Detailed Description	Waste Volume, litres	Typical Dimensions, mm	Additional Information
200l Poly Lined Drum	200 litre steel drum with polyethylene liner	200-220	610 Ø × 870 h	Typically 6-8mm thick polyethylene liner for CHILW
400l/500l Drum	400 / 500 litre steel drum	400 / 500	715 Ø × 1067 h	Typically 1.1-1.5mm wall thickness
400/500l Grout Lined Drum	Steel drum with grout annulus and capping.	400 / 500	780 Ø × 1150 h	Used for final disposal
Small Crate	1 m <sup>3</sup> to 2 m <sup>3</sup> volume (e.g. 1m <sup>3</sup> box)	1000 – 2000	1m <sup>3</sup> box: 1090 × 915 × 1215 (h)	Typically 1-5mm wall thickness
Large Crate	> 2 m <sup>3</sup> volume (e.g. ISO freight container (20m <sup>3</sup> ))	2000 – 8000	Standard Waste Box (SWB): 1650×1310×1000 Standard Large Box (SLB-2): 1600×1680×2590	

### 1.1.3 Matrix Characteristics

The waste matrix categories identified in Table 2 are considered to represent the range of commonly encountered real waste matrices and will be referred to when defining the performance values.

When waste containers are filled with waste there is generally some manual compaction of soft wastes, and this has been assumed to be the case in the density ranges quoted in Table 2. In the performance values tables that follow, if the density has not been quoted then it can be assumed to be the typical value as indicated in Table 2. If different to the typical value, or if the performance value pertains to a particular range (e.g. <0.4 g.cm<sup>-3</sup>), then the specific value or range is indicated in the performance value table.

In some applications, the waste processing involves mechanical “pre-compaction” of the waste during the filling of the waste container, which typically doubles the density of the waste above the values quoted in Table 2. The term “pre-compaction” has been used to differentiate this process from the “compaction” of the overall waste container after it has been filled with waste. The compaction of the overall waste container, which results, for example, in pucks if the containers are drums, normally precedes the loading of the compacts into a larger container, which is then generally filled with grout in readiness for long term storage or disposal.

Note that the first column of Table 2 assigns a name to each matrix category that is used throughout this report.

**Table 2:** Waste Matrix Categories for Performance Values

Waste Matrix Name	Waste Matrix Description	Typical waste density range (no pre-compaction), g cm <sup>-3</sup>	Additional Information
Combustibles	Soft combustible waste (low PVC fraction). Typically paper and plastics.	0.05 – 0.2	Low PVC fraction defined as <10% by weight.
PVC	Soft combustible waste (high PVC fraction). Typically paper and plastics.	0.05 – 0.2	High PVC fraction defined as >10% by weight.
Metals	Metallic waste varying from small items (e.g. tools) to metal sheeting or plant items	0.3 – 2.0	
Lead	Lead shielding materials (sheets / bricks)	1.0 – 3.0	
Graphite	Graphite wastes	0.2 – 0.5	
Mixed	Mixed waste (metallic / soft) e.g. lab waste, dismantled gloveboxes.	0.05 – 0.5	PVC content can be high or low.
Filters	Filters	n/a	Filters are not normally pre-compacted.



Waste Matrix Name	Waste Matrix Description	Typical waste density range (no pre-compaction), g cm <sup>-3</sup>	Additional Information
Ash	Ash from incineration of soft waste	1.0 – 3.0	Ashes from combustion of filters.
Sludge	Sludge	1.0 – 2.0	
Bitumen / concrete	Waste solidified in bitumen or concrete for final disposal	1.5 – 3.0	
Debris	Decommissioning waste (soil / sand / rubble / concrete / debris / metals)	0.5 – 2.0	

Note that there may be other “special” matrix materials present in the waste that are problematic for certain NDA techniques even when present in small quantities, for example materials that are used for radiation shielding, such as lead or boron. Commonly such materials are excluded from the waste stream by pre-sorting and / or waste management procedures, because of the known problems presented by these materials to the assay equipment. The normal practice is to treat drums containing these materials as special cases, which require non-standard measurements or specific calibrations and expert offline analysis, or alternatively to argue that the frequency of occurrence is so low that the biases to individual results are acceptable. Because of these issues, “special” matrix materials have generally not been

considered when determining the performance values presented later in this report.

In addition to the types of matrix materials present, the degree of heterogeneity of the waste may also affect the performance of the assay technique. An extreme example of heterogeneity is a partially filled waste container. Discrete shielding objects can also be important, for example if a large proportion of the total mass is in a small piece of lead. Note that real waste is almost always heterogeneous to some extent (see Figure 1 below for some example Real-Time Radiography (RTR) images of real waste drums), although there are exceptions where the matrix can be considered homogeneous (e.g. incinerator ash, or when the waste has been sorted).

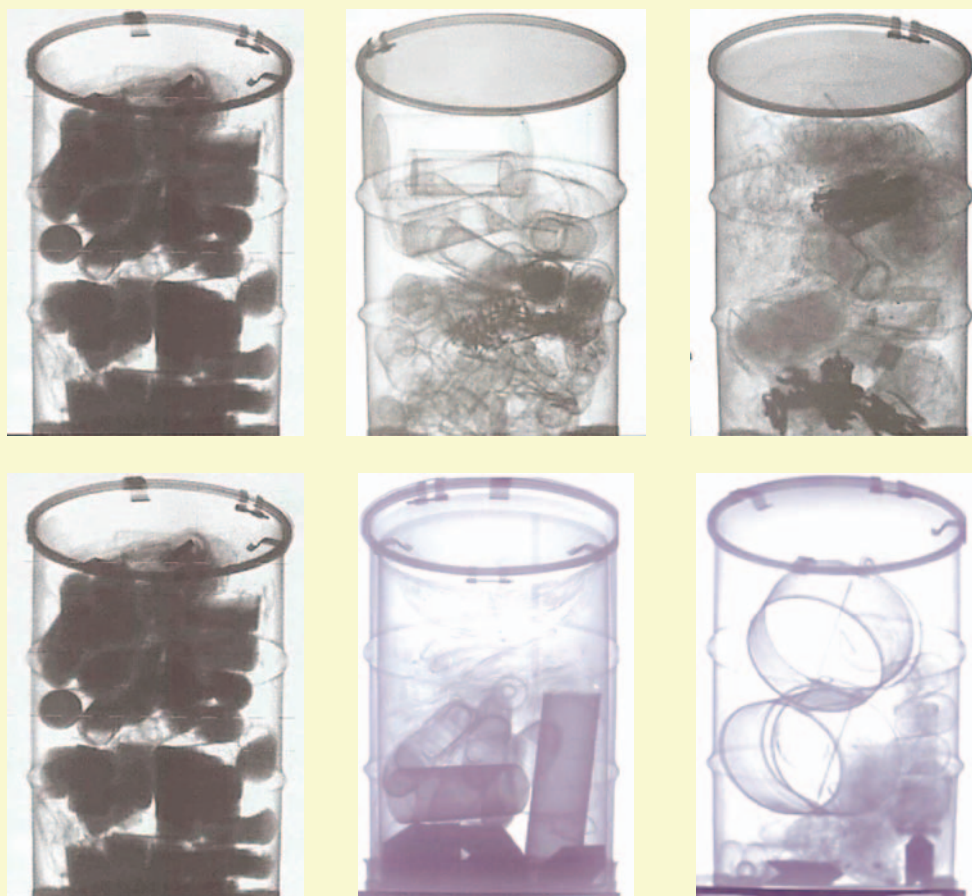


Figure 1: Example RTR Images of 200 Litre Waste Drums

Waste assay systems are normally calibrated using surrogate materials and the waste containers are homogeneously filled. Re-entrant tubes are usually positioned in the waste matrix at defined positions to allow known test sources to be introduced at reproducible locations. This allows the response of the systems to be characterised for a range of matrices and source distributions. The waste matrices are usually selected to represent the full range of the expected properties of the real waste forms; however, there are limitations in the validity of this approach because of the heterogeneity of real waste matrices. However, for the purposes of this performance values report, it has been assumed that the assay systems have been calibrated using representative waste matrices, and any bias due to the heterogeneity expected in typical real wastes is assumed to be negligible or has been taken into consideration within the reported total measurement uncertainty. This shows the importance of documenting the calibration assumptions.

#### 1.1.4 Calibration Philosophy and Assumptions

The calibration philosophy of the waste assay systems is also an important consideration, since the primary purpose of the measurement may not be SNM accountancy / Safeguards.

Assay systems generally report a best estimate result and quote the standard measurement uncertainty (usually a combination of statistical and systematic terms). If the measurement result is being used for nuclear (criticality) safety, then the calibration will tend to be pessimistic, particularly if any assumptions need to be made to cover limitations in the measurement technique. This is required to avoid significant underestimation of the fissile content, which would increase the criticality hazard in subsequent processing or handling of the waste.

An example of this is the U235 mass calibration of the drum monitors used to measure the U235 content of historical waste drums. The U235 mass measured by the HRGS system may be overestimated by a factor of 50 or more [7], due to pessimistic assumptions regarding the amount of self-shielding, the radial distribution of the uranium, and the system design with a large field-of-view collimator.

Clearly, assay systems that are calibrated in this way do not give meaningful performance information for Safeguards, since the SNM content is likely to be grossly overestimated. The performance values presented in this report are therefore for a “best estimate” type of measurement, where the calibration has not been made deliberately pessimistic to avoid unsafe underestimation. However, it should be noted that the measurement techniques do have limitations, and that assumptions may need to be made in the calibration to account for these. The validity of these assumptions will clearly affect the accuracy of the measurement, and any assumptions that may need to be made for a particular

measurement technique have therefore been noted in the performance values tables.

#### 1.1.5 Instrument System Design and Application Regime

Generally the design of the instrument system will depend on what the instrument is required to measure (and the required performance), how it will be used and any other constraints such as the environment where the instrument will operate. Clearly the design of the instrument system and how the chosen measurement technique has been implemented will significantly impact on the measurement performance.

Some examples of how the design influences the performance are given below:

- The detection efficiency of the measurement chamber in different passive neutron counters may vary from say 5% to 50%, depending on the number of detectors and detailed design. The efficiency is normally dictated by the performance requirements (i.e. detection limit and accuracy) which are in turn dictated by the primary end-use application.
- There are different options for matrix correction in both neutron and gamma based measurement techniques. The system may not perform matrix correction at all and assume a fixed value, or the correction may be very simple (e.g. based on the container weight and an assumed fill volume and material). More sophisticated techniques include the use of transmission sources for gamma systems, or an add-a-source or flux probes for neutron measurements.
- The instrument system may include more advanced data acquisition and processing that improve the assay performance. An example of this is the use of imaging algorithms to determine the spatial distribution of the SNM and reduce the corresponding uncertainty component.
- There may be constraints on the measurement time or throughput demanded by the end user, which dictates a higher efficiency design in order to achieve the same performance in a shorter measurement time.

For this performance values report, it is intended that, where appropriate, a range of performance values will be quoted reflecting the different performance that would be obtained for a “typical” system and a “high-end” system. The “typical” system would represent the majority of systems operating in-the-field, and the “high-end-system” would represent the state-of-the-art. Note that all the performance values are for in-the-field systems in normal operating conditions as opposed to theoretical performance values for laboratory type equipment.

The measurement time will affect the measurement performance, with the random uncertainty component

increasing at lower count rates. The performance values that are provided are generally be quoted for the standard measurement time for a particular assay system / measurement technique. Where appropriate and if the data are available, performance values for a range of counting times may be quoted.

### 1.1.6 Expert Analysis

A further issue that can influence the performance of an assay system is the use of non-standard or expert analysis, which is often made possible when the results from diverse measurement techniques are employed and can be intelligently combined. Sometimes these features may form part of an integrated assay system, but often results are analysed offline by a waste assay expert.

An example of how results from different assay techniques are combined to obtain the best performance over a wide range of plutonium mass is use of a combined active and passive neutron assay system. The active neutron measurement (using Differential Die-Away (DDA) or californium shuffler) is applied at low plutonium masses since the active measurement has the best detection limit and hence accuracy in this range, but above a certain plutonium mass (typically 5 to 10g) the passive neutron measurement (using PNCC) is preferred, since the PNCC measurement will be more accurate (the active measurement is less accurate due to sample self-shielding).

A common scenario requiring offline expert analysis of results is when the waste has unusual characteristics and the performance of the standard assay routines is biased or poor as a consequence. The improvements in assay results that can be obtained from expert offline review are not easy to quantify as they will be highly dependent on the specific measurement conditions; as this normally does not feature in the normal automated measurement process, offline review is not considered further in this report.

## 1.2 Origin and Structure of the Data

The NDA performance values reported in this paper have been evaluated from data and information of different origin, such as:

- published performance data from in-the-field operational systems;
- intercomparison exercises carried out with appropriate reference materials with the aim of assessing NDA performance.

In general, published or referenced performance data has been quoted in the text since this has an inherent level of quality assurance. The appropriate references have been included with the performance values (see Appendix 1). In the absence of published data, information has been gathered and assessed by experts within the NDA Working Group before being included.

The performance values of interest for nuclear materials accountancy are the measurement accuracy, expressed in terms of a total measurement uncertainty and the limit of detection. The definition of these terms used in this report are consistent with international best practice [3].

The total measurement uncertainty is a combination of both random and systematic uncertainties.

- The random (or Type A) uncertainty component is a measure of the repeatability of a result under constant conditions. A Type A uncertainty is defined as one that can be evaluated by statistical means. For NDA measurements this is largely dependent on the counting time (or number of repeat measurements). The random uncertainty included within the total measurement uncertainty values quoted in the performance values tables are relative to the standard or typical measurement times used for the measurement technique / system.
- The systematic (or Type B) uncertainty component arises from a number of sources and cannot be determined by repeated measurements. They result in assay bias which is constant under fixed conditions. Some examples of components that contribute to the overall systematic uncertainty are listed below:
  - Variability in background radiation.
  - The accuracy of the fissile mass value / isotopic composition of the calibration standards.
  - The effect of differences in the physical / chemical form of the real waste material compared to the reference (calibration) standards (e.g. self-shielding).
  - The effects of the waste matrix on the instrument response.
  - The effects of non-uniform (heterogeneous) distribution of fissile material within the waste container.
  - Sample effects such as self-shielding, self-multiplication and variable chemical forms of SNM.

The different systematic components are normally combined to determine the total systematic uncertainty, and this has been included within the total measurement uncertainty values that are reported in the performance values tables.

The total measurement uncertainty is generally assumed to correspond to the quadratic sum of the random and systematic components, assuming that the two types of uncertainty are independent.

The total measurement uncertainty values given in the performance values tables are relative (not absolute) uncertainties, quoted to one standard deviation (i.e. expansion or coverage factor  $k = 1$ ). However, it is noted that waste assay systems are normally configured to report the measurement uncertainty at the 2 sigma confidence level.

Note that the total measurement uncertainty values have been reported in the performance values tables in this report rather than separately recording the random and systematic components, as was the case in the previous evaluations (Reference [1], [2]). This is because the systematic uncertainty component normally dominates for waste assay applications, particularly in the SNM mass range pertaining to Safeguards. Consequently increases in the counting time do not generally result in a significant improvement in the overall accuracy, as the random component is less important, and only the total measurement uncertainty has therefore been included to reduce the data presented.

The detection limit is defined as the minimum amount of the measured quantity that can be measured and reported with 1 sigma confidence that it is greater than background radiation levels.

As discussed in Section 1.1, to be consistent with previous ESARDA performance value evaluations the performance values data is organised at the highest level by the type of SNM present. The following categories have therefore been defined:

- Solid Plutonium-bearing wastes.
- Solid Uranium-bearing wastes.
- Assay of fissile material in Spent Fuel wastes.

The different measurement techniques applicable to each of the SNM categories will be discussed separately, and performance values are presented in the form of tables. The performance values tables are presented as separate

tables for each relevant waste container type (as defined in Table 1), with the individual tables structured such that the waste matrix types (as defined in Table 2) are in separate rows and the mass of SNM in separate columns. Where performance data is available, the appropriate entry in the table provides the relative random uncertainty ( $r$ ) and systematic uncertainty ( $s$ ) components.

Note that it has not been possible to obtain performance values for each measurement technique for all the container types identified in Section 1.1.2 and waste matrices listed in Section 1.1.3, because in several cases the waste / container types are not relevant for a particular application and the information therefore does not exist. For example, calorimetry measurements have not been applied to large waste crates because of practical limitations, and it is clearly not appropriate to estimate the performance of such a system. Also, since this report presents the information that is available from waste assay systems that are currently in operation, and there are gaps in the coverage of waste / container types because, although the application of the technique is relevant to a particular waste / container type, performance data is not available. Table 3 and Table 4 below identify combinations of waste / container types that are not relevant to a particular application and also summarise the performance information that has been obtained and is presented later in this report.

The following colour coding is used throughout these tables:



**Table 3:** Containers and Waste Matrix Types Applicable to Solid Wastes (Pu and U)

		Waste Container Type										
		Waste Sorting Tray	Bag / Piece	Small Can	Poly Bin	100l Drum	200l Drum	200l Poly Lined Drum	400/500l Drum	400/500l Grouted Drum	Small Crate	Large Crate
Waste Matrix Type	Combustibles		√Pu √U	√Pu √U		√Pu √U	√Pu √U					
	PVC		√Pu √U	√Pu √U		√Pu √U	√Pu √U					
	Metals		√Pu √U			√Pu √U	√Pu √U					√Pu √U
	Lead			√Pu √U								
	Graphite											
	Mixed		√Pu √U	√Pu √U		√Pu √U	√Pu √U	√U	√Pu √U			√Pu √U
	Filters		√Pu √U				√Pu √U		√Pu √U			
	Ash											
	Sludge						√Pu					
	Bitumen / concrete			√Pu √U			√Pu			√Pu √U		
	Debris						√Pu					√Pu

**Table 4:** Containers and Waste Matrix Types Applicable to Spent Fuel Wastes

		Waste Container Type										
		Waste Sorting Tray	Bag / Piece	Small Can	Poly Bin	100l Drum	200l Drum	200l Poly Lined Drum	400/500l Drum	400/500l Grouted Drum	Small Crate	Large Crate
Waste Matrix Type	Combustibles											
	PVC											
	Metals	√U									√Pu √U	
	Lead											
	Graphite											
	Mixed											
	Filters											
	Ash											
	Sludge											
	Bitumen / concrete											
	Debris											

  = Container type / matrix not applicable

  = Container type / matrix is applicable, but no performance data available

 √Pu = Container type / matrix is applicable and Plutonium measurement performance data is available

 √U = Container type / matrix is applicable and Uranium measurement performance data is available

It can be seen from the above tables that the majority of the performance information that has been obtained and is presented in this report is for solid waste containing plutonium and/or uranium. The following points are noted regarding the available performance data for solid uranium and plutonium bearing wastes:

- No performance data was obtained for the “Waste Sorting Tray”, “Polythene Bin”, “200l Polythene Lined Drum” and “Small Crate” container types.
- No performance data was obtained for the “Ash” and “Graphite” matrix types.
- Despite these gaps in the performance information, the data that has been obtained does represent a good coverage of the range of container types and waste matrix types that might be encountered. It is worth noting that in many cases the missing information can be inferred from the data that is available – for example, the performance for a small crate is likely to be similar to that of a large 400/500l drum.
- For large container sizes greater than say 200 litres, the systematic uncertainties begin to increase very rapidly, so that the measurement uncertainty can increase almost exponentially with container dimensions.

In contrast only a limited amount of performance data was obtained for spent fuel wastes, reflecting the small number of waste assay systems currently in use for the measurement of this type of waste.

## 2. Techniques for Plutonium Bearing Solid Wastes

This section discusses characteristic features and performance of NDA techniques utilised for the determination of the amount and isotopic composition of wastes contaminated with plutonium-bearing solid materials. Some of these techniques are also applicable to uranium bearing solid wastes, as discussed in Section 3. By far the most common NDA techniques applied for the determination of the total amount of plutonium in a waste container are Passive Neutron Coincidence Counting (PNCC) combined with High Resolution Gamma Spectrometry (HRGS), but several other techniques are also applicable.

The features and performances of the following techniques are examined:

- High Resolution Gamma Spectrometry (HRGS) for Determination of Plutonium Isotopic Composition.
- High Resolution Gamma Spectrometry (HRGS) for Plutonium Mass Determination.
- Low Resolution Gamma Spectrometry (LRGS) for Plutonium Mass Determination.
- Passive Neutron Counting Techniques for Plutonium Mass Determination.
- Calorimetry for Plutonium Mass Determination.

### 2.1 High Resolution Gamma Spectrometry (HRGS) for Determination of Plutonium Isotopic Composition

#### 2.1.1 Fundamentals

Measurement of the mass of plutonium often requires measurement of the isotopic abundances, in order to convert the measured parameter (e.g. Pu240 effective mass in the case of PNCC) to the total plutonium mass. HRGS is normally used to determine the plutonium isotopic abundances in NDA measurements. The technique, which is based on measurement of isotope-specific gamma rays emitted by the plutonium isotopes, has the advantage of not requiring calibration with physical standards. The underlying intrinsic calibration approach relies on fundamental nuclear data such as isotope half-lives, gamma emission probabilities and correlation between isotopes with characteristic gamma emissions and those with no measurable emissions.

Plutonium isotopic abundance measurements by HRGS can be made on virtually all kinds of plutonium samples containing plutonium, with a wide range of plutonium masses (from fractions of a gram to kilogram size samples). The method only requires that the gamma rays from the sample are detected for analysis. In waste assay the presence of matrix materials that attenuate the gamma rays makes the plutonium isotopic measurement more challenging: because the isotopic analysis codes rely upon peak ratios the presence of waste materials does not bias the measurement, but the additional attenuation increases the minimum plutonium masses that can be accurately assayed.

In general, proprietary software codes are used to analyse the gamma spectra and determine the plutonium isotopic abundances. The most commonly used example is the Multi-Group Analysis (MGA) code, which usually exploits the 94-104keV region of the gamma spectrum for the isotopic analysis (Reference [11]). Since this spectral region contains the most abundant plutonium gamma and X-rays detectable in a gamma spectrum from plutonium in the presence of some Am241, fairly precise isotope abundance determinations are feasible with gamma spectra accumulated in relatively short counting times (i.e. 15-30min), assuming an appreciable mass of plutonium is present (i.e. gram quantities and above). The MGA code can also operate in High Energy mode, which takes advantage of several higher energy, and therefore more penetrating, gamma emissions.

Another software code in widespread usage for plutonium isotopic analysis is called FRAM (Reference [12]). This code makes use of the more energetic but less abundant gamma rays between 120 and 660keV, which provides some advantages in cases of stronger sample shielding such as in waste assay. The FRAM code is also capable of analysing the low energy region similar to MGA, although the main difference between the two codes is that the energy regions used by MGA are hard coded (and the user must

choose whether to analyse in Low Energy or High Energy mode), whilst those used by FRAM are user defined and therefore more adaptable.

Some advantages and disadvantages of the MGA and FRAM codes with respect to waste assay are summarised in the following table:

**Table 5:** Main Advantages / Disadvantages of MGA and FRAM for Plutonium Isotopic Analysis in Waste

	Advantages	Disadvantages
<b>MGA</b>	<ul style="list-style-type: none"> <li>Narrow energy range (94 – 120keV) in Low Energy mode means results less dependent on accuracy of efficiency fit and less prone to interference.</li> <li>Uses most intense region of spectrum so should give shortest measurement time.</li> <li>High energy mode can be selected when there is significant attenuation.</li> <li>Simple to use.</li> </ul>	<ul style="list-style-type: none"> <li>Low energy range which is more easily attenuated.</li> <li>Limited scope for user configuration and handling of unusual situations or poorer quality spectra.</li> </ul>
<b>FRAM</b>	<ul style="list-style-type: none"> <li>Energy ranges are not hard coded (like MGA) so more adaptable to cases where there is significant attenuation, interferences, unusual situations or poorer quality spectra.</li> </ul>	<ul style="list-style-type: none"> <li>If a broad energy range is used, the accuracy of results will be highly dependent on the efficiency fit.</li> <li>Requires a more experienced user.</li> </ul>

A new code called IGA has also recently been developed by CEA in collaboration with AREVA (Reference [13]). This code makes use of the whole gamma spectrum, and does not restrict itself to smaller regions, and is fully configurable like FRAM. An advantage of the software is that there are no constraints on the experimental setup (i.e. energy range, gain, resolution) or restrictions on the gamma peaks and isotopes that can be present. The code is now commercially available, but to date limited in-the-field performance data is available. A number of other isotopics codes are also available (e.g. Banham) which can be used as alternatives to those already mentioned. A more detailed overview of the current status of isotopic analysis codes can be obtained from the proceedings of the recent International Workshop on Isotopic Analysis Codes (Reference [15]).

A limitation of the gamma spectrometric approach is the inability to directly measure Pu242, because of the absence of a detectable gamma-ray. Therefore recourse has to be made to isotope correlation techniques to determine the abundance of this isotope. This limits the overall accuracy of the plutonium isotopic measurement, particularly if the fraction of Pu242 is significant.

Another important consideration for waste assay is sample homogeneity. The accuracy of the plutonium isotopic analysis relies upon the gamma spectrum being representative of the plutonium in the sample. In waste assay it is possible that the waste item contains a heterogeneous distribution of plutonium with different isotopic compositions present in the same waste container. In such cases the isotopic analysis may be biased, with the amount of bias depending on the relative positions, masses and isotopic compositions of the Plutonium in the waste container.

### 2.1.2 Instrumentation

Reliable plutonium isotopic analysis requires high-quality gamma spectra with good energy resolution. This is particularly true if the isotopic analysis software uses the most complex parts of the plutonium spectrum (e.g. 94 – 120keV), but even when higher energy regions are used, there is still a requirement for good resolution so overlapping peaks can be resolved. The most common form of detector technology used for HRGS is High Purity Germanium (HPGe). HPGe detectors typically have energy resolution (FWHM) of between 0.5 to 1.2 keV at 122 keV and 1.7 to 2.3 keV at 1332 keV. For the appropriate detector size, compromises often have to be made between desired energy resolution and detector efficiency because often the same detector is used for measurement of other (non-SNM) gamma-emitting isotopes, which have different performance requirements.

All HPGe detectors have to be cooled to a temperature below 100K (-173°C) in order for them to operate, and this is traditionally achieved using liquid nitrogen. Electrical refrigeration can also be used and the technology is now mature enough to offer a reliable and cost-effective alternative to liquid nitrogen.

Improvements have been made in recent years in semiconductor detectors operating at room temperatures or with Peltier cooling such as Cadmium-Telluride (CdTe) or Cadmium-Zinc-Telluride (CdZnTe) detectors, and in the development of analysis codes applicable to spectra acquired with these detectors (Reference [16]). However, because of the inferior energy resolution and poorer energy response function, it is unlikely that this type of intermediate resolution detector will become a full substitute for

HPGe-based spectroscopy in the foreseeable future, particularly for waste assay applications.

### 2.1.3 Performance Values

Performance data for the measurement of the plutonium isotopic composition in waste is reported in Table 6 for three

different systems. The isotopic analysis code used is identified in the table in addition to the system configuration. Note that in all three cases the performance data is for the measurement of the percentage Pu240 effective (%Pu240eff), since the isotopic result is combined with a measurement of the effective Pu240 mass via passive neutron counting in subsequent analysis to determine the total plutonium mass.

**Table 6:** Performance Values for Measurement of Plutonium Isotopic Composition (%Pu240eff) using HRGS

System Information	Isotopic Analysis Code	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
				LOD (1 $\sigma$ )	TMU (1 $\sigma$ )		
Single fixed HPGe (planar detector)	MGA	100l & 200l drum	Combustibles, PVC or Metals (< 0.4 g.cm <sup>-3</sup> )	-	2 - 12%	2700 s	1
Single scanning HPGe (e ~ 25%)	MGA	200l drum	Filters	-	4.6%	1200 s	23
			Combustibles, PVC, Mixed, Metals, Filters	-	7.7%	1200 s	

Note 1 The uncertainties given by MGA on the isotopic abundances of the even isotopes depend on the counting statistics, the Pu mass, the nature of the matrix, the type of plutonium (burn-up). The range of TMU is given for PuO<sub>2</sub> powder in retention in combustible matrices (density <0.4 g/cm<sup>3</sup>), with a Pu mass range between 150 mg and 150 g, and Pu239 abundance range between 55 and 90%.

The following points are noted regarding the results in Table 6:

- The TMU is significantly higher for the system using the scanning moderate efficiency detector, due to the reduced spectrum quality. I.e. on account of the lower efficiency detector combined with the scanning which means that the plutonium is only in the field of view for part of the measurement.
- The TMU values for the scanning system are more typical of general plutonium waste assay systems.

Although no detection limit values were provided for these systems, reliable plutonium isotopic analysis in 200 litre waste drums is typically only possible for gram quantities of plutonium or higher. However, it is noted that the lower mass limit at which the isotopic analysis will give reliable results is dependent on the measurement geometry, detector efficiency and measurement time. At low plutonium masses the isotopic analysis may fail completely due to the acquired spectrum not meeting one or more of the code requirements (e.g. missing peaks, poor resolution or insufficient counts in the spectrum), or alternatively the analysis may generate a result but indicate that it has poor quality (note the quality of the result can usually be assessed by means of a reported "goodness-of-fit" parameter). It is common for the waste assay system software to use the "goodness-of-fit" parameter to decide whether the reported isotopic results are reliable, and if the results are not considered reliable or if the isotopic analysis has failed, then a default isotopic composition will normally be applied.

Additionally, the performance of HRGS systems measuring the plutonium isotopic composition within simulated waste drums was evaluated in a recent round-robin exercise performed in France (Reference [21]). Plutonium measurements were performed on three 118 litre simulated waste drums containing soft waste, although one drum included some added metal plates. The total mass of plutonium present in each drum was in the range 100 to 200 mg.

Note that the data from the intercomparison exercise are different to that presented in the previous table (and the general structure of the other performance values tables), because only the measurement technique employed is recorded (in this case HRGS) and no specific details regarding the type and configuration of the instrument systems being used was provided (although the number and location of participants is known). For this reason it is considered appropriate to report these data as a separate table to illustrate the performance that is achievable in the field using HRGS to measure the plutonium isotopic composition.

The results from the French intercomparison exercise are summarised below in Table 7. Note that the average % bias and % standard deviation ("Stdev") obtained from the results reported by the different participating laboratories are presented, and these are in relative rather than absolute terms. The results are presented for each isotope rather than just the %Pu240eff (as in Table 6), and there were eight participating laboratories for the first simulated waste drum and nine for the second and third drums.

**Table 7:** Performance Values for Plutonium Isotopic Composition Measurement using HRGS from French Intercomparison Exercise – EQRAIN 4 (Reference [21])

Simulated Waste Drum	%Pu238		%Pu239		%Pu240		%Pu241		%Pu242	
	Average Bias	Stdev (1 $\sigma$ )	Average Bias	Stdev (1 $\sigma$ )	Average Bias	Stdev (1 $\sigma$ )	Average Bias	Stdev (1 $\sigma$ )	Average Bias	Stdev (1 $\sigma$ )
118 litre, Soft waste (90% PVC)	-11%	13%	1%	2%	-4%	7%	-5%	5%	9%	27%
118 litre, Soft waste	-4%	7%	0.2%	3%	-6%	9%	-8%	5%	319%	327%
118 litre, Soft waste + Metals	-1%	2%	-1%	2%	-1%	3%	-4%	5%	309%	336%

The following points are noted regarding the results in Table 7:

- The results are worse for Pu242 because it is not directly measurable and the isotopic percentage must be obtained based on correlations with the other isotopes. Consequently the Pu242 results are less accurate and more variable.
- The average bias and standard deviation for the other isotopes is typically less than 10%.
- To compare directly with Table 6, the average bias and standard deviation in the %Pu240eff has been estimated for the three drums, based on the reported Pu238, Pu240 and Pu242 results. The average biases in the %Pu240eff are -2%, 3% and 12%, and the standard deviations are 8%, 17% and 15% respectively.
- The %Pu240eff standard deviations computed from the data in Table 7 are slightly higher than the TMU values reported in Table 6, but are considered to be a more realistic representation of the typical performance obtained when measuring the plutonium isotopic composition in waste using HRGS because of the source of the data.

## 2.2 High Resolution Gamma Spectrometry (HRGS) for Plutonium Mass Determination

### 2.2.1 Fundamentals

The plutonium content of a waste item can be quantified by measurement of the characteristic gamma-ray emission. The complex and generally unknown nature of the gamma spectra acquired during waste measurements (arising from

the unknown mix of radionuclides that may be present due to the unknown fuel history and burnup) means that HRGS is generally the preferred gamma technique.

In certain circumstances it may be acceptable to apply lower resolution techniques: For example, gross gamma counting or LRGS can be used to measure the plutonium content when the isotopic composition is well-known and only a simple mixture of non-interfering radionuclides is present (as discussed in Section 2.3). Intermediate resolution detectors such as Cadmium-Zinc-Telluride (CdZnTe) are not currently considered suitable for plutonium waste assay because of limitations in the size of detector crystal that can be grown, which limits their applicability to measurements of large quantities of plutonium (e.g. product cans or spent fuel measurements).

It is also possible to perform an indirect measurement of the plutonium content. That is, measurement of another radionuclide commonly present in the waste such as Cs137, and using a “fingerprint” (usually derived from chemical sampling of the waste and/or knowledge of the waste generating process and plants) to relate the Cs137 activity to the plutonium mass. However, these indirect measurements are generally only used for spent fuel wastes, where the intense gamma emissions from fission and activation products completely mask the characteristic gamma emissions from plutonium, as discussed further in Section 4.1.

Table 8 lists some of the key gamma-rays characteristic of plutonium.



**Table 8:** Principal Characteristic Gamma-rays from Plutonium and Am241 (Reference [17])

Isotope	Energy (keV)	Intensity (gammas/s-g)	Isotope	Energy (keV)	Intensity (gammas/s-g)
Pu238	152.7	$6.1 \times 10^6$	Am241	59.6 125.3 146.6 169.6 208.0 267.5 322.5 332.3 335.4 368.6 376.6 419.2 619.0 662.4	$4.5 \times 10^{10}$ $5.2 \times 10^6$ $5.8 \times 10^5$ $2.2 \times 10^5$ $1.0 \times 10^6$ $3.3 \times 10^4$ $1.9 \times 10^5$ $1.9 \times 10^5$ $6.3 \times 10^5$ $2.7 \times 10^5$ $1.7 \times 10^5$ $3.6 \times 10^4$ $7.5 \times 10^4$ $4.6 \times 10^5$
Pu239	129.3	$1.4 \times 10^5$			
	203.5	$1.3 \times 10^4$			
	332.8	$1.1 \times 10^4$			
	345.0	$1.3 \times 10^4$			
	375.0	$3.6 \times 10^4$			
	380.2	$7.0 \times 10^3$			
	382.8	$5.9 \times 10^3$			
	413.7	$3.5 \times 10^4$			
	451.5	$4.3 \times 10^3$			
	640.1	$1.8 \times 10^2$			
	645.0	$3.3 \times 10^2$			
Pu240	160.3	$3.4 \times 10^4$			
	642.5	$1.1 \times 10^3$			
Pu241	148.6	$7.1 \times 10^6$			
	160.0	$2.6 \times 10^5$			
	164.6	$1.7 \times 10^6$			
	208.0	$2.0 \times 10^7$			
	267.5	$6.8 \times 10^5$			

Generally measurement of the plutonium content will be based upon a characteristic gamma-ray from Pu239, since Pu239 is normally the most abundant isotope present and it emits several relatively high intensity gamma-rays, some of which are not normally interfered with by the other radionuclides that may be present. The most common choice is the 413.7 keV gamma-ray, which has the advantage of being at a relatively high energy and therefore can penetrate through more dense wastes. An alternative option is the 129.3 keV gamma-ray, which is four times more abundant and can therefore lead to improved detection limits if the waste density is low.

The count rate in the characteristic gamma peak is converted to isotope mass and then combined with a measurement of the isotopic composition to determine the total plutonium mass. A description of the determination of the plutonium isotopic composition using HRGS and the associated performance values is provided in Section 2.1.

To achieve quantitative assay of the plutonium content using HRGS the system must be calibrated for the specific measurement geometry. To obtain an accurate result, corrections must be determined and applied for the following:

- the distribution of plutonium within the waste item,

- attenuation of the measured gamma-ray signal within the waste matrix,
- self-attenuation of the measured gamma-ray signal within the plutonium,
- the isotopic composition of the plutonium.

In the case of waste assay, all the above factors are generally unknown and corrections must be assumed or determined via measurement. The larger volume of the waste item necessitates a larger system, with perhaps multiple HRGS detectors or a scanning arrangement, and the potential for significant attenuation of the gamma-ray signal in the waste matrix necessitates some form of matrix correction.

An HRGS detector is usually located within a collimator. The collimator serves both to reduce the effects of background radiation and to provide a well defined field-of-view. The field-of-view can be optimised to match the size of the object being measured or it can be deliberately set to a smaller segment of the object. In the latter case, multiple measurements are made to achieve complete coverage of the object being measured, and by applying different efficiency calibrations and attenuation corrections to each segment, the uncertainty due to geometrical effects can be reduced. For plutonium assay, it is common for the

collimator to incorporate a thin cadmium “window” between the detector crystal and waste item (typically 0.5 mm to 1 mm thick), to reduce the count rate from intense Am241 59.5 keV gamma emission, avoiding excessive dead-time in the gamma counting nucleonics.

To correct for attenuation in the waste matrix, several approaches are possible as discussed in more detail in Section 2.2.2. The most common approach for accurate assay

is to use an external gamma source to measure the transmission of gamma-rays through the waste item to determine the required attenuation correction at the gamma energy of interest.

Corrections for self-attenuation are more difficult to perform due to the high density of plutonium (particularly if in metallic form) which means the effect can be significant in physically small “lumps”.

Benefits of HRGS	Limitations of HRGS
<ul style="list-style-type: none"><li>• Allows measurement of complex mixtures of gamma emitters.</li><li>• May be used to detect unexpected radionuclides or to interpret unknown radionuclide mixtures.</li><li>• Energy dependant matrix correction can be applied to the measurements.</li><li>• Can provide direct measurement of the plutonium isotopic composition.</li></ul>	<ul style="list-style-type: none"><li>• Detectors must be operated at liquid nitrogen temperatures requiring a ready supply of liquid nitrogen or electrical coolers.</li><li>• Can be prone to interference in electrically noisy environments.</li><li>• Problem when high density and/or high Z matrix present.</li><li>• Can be prone to fast neutron damage when exposed for prolonged periods to high neutron emitting samples.</li></ul>

2.2.2 Instrumentation

The HRGS instruments that are used for plutonium mass determination can be divided into three different configurations; Fixed Detector, Segmented Gamma Scanner (SGS) and Tomographic Gamma Scanner (TGS). The first two of these types of system may also have single or multiple HRGS detectors installed to improve the measurement performance (i.e. increase the precision of the peaks in the gamma spectra for a given measurement time).

**Fixed Detector** – The HRGS detector (or detectors if more than one is included) remain static during the measurement, having a fixed field-of-view of the waste container. If more than one detector is installed, the detectors will normally be collimated such that each has a partial view of the waste item. The waste container is normally rotated during the measurement to minimise the uncertainty due to heterogeneity of the waste matrix and source distribution.

**Segmented Gamma Scanner (SGS)** – In this type of system, either the HRGS detector(s) or waste container are moved vertically (scanned) relative to the field-of-view of the HRGS detector(s), which is (are) collimated to view only a thin vertical segment of the waste item. The advantage of scanning the whole waste item in this way is that the uncertainty due to heterogeneity of the source distribution and waste matrix is reduced compared to a fixed detector system, in which a larger volume of the waste item is viewed by the detector(s).

**Tomographic Gamma Scanner (TGS)** – The TGS is similar to an SGS except that the waste container is also translated in addition to the vertical scanning of the HRGS detector(s). This allows a three-dimensional image of the

gamma emission (and matrix attenuation if an external transmission source is used) to be constructed, further reducing the uncertainty caused by variations in waste matrix density and source distribution.

The method of correction for attenuation of the gamma-ray signal in the waste matrix may also be different, depending on the performance requirements of the instrument system. Three different approaches are commonly applied; transmission source, Differential Peak Attenuation (DPA) and weight correction.

**Transmission Source** – A transmission source is often used to directly measure the attenuation of the gamma-ray signal in the waste matrix using an external gamma source. The source is mounted opposite the HPGe detector on the other side of the waste container, and the transmission fraction for the gamma-rays from the source through the matrix is measured. The transmission source is selected to ensure that the gamma-ray energies span the range of interest for the measurement. Eu152 is a common choice, because it emits several intense gamma-rays in the energy range 122-1408 keV covering both plutonium, uranium and other beta/gamma isotopes (such as Co60 and Cs137) that may be present. The transmission source measurement is normally performed as a separate measurement, thereby increasing the total assay time. In SGS systems, the transmission source is scanned over the full height of the waste container, allowing different transmission corrections to be applied at different heights in the waste container. However, since no information on the radial distribution of plutonium is obtained, a particular radial distribution must be assumed (usually uniform). In a TGS system, the assumption of radial uniformity is avoided



because the TGS transmission measurements are used to build up a three-dimensional image of the waste matrix attenuation. In very dense waste matrices, the gamma peaks from the transmission source may fail to be detected, and this technique is therefore generally only applicable to waste matrix densities below about 1 g/cm<sup>3</sup>.

**Differential Peak Attenuation (DPA)** – The DPA technique utilises the fact that plutonium emits several intense gamma-rays across a range of different gamma energies, and relies on being able to separately resolve gamma-rays from the same isotope. After correction for differences in the intrinsic detection efficiency of the HPGe detector, the ratios of the measured gamma peak intensities ought to be the same as the relative abundances (if there is no attenuation). However, if there is attenuation of the gamma-ray signal, the difference in attenuation with gamma energy will bias the measured gamma peak ratios. The measured difference in the gamma peak ratios can therefore be used to calculate the amount of attenuation of the gamma-ray signal. This technique can be accurate if the measured gamma peaks are precise, the distribution of plutonium is close to a uniform distribution and there are no complications such as a large lump of plutonium being shielded by a large number of smaller lumps. If, however, the plutonium is not uniformly distributed, the DPA analysis will be biased towards the plutonium that is closer to the detector (in a more efficient position). The bias will also be worse at higher waste matrix densities. The technique is also powerful for light matrices but applying a self-attenuation correction factor for lumps of plutonium.

**Density Correction** – The simplest but least accurate approach is to calculate the matrix density using the weight of the waste container (which is usually known or is easily measured) combined with an estimate of the container fill height (generally unknown). In order to determine the correction to apply to the measured gamma signal, the elemental composition of the material present in the waste container must also be known or assumed. For plutonium measurements, the gamma energy used to quantify the

plutonium is generally the 413.7 keV gamma peak from Pu239, which is high enough in energy such that incorrect assumptions about the matrix material will have a small (and almost negligible) effect. Again, the accuracy of the matrix correction will depend on how closely the true distribution of plutonium is to that assumed. This approach also does not account for local variations in matrix density (heterogeneity), differences in the container fill height, or the presence of small massive objects within the container.

The other major source of uncertainty in the measurement of the plutonium content is self-attenuation (or self-shielding) within the plutonium itself. The magnitude of this effect depends on a number of factors such as the density of the plutonium, the size / shape of the “lump”, the chemical form and the gamma-ray energy being measured. The instrument system may employ a correction for this effect which is referred to as “Lump Correction”. The methods employed generally rely upon measurement of several different gamma-rays from plutonium across a range of gamma energies, making some correction for the attenuation within the matrix materials. An example self-attenuation correction is the “Infinite Energy Extrapolation Method” developed by Jean Morel (Reference [18]). In this technique, an apparent mass of plutonium (which implies the previous determination of the isotopic abundances of Pu239 and Pu241) is calculated at each available gamma energy (in the range 120 – 500 keV) and corrected for matrix attenuation using a transmission measurement. The Pu mass is plotted against the inverse of the Pu239 or Pu241/U237 gamma energy, and by extrapolating back to infinite energy (where the attenuation will be zero), it is possible to determine the self-attenuation corrected mass. The quoted accuracy of this technique is  $\pm 15\%$ .

In some systems, accurate knowledge of the waste stream precludes the presence of significant lumps of plutonium, and in these cases no correction is required. It may also be possible to use process knowledge to place realistic bounds on the range of the possible lump correction factors, and use this information in the Total Measurement Uncertainty budget to account for the potential bias.

**Segmented Gamma Scanner (SGS)****Tomographic Gamma Scanner (TGS)****Figure 2:** Example Instrument Systems Employing HRGS**2.2.3 Performance Values**

Performance data for the measurement of the plutonium mass in waste using HRGS is reported in Table 9. The performance is reported for several different types of system, including both fixed detectors, SGS and TGS systems. In addition to the system configuration, information has been included on the matrix correction employed and any other special algorithms or features that are used to improve the system performance.

Note that the information from instrument systems with similar configurations and performance has been combined where possible, by combining the data and reporting

a range of performance values to span the reported values for individual systems. The information has been organised with the least complex instrument systems (i.e. fixed detector(s)) listed first in the table.

Note that in several cases the reported values are for more than one container size (e.g. 100l & 200l drums or 200l & 400l drums). This indicates that the system(s) is capable of measuring both sizes, and unless stated otherwise, the reported performance values are applicable to both container sizes.

Note that the Limits of Detection (LODs) in Table 9 are in terms of Pu239 mass and not total Pu mass.

**Table 9:** Performance Values for Pu239 Mass Measurement using HRGS

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1σ), g Pu239	TMU (1σ)		
Fixed Detector Systems						
<b>Geometry:</b> Fixed detector <b>Detectors:</b> 1 x BEGe <b>Matrix Correction:</b> Density <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> NO <b>Notes:</b> Integrated with PNCC	200l Drum	Debris	-	± 10-12%	2100 s	11, 14, 15
		Homogeneous Solids	-	± 16-19%		
		Combustibles	10-20 mg	-		
<b>Geometry:</b> Fixed detector <b>Detectors:</b> 1 x HPGe (e = 80%) <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> NO <b>Validity domain:</b> Pu surface mass < 3 g/cm² <b>Notes:</b> SGS for Transmission using HPGe (e = 20%)	100l & 200l Drums	PVC / Metals (<0.4 g.cm <sup>-3</sup> )	-	± 15% on the Pu mass	1800 s to several hours <sup>Note 1</sup>	2
Segmented Gamma Scanners (Single Detector)						
<b>Geometry:</b> SGS <b>Detectors:</b> 1 x HPGe (e = 10%) <b>Matrix Correction:</b> Density <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b>	200l & 400l Drums	Mixed (0.03 to 0.6 g.cm <sup>-3</sup> )	-	No lumps: ± 4-32% (12%) (homo) <sup>Note 2</sup> > 63% (hetero) With lumps: ± 4-45% (homo) > 70% (hetero)	6000 s	4
<b>Geometry:</b> SGS <b>Detectors:</b> 1 x BEGe <b>Matrix Correction:</b> Density & Transmission <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> NO <b>Notes:</b>	200l Drum	Combustibles (0.1 g.cm <sup>-3</sup> )	-	± 18-28%	1800 s	6, 13
		Graphite (0.25 g.cm <sup>-3</sup> )	-	± 21-29%		
		Metals (0.5 g.cm <sup>-3</sup> )	-	± 28-31%		
		Combustibles (0.02 g.cm <sup>-3</sup> )	26-55 mg	-		
<b>Geometry:</b> SGS <b>Detectors:</b> 1 x HPGe <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b>	200l Drum	Combustibles / PVC (<0.2 g.cm <sup>-3</sup> )	<4 mg (129keV) <15 mg (414keV)	± 15% (414keV)	10800 s	30
		Mixed (low Z) (<0.5 g.cm <sup>-3</sup> )	-	± 65% (414keV)		

<sup>Note 1</sup> The count time depends on the Pu mass to be measured (from some hundred milligrams to some hundred grams of Pu), the form and the distribution of Pu and the nature of the matrix. For instance, 1g of Pu (PuO<sub>2</sub> powder form) in a 0.2 g/cm<sup>3</sup> combustible matrix can be measured in 30 minutes whereas 100 mg of Pu (PuO<sub>2</sub> powder form) in a 0.2 g/cm<sup>3</sup> combustible matrix needs several hours of acquisition.

<sup>Note 2</sup> Value in brackets assumes negligible counting statistics. "Homo" is an abbreviation of homogenous, and "Hetero" is an abbreviation of heterogeneous.

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1σ), g Pu239	TMU (1σ)		
Segmented Gamma Scanners (Multiple Detectors)						
<b>Geometry:</b> SGS <b>Detectors:</b> 3 or 4 x BEGe <b>Matrix Correction:</b> Density & Transmission <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> YES (15cm steel) <b>Notes:</b>	200l Drum	Combustibles (0.1 g.cm <sup>-3</sup> )	-	± 17%	600 s	7, 9, 10
		Graphite (0.25 g.cm <sup>-3</sup> )	-	± 20%		
		Metals (0.5 g.cm <sup>-3</sup> )	-	± 27%		
		Combustibles (0.02 g.cm <sup>-3</sup> )	2-4 mg	-	600 s	
		Combustibles (0.02 g.cm <sup>-3</sup> )	0.2 - 0.4 mg	-	1200 s	
		Debris (1.6 g.cm <sup>-3</sup> )	27 mg	-	600 s	
		Concrete (1.9 g.cm <sup>-3</sup> )	10 mg	-	1200 s	
<b>Geometry:</b> SGS <b>Detectors:</b> 3 x HPGe (e = 20%) <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> YES (15cm steel) <b>Notes:</b>	200l Drum	Combustibles / PVC (0.1 g.cm <sup>-3</sup> )	5 mg (in 900s)	+14% -11% (few g Pu239) +12% -8% (~100g Pu239)	1200 s	28
		Mixed / Metals (0.6 g.cm <sup>-3</sup> )	9 mg (in 900s)	+18% -9% (few g Pu239) +21% -13% (~100g Pu239)	1200 s	
		Mixed / Filters	-	± 13% (homo)	1800 s	5
	<b>Geometry:</b> SGS <b>Detectors:</b> 4 x HPGe (e = 18%) <b>Matrix Correction:</b> Transmission & Density <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> NO <b>Notes:</b> LRGS for Transmission	200l Drum	Debris (0.3 g.cm <sup>-3</sup> )	-	± 27%	3600 s
Mixed (0.4 g.cm <sup>-3</sup> )			20 mg	-		
Large Crate (SWB)		Debris (0.3 g.cm <sup>-3</sup> )	-	± 40%		
		Mixed (0.13 g.cm <sup>-3</sup> )	32 mg	-		
Large Crate (SLB-2)		Debris (0.3 g.cm <sup>-3</sup> )	-	± 50%		
		Mixed (0.17 g.cm <sup>-3</sup> )	51.6 mg	-		
Tomographic Gamma Scanners (Single Detector)						
<b>Geometry:</b> TGS <b>Detectors:</b> 1 x HPGe (e = 50%) <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b> Separate HRGS for isotopic determination, Cd109 rate loss correction	200l Drum	Debris (0.05 g.cm <sup>-3</sup> )	0.28 g	± 6.5%	3600 s	32

The following points are noted regarding the results in Table 9:

- For the systems measuring 200l drums, similar performance values are reported for all systems regardless of their complexity. It might be expected that the more complex (and therefore more expensive) systems would yield better performance. It is believed that this consistency of performance is likely to reflect the usage of the systems; that is, the least complex systems are generally used for better defined waste streams (i.e. low density, segregated and mostly homogeneous), whereas the higher complexity systems are used for more matrices with a greater range of densities and degree of heterogeneity.
- As expected, both the detection limit and TMU performance is significantly worse for large crates compared to 200 litre drums. The performance of the system designed to measure both large crates and 200l drums also has poorer performance for 200l drum measurements compared to the systems designed only to measure 200l drums. Clearly, the 200l drum measurement performance has been compromised in order to assay a wider range of container sizes.

Additionally, the performance of HRGS systems measuring the plutonium mass within simulated waste drums was evaluated in a recent round-robin exercise performed in France (Reference [21]). Plutonium measurements were performed on three 118 litre simulated waste drums containing soft waste, although one drum included some added metal plates. The total mass of plutonium present in each drum was in the range 100 to 200 mg.

Note that the data from the intercomparison exercise are different to that presented in the previous table (and the general structure of the other performance values tables), because only the measurement technique employed is recorded (in this case HRGS) and no specific details regarding the type and configuration of the instrument systems being used was provided (although the number, in this case nine, and location of participants is known). For this reason it is considered appropriate to report these data as a separate table to illustrate the performance that is achievable in the field using HRGS to measure the plutonium mass.

The results from the French intercomparison exercise are summarised below in Table 10. Note that the average % bias and % standard deviation obtained from the results reported by the different participating laboratories are presented. Although the intercomparison included results for all plutonium isotopes, only the Pu239 results are presented below to allow direct comparison with Table 9.

**Table 10:** Performance Values for Pu239 Mass Measurement using HRGS from French Intercomparison Exercise – EQRAIN 4 (Reference [21])

Simulated Waste Drum	Pu239 Mass	
	Average Bias	Standard Deviation (1σ)
118 litre, Soft waste (90% PVC)	-28%	35%
118 litre, Soft waste	13%	18%
118 litre, Soft waste + Metals	31%	35%

The following point is noted regarding the results in Table 10:

- The percentage values in Table 10 are comparable with the reported TMU values in Table 9 and are considered to represent the typical accuracy that is achievable using this measurement technique.

**2.3 Low Resolution Gamma Spectrometry (LRGS) for Plutonium Mass Determination**

*2.3.1 Fundamentals*

As discussed in Section 2.1.1, LRGS can be used to directly measure the plutonium content when the isotopic composition is known and the gamma spectrum is relatively simple, with only a few non-interfering radionuclides present. This is because the most common detectors used in LRGS systems are thallium-activated sodium iodide [NaI(Tl)], which have an energy resolution (FWHM) of 40-60 keV at the Cs137 energy of 662 keV. For comparison, the resolution of HRGS detectors at this energy is typically 1.5 keV. The poorer resolution of NaI detectors means that it is not possible to perform accurate plutonium isotopic analysis and correct for the presence of interfering gamma peaks.

LRGS detectors do have the advantage over HRGS detectors of not requiring cooling to liquid nitrogen temperatures, although the associated nucleonics are sensitive to drifts in temperature and generally require a temperature controlled environment or compensation for temperature-induced gain drift. LRGS detectors are also cheaper and can generally be made larger, meaning that high detection efficiency can be obtained. The choice of detector size and type (i.e. scintillator material) is dependent on the application, since larger detectors are used when a high counting efficiency is required or when the gamma-rays of interest are high energy and are unlikely to deposit all of their energy in a smaller crystal. Alternative scintillator materials such as caesium iodide (CsI) and bismuth germanate (BGO) offer better stopping power for a given crystal size, but have poorer light transmission compared to NaI. BGO is useful for gamma measurements in high



ambient neutron backgrounds as the material is relatively insensitive to neutron capture.

For plutonium assay, the large number of gamma-rays emitted means that different options may be used when setting up a Region of Interest (ROI) for analysis of the LRGS gamma spectrum. For example:

- (i) Measurement of the intense low energy gamma-ray at 59.5 keV from Am241, which is then related to the total plutonium content using prior knowledge of the plutonium isotopic composition.

- (ii) Selection of a higher energy ROI in the 200 keV to 450 keV energy region, which has the advantage of being more penetrating in denser wastes. Depending on the characteristics and origin of the plutonium, it may also be possible to select a region where the count rate shows a relatively low dependence on the isotopic composition (because the region contains a mixture of peaks associated with Pu239, Pu241 and Am241).

For waste assay, Option (ii) is likely to give a more accurate measurement because of the higher gamma energy.

Benefits of LRGS	Limitations of LRGS
<ul style="list-style-type: none"><li>• Relatively inexpensive detectors and nucleonics.</li><li>• High detection efficiency can be obtained.</li><li>• Room temperature operation.</li><li>• Low maintenance.</li></ul>	<ul style="list-style-type: none"><li>• Poor resolution means isotopic composition must be known and only a simple mixture of non-interfering radionuclides is present.</li><li>• Unexpected interferences may not be detected and may cause bias.</li><li>• Nucleonics susceptible to temperature-induced gain drift.</li><li>• Poor resolution limits use of advanced matrix correction techniques such as transmission sources or differential peak absorption.</li></ul>

2.3.2 Instrumentation

LRGS instruments are simpler than their HRGS counterparts because of the lack of special cooling requirements.

Standard 3 inch diameter, 3 inch long, cylindrical NaI(Tl) crystals are a common choice as they give good all round performance for the detection of gamma-rays in the range 30 keV to 2 MeV and are therefore suitable for plutonium assay.

It is common to incorporate an Am241 “seed” within the detector crystal to provide gain stabilisation of the gamma spectrum to cope with ambient temperature fluctuations. The alpha particles emitted by Am241 result in a peak in the spectrum with a gamma equivalent energy of approximately 3 MeV. The position of this peak is monitored for drift and the amplifier gain automatically adjusted to compensate.

The configuration of detectors will depend on the size of the item being measured and other constraints such as the measurement time and performance requirements. Similar options to those discussed in Section 2.2.2 for HRGS can be implemented, but as LRGS systems are less commonly

applied to assay waste containing special nuclear materials, the technology is less well developed. Several systems employing multiple fixed detectors are known to be used for this application, but it is not known whether there are any scanning detector systems in use.

Similar to HRGS systems, LRGS detectors are usually mounted within collimators to provide background shielding and to limit the detector field of view to part or the entire waste container. A cadmium shield may again be incorporated to reduce the count rate from the intense Am241 59.5 keV gamma emission and avoid excessive dead-time.

LRGS systems also generally use weight correction to compensate for attenuation of the measured gamma signal by the waste matrix. This is because the poor detector resolution makes it difficult to select a suitable source for transmission source correction or to apply the differential peak absorption technique.

2.3.3 Performance Values

Performance data has been obtained for an LRGS system measuring plutonium waste in a variety of waste containers.

**Table 11:** Performance Values for Measurement of Pu239 Mass using LRGS

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ ), g U235	TMU (1 $\sigma$ )		
<b>Geometry:</b> 3x fixed NaI detectors  <b>Matrix Correction:</b> Weight (based on known matrix & fill height)  <b>Lump Correction:</b> None  <b>Notes:</b>	200l Drum	Mixed	-	$\pm 25\%$	300 s	36
	Waste Bag	PVC	-	$\pm 6\%$		
	Waste Bag	Metals	-	$\pm 17\%$		
	Waste Bag	Filters	-	$\pm 7\%$		

## 2.4 Passive Neutron Counting Techniques for Plutonium Mass Determination

### 2.4.1 Fundamentals

Measurement of the plutonium content using passive neutron counting techniques relies upon the detection of neutrons emitted from plutonium. The even numbered plutonium isotopes (i.e. Pu238, Pu240 and Pu242) have a relatively high neutron emission rate from spontaneous fission, resulting in bursts of a small number of “coincident” neutrons. Furthermore, most plutonium isotopes (the exception being Pu241) and Am241 (a decay product of Pu241), decay by alpha particle emission resulting in the emission of single “random” neutrons through ( $\alpha$ , n) interactions with any surrounding low atomic number materials (e.g. oxygen if the plutonium is in oxide form).

In passive neutron counting, the assayed quantity is usually reported as an effective (or equivalent) Pu240 mass,  $m_{\text{eff}}$ , which is the mass of Pu240 that would give the same response as that which was measured. The measured response includes contributions from all the spontaneously fissionable isotopes of plutonium (i.e. Pu238, Pu240 and Pu242), as well as any other spontaneously fissionable radionuclides that may be present (e.g. Cm242, Cm244, Cf252, and U238).

For the interpretation of passive neutron counting data using the point model<sup>1</sup>, four variables are usually considered:

- Effective mass of Pu240,  $m_{\text{Pu240eff}}$ .
- Neutron leakage multiplication,  $M_L$ , which arises when neutrons from either spontaneous fission or ( $\alpha$ , n) reactions induces another fission in the sample.
- Efficiency of the counting system,  $\epsilon$ .

<sup>1</sup> The Bohnel point model assumes that the detection efficiency and probability of spontaneous fission are uniform across the sample volume (i.e. equivalent to assuming all neutrons are emitted from one point). The detection efficiency assumption is usually addressed by careful design of the measurement chamber.

- Ratio of neutrons produced by ( $\alpha$ , n) reactions to those produced by spontaneous fission,  $\alpha$ .

In order to fully characterise the measured waste item, at least four measurable quantities are needed to determine these four unknowns, otherwise assumptions for some of these parameters must be made. When assaying plutonium in waste items, all four of these parameters may be unknown and variable due to a lack of knowledge of the plutonium characteristics and the large volume of the waste item plus potential presence of neutron interfering materials.

Background neutrons from sources external to the assay system can be an issue in passive neutron counting systems, particularly in waste plants which may have other waste containers stored nearby. Passive counting systems usually perform regular background measurements with an empty measurement chamber and subtract the measured background from the count rates measured with the waste item present. This approach is acceptable when the background is fairly constant, but if the background radiation fluctuates, for example due to movements of waste items in the vicinity, then variations in the background will increase the total measurement uncertainty. Performing empty chamber background measurements also does not account for background neutrons arising from cosmic-ray interactions with high atomic number materials in the waste item (and waste container itself). This source of background can be significant if a low detection limit is a priority or if the waste item contains large masses of high atomic number materials (e.g. lead bricks).

The detectors most commonly used in passive neutron waste assay systems are He3 proportional detectors which are sensitive to thermal neutrons. These are housed in polythene slabs which serve to moderate the fast neutrons emitted from the sample to thermal energies which can be detected. The measurement chamber which holds the sample is surrounded by a number of these slabs to increase the detection efficiency. The inner surface of the measurement chamber is often lined with a layer



of cadmium which has a high capture cross section for thermal neutrons. This serves to prevent thermalised neutrons from re-entering the sample chamber and inducing further fission events in the sample. The inclusion of this liner is also often a requirement to allay the possibility of an unforeseen criticality event.

2.4.2 *Passive Total Neutron Counting (TNC)*  
*Instrumentation*

In Total Neutron Counting (TNC), all neutrons are counted and no attempt is made to discriminate between those which originate from spontaneous fission or ( $\alpha$ , n) interactions with surrounding materials. In order to determine the plutonium mass, knowledge is required of the  $\alpha$  ratio, leakage multiplication ( $M_L$ ), detection efficiency ( $\epsilon$ ) and isotopic composition. Background correction is normally carried out by subtracting the count rate measured in a back-

ground measurement performed immediately before or after the waste item measurement.

For samples which are well characterised, TNC can provide an accurate, simple, rapid and cost effective solution. However, large errors can occur when attempting to assay unknown samples particularly if the ( $\alpha$ , n) component is large due to the presence of light elements such as fluorine, oxygen, boron or aluminium.

In waste assay, TNC is particularly suited to point-of-origin measurements of waste bags, for example, where good knowledge of the characteristics of the plutonium is available.

TNC is also valuable for assay of radioactive species where one source of neutrons dominates, for example uranium hexafluoride ( $UF_6$ ).

Benefits of TNC	Limitations of TNC
<ul style="list-style-type: none"><li>Simple and robust electronics and data processing.</li><li>Generally superior statistical precision compared to other neutron counting techniques.</li><li>Relatively unaffected by the presence of high density and/or high Z materials.</li></ul>	<ul style="list-style-type: none"><li>If the sample characteristics are not well known, can result in a large over or under estimate of the plutonium mass.</li><li>Requires knowledge of the isotopic composition.</li><li>Requires knowledge of the chemical composition (affects <math>\alpha</math>) and / or matrix materials.</li><li>Requires knowledge or determination of the sample leakage multiplication (<math>M_L</math>) to provide accurate plutonium mass.</li><li>Sensitive to background fluctuations.</li><li>Sensitive to changes in efficiency (<math>\epsilon</math>) due to moderators / absorbers in chamber.</li><li>Highly dependent on system calibration, which ideally should be carried out under as realistic conditions as possible.</li></ul>

- Measures total plutonium mass of waste packages (bags) at point of origin, to facilitate optimum filling of waste drum.
- Suitable for waste packages containing hard, soft or mixed hard and soft waste.
- Uses total neutron counting combined with an assumed isotopic and chemical composition.
- Operating range: ~0 to 1 kg plutonium.
- Measurement time: Typically 100 s.
- Limit of detection: Typically 100 mg plutonium.
- Total Measurement Uncertainty: Typically  $\pm 10\%$ . ( $1\sigma$ )



Figure 3: Example Instrument System Employing TNC – Portable Packet Monitor

### 2.4.3 Passive Total Neutron Counting (TNC) Performance Values

Performance data for the measurement of the plutonium mass in waste using TNC is reported in Table 12. Perfor-

mance data has only been obtained for a single measurement system used to measure waste bags.

**Table 12:** Performance Values for Measurement of Total Plutonium Mass using TNC

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ )	TMU (1 $\sigma$ )		
<b>Detection Efficiency:</b> 2% <b>Matrix Correction:</b> None (assumes PVC @ 0.3 g.cm <sup>-3</sup> ) <b>Notes:</b> Assumed isotopics, Assumed (a, n), assume no multiplication	Bag	Combustibles, PVC, Metals, Mixed	< 1g <sup>1</sup>	± 20% <sup>Note 1</sup>	100s	19

<sup>Note 1</sup> Performance values dependent on validity of the assumptions made in the calibration (i.e. plutonium isotopics, chemical form and multiplication).

As noted above, the performance of TNC is highly dependent on validity of the calibration assumptions (i.e. plutonium isotopic composition, chemical form and multiplication). These systems are therefore used for measurements at the point of waste generation, where the characteristics of the waste are very well defined. For measurements of waste containing mixed waste streams or waste of unknown origin, the validity of the assumptions will be challenged and the performance significantly worse than that quoted in Table 12.

### 2.4.4 Passive Neutron Coincidence Counting (PNCC) Instrumentation

In Passive Neutron Coincidence Counting (PNCC), electronic circuitry is used to discriminate between the coincident (time-correlated) neutrons produced in bursts from spontaneous fission events, and random neutrons produced in ( $\alpha$ , n) and other reactions. This discrimination permits PNCC measurements to be made even in the presence of high ( $\alpha$ , n) or high ambient background conditions.

In plutonium, the spontaneous fission neutrons originate from the even number plutonium isotopes (i.e. Pu238, Pu240 and Pu242) and any other spontaneous fission radionuclides present (e.g. Cm242, Cm244, Cf252 and U238). The Pu240 effective mass result can be defined as follows, taking into account the different neutron multiplicities and energy spectra of the plutonium isotopes:

$$\text{Pu240 effective mass} = (2.58 \times \text{Pu238}) + \text{Pu240} + (1.71 \times \text{Pu242})$$

A reference listing the Pu240 effective coefficients for the curium, californium, uranium and plutonium spontaneous fission isotopes is Reference [19].

When assaying the plutonium content of waste, the plutonium masses are generally small and leakage multiplication

is generally a small effect. However, leakage multiplication can become significant if large plutonium masses are encountered (Reference [5]).

PNCC can be combined with TNC, allowing two of the four unknown parameters ( $m_{\text{Pu240eff}}$ ,  $\epsilon$ ,  $a$  and  $M_L$ ) to be determined. The two measured signals are often referred to as “Totals” and “Reals”.

- For example, PCM waste assay systems may be configured so that the plutonium mass and multiplication are solved using a known detection efficiency (from calibration combined with a measured correction for neutron moderation in the waste matrix) and an assumed a ratio (i.e. assuming PuO<sub>2</sub>) – this is the Krick-Ensslin Method.
- Alternatively, the “Reals” count rate can be converted directly to a Pu240eff mass using a calibration factor and a measured correction for the effect of the waste matrix. This approach assumes multiplication is as per the calibration factor (note this gives overestimate) and a makes no contribution to the “Reals” rate. Although (a, n) neutrons are single and therefore do not directly contribute to the Reals, they do contribute to multiplication and the subsequent bursts of neutrons from induced fission can still overestimate the plutonium content if not taken account of in the calibration.

For impure and heterogenous materials, such as residues (e.g. glovebox sweepings) and waste, corrections for matrix, leakage multiplication and other effects can become significant and the two measured count rates may not be sufficient for accurate assay.

In PNCC systems, background is again usually corrected by performing an empty chamber measurement before or after the waste item measurement. Background interference from external neutron sources is a lesser effect in PNCC

systems compared to TNC systems, since an external source of neutrons is much more likely to result in uncorrelated neutron events. In systems where a low detection limit is required, the “Reals” from cosmic-ray induced neutron events is generally the largest background component.

In PCM waste assay, matrix effects can significantly change the passive neutron detection efficiency. Two different approaches are commonly applied for determining the required efficiency correction in PNCC instruments: (i) Flux Probe or Ring Ratio and (ii) Add-A-Source.

#### (i) Flux Probe / Ring Ratio:

A separate group of bare or under-moderated neutron detectors are used to measure the signal relative to the main detector group, and this signal is related to the amount of neutron moderation within the waste matrix during the measurement. Flux Probes are normally bare He3 detectors inside the measurement chamber. The Ring Ratio

uses an under-moderated detector group on the same side of the cadmium absorber as the main detector ring, comparing the responses of the two groups (the relative efficiencies of the two groups is dependent on the neutron energy and hence the matrix). Alternatively, two or more “rings” of He3 detectors, embedded in the polyethylene walls by different amounts, are compared.

#### (ii) Add-A-Source:

An external (Cf252) neutron interrogation source is introduced and the neutron signal measured with the waste item present is compared to that measured for an empty container to determine the amount of neutron moderation / absorption. In some cases, multiple measurements are performed at different source positions to characterise heterogeneous waste items.

A comparison of these two matrix correction techniques commonly used in PNCC systems is provided in Table 16.

**Table 13:** Comparison of Flux Probe / Ring Ratio and Add-A-Source PNCC Matrix Corrections

	Advantages	Disadvantages
<b>Flux Probe / Ring Ratio</b>	<ul style="list-style-type: none"> <li>No extra measurement required.</li> <li>No neutron source required</li> <li>Not biased by heterogeneous mixtures of plutonium and neutron absorbers / moderators.</li> </ul>	<ul style="list-style-type: none"> <li>Flux probe ratio differs for different absorbing materials, so need to assume a matrix type.</li> <li>Only reliable above a certain count rate, so default correction required at low plutonium masses.</li> </ul>
<b>Add-A-Source</b>	<ul style="list-style-type: none"> <li>Sensitive to effect of both moderators and absorbers (provided within calibration range).</li> <li>Will give correction even at low plutonium masses.</li> <li>Can interrogate at multiple positions to obtain spatial matrix information.</li> </ul>	<ul style="list-style-type: none"> <li>Requires extra measurement (s).</li> <li>Cf252 source needs shielding and has maintenance / replacement costs.</li> <li>Potentially biased by heterogeneous mixtures of plutonium and neutron absorbers / moderators.</li> <li>More complex.</li> </ul>

Benefits of PNCC	Limitations of PNCC
<ul style="list-style-type: none"> <li>Requires no knowledge of chemical composition, but very high (<math>\alpha, n</math>) levels (e.g. fluorides) may cause problems due to pile up of accidental coincidences and subsequent poor statistical precision.</li> <li>Relatively insensitive to background neutron radiation conditions.</li> <li>Relatively unaffected by the presence of high density and/or high Z materials.</li> <li>Abundant electronics and standard data processing algorithms.</li> <li>Knowledge required of the waste/stream parameters is generally less than TNC.</li> <li>Generally more accurate than TNC (although TNC can be more accurate if uncertainties are dominated by spatial effects, since TNC is proportional to <math>\epsilon</math> and PNCC is proportional to <math>\epsilon^2</math>).</li> </ul>	<ul style="list-style-type: none"> <li>Poorer precision than TNC.</li> <li>Other spontaneous fission emitters will cause a potential overestimate of plutonium content.</li> <li>Requires knowledge of isotopic composition because no discrimination is possible.</li> <li>Requires knowledge of and correction for leakage multiplication.</li> <li>Higher sensitivity to changes in efficiency due to neutron absorbers/moderators or variation of the position of the plutonium within chamber (varies with <math>\epsilon^2</math>). This limitation is minimised in the design of most systems, as well-designed measurement chambers will have very flat “empty” response profiles.</li> <li>Highly reliant upon the performance of an appropriate system calibration.</li> </ul>

- Final measurement of waste drums (200 or 400 litre) prior to storage or processing for repository disposal.
- Flux Probe or Add-a-Source matrix correction to determine properties of waste matrix.
- Uses PNCC combined with HRGS to determine plutonium isotopic composition.
- Neutron detection efficiency typically in range 10% to 30% (depends on chamber design and number of neutron detectors).
- Operating range: ~0 to 1 kg plutonium.
- Measurement time: Typically 1200 s.
- Limit of detection: Typically 10 mg plutonium.
- Total Measurement Uncertainty: Typically  $\pm 10\%$  ( $1\sigma$ ).



**Figure 4:** Example Instrument System Employing PNCC – Drum Monitor

#### 2.4.5 Passive Neutron Coincidence Counting (PNCC) Performance Values

Performance data for the measurement of the Pu240 effective mass in waste using PNCC is reported in Table 14. The performance is reported for several different systems with a range of detection efficiencies. In addition to the system configuration, information has been included on the matrix correction employed and any other special algorithms or features that are used to improve the system performance.

Note that the information from instrument systems with similar configurations and performance has been combined where possible, by combining the data and reporting

a range of performance values to span the reported values for individual systems. The information has been organised so that the lowest detection efficiency systems appearing first in the table.

Note that in several cases the reported values are for more than one container size (e.g. 100l & 200l drums or 200l & 400l drums). This indicates that the system(s) is capable of measuring both sizes, and unless stated otherwise, the reported performance values are applicable to both container sizes.

Note that the Limits of Detection (LODs) in Table 14 are in terms of Pu240 effective mass and not total Pu mass.

**Table 14:** Performance Values for Measurement of Pu240 Effective Mass using PNCC

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD ( $1\sigma$ ), g Pu240eff	TMU ( $1\sigma$ )		
<b>Detection Efficiency:</b> 15% <b>Matrix Correction:</b> None (assumed worst case) <b>Notes:</b> Count to precision algorithm, Integrated HRGS	Waste Items	Soft waste	0.2 g	11.3%	Typically 180 s	20
		Hard waste		9.5%	Typically 180 s	
<b>Detection Efficiency:</b> 14% <b>Matrix Correction:</b> AAS <b>Notes:</b>	100l & 200l Drums	PVC / Metals ( $<0.4 \text{ g.cm}^{-3}$ )	12 mg	$\pm 15\%$	2700 s	1

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ ), g Pu240eff	TMU (1 $\sigma$ )		
<b>Detection Efficiency:</b> ~20% <b>Matrix Correction:</b> MOD index from DDA measurement <b>Notes:</b> Integrated with DDA system, Imaging algorithms	200l Drums	Debris	22 - 209 mg	-	600 s	12
<b>Detection Efficiency:</b> 26% <b>Matrix Correction:</b> AAS <b>Notes:</b> Integrated with DDA system, Imaging algorithms	200l & 400l Drums	Combustibles / PVC	-	$\pm 3\%$	3600 s	29
		Concrete	-	$\pm 40\%$		
<b>Detection Efficiency:</b> ~30% <b>Matrix Correction:</b> AAS <b>Notes:</b> Integrated HRGS	200l Drums	Combustibles (0.1 g.cm <sup>-3</sup> )	15 - 17 mg	-	2100 s	11, 14, 15
		Debris	-	$\pm 9\text{-}14\%$		
		Mixed	-	$\pm 24\text{-}26\%$		
<b>Detection Efficiency:</b> 30% <b>Matrix Correction:</b> AAS <b>Notes:</b> Cosmic-ray background correction using PNMC	200l Drums	Combustibles / PVC (0.1 g.cm <sup>-3</sup> )	1.6 mg	$\pm 4\%$	1200 s	27
		Mixed / Metals (0.6 g.cm <sup>-3</sup> )	3.4 mg	$\pm 4\text{-}5\%$		
<b>Detection Efficiency:</b> 45% <b>Matrix Correction:</b> Flux Probe <b>Notes:</b> Cosmic-ray background shielding, Integrated with Cf Shuffler	200l Drums	Combustibles, PVC, Metals, Lead, Filters, Mixed, Rubble (<0.1 - 1 g.cm <sup>-3</sup> )	0.6 mg <sup>Note 1</sup>	$\pm 14\%$	1200 s	26
<b>Detection Efficiency:</b> 30% <b>Matrix Correction:</b> AAS <b>Notes:</b> Cosmic-ray background correction	200l Drums	Low Moderating	-	$\pm 13\%$	3600 s	8
		High Moderating	-	$\pm 63\%$		
		Mixed (0.02 g.cm <sup>-3</sup> )	4.4 mg	-		
	Large Crate (SWB)	Low Moderating	-	$\pm 12\%$		
		High Moderating	-	$\pm 80\%$		
		Mixed (0.2 g.cm <sup>-3</sup> )	4.8 mg	-		
	Large Crate (SLB-2)	Low Moderating	-	$\pm 15\%$		
		High Moderating	-	$\pm 86\%$		
		Mixed (0.2 g.cm <sup>-3</sup> )	12.4 mg	-		

Note 1 For average matrix and uniform source distribution.

The following points are noted regarding the results in Table 14:

- The detection limit is better for the higher detection efficiency systems, which is to be expected. The best reported detection limit of 0.6 mg Pu240eff was achieved with a 45% efficient system. This system also incorporates a 1.4 m thick cosmic-ray induced neutron

background shield. This performance is comparable with that of the DDA active neutron interrogation systems in Table 23.

- The TMU performance is similar for all systems regardless of detection efficiency.
- For matrices with a low moderator content, the TMU can be as low as  $\pm 5\%$ . For high moderating matrices



(e.g. concrete) the TMU is approximately an order of magnitude higher at  $\pm 50\%$ .

- As expected, the detection limits and TMU are higher for measurements of large crates. The performance of the system designed to measure both large crates and 200l drums also has poorer performance for 200l drum measurements compared to the systems designed only to measure 200l drums. Clearly, the measurement performance has been compromised in order to accommodate a wider range of container sizes.

Additionally, the performance of PNCC systems measuring the plutonium mass within simulated waste drums was evaluated in a recent round-robin exercise performed in France (Reference [21]). Plutonium measurements were performed on three 118 litre simulated waste drums containing soft waste, although one drum included some added metal plates. The total mass of plutonium present in each drum was in the range 100 to 200 mg.

Note that the data from the intercomparison exercise are different to that presented in the previous table (and the general structure of the other performance values tables), because only the measurement technique employed is recorded (in this case PNCC) and no specific details regarding the type and configuration of the instrument systems being used was provided (although the number, in this case five, and location of participants is known). For this reason it is considered appropriate to report these data as a separate table to illustrate the performance that is achievable in the field using PNCC to measure the plutonium mass.

The results from the French intercomparison exercise are summarised below in Table 15. Note that the average % bias and % standard deviation obtained from the results reported by the different participating laboratories are presented. Although the intercomparison included results for all plutonium isotopes, only the Pu239 results are presented below to allow direct comparison with Table 14.

**Table 15:** Performance Values for Plutonium Mass Measurement using PNCC from French Intercomparison Exercise – EQRAIN 4 (Reference [21])

Simulated Waste Drum	Plutonium Mass	
	Average Bias	Standard Deviation ( $1\sigma$ )
118 litre, Soft waste (90% PVC)	3%	19%
118 litre, Soft waste	-13%	6%
118 litre, Soft waste + Metals	-9%	12%

The following points are noted regarding the results in Table 15:

- The percentage values in Table 15 are comparable with the reported TMU values in Table 14 and are considered to represent the typical accuracy that is achievable using this measurement technique.

#### 2.4.6 Passive Neutron Multiplicity Counting (PNMC) Instrumentation

The neutron multiplicity technique is able to differentiate between coincident neutron events involving different numbers of neutrons. Complex electronic circuitry is used to determine the number of times singles, double coincidences and triple coincidences are observed. It is not practical to measure higher order multiplicities with sufficient precision, but higher order bursts of neutrons from cosmic-ray events can be filtered out. PNMC requires high and uniform neutron detection efficiency to precisely measure triples (i.e. efficiency  $> 30\%$  typically).

For impure and heterogeneous materials, such as residues or waste, where corrections for multiplication, matrix or other effects become significant, having three measured count rates permits reliable and accurate plutonium assay.

For waste assay, PNMC is able to determine an effective detection efficiency that best matches a distribution of sources, but errors can arise and careful interpretation is required. For example, it is possible that the observed signal from a plutonium source at one location can dominate (and mask) that from an equal mass at another location, leading to an erroneous result for the plutonium mass.

PNMC is more commonly applied to Safeguards measurements (i.e. product cans) due to the improved measurement geometry (i.e. small chamber size, large sample and non-interfering matrix).

Benefits of PNMC	Limitations of PNMC
<ul style="list-style-type: none"> <li>Chemical composition, leakage multiplication or system calibration may be considered variables (depending upon analysis approach taken).</li> <li>Impure samples such as mixed oxide wastes can be assayed.</li> <li>Cosmic-ray spallation effects can be rejected as the high order multiplicity bursts.</li> <li>Can determine three of the four unknown quantities (<math>a</math>, <math>ML</math>, <math>\epsilon</math>, <math>m_{eff}</math>) using the three measured quantities (Totals, Doubles and Triples). However, this requires careful interpretation. Under certain conditions, provide more accurate results than PNCC.</li> </ul>	<ul style="list-style-type: none"> <li>Sensitive to high neutron backgrounds due to use of totals in the multiplicity algorithms.</li> <li>Complex electronics and data processing techniques.</li> <li>Lower precision than total or coincident neutron counting – requires very high and uniform efficiency (<math>&gt;30\%</math>) to achieve the required performance with good background shielding and long count times.</li> <li>Requires knowledge of isotopic composition.</li> <li>High sensitivity to changes in efficiency (varies with <math>\epsilon^2</math> for doubles and <math>\epsilon^3</math> for triples), but this can be maintained as a variable in system algorithms.</li> <li>Still requires knowledge of at least one parameter (either <math>a</math>, <math>\epsilon</math>, or <math>ML</math>).</li> <li>Dead-time corrections have much higher importance and can be accuracy limiting factor at high count rates.</li> </ul>

- Measurement of plutonium content of waste drums.
- High detection efficiency ( $\epsilon = 31\%$ ) and low background design (low-Z materials in chamber).
- Uses PNMC combined with HRGS to determine plutonium isotopic composition.
- Patented time-stamping neutron counting electronics.
- Ring Ratio matrix correction.
- Measurement time: 3600s
- Limit of detection: 0.8mg Pu240eff
- Total Measurement Uncertainty:  $\pm 10\%$  ( $1\sigma$ )



**Figure 5:** Example Instrument System Employing PNMC – Drum Monitor

#### 2.4.7 Passive Neutron Multiplicity Counting (PNMC) Performance Values

Performance data for the measurement of the plutonium mass in waste using PNMC is reported in Table 16. Perfor-

mance data has only been obtained for a single measurement system used to measure 200 litre waste drums.

**Table 16:** Performance Values for Measurement of Pu240 Effective Mass using PNMC

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD ( $1\sigma$ )	TMU ( $1\sigma$ )		
<b>Detection Efficiency:</b> 31% <b>Matrix Correction:</b> Ring Ratio <b>Notes:</b> Integrated HRGS, Low background design	200l Drum	Non-Interfering Matrix	-	$\pm 7\%$	3600 s	23
		Combustibles, PVC, Mixed, Metals, Filters	$< 0.8 \text{ mg}$ <sup>Note 1</sup>	$\pm 10\%$		

<sup>Note 1</sup> Worst case matrix and source position.



The performance of the PNMC system is comparable with that of the PNCC systems reported in Table 14 in terms of TMU. This particular system has a very low detection limit, which is due to the high detection limit of 31%, low background design (minimisation of high atomic number materials in the measurement chamber construction) and a relatively long count time of 1 hour.

## 2.5 Calorimetry for Plutonium Mass Determination

### 2.5.1 Fundamentals

Calorimetry determines the amount of plutonium in an item by measurement of the heat produced by the radioactive decay of the plutonium and associated isotopes. Plutonium isotopes decay emitting  $\alpha$ ,  $\beta$  and  $\gamma$  particles, and the  $\alpha$  and  $\beta$  particles interact with the surrounding material and lose their energy in the form of heat. The amount of heat generated (specific power, expressed in mW/g) by a particular isotope is proportional to the type and energy of the particle emitted and the decay rate of the isotope. The specific thermal power values for the plutonium isotopes and Am241 can be shown in Table 17.

**Table 17:** Specific Thermal Power Values (Reference [5])

Isotope	Main Decay Mode	Specific Power (mW/g)
$^{238}\text{Pu}$	$\alpha$	567.57
$^{239}\text{Pu}$	$\alpha$	1.9288
$^{240}\text{Pu}$	$\alpha$	7.0824
$^{241}\text{Pu}$	$\beta$	3.412
$^{242}\text{Pu}$	$\alpha$	0.1159
$^{241}\text{Am}$	$\alpha$	114.2

In order to determine the total plutonium mass, the heat output measurement from the calorimeter must be combined with a measurement of the plutonium isotopic composition, which is usually obtained from HRGS as discussed in Section 2.1.

Calorimetry is potentially a very accurate measurement technique as no corrections are required for neutron multiplication effects, plutonium distribution, matrix effects or chemical form. Indeed, the technology is widely used for Safeguards measurements of process materials.

However, for plutonium assay in waste, the relatively large container size, small mass of plutonium and large mass of matrix material mean that calorimetry is likely to be impractical due to the long measurement times that would be required. The large volume of the calorimeter, small plutonium mass and large amount of matrix material means that it takes a long time for the heat flow to reach equilibrium, such that the heat flow from the waste item (i.e. the property measured by the calorimeter) is equal to the rate of heat production from the radioactive decay of the plutonium and associated isotopes. It is likely that the measurement times for typical plutonium waste drums would be significantly in excess of 8 hours.

Reference [20], which was published in 2000, concludes that calorimetry could be applied to waste measurements and that large volume calorimeters suitable for standard 200 litre waste drums will be built. However, no known calorimetry systems are known to be currently in use for waste assay and no performance data has been received to date.

### 2.5.2 Calorimetry Performance Values

No performance data have been obtained for calorimetry systems for waste measurements.

Benefits of Calorimetry	Limitations of Calorimetry
<ul style="list-style-type: none"> <li>No correction required for neutron multiplication effects, plutonium distribution, matrix effects or chemical form.</li> <li>Very accurate assay of heat producing material.</li> <li>Do not need expensive fissile calibration sources, can use electrical standards.</li> <li>Simple technology.</li> </ul>	<ul style="list-style-type: none"> <li>Requires separate isotopic analysis by HRGS which will limit the overall accuracy.</li> <li>Only useful for plutonium and Am241 assay.</li> <li>Long assay time required to reach thermal equilibrium (many hours for waste assay).</li> <li>Can require temperature controlled environment.</li> <li>Tend to be bulky and not very mobile.</li> </ul>

### 3. Techniques for Uranium-Bearing Solid Wastes

This section discusses characteristic features and performances of NDA techniques utilised for the measurement of wastes contaminated with uranium-bearing solid materials.

The features and performances of the following techniques are examined:

- High Resolution Gamma Spectrometry (HRGS) for the Determination of the U235 Content.
- Low Resolution Gamma Spectrometry (LRGS) for the Determination of the U235 Content.
- Active Neutron Techniques for the Determination of the U235 Content (i.e. Differential Die-Away and Cf252 Shuffler).

#### 3.1 High Resolution Gamma Spectrometry (HRGS) for the Determination of the U235 Content

##### 3.1.1 Fundamentals

The measurement of the U235 content in waste items using HRGS is the same technique discussed in Section 2.2.1 for quantitative plutonium assay. However, in the case of U235 assay, there are fewer characteristic gamma-rays (see Table 18) and they are at all at a relatively low energy.

**Table 18:** Principal Characteristic Gamma-rays from U235 (Reference [17])

Isotope	Energy (keV)	Intensity (gammas/s-g)
U235	143.8	$7.8 \times 10^3$
	163.4	$3.7 \times 10^3$
	185.7	$4.3 \times 10^4$
	202.1	$8.0 \times 10^2$
	205.3	$4.0 \times 10^3$

The most abundant gamma-ray at 185.7 keV is normally used for quantitative U235 assay in waste. The low energy of this gamma-ray compared to the 413.7 keV gamma-ray used to assay Pu239 means that, in general, the assay of U235 will be less accurate than the assay of Pu239. The measurement uncertainty will generally be higher due to the increased attenuation in the waste matrix and any “lumps” of uranium present (i.e. sample self-attenuation).

Correction for self-attenuation in “lumps” of uranium is technically challenging due to the narrow energy range of the characteristic U235 gamma-rays. It is therefore common not to apply a correction for sample self-attenuation in U235 waste assay, and to either assume zero self-attenuation or include an appropriate factor in the calibration to account for the self-attenuation. This assumption may be supported by historical waste records or waste sampling.

Techniques to correct for self-attenuation in uranium lumps have been developed and successfully applied to U235 waste assay. One example uses a Differential Peak Absorption (DPA) analysis of the characteristic U235 gamma peaks – Reference [7]. An alternative method uses gamma-rays from both U235 and U238 and requires knowledge of the uranium enrichment – Reference [8].

The good resolution of HRGS detectors permits the measurement of U235 in the presence of other radionuclides such as plutonium. However, the presence of Ra226 in waste can be problematic since this radionuclide emits a characteristic gamma-ray at 186.0 keV which directly interferes with the U235 185.7 keV gamma-ray.

Measurement of the 185.7 keV gamma-ray allows direct quantification of the U235 mass and it is not normally necessary to determine the total uranium mass by combining the U235 mass with information on the uranium enrichment. However, it is noted that the uranium enrichment can easily be measured using the same HRGS system by analysing the spectra with an isotopic analysis code in a similar way to the plutonium isotopic analysis discussed in Section 2.1. The isotopic analysis codes discussed in Section 2.1 (i.e. MGA, PC/FRAM and IGA) can be applied to uranium isotopic analysis as well as plutonium.

##### 3.1.2 High Resolution Gamma Spectroscopy (HRGS) Instruments

The information presented in Section 2.2.2 also applies to quantitative U235 assay.

##### 3.1.3 Performance Values

Performance data for the measurement of the U235 mass in waste using HRGS is reported in Table 19. The performance is reported for several different types of system, including both fixed detectors and SGS systems. In addition to the system configuration, information has been included on the matrix correction employed and any other special algorithms or features that are used to improve the system performance.

Note that the information from instrument systems with similar configurations and performance has been combined where possible, by combining the data and reporting a range of performance values to span the reported values for individual systems. The information has again been organised with the least complex instrument systems (i.e. fixed detector(s)) appearing first in the table.

Note that in several cases the reported values are for more than one container size (e.g. 100l & 200l drums or 200l & 400l drums). This indicates that the system(s) is capable of measuring both sizes, and unless stated otherwise, the reported performance values are applicable to both container sizes.

**Table 19:** Performance Values for U235 Mass Measurement using HRGS

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1σ)	TMU (1σ)		
Fixed Detector Systems						
<b>Geometry:</b> Fixed detector <b>Detectors:</b> 1 x HPGe (e = 19%) <b>Matrix Correction:</b> Density <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b> ISOCS absolute efficiency modelling	100l & 200l Drums	Low Density	0.15 g	U235 mass (<3g): ± 28%	1000 – 2000 s	31
		High Density	0.43 g	U235 mass (3-100g): ± 19%		
<b>Geometry:</b> Fixed detector <b>Detectors:</b> 1 x HPGe (e = 80%) <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> NO <b>Validity domain:</b> U surface density < 3 g/cm <sup>2</sup> U238 daughter g line at 1001 keV > 300 cps <b>Notes:</b> SGS for Transmission using HPGe (e = 20%)	100l & 200l Drums	PVC / Metals (<0.4 g.cm <sup>-3</sup> )	-	± 20% on U mass	900 s to several hours <sup>Note 1</sup>	3
<b>Geometry:</b> Fixed detector <b>Detectors:</b> 4 x HPGe <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b>	200l Drums	Empty (0.1 g.cm <sup>-3</sup> )	-	± 36%	120 s	25
		Combustibles / PVC (0.4 g.cm <sup>-3</sup> )	-	± 45%		
		Graphite (0.4 g.cm <sup>-3</sup> )	-	± 37%		
		Concrete (1 g.cm <sup>-3</sup> )	-	± 83%		
<b>Geometry:</b> Fixed detector <b>Detectors:</b> 2 x HPGe <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b>	Large Crate	Mixed (Gloveboxes)	3-5 g	± 30% <sup>Note 2</sup>	1800 s	22
Segmented Gamma Scanners (Single Detector)						
<b>Geometry:</b> SGS <b>Detectors:</b> 1 x BEGe <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b> Integrated with PNCC system	200l & 400l Drums	Combustibles, PVC, Mixed, Metals, Rubble, Lead	2 g	± 10% <sup>Note 3</sup>	960 s	21

<sup>Note 1</sup> The count time depends on the U mass to be measured (from some grams to some kilograms), the enrichment of U235 (depleted, natural or low enriched uranium), the distribution of U, and the nature of the matrix. For instance, 4 kg of U (UO<sub>2</sub> 4% of U235, powder form) in a 0.4 g/cm<sup>3</sup> metallic matrix can be measured in 15 minutes whereas 15g of U (UO<sub>2</sub> 1.5% of U235, powder form) in a 0.2 g/cm<sup>3</sup> combustible matrix can be measured in 90 minutes.

<sup>Note 2</sup> Calibration assumes factor of 8 correction for self-absorption in uranium lumps, reported TMU does not include this potential overestimation.

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1σ)	TMU (1σ)		
<b>Geometry:</b> SGS <b>Detectors:</b> 1 x HPGe (e = 10%) <b>Matrix Correction:</b> Density <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> YES (15cm steel) <b>Notes:</b>	200l & 400l Drums	Mixed (0.03 to 0.6 g.cm <sup>-3</sup> )	30 mg (0.03 g.cm <sup>-3</sup> ) To 210 mg (0.6 g.cm <sup>-3</sup> )	No lumps: ± 4-32% (12%) (homo) <sup>Note 4</sup> > 63% (hetero) With lumps: ± 4-45% (homo) > 70% (hetero)	6000 s	4
<b>Geometry:</b> SGS <b>Detectors:</b> 1 x HPGe <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> NO <b>Shielded Cavity:</b> NO <b>Notes:</b>	200l Drum	Combustibles / PVC (<0.2 g.cm <sup>-3</sup> )	<10 mg	± 35%	10800 s	30
		Mixed (low Z) (<0.5 g.cm <sup>-3</sup> )	-	± 200%		
Segmented Gamma Scanners (Multiple Detectors)						
<b>Geometry:</b> SGS <b>Detectors:</b> 3 x HPGe (e = 20%) <b>Matrix Correction:</b> Transmission <b>Lump Correction:</b> YES <b>Shielded Cavity:</b> YES (15cm steel) <b>Notes:</b>	200l Drum	Combustibles / PVC (0.1 g.cm <sup>-3</sup> )	2.7 mg (in 900s)	+25% -1% (1g HEU) <sup>Note 3</sup> +70% -1% (20g HEU) <sup>Note 3</sup>	1200 s	28
		Mixed / Metals (0.6 g.cm <sup>-3</sup> )	10.5 mg (in 900s)	+55% -2% (1g HEU) <sup>Note 3</sup> +160% -1% (20g HEU) <sup>Note 3</sup>	1200 s	
		Mixed / Filters	-	± 13% (homo)	1800 s	5

<sup>Note 3</sup> Value assumes negligible counting statistics.

<sup>Note 4</sup> With no lump correction and with self-attenuation due to possible lumps included in TMU.

The following points are noted regarding the performance values in Table 19:

- For 200l drum systems, similar TMU performance values are reported for all systems regardless of their complexity. It might be expected that the more complex (and therefore more expensive) systems would yield better performance. The reason for this is considered to be related to the usage of the systems; that is, the least complex systems are generally used for better defined waste streams (i.e. low density, segregated and predominantly homogeneous), whereas the higher complexity systems are used for more matrices with a greater range of densities and degree of heterogeneity.
- The detection limit performance is better for the systems employing multiple detectors as expected.

Additionally, the performance of HRGS systems measuring the U235 mass within simulated waste drums was evaluated in a recent round-robin exercise performed in France (Reference [21]). Uranium measurements were performed on a single 223 litre simulated waste drum containing soft waste. The total mass of U235 present in the drum was 0.56g.

Note that the data from the intercomparison exercise are different to that presented in the previous table (and the general structure of the other performance values tables), because only the measurement technique employed is recorded (in this case HRGS) and no specific details regarding the type and configuration of the instrument systems being used was provided (although the number, in this case seven, and location of participants is known). For this reason it is considered appropriate to report these data as a separate table to illustrate the performance that is achievable in the field using HRGS to measure the U235 mass.

The results from the French intercomparison exercise are summarised below in Table 20. Note that the average % bias and % standard deviation obtained from the results reported by the different participating laboratories are presented. Although the intercomparison also included results for U234 and U238, only the U235 results are presented below to allow direct comparison with Table 19.

**Table 20:** Performance Values for U235 Mass Measurement using HRGS from French Intercomparison Exercise – EQRAIN 4 (Reference [21])

Simulated Waste Drum	U235 Mass	
	Average Bias	Standard Deviation ( $1\sigma$ )
223 litre, Soft waste	-8%	22%

The following point is noted regarding the results in Table 20:

- The percentage bias and standard deviation is comparable with the reported TMU values in Table 19 and are considered to represent the typical accuracy that is achievable using this measurement technique.

### 3.2 Low Resolution Gamma Spectrometry (LRGS) for U235 Mass Determination

#### 3.2.1 Fundamentals

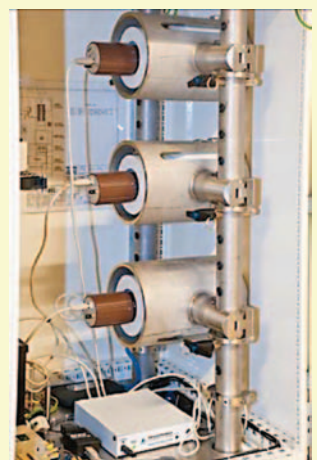
The measurement of the U235 content in waste items using LRGS is the same technique discussed in Section 2.3.1 for quantitative plutonium assay. However, in the case of U235 assay, the fewer characteristic gamma-rays, which are all at a relatively low energy (see Table 18) compared to the principal characteristic U238 gamma-rays at 742.8 keV, 766.4 keV, 786.3 keV and 1001.0 keV (from Pa234m, a daughter product) also likely to be present, means the U235 measurement is better suited to LRGS compared to plutonium.

In the case of U235 assay, it is common to set up an analysis region of interest in the gamma spectrum that encompasses the U235 gamma emissions between 140 and 210 keV.

#### 3.2.2 Instrumentation

The information presented in Section 2.3.2 for plutonium assay using LRGS also applies to U235 assay.

- Measures U235 content of waste packages (bags) or 200 litre drums in support of drum filling operations.
- Calibrated for waste packages containing soft waste, ash and metals.
- Uses 3 fixed 3" by 3" NaI(Tl) LRGS detectors
- Measurement time: 300s
- Limit of detection: 0.1g U235
- Total Measurement Uncertainty:
  - Soft waste packages  $\pm 10\%$  ( $1\sigma$ )
  - Heavy items (<80kg)  $\pm 26\%$  ( $1\sigma$ )



**Figure 6:** Example Instrument System Employing LRGS – LRGS DrumScan



### 3.2.3 Performance Values

Performance data for the measurement of the U235 mass in waste using LRGS is reported in Table 21. The performance is reported for a two similar LRGS systems used to measure bagged waste packages and 200 litre drums. In

addition to the system configuration, information on the implementation of the LRGS technique is also included.

Note that the Limits of Detection (LODs) in Table 21 are in terms of U235 mass.

**Table 21:** Performance Values for Measurement of U235 Mass using LRGS

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ ), g U235	TMU (1 $\sigma$ )		
<b>Geometry:</b> 3x fixed NaI detectors <b>Matrix Correction:</b> Weight (based on known matrix & fill height) <b>Lump Correction:</b> None <b>Notes:</b>	Waste Bag or 200 litre drum	Soft waste, Ash, Metals, Cellulosic	0.1 g	Packages: Soft waste = $\pm 9.6\%$ Heavy items (<80 kg) = $\pm 25.9\%$ Upper weight limit (200 kg) = $\pm 37.7\%$	300 s	35
<b>Geometry:</b> 3x fixed NaI detectors <b>Matrix Correction:</b> Weight (based on known matrix & fill height) <b>Lump Correction:</b> None <b>Notes:</b>	200l Poly Lined Drum	Mixed	-	Hard Waste / Rubble = $\pm 30\%$	300 s	36
	Waste Bag	PVC	-	$\pm 6\%$		
	Waste Bag	Metals	-	$\pm 25\%$		
	Waste Bag	Filters	-	$\pm 7\%$		

## 3.3 Active Neutron Techniques for the Determination of the U235 Content

### 3.3.1 Fundamentals

Fissions can be induced in fissile nuclides such as Pu239, Pu241 and U235 using an external neutron or gamma ray interrogation source. The neutrons produced in the induced fission reactions can then be counted to determine the fissile mass present.

Active neutron techniques have the advantage of being a direct measurement of the fissile content of the waste item, which is the parameter of interest for nuclear safety measurements. However, these techniques are unable to distinguish between the individual fissile nuclides and the measured result is usually expressed in terms of an effective mass of Pu239 or U235 (i.e. the same mass of Pu239 or U235 that would give a signal equivalent to that which was measured). It is noted that detection limits are higher for U235 compared to Pu239 in Differential Die-Away (DDA) systems (i.e.  $\approx 1.6\times$  higher) on account of the lower U235 thermal neutron fission cross-section. Similarly for Active Neutron Coincidence Counting (ANCC) systems, the detection limits are higher for U235. For

Californium-252 Shuffler systems, the detection limits are higher for Pu239, due to the lower delayed neutron yield compared to U235.

A general limitation of active neutron techniques is the potential underestimation of the true fissile mass in "lumps" of fissile material due to self-shielding, which arises as a result of the low penetrability of interrogation thermal neutrons. This is similar to self-attenuation when gamma systems are used for direct quantification of the plutonium or uranium mass.

Three different active neutron techniques are considered applicable to waste measurements:

- Active Neutron Coincidence Counting (ANCC).
- Differential Die-Away (DDA).
- Californium-252 Shuffler (Cf252 Shuffler).

### 3.3.2 Active Neutron Coincidence Counting (ANCC) Instruments

Active Neutron Coincidence Counting (ANCC) systems use a random neutron source for the neutron interrogation, for example an (a, n) source such as Am-Li or a ( $\gamma$ , n) source such as Sb-Be. Since these neutron sources



produce random (i.e. not time-correlated) neutrons, the induced fission response can be distinguished by neutron coincidence counting. In practice, similar neutron counting electronics to that in PNCC systems can be used. The design of the ANCC measurement chamber is also very similar to a PNCC system, with modification to allow the neutron source to be inserted. Note that Active Neutron Multiplicity Counting (ANMC) is also possible, in which multiplicity counting is used instead of coincidence counting.

ANCC (or ANMC) systems are normally applied to uranium measurements and there are no known plutonium waste assay systems in operation that use this technique. The reason is that for plutonium measurements, the performance of an ANCC system will be no better than a PNCC system. ANCC systems are commonly used for measurements of uranium product wastes or for the characterisation of unirradiated fuel assemblies (i.e. fuel collars). ANCC measurements do not prove very useful when there is a high passive coincidence signal, such as in high plutonium mass samples or irradiated fuel when curium is likely to be present. Detection limits are typically at the gram level. Note that for ANCC systems, the lower limit of detection does not improve with increasing source strength, as the

increased accidental coincidence rate tends to balance the increased “true” signal strength.

ANCC systems can also be used with a cadmium liner inside the measurement cavity. In this mode of operation, thermal neutrons cannot interrogate the sample. This restricts the utilised interrogating flux to the higher energy neutrons and hence the penetrability of the utilised flux in self-shielded “lumps” of fissile material is improved. A second benefit is that leakage multiplication effects are reduced, since induced fission neutrons from the sample cannot be reflected back as thermal neutrons from the walls of the cavity. The disadvantage of this mode of operation is that the fission cross-sections are much lower for non-thermal neutrons, so the detection limit is much higher and the count times are longer to achieve the required precision. This mode of operation is often referred to as “fast” mode, with the standard (no cadmium) mode being referred to as “thermal” mode. Note that there is no advantage in using the “fast” mode when the sample contains neutron moderating materials, since the interrogation flux will thermalise, biasing the response of the system. In cases where there is significant moderation in the sample, then it is better to use the “thermal” mode of operation, since the response of the system will suffer a smaller bias.

Benefits of ANCC	Limitations of ANCC
<ul style="list-style-type: none"><li>• Direct measure of fissile isotopes (as long as there is no interference from spontaneously fissionable nuclides).</li><li>• Useful for U235 measurements. The neutron energy spectrum of the (Am-Li) interrogating source is predominantly below the threshold for fast neutron fission in U238 (i.e. mean energy of source neutrons ~400keV), so there is only a small contribution from fast fission of U238 in the measured signal.</li><li>• Can be operated in “fast” mode (cadmium liner) to improve the penetrability of the interrogation flux in self-shielding lumps.</li><li>• Long half-life sources (Am241 based) may be used.</li><li>• Can utilise conventional multiplicity electronics to gain further sample characterisation data.</li></ul>	<ul style="list-style-type: none"><li>• Lumps of fissile material can cause underestimation of the true fissile mass, due to self-shielding as a result of the low penetrability of interrogation thermal neutrons.</li><li>• Accidental coincidence rates may be high from installed neutron sources – increases the statistical uncertainty.</li><li>• Raised dose to operators from constantly exposed sources, if design does not take this risk into account.</li><li>• Difficulty in obtaining purely random neutron sources.</li><li>• Count times and detection limits are higher in “fast” mode.</li></ul>

3.3.3 Performance Values

Performance data for the measurement of the U235 mass in waste using ANCC is reported in Table 22. The performance is reported for a single system used to measure bagged waste packages. In addition to the system config-

uration, information has been included on the matrix correction employed and any other special algorithms or features that are used to improve the system performance.

Note that the Limits of Detection (LODs) in Table 22 are in terms of U235 mass.

**Table 22:** Performance Values for Measurement of U235 Mass using ANCC

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ ), g U235	TMU (1 $\sigma$ )		
<b>Detection Efficiency:</b> ~39% <b>Matrix Correction:</b> None <sup>Note 1</sup> <b>Lump Correction:</b> Fast mode of operation <sup>Note 2</sup> <b>Notes:</b> Uses 2 x AmLi sources ~10 <sup>5</sup> ns <sup>-1</sup> .	Waste Bag	Combustibles, PVC, Metals, Filters, Residues	Thermal mode: 0.2 – 0.7g Fast mode: 12 g	Thermal mode: $\pm 9.5\%$ Fast mode: $\pm 15\%$	1800s	34

<sup>Note 1</sup> Uses a specific calibration based on known matrix type.

<sup>Note 2</sup> Fast mode is used for large U235 mass packages where self-shielding could be significant, by adding a cadmium liner to the well.

The following point is noted regarding the performance information in Table 22:

- As expected the performance is significantly worse in fast mode compared to thermal mode due to the lower count rates.

### 3.3.4 Differential Die-Away (DDA) Instruments

The DDA technique normally uses a high intensity deuterium-tritium (D-T) neutron generator as the interrogation neutron source, which produces short (~10  $\mu$ s), intense (~10<sup>6</sup> neutrons per pulse) bursts of 14 MeV fast neutrons. These fast neutrons are quickly thermalised in an external moderator assembly or the walls of the measurement chamber. A thermal neutron flux then interrogates the waste item in the measurement chamber for several milliseconds, inducing fissions in any fissile material in the waste item. The prompt, fast fission neutrons are detected in fast neutron detector packages surrounding the waste item, which are usually He3 proportional counters embedded in polyethylene and wrapped in cadmium to exclude the thermal

neutron flux. The signal that is measured in a time interval after the neutron pulse after the fast neutrons from the generator has died away but the thermal flux still persists in the measurement chamber is proportional to the quantity of fissile material within the waste item.

The measured quantity is effective mass of U235 (or Pu239): i.e.

$$U235_{\text{eff}} = U235 + (1.61 \times Pu239) + (2.21 \times Pu241)$$

A reference listing the U235 effective coefficients for the fissile nuclides is Reference [19].

DDA is a potentially a very sensitive measurement technique and can measure down to milligram quantities in the waste item by virtue of large thermal neutron fission cross-sections. The lower limit of detection is inversely proportional to the interrogating neutron flux. DDA is also well-suited to high background assay applications (e.g. irradiated fuel with high curium content) that demand a high signal to background ratio.

Benefits of DDA	Limitations of DDA
<ul style="list-style-type: none"> <li>Direct measure of total fissile mass, the quantity of interest for criticality safety.</li> <li>Highest sensitivity measurement (milligram quantities of Pu239 or U235) due to large signal-to-background ratio.</li> <li>High neutron interrogating flux permits measurements in high passive backgrounds (e.g. spent fuel wastes with plutonium/curium content).</li> <li>Large interrogation flux permits good penetration of matrix and measurement of matrix properties.</li> <li>Large signal allows sophisticated data processing, e.g. imaging, to reduce uncertainties.</li> <li>Biological dose rate from neutron source can be controlled by pulsing, reducing the need for large "source store".</li> <li>Can tailor interrogating flux to optimise performance for particular sample types or waste matrices.</li> </ul>	<ul style="list-style-type: none"> <li>Lumps of fissile material can cause underestimation of the fissile mass, due to self-shielding as a result of the low penetrability of interrogation thermal neutrons.</li> <li>Potentially large matrix effects requiring a complex correction technique when applied to waste assay.</li> <li>Extensive calibration required (approximately twice as much compared to a PNCC system).</li> <li>Cannot discriminate between plutonium and uranium in mixed streams.</li> <li>High cost and maintenance burden of neutron generator.</li> <li>Complex control systems and algorithms.</li> </ul>

- Measures the fissile (Pu239eff) and fertile (Pu240eff) mass of drummed waste.
- Neutron generator output  $10^8$  n/s.
- Combined with PNCC system ( $\epsilon = 26\%$ ).
- Add-a-Source (AAS) and flux monitor matrix corrections.
- Neutron imaging algorithm to reduce uncertainty.
- Measurement time = 15 minutes.
- Detection limits ( $1\sigma$ ): 0.8 to 15 mg Pu239eff.
- Total Measurement Uncertainty ( $1\sigma$ ):
  - Light Matrix  $\pm 6\%$ .
  - Cement Matrix  $\pm 40\%$ .



**Figure 7:** Example Instrument System Employing DDA – PANWAS

### 3.3.5 Performance Values

Performance data for the measurement of the total fissile mass in waste using DDA is reported in Table 23. The performance is reported for two systems used to measure drummed waste. In addition to the system configuration, information has been included on the matrix correction employed and any other special algorithms or features that are used to improve the system performance.

Note that in several cases the reported values are for more than one container size (e.g. 200l & 400l drums). This indicates that the system(s) is capable of measuring both sizes, and unless stated otherwise, the reported performance values are applicable to both container sizes.

Note that the Limits of Detection (LODs) in Table 23 are in terms of U235 effective mass.

**Table 23:** Performance Values for Measurement of Total Fissile Mass using DDA

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD ( $1\sigma$ ), mg U235eff	TMU ( $1\sigma$ )		
<b>Detection Efficiency:</b> ~20% <b>Matrix Correction:</b> ABS & ABSMOD <sup>Note 1</sup> <b>Lump Correction:</b> NO <b>Notes:</b> Imaging algorithms, Neutron generator output $\sim 10^8$ ns <sup>-1</sup>	200l Drums	Debris	2.3 mg - 19.2 mg U235eff	-	600s	12
<b>Detection Efficiency:</b> 26% <b>Matrix Correction:</b> ABS index combined with AAS <b>Lump Correction:</b> NO <b>Notes:</b> Integrated with DDA system, Imaging algorithms, Neutron generator output $\sim 10^8$ ns <sup>-1</sup>	200l & 400l Drums	Combustibles / PVC	1.2 mg - 22.8 mg U235eff	$\pm 6\%$ <sup>Note 2</sup>	900s	29
		Concrete		$\pm 40\%$ <sup>Note 2</sup>		
	400l Grouted Drum	Concrete / Neoprene / Wood	45 mg U235eff	-		

<sup>Note 1</sup> Active and passive matrix corrections are derived from the active neutron measurement using flux probes and fast neutron detectors.

<sup>Note 2</sup> Assuming no self-shielding.

The following points are noted regarding the performance information in Table 23:

- The two systems have similar performance with respect to detection limit.
- The detection limits that can be achieved with a DDA system are better than those achievable with HRGS (see Table 9 and Table 19) and PNCC (see Table 14) for equivalent waste matrices and count times. [Note that since a PNCC system is measuring the Pu240 effective mass, the detection limits results in Table 14 should be multiplied by approximately 4 to represent typical reactor grade plutonium to account for the lower Pu240 abundance before comparing with the DDA and HRGS results].
- The TMU is significantly higher for the concrete waste matrix due to the high hydrogen content.

### 3.3.6 Californium-252 Shuffler Instruments

In a Californium Shuffler, a Cf252 neutron source on a source transfer mechanism is used as the interrogation source. The source is transferred rapidly from a shielded

enclosure to irradiate the waste item for a period of typically a few seconds. The neutrons from the source are thermalised in the measurement chamber and produce fissions within any fissile material present. After this period the interrogation source is rapidly returned to its shielded location. Note that because of the time taken to retract the source from the irradiation position to the shielded position (typically 0.5 s to 1 s), it is not possible to measure the prompt neutron signal (as measured by a DDA system), and instead the less abundant (i.e.  $\approx 100\times$  less yield) delayed neutron is measured. The measured delayed neutron signal is proportional to the mass of fissile material present.

Shufflers are generally used at higher fissile masses where the high sensitivity of DDA is not needed.

Shuffler chambers are often lined with cadmium for all or part of a measurement to stop thermalised neutrons from entering the item. This reduces potential self-shielding problems by increasing mean interrogation neutron energy.

However, this increase in accuracy is obtained at the expense of a significant reduction in sensitivity.

Benefits of Cf252 Shuffler	Limitations of Cf252 Shuffler
<ul style="list-style-type: none"> <li>• Direct measure of total fissile mass, the quantity of interest for criticality safety.</li> <li>• Chamber characteristics are also suitable for PNCC and so a combined passive/active chamber is often employed.</li> <li>• Large interrogation flux permits good penetration of matrix and measurement of matrix properties</li> <li>• For fissile masses well above the LLD, the large signal allows sophisticated data processing, e.g. imaging, to reduce geometrical uncertainties.</li> <li>• Capability to "tailor" the interrogating spectrum for optimised performance in specific sample / matrix types.</li> </ul>	<ul style="list-style-type: none"> <li>• Affected by self-shielding in lumps of fissile material, slightly less effect than DDA. Using a cadmium liner with a shuffler greatly reduces the effect of self shielding.</li> <li>• Potentially large matrix effects requiring a complex correction technique when applied to waste assay.</li> <li>• Extensive calibration required (approximately twice as much compared to a PNCC system).</li> <li>• Cannot discriminate between plutonium and uranium in mixed streams.</li> <li>• Elevated passive background will worsen the detection limit performance (extremely difficult to perform shuffler measurements in the presence of curium).</li> <li>• Cost and maintenance burden of source transfer and shield systems.</li> <li>• High costs involved in maintaining and renewing the Cf252 source (half-life of 2.6 years), although may be much lower than an equivalent DDA system.</li> <li>• Dose uptake to operators when replacing Cf252 sources</li> <li>• Complex control systems and algorithms.</li> </ul>

### 3.3.7 Performance Values

Performance data for the measurement of the total fissile mass in waste using a Cf Shuffler is reported in Table 24. The performance is reported for two different systems,

one used to measure drummed waste and the other canned waste. In addition to the system configuration, information has been included on the matrix correction employed and any other special algorithms or features that are used to improve the system performance.

**Table 24:** Performance Values for Measurement of Total Fissile Mass using Cf Shuffler

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ )	TMU (1 $\sigma$ )		
<b>Detection Efficiency:</b> 10% <b>Matrix Correction:</b> Not Specified <b>Lump Correction:</b> YES <b>Notes:</b> Cf252 source strength = $2 \times 10^9 \text{ ns}^{-1}$	Cans	Combustibles, PVC, Metals, Debris, Lead, Concrete	~0.5g U235eff	$\pm 24\%$ ( $>10\text{l can}$ ) <sup>Note 1</sup>	2700s	24
<b>Detection Efficiency:</b> 45% <b>Matrix Correction:</b> Flux Probes <b>Lump Correction:</b> NO <b>Notes:</b> Multi-height irradiation for U235 vertical segmentation, Concrete background shielding, Cf252 source strength = $\sim 10^8 \text{ ns}^{-1}$	200l Drums	Combustibles, PVC, Metals, Lead, Filters, Mixed, Rubble ( $<0.1 - 1 \text{ g.cm}^{-3}$ )	$<0.3\text{g U235eff}$ <sup>Note 2</sup>	$\pm 27\%$	200s	26

<sup>Note 1</sup> Excludes lump correction uncertainty component which can be a few 100%.

<sup>Note 2</sup> For average matrix and uniform source distribution.

The following points are noted regarding the performance information in Table 24:

- The two systems have similar detection limits despite the different applications and container sizes being measured. The second system has a neutron interrogation source that is approximately 10 times weaker but has a much higher neutron detection efficiency to compensate.
- The TMU values of the two systems are similar despite the big difference in detection efficiency.

#### 4. Techniques for Spent Fuel Wastes

This section discusses characteristic features and performance of NDA techniques for the determination of the amount of SNM present in spent fuel waste arisings.

In spent fuel the characteristic radiation signatures from plutonium and uranium are completely masked by the radiation from fission product (e.g. Cs134, Cs137 and Eu154), activated structural components (e.g. Co58, Co60 and Mn54) and transuranic elements that have built up (e.g. Cm242 and Cm244). The intense gamma emissions from the fission and activation products prevent direct measurement of the plutonium and uranium gamma rays, meaning that quantification of the plutonium and uranium mass or determination of the plutonium and uranium isotopic composition is not possible. Similarly, the intense neutron emissions from spent fuel, which are mainly from Cm242 and Cm244, preclude measurement of the passive neutron emissions from plutonium and direct measurement of the plutonium content by passive neutron counting techniques is not possible. This means that most of the

NDA techniques discussed earlier in this report cannot be applied to spent fuel waste arisings to directly measure the required fissile mass.

It is possible, however, to use active neutron interrogation to determine the amount of fissile material in spent fuel wastes, provided that a large enough interrogation source is available to induce a measurable signal above the high background radiation levels.

The fissile mass of spent fuel wastes can also be estimated indirectly by measurement of a radiation signature, for example the activity of a fission product (e.g. Cs137) or the total neutron emission. This indirect measurement relies upon determining a relationship between the measured radiation and the fissile mass, which is usually estimated by means of a reactor fuel depletion code (which models the behaviour of nuclear fuel in the reactor) such as FISPIN and ORIGEN. Normally the correlation requires knowledge of the fuel irradiation (or burnup), cooling time and initial enrichment, which may be known in advance or can be estimated by measurement of characteristic radiation signatures. Clearly the accuracy of these indirect measurements will be dependent on the accuracy of the knowledge of the fuel characteristics. Only indirect measurements using the fission product activities (measured with HRGS) are considered further in this section. Although indirect measurements using the total neutron emission are also possible, no performance data has been received for spent fuel waste measurements of this type and therefore the technique has not been considered further.



The features and performances of the following techniques are examined in this section:

- High Resolution Gamma Spectrometry (HRGS) for Indirect Measurement of Uranium Mass from the Fission Product Activity.
- Active Neutron Interrogation using Differential Die-Away (DDA) for Fissile Mass Determination.

4.1 High Resolution Gamma Spectrometry (HRGS) for Indirect Measurement of Uranium Mass from the Fission Product Activity

4.1.1 Fundamentals

The complex gamma spectra that are obtained from spent fuel measurements require HRGS detectors to resolve the gamma peaks of interest. An example gamma spectrum from spent fuel is shown in Figure 8.

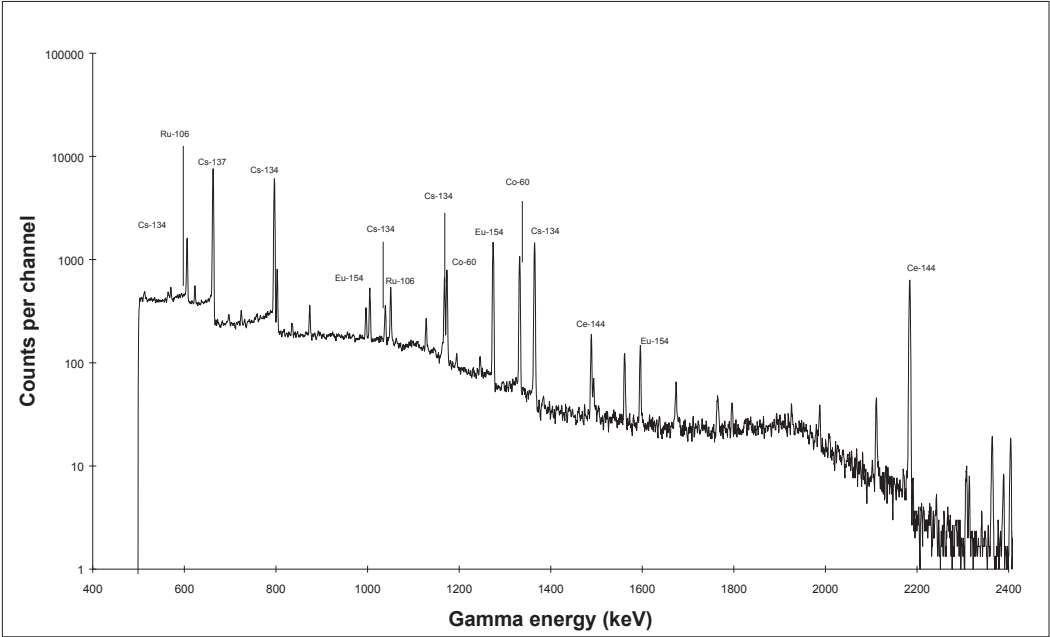


Figure 8: Example Gamma Spectrum from Spent Fuel

Table 25 lists some of the gamma-rays typically present in spent fuel gamma spectra. The nuclides which appear in

brackets in the first column of the table are short-lived daughter products that emit the characteristic gamma-rays listed.

Table 25: Principal Gamma-rays Present in Spent Fuel Gamma Spectra (Reference [5])

Nuclide	Origin	Half-Life	Gamma Energy, keV	Branching Ratio, %
Zr95	Fission product	64 d	724.2	43.1
			756.7	54.6
Nb95	Fission product	35 d	765.8	99.8
Ru106 (Rh106)	Fission product	366 d	622.2	9.8
			1050.5	1.6
Cs134	Fission product	2.1 yr	604.7	97.6
			795.8	85.4
			801.8	8.7
			1167.9	1.8
			1365.1	3.0
Cs137 (Ba137)	Fission product	30.2 yr	661.6	85.1



Nuclide	Origin	Half-Life	Gamma Energy, keV	Branching Ratio, %
Ce144 (Pr144)	Fission product	285 d	696.5	1.3
			1489.2	0.3
			2185.6	0.7
Eu154	Fission product	8.5 yr	996.3	10.3
			1004.8	17.4
			1274.4	35.5
Mn54	Activation product	312 d	834.8	100.0
Co58	Activation product	70 d	811.1	99.0
Co60	Activation product	5.3 yr	1173.2	100.0
			1332.5	100.0

Cs137 is the most commonly used indicator of fuel irradiation because its neutron absorption cross-sections are negligible, its yields from U235 and Pu239 are approximately equal, and its 30 year half-life is long compared to the time spent in the reactor, making a correction for reactor power history unnecessary (Reference [5]). As a consequence the Cs137 concentration is approximately proportional to the fuel irradiation. The characteristic gamma-ray emission from Cs137 at 662 keV<sup>2</sup> is also energetic enough to escape from bulk fuel (if present in the waste) and give good penetration through the waste materials. Measurement of the Cs137 activity can therefore be related to the fuel (and hence fissile) mass if the fuel irradiation, cooling time and initial enrichment are known.

However, if the fuel has been subject to dissolution, Cs137 is preferentially leached from the fuel due to its high solubility, and therefore the amount of Cs137 present in the spent fuel waste may not be proportional to the residual fuel mass. Furthermore, the increased mobility of Cs137 means that it can migrate into the fuel cladding material whilst in the reactor, and if this occurs the Cs137 content of the spent fuel waste arisings will again not be proportional to the residual fuel mass.

If the fuel is relatively short-cooled, then it may be possible to use the characteristic gamma ray from Ce144 at 2186keV<sup>3</sup>; however the short half life of 285 days may prevent this. Ce144 is less susceptible to leaching from the fuel and the higher energy of the gamma emission will give better penetration through the waste materials. Although having longer half lives, Cs134 and Eu154 will give a less reliable indirect measure of the fuel content because they are mainly produced by neutron capture reactions rather than heavy element fissions.

Activity ratios can also be used to estimate the fuel irradiation, for example Cs134 /Cs137 and Eu154/Cs137, since these ratios are approximately proportional to the burnup. Ratios are easier to determine than absolute activities, since only the relative detection efficiency at the different gamma energies is required. However, the different radioactive decay rates of the nuclides must be taken into account by applying a cooling time correction.

#### 4.1.2 HRGS Instruments

The main difference compared to the HRGS instruments described elsewhere in this report is the high gamma activity of the spent fuel waste and the high count rates that are generally encountered. This means that lower efficiency HPGe detectors are normally used and the detector will typically be positioned further from the waste item (i.e. >2 m compared to <1 m). It is also common for the detector to be located in a collimator to restrict the field of view to only a portion of the waste item, and to include either fixed or actuator driven attenuators to reduce the count rate further. The associated counting electronics will also be designed to process higher throughputs and may include verification of the dead time correction.

#### 4.1.3 Performance Values

Performance data for the measurement of the uranium mass in spent fuel waste from a HRGS measurement of the fission product activity is reported in Table 26. The performance is reported for a system used to measure the amount of uranium on a tray containing swarf generated from the decanning of Magnox fuel assemblies prior to re-processing.

<sup>2</sup> This gamma-ray is actually from Ba137m, a very short-lived daughter product of Cs137.

<sup>3</sup> This gamma-ray is actually from Pr144m, a very short-lived daughter product of Ce144.

**Table 26:** Performance Values for Measurement of Uranium Mass in Spent Fuel Waste from HRGS Measurement of Cs137 Activity

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ )	TMU (1 $\sigma$ )		
<b>Detector Configuration:</b> Single HPGe ( $\epsilon = 15\%$ ) <b>Matrix Correction:</b> None (assumed) <b>Lump Correction:</b> Yes <b>Notes:</b>	Waste Sorting Tray	Swarf (from Magnox fuel decanning)	-	$\pm 10\%$	60s	33

## 4.2 Active Neutron Interrogation using Differential Die-Away (DDA) for Fissile Mass Determination

### 4.2.1 Fundamentals

A description of active neutron interrogation techniques is provided in Section 3.3.1. Active neutron interrogation can also be used to determine the fissile content of spent fuel wastes, provided a strong enough neutron interrogation source is available to overcome the high neutron background. Although the Cf252 shuffler technique can be applied, the Differential Die-Away (DDA) technique is more commonly used for spent fuel wastes because of the higher signal to background that can be obtained by counting the prompt neutrons from induced fission reactions rather than delayed neutrons (Cf252 Shuffler).

Although no performance data has been obtained for inclusion in this report, it is noted that Cf252 Shuffler systems have also been used to measure spent fuel wastes using a high intensity Cf252 interrogation source.

### 4.2.2 Differential Die-Away (DDA) Instruments

The high gamma and neutron background radiation emissions from spent fuel wastes require some changes to the design of the DDA instruments described in Section 3.3.3.

A higher intensity neutron source is usually required to induce a measurable signal above the high passive neutron background from curium. The typical DDA instruments in Section 3.3.3 that are used for low to intermediate level wastes utilise a Deuterium-Tritium (D-T) neutron generator with a neutron output in the region of  $10^8 \text{ ns}^{-1}$ , operating at a pulse frequency of 50 Hz or 100 Hz. This means that the neutron output per pulse is of the order of  $10^6 \text{ n.pulse}^{-1}$ .

For measurements of spent fuel, a neutron output per pulse approximately 10 times higher than this (i.e.  $\sim 10^7 \text{ n.pulse}^{-1}$ ) is generally required in order to induce a sufficient signal above the neutron background. Higher output D-T neutron generators are available, but they are considerably more expensive and tend to be less reliable than the lower output systems. Alternative neutron sources such as linear accelerators (for example, accelerating deuterium into a beryllium target) and cyclotrons (for example, accelerating protons into a beryllium target) have also been successfully used in DDA systems measuring spent fuel wastes (Reference [14]).

The high gamma dose rate from the spent fuel wastes also causes problems for the neutron detectors, since multiple gamma interactions in the neutron detector (which each deposit only a small amount of energy compared to a neutron interaction) can “pile-up” to appear as neutron events in the detection electronics. To avoid this problem, several cm of lead shielding is required to reduce the gamma dose rate to the neutron detectors. However, the presence of lead in the DDA system serves to prolong the thermal neutron lifetime of the measurement region, significantly changing the performance characteristics. The longer thermal neutron lifetime means that the detection limits will be higher and the neutron generator pulse frequency must be reduced.

### 4.2.3 Performance Values

Performance data for the measurement of the total fissile mass in spent fuel waste using DDA is reported in Table 27. The performance is reported for one system used to measure the fissile content of a basket containing the metal waste from the shearing of AGR, PWR or BWR fuel assemblies (which are commonly known as hulls).

**Table 27:** Performance Values for Measurement of Total Fissile Mass in Spent Fuel Waste using DDA

System Information	Container Type	Waste Matrix	Performance Values		Count Time	System Number (Appendix 1)
			LOD (1 $\sigma$ )	TMU (1 $\sigma$ )		
<b>Detection Efficiency:</b> Not given <b>Matrix Correction:</b> Flux probes and fuel type dependent calibration. <b>Lump Correction:</b> NO <b>Notes:</b> Uses TNC to infer Safeguards results <sup>Note 1</sup> . High output neutron generator $\sim 10^7$ neutrons / pulse.	Small Crate <sup>Note 2</sup>	Metals	2-3g U235eff	$\pm 10\%$ <sup>Note 3</sup>	4200s	18

<sup>Note 1</sup> U235 mass, Total Pu Mass, Fissile Pu Mass are inferred from fissile mass measurement using algorithms derived from FISPIN data.

<sup>Note 2</sup> Actually a basket of diameter "70cm and fill height ~2m.

<sup>Note 3</sup> Fissile mass generally overestimated due to pessimistic assumptions on the distribution in the calibration. Amount of overestimation typically " 26% for PWR/BWR fuels and " 16% for AGR fuel.

Comparing the performance of this DDA system with those in Table 23, it is evident that this system has a much higher detection limit due to the higher background (neutron and gamma) from spent fuel. In this case the TMU is relatively low due to the use of specific calibrations for each fuel type, although it is noted that the calibration makes several (pessimistic) assumptions regarding the distribution and self-shielding in the fuel, which will potentially bias the measurement result, and this bias is not included in the reported TMU.

## 5. Conclusions

This report presents performance values for Non-Destructive Assay (NDA) systems used to measure the plutonium and/or U235 content of waste items. The detection limit and Total Measurement Uncertainty (TMU) performance values have been obtained from systems that are operating in-the-field and all values presented have been evaluated and are endorsed by the members of the ESARDA NDA Working Group.

It is intended that all the measurement techniques that are applicable to waste assay, and all the container types and waste matrices that are in usage are represented in this report, and good coverage has been obtained for the techniques that are commonly used. However, there are some gaps where verifiable performance information could not be obtained for some of the less widely used techniques, containers and waste matrices. As expected, most information was obtained for systems employing the HRGS and PNCC techniques as these are the most commonly used for measurements of Special Nuclear Material (SNM).

Performance values were obtained for a wide variety of measurement systems employing different radiometric techniques and having different levels of sophistication; for example, the HRGS systems in this report range from single, fixed detector configurations to multiple detector Segmented Gamma Scanners (SGS) and Tomographic Gamma Scanners (TGS). Interestingly, similar performance values were obtained for all the HRGS systems regardless of their complexity. The reason for the consistency of performance reflects the usage of the systems; that is, the least complex systems are generally used for better defined waste streams (i.e. low density, segregated and mostly homogeneous), whereas the higher complexity systems are used for more matrices with a greater range of densities and degree of heterogeneity.

In the case of PNCC systems, as expected better detection limit performance was obtained for systems with higher neutron detection efficiency, however the TMU performance was similar irrespective of detection efficiency.

Where possible, performance values are presented for different types of waste container (ranging from small cans or bags to large crates) and for different waste matrices, since the performance of many of the techniques is highly dependent on the waste type and container size. As expected the performance is worse for the larger container sizes and more challenging waste matrices, with higher detection limits and total measurement uncertainty.

These performance values which represent the performance that is currently achievable for waste assay will be useful to the Safeguards community.

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## Appendix 1 – List of Systems used for Performance Values

References for the performance data for each system are listed below the table.

System Number	System Name	Supplier (Product Name if Applicable)	Location (Site / Country)	Performance Value References
1	FUNE (Fut NEutron)	IRSN	IRSN/DEND/SATE, France	[P1][P2][P3][P4][P5]
2	PLUM (PLUtonium Masse)	IRSN	IRSN/DEND/SATE, France	[P1][P3][P4][P6][P7][P8]
3	MAPU	IRSN	IRSN/DEND/SATE, France	[P3][P9]
4	Waste assay system 3AX	SCK•CEN	SCK•CEN waste building, Belgium	Internal reports
5	Low-level waste assay system	Canberra Q <sup>2</sup>	SCK•CEN waste building, Belgium	Internal reports
6	MCS SGS	Canberra SGS	Savannah River Site, USA	[P10][P11]
7	MCS IQ3	Canberra IQ <sup>3</sup>	Savannah River Site, USA	[P12][P13]
8	Savannah River Box Counter (SRBC)	Canberra Large Box Counter	Savannah River Site, USA	[P14][P15][P16][P17][P18][P19]
9	SGRS	Canberra Q <sup>2</sup> variant	Idaho National laboratory, USA	[P20][P21]
10	WAGS	Canberra IQ <sup>3</sup> variant	Idaho National laboratory, USA	[P22][P23]
11	HENC	Canberra HENC	Idaho National laboratory, USA	[P24][P25]
12	DWAS	Pajarito Scientific Corp Imaging Passive/Active Neutron Counter	Oak Ridge National laboratory – Melton Valley Site, USA	[P26][P27]
13	Mobile SGS-01	Canberra SGS	Oak Ridge National laboratory – Melton Valley Site, USA	[P28][P29]
14	HENC 1	Canberra HENC	Los Alamos National Laboratory TA 54, USA	[P30][P31]
15	HENC 2	Canberra HENC	Los Alamos National Laboratory TA 54, USA	[P32][P33]
16	WTC HRGS and PNCC Systems	Babcock (PNCC) & Canberra (HRGS)	Sellafield, Waste Treatment Complex, UK	Internal reports
17	SMP Drum Monitor	A.N Technology Ltd (ANTECH)	Sellafield, SMP, UK	Internal reports
18	Thorp Hulls Monitor	Babcock	Sellafield, Thorp, UK	Internal reports
19	TRU-D® Plutonium Packet Monitor	Babcock	Various (Mainly UK)	Internal reports
20	TRU-D® Piece Monitor	Babcock	Sellafield, UK	Internal reports
21	Drigg Retrievals Project Drum Monitor	Canberra UK	LLWR, B746/B720, UK	Internal reports
22	B748 Crate Monitor	Babcock	LLWR, B748, UK	Internal reports
23	Waste Drum Assay System (WDAS)	Babcock TRU-D® Drum Monitor & DrumScan®	RRP, Rokkasho, Japan	[P34]
24	HEC NI	Canberra UK	Harwell, RHILW Solid waste plant, UK	Internal reports
25	NDA19 Segmented Gamma Scanner	Canberra	Dounreay, WRACS, UK	[P35]
26	NDA20 Combined PNCC / Cf Shuffler	Canberra UK	Dounreay, WRACS, UK	[P35][P36] & Internal reports
27	SILWAS HENC	Canberra HENC	AWE Plc, UK	Internal reports
28	SILWAS AQ2	Canberra AQ <sup>2</sup>	AWE Plc, UK	Internal reports
29	PANWAS (Passive Active Neutron Waste Assay System)	Canberra	Nucleco, Rome, Italy	Internal reports
30	SGS	Canberra	Nucleco, Rome, Italy	Internal reports
31	ISOCS	Canberra ISOCS		Internal reports
32	PTGS	ANTECH TGS	LANL, USA	[P37][P38][P39]
33	Swarf Inventory Monitor	Babcock	Sellafield, UK	Internal reports
34	NDA23 Active Well Coincidence Counter	Canberra UK	Dounreay, UK	[P40][P41] & Internal reports
35	NDA34 Package Counter	Babcock	Dounreay, UK	Internal reports
36	DrumScan LRGS MkII (2 systems)	Babcock	AWE Plc, UK	Internal reports



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