Measurement Good Practice Guide No. 34

Radiometric Non-Destructive Assay

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ABSTRACT

This document is intended to promote good practice on the specification, supply, operation and maintenance of equipment used for radiometric non-destructive assay (NDA) of fissile and other radioactive materials.

The various techniques, associated applications and their limitations are described. Guidance is given on methods of good practice for calibration of NDA systems and the treatment of uncertainties in NDA measurements. Recommendations are made for certification, performance demonstration and testing; competence and responsibility; documentation and the control of modification.

Acknowledgements

Contents

Glossary

The following terms are used in this document:

Accidentals count See *Reals count.*

Accuracy The accuracy of a measurement system is the degree of closeness of measurement of a quantity to its actual (true) value.

Benchmarking The results of measurements or tests carried out under reference conditions against which the results of other measurements, tests or modelling predictions may be compared.

Bias A constant error that produces a consistent deviation from the true value.

Calibration Procedure to establish a quantitative relationship between the response of an instrument and the quantity to be measured. Performed using analytical techniques or measurements involving one or more reference standards under a set of reference conditions.

Calibration Check Procedure carried out periodically to verify the validity of the original calibration parameters.

Characterisation Used interchangeably with *Calibration.*

Calibration Factor Reciprocal of response. Factor by which a reading is multiplied to obtain the quantity being measured.

Californium Shuffler An active neutron interrogation device involving rapid exposure and removal (shuffling) of a californium neutron source.

Confidence Level Number expressing degree of confidence in the result; usually expressed as a percentage.

Coverage Factor Number that is multiplied by the combined standard uncertainty to give an expanded uncertainty for a given confidence level.

Detection Limit The smallest quantity of radioactive material which, under defined operating conditions, can be quantified with a specified degree of confidence. Also commonly referred to as *Minimum Detectable Activity (MDA)* or *Minimum Detectable Quantity (MDQ).*

Error A deviation from the true value.

Expanded Uncertainty The standard uncertainty (or combined standard uncertainty) multiplied by a coverage factor to achieve a given confidence level.

Functional Check See *Operational Check.*

List Mode Type of event-by-event data acquisition used in some counting systems.

Matrix The material surrounding or holding the substance being assayed. This material may significantly affect the response of the system by attenuation, absorption and other effects on radiation emerging from or entering the substance.

Minimum Detectable Activity (MDA) or *Minimum Detectable Quantity (MDQ)*	See *Detection Limit.*
Non-Destructive Assay (NDA)	The observation of spontaneous or stimulated (induced) radioactive emissions, suitably interpreted to estimate the amount of one or more nuclides of interest in the item being assayed, without affecting the physical or chemical form of the material.
Operational Check	A frequent test performed to check for faults within the instrument. Also known as *Functional Check.*
Reals count	In a neutron coincidence counting system, a *Real* coincidence event occurs when the system registers two or more neutron detections within a specified time window and when this has arisen due to the detection of neutrons arising from the same emission (e.g. spontaneous fission) event. This is in contrast to an accidental coincidence between neutrons (the *Accidentals count*) arising from separate emission events that happen to arrive at the detection system at the same time.
Reference Standard	A physically and chemically stable item for which the attributes of interest are well-characterised and traceable to primary standards, and for which the other properties affecting the measurement technique are known. Reference standards are used in a measurement system to establish the relationship between the basic instrument response and the attribute(s) of interest.
Repeatability	The degree of agreement between repeated measurements of the same quantity under unchanged measurement conditions. Also referred to as *Reproducibility.*
Reproducibility	See *Repeatability.*
Response	The ratio of the observed reading of an instrument to the true value of the quantity producing that reading.
Scheduled Maintenance	A test of the equipment performed at periodic intervals.
Self-shielding	Attenuation and absorption of emitted radiation by the substance emitting the radiation.
Sensitivity	The magnitude of the detected signal in a measurement system relative to the quantity of radioactive material.
Setting to Work	The determination of the measurement system's characteristics and operating parameters for the defined assay envelope.
Solid Angle	The area of the segment of unit sphere covered by the projection of the object onto that sphere.
Standard Deviation	The positive square root of the *Variance.*
Standard Uncertainty	Uncertainty of a measurement expressed as a margin equivalent to plus or minus one standard deviation.

Tomography	Cross-sectional imaging technique involving the measurement of radiation from and/or the irradiation of a sample from different angles and using algorithms to construct a 2-D/3-D image.
Totals count	In a neutron counting system, this is the total number of neutron detection events recorded, irrespective of the time correlations between individual counting events.
Traceability	The ability to relate measurements to appropriate national or international standards through an unbroken chain of calibrations carried out in a technically sound manner.
Uncertainty	Parameter, associated with the result of a measurement, which characterises the dispersion of the values that could reasonably be attributed to the quantity being measured.
Variance	A term used to describe the dispersion of a set of observations with respect to its arithmetic mean. Equal to the mean square deviation from the arithmetic mean.
Working Standard	A physically and chemically stable item containing material that is measured as part of a measurement control programme to test system stability. Produces an instrument response characteristic of items being assayed in the measurement system. The material may bear no physical or chemical resemblance to those items.

The following acronyms and symbols appear in this document:

α	Ratio of random-to-spontaneous fission neutron production rates for non-multiplying plutonium bearing material.
AAS	Add-A-Source (matrix correction technique for PNCC); also known as Matrix Interrogation Source (MIS).
ANCC	Active Neutron Coincidence Counting: a neutron technique based on discriminating between neutrons from fission induced by an external source and the interrogating radiations.
BGO	Bismuth germanate; a scintillator material used for gamma-ray detection and energy measurement.
CdZnTe or CZT	Cadmium zinc telluride; a semiconductor material used for gamma-ray detection and energy measurement.
CsI(Tl) or *CsI*	Thallium-activated caesium iodide; a scintillator material used for gamma-ray detection and energy measurement.
DDA	Differential Die-Away: a neutron technique based on the build-up and decay of a thermal neutron interrogating flux following the injection of a short pulse of neutrons.
E	Efficiency of detector system.
EMC	Electro-Magnetic Compatibility.
HEU	Highly Enriched Uranium.

HPGe	High Purity Germanium; used for gamma-ray detection and energy measurement.
HRGS	High Resolution Gamma Spectrometry; a technique based on measurement of the γ-ray spectrum from a sample using a high resolution detector.
ILW	Intermediate Level Waste.
LLD	Lower Limit of Detection.
LLW	Low Level Waste.
LRGS	Low Resolution Gamma Spectrometry; a technique based on measurement of the γ-ray spectrum from a sample using a low-resolution detector.
m_{eff}	Effective mass of plutonium (or other nuclear material).
MCNP	Monte Carlo N-Particle. A Monte Carlo radiation transport modelling code widely used to support the characterisation and calibration of systems / processes.
M_L	Neutron leakage multiplication.
MDA	Minimum Detectable Activity.
MDQ	Minimum Detectable Quantity.
MIS	Matrix interrogation source (see AAS).
MOX	Mixed (plutonium and uranium) Oxide; a reactor fuel type.
NaI(Tl) or *NaI*	Thallium-activated sodium iodide; a scintillator material used for gamma-ray detection and energy measurement.
PNCC	Passive Neutron Coincidence Counting; a neutron technique based on discriminating between time-correlated neutrons from spontaneous fission materials and random neutrons from other sources.
PNMC	Passive Neutron Multiplicity Counting; a neutron technique based on the measurement of singles, double coincidences and triple coincidences of neutrons emitted from a sample.
$^{240}Pu_{eff}$ or $^{240}Pu_{eq}$	^{240}Pu effective mass (or ^{240}Pu equivalent mass).
RTR	Real time radiography.
SF	Spontaneous fission.
SGS	Segmented Gamma Scanner; a technique involving the measurement of γ-rays from successive segments of a sample.
TCA	Time correlated analysis; data analysis method used in certain types of neutron coincidence counters.
TGS	Tomographic Gamma Scanner; a similar technique to SGS in which spatial emission and attenuation maps are determined.

TNC	Total Neutron Counting; a technique involving the measurement of all neutrons emitted by a sample.
$^{total}Pu$, ^{tot}Pu or Pu_{total}	Total plutonium mass.
$^{235}U_{eff}$	^{235}U effective mass.
XRF	X-ray Fluorescence; a technique based on the use of an external photon source to excite fluorescent X-rays in a sample.

Foreword

A forum of experts representing the interests of all facets of the non-destructive assay (NDA) community has been brought together as a working group to review and update a document that provides general guidance for making non-destructive assay measurements of high quality and to help standardise good practice across the industry. The test procedures recommended in this document are general methods based on current accepted good practice and are not intended to be a standard.

The working group has membership with participants from the following groups:

- Instrument suppliers
- Expert users
- Technical support specialists
- Scientific community (standards-setting organisations)

This guide should prove useful to professionals involved in many different areas including radioactive waste activity measurements, fissile material quantification for fuel reprocessing process control, and nuclear material accountancy for international safeguards, as well as supporting criticality control.

The guide has been revised to encompass all aspects of the measurement process, and so should be valuable to anyone involved in any aspect of NDA. Regulators, users, technical specialists, programme managers, operations managers, equipment designers and suppliers should all find useful information within.

Introduction

1

IN THIS CHAPTER

■ Introduction

A variety of radiometric non-destructive assay (NDA) techniques are routinely used in the nuclear industry for quantifying the amount of radioactive materials in packages and containers for various important safety and accountancy purposes. For example, the results of NDA measurements have been used to make accountancy declarations for inventory and security purposes, and to demonstrate compliance with criticality safety rules.

This Good Practice Guide provides recommended procedures for the development, general examination, testing, characterization, calibration and operation of NDA equipment. The guide is intended to assist users and employers in gaining confidence in their arrangements, and is aimed at key applications in the nuclear industry, such as:

- Sentencing
- Radiological Characterization
- Process Control
- Process Measurement
- Hold-up
- Accountancy Control
- Radiological Safety
- Criticality
- Security
- Safeguards
- Quality Control.

The guide is not intended to cover:

- Non-destructive examination for the purposes of testing the structural integrity of plant or equipment, or for security purposes, e.g. the use of X-ray equipment in airports or use of radiometric equipment border control / security checks
- Nuclear techniques for medical purposes
- Radiological protection equipment, whether portable or installed
- Environmental or emergency response monitoring.

It is intended that this guide will contribute to the continuing professional development of practitioners as well as assisting organisations in meeting their commitment to ensure that arrangements for the use of such equipment are adequate. As a result of operational experience in the use of NDA equipment in the field, it has been recognised by employers and regulators that there are advantages in setting out clearly defined minimum practices and expectations in the use of such techniques. This should encourage consistent approaches between different organisations and their applications. It should be noted however that this good practice guide does not constitute a standard.

The Lifecycle

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IN THIS CHAPTER

- ■ The Lifecycle

Engineering projects follow lifecycles; for a NDA system this generally encompasses conception through to decommissioning and ultimate disposal of the component parts. Each phase within the lifecycle has clearly defined objectives and is (usually) governed by a set of procedures and processes.

The lifecycle is often depicted in the form of a model, which shows us not only the stages within the lifecycle, but also the way in which they relate to one another. A simple lifecycle model is depicted below:

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        ┌─────────────────────────────┐
        │     Define Requirements     │
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        ┌─────────────────────────────┐
        │     Supply and Test System  │
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        │      Commission System      │
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        ┌─────────────────────────────┐
        │   Operate and Maintain System │
        └─────────────────────────────┘
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        │     Decommission System     │
        └─────────────────────────────┘
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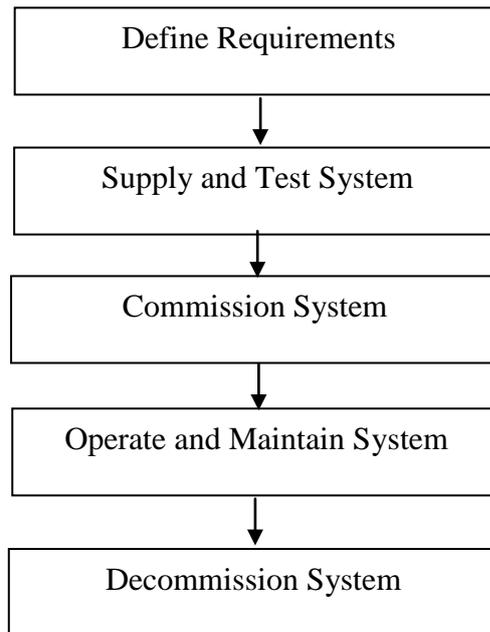
Within this simple model, the objectives of the various phases would be:

Define System Requirements: to fully understand the need and convert it into a complete and unambiguous specification for the system.

Supply and Test System: to design, build and deliver a system that demonstrably performs the functions and otherwise meets the requirements as defined; to reduce the risk that the system fails to deliver its requirements once integrated into its operational environment.

Commission System: a complete demonstration that the system performs the functions for which it has been supplied and otherwise meets the requirements as defined, having been integrated into its operational environment; to set the system to work.

Operate and Maintain System: to use the system to meet the business need and to ensure that the system continues to perform in the required manner.

Decommission System: to remove the system from operation and safely dispose of it or its components.

Over the years, a number of different lifecycle models have been developed, largely because deficiencies have existed which, through use and experience, have been recognised and rectified. All lifecycles have the same basic stages as the lifecycle shown above; some decompose the stages further i.e. there are more stages; some include additional verification and validation activities; however, the biggest difference is to be found in the way in which the phases relate to one another. A few common lifecycle models are depicted below.

The Waterfall Model

The Incremental Development Model

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│   Define Requirements   │◄───────────┐
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┌─────────────────────────┐            │
│      Design System      │            │
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┌─────────────────────────┐            │
│   Implement System      │            │
│        Design           │            │
└─────────────────────────┘            │
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┌─────────────────────────┐            │
│      Test System        │            │
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          ◇ Deploy    No              │
          ◇ System? ──────────────────┘
             │
            Yes
             ▼
┌─────────────────────────┐
│    Commission System    │
└─────────────────────────┘
             │
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┌─────────────────────────┐
│   Operate and Maintain  │
│         System          │
└─────────────────────────┘
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┌─────────────────────────┐
│   Decommission System   │
└─────────────────────────┘
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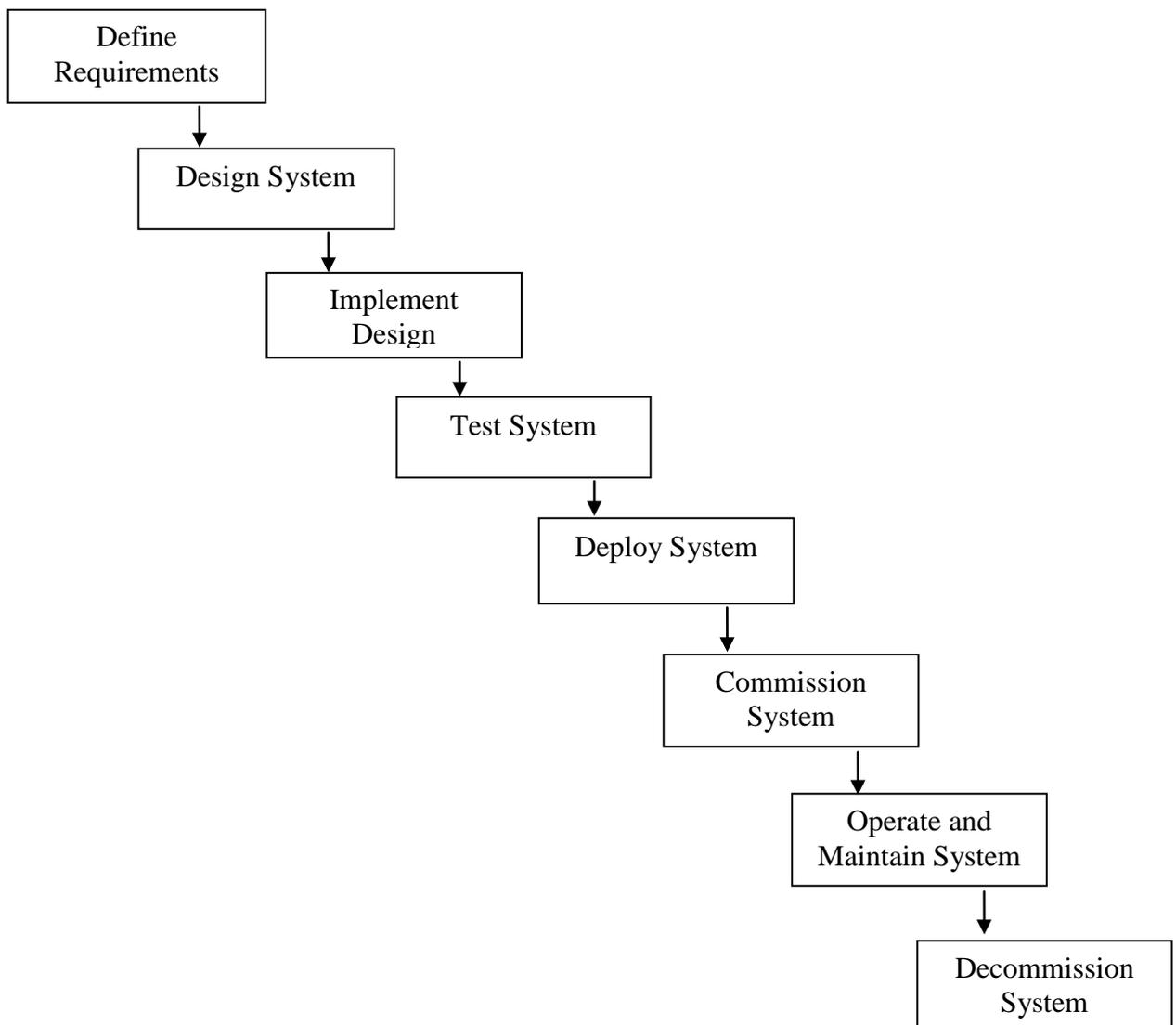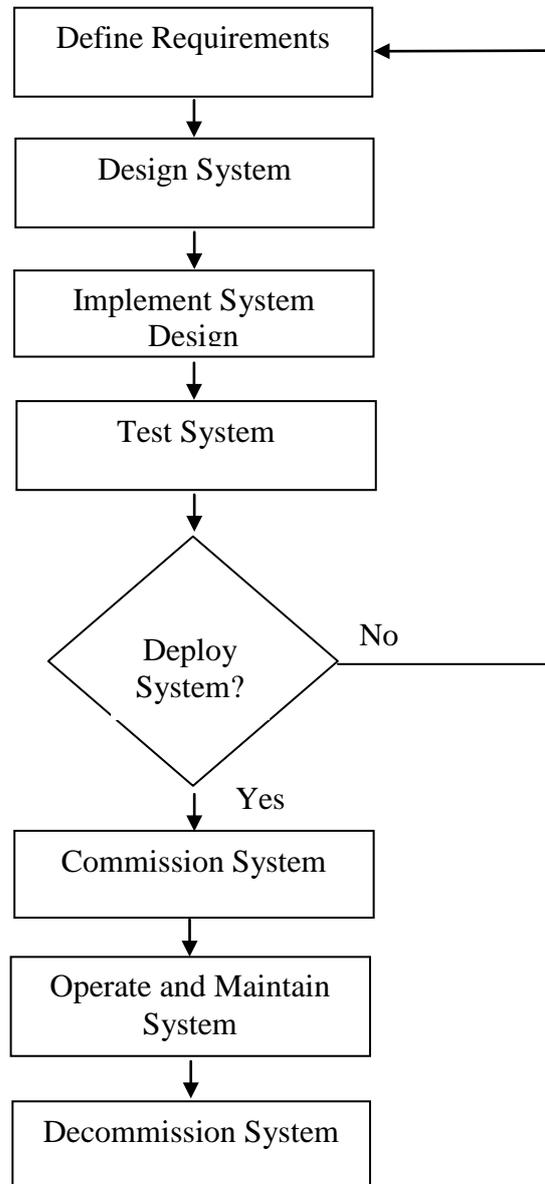
The V Model
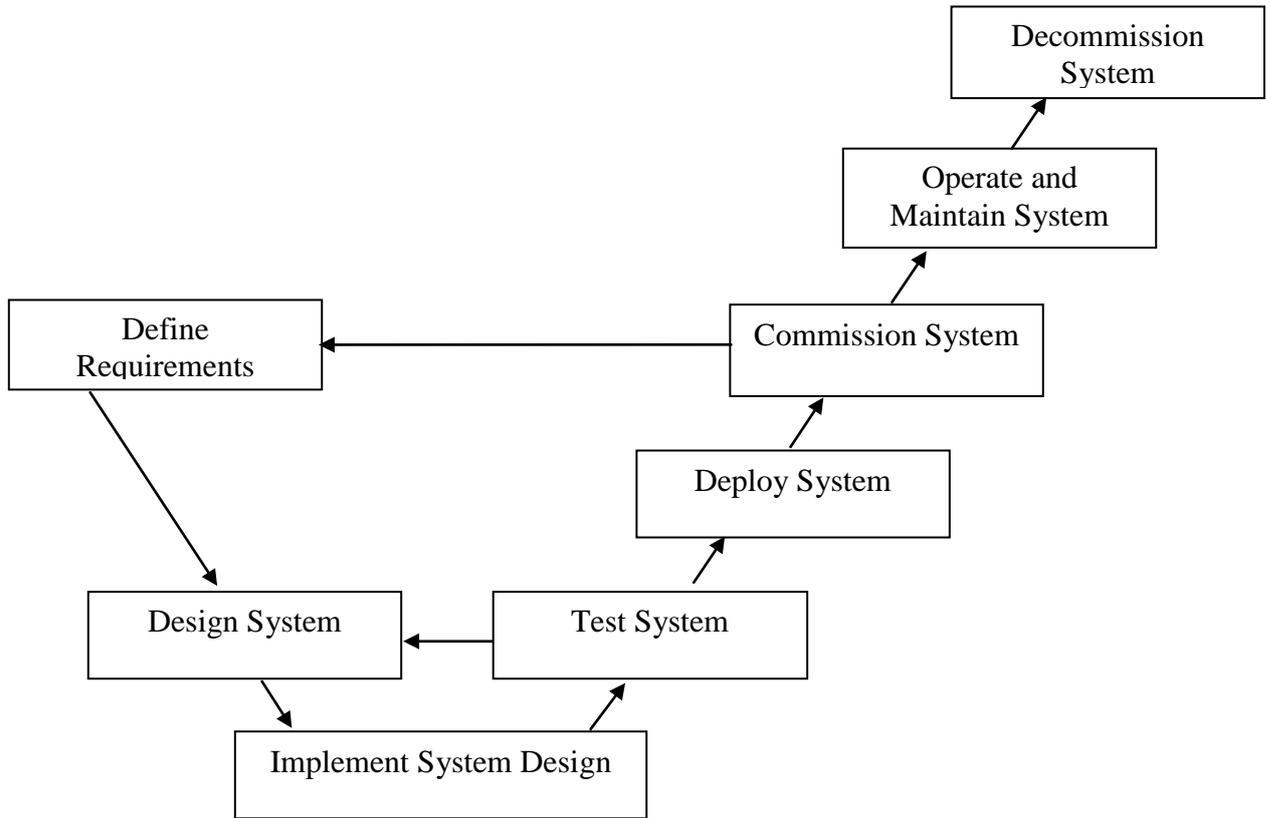
Each of these lifecycle models has different traits and is best suited to different scenarios. For example, the Waterfall Model assumes that each stage can be, and is, completed before the next stage begins; once a stage is completed, it is not returned to. This model is best suited to applications where the requirements of the system are well understood and there is limited complexity. Often this is because the job has been done many times before.

Conversely, the Incremental Development Model not only permits, but encourages iteration; each phase can be revisited as many times as is necessary. This model is best suited to novel applications where the solution may not be obvious.

The V Model places a greater emphasis on testing (verification and validation) activities. This model is often associated with enhanced robustness or integrity and is used where the system is to be used in an application where a high degree of reliance is to be placed on its performance; within the industry, this has become the preferred lifecycle model where a safety or safety related requirement has been specified.

Other lifecycle models exist and it is not the objective of this section to preclude the use of these; it is suggested that, for any given system, the lifecycle model should be selected and justified based on application and complexity. It is not uncommon to find that a project uses a combination of lifecycle approaches. The remainder of the sections within this guide are not specific to any lifecycle and are intended to provide guidance as to what should be considered during each stage of the lifecycle.

Although the lifecycle models described above refer to "systems", the same principles apply for measurement projects or tasks involving mobile assay equipment. In these cases the commissioning and testing activities may be less onerous than for projects involving major new assay system installation(s), because the portable equipment is typically maintained in a state of readiness for measurement tasks in any case. However, it is equally important to identify and define the requirements of such measurement tasks and follow an appropriate lifecycle of system specification, calibration, testing, method validation, quality control, checking and validation of the final results.

- ## **Define Requirements**

The first stage in any system lifecycle is 'Define Requirements'. The objective of this stage is to understand the need and convert it into a complete, consistent and unambiguous specification for the system. The specification must be understandable at all stages of the lifecycle, and by those involved at each stage. As such, the focus is on deciding what the system should do, how well it should do it and under what conditions. How the system will achieve all of this is considered at subsequent stages within the lifecycle.

It is good practice to understand that any number of different parties (often referred to as stakeholders) may have expectations about what the system should do and why it should do it; these need to be elicited and understood before the system requirements can be fully specified. Stakeholders commonly include different groups within the same organisation, as well as external groups such as the regulators and the general public; the various stakeholder groups need to be identified.

It should be understood that the lifecycle models depicted above are (intentionally) simplistic. Lifecycles for many real non-destructive assay systems are more complex, certainly in the context of adding further detail to the activities to be undertaken. It is not uncommon to further de-compose the 'Define Requirements' stage of the lifecycle.

Typically, activities such as:

- Optioneering
- Feasibility Studies
- Modelling
- Prototyping

are all activities that may be undertaken to elicit and fully understand the system requirements.

The importance of this phase of the lifecycle should not be underestimated – many so-called system 'failures' can be attributed to inadequate or incorrect specification or to misunderstandings between specifiers, suppliers and operators. As a general rule, the later problems are detected, the more time consuming and expensive they are to overcome.

Problems commonly encountered include: not all stakeholders are identified; stakeholders are unable to explicitly state their expectations; the application domain is often complex, or even unfamiliar; stakeholder requirements are inconsistent and may even conflict. These difficulties need to be overcome before the system requirements can be specified.

In general, system requirements fall into two categories: functional requirements and non-functional requirements.

• **Functional Requirements**

Functional requirements define what the system is supposed to do and generally take the form "the system *shall* [requirement]". Functional requirements describe the functions of the system (i.e. what the system is required to do) and should be implementation independent.

Functional requirements should also describe anything the system is not supposed to do in this case, they tend to take the form "the "system *shall not* [requirement]". Often this type of functional requirement pertains to fault conditions. Again they should be implementation independent. For example:

- The system *shall* return the ^{137}Cs activity of the sample
- The system *shall* return the total plutonium content of the sample
- The system *shall* return the % ^{235}U enrichment of the sample
- The system *shall* return the total fissile mass (^{235}U+total Pu) of the sample
- The system *shall* return the measurement uncertainty to 2 standard deviations.

- The system *shall not* make a measurement if a detector has failed.

• **Non-functional Requirements**

Non-functional requirements are often called qualities. Other common terms include constraints, quality attributes, quality goals, quality of service requirements and non behavioural requirements.

Non-functional requirements define what the system is supposed to be and generally take the form "the system *shall be* [requirement]" and can be divided into two main categories:

1) Execution qualities, such as security and usability, which are observable at run time
2) Evolution qualities such as integrity, testability, maintainability which are embodied within the static nature of the system.

For example:

*   The limits of detection for ^{235}U *shall be* 50g across the range of samples
*   The system *shall be* required to interrogate samples having the following range of physical form
*   The system *shall be* required to interrogate samples having the following range of activity distribution
*   The system *shall be* capable of assaying 20 samples per 8 hours of operation
*   The availability of the system *shall be* 95%
*   A functional test *shall be* performed every 8 hours
*   The system *shall not be* implemented using a Windows 3.11 platform.

Often specification of non-functional requirement is more difficult and time consuming than specification of functional requirements.

Requirements should be documented in the form of a system specification document. When constructing such a document, it is good practice to provide background as to why the system is needed, in addition to specifying the system requirements.

The remainder of the lifecycle is dedicated to ensuring that the system is designed, delivered, operated and maintained so as to demonstrably deliver the requirements, as specified. Clearly, specification of requirements is key to success; therefore, the requirements specification should be agreed by all stakeholders before delivery commences.

NDA systems will sometimes be called upon to perform safety function(s) derived from safety case or risk assessment, for activities or operations involving nuclear material. However, it is not the intent of this guide to be prescriptive as to the supply of such systems; certainly, it is not intended that adherence to the good practices promoted herein will provide an automatic method of compliance with those standards which address functional safety or safety instrumented systems.

Measurement Techniques and Applications

3

IN THIS CHAPTER

- ■ Neutron Techniques
- ■ Gamma Techniques
- ■ Calorimetry
- ■ Other Techniques

This section describes the basic physics of the various NDA techniques available. In addition, the key features of the assay systems in common use are described, with an appraisal of their benefits and limitations with respect to both technical and operational issues.

The selection of assay technique(s) for a particular application depends on a large number of factors relating to the effectiveness of the techniques –

- Provenance and history of material to be assayed
- Physical characteristics of the item being assayed (shape/size/composition of container and physical composition of the contents - materials, density, homogeneity etc)
- Chemical composition of the material to be assayed
- Isotopic composition of the material to be assayed
- Physical parameters which are required to be measured
- Lower Limit of Detection requirements
- Upper limit activity/mass requirements.

and to issues relating to practical implementation -

- Throughput requirements
- Assay accuracy requirements
- Plant geometrical / space constraints
- Available financial resources
- Available operational expertise
- Available measurement technology.

Due to the large number of variables, the criteria for selection of an assay technique can be complex. This section provides some guidance to assist with the decision making process for selection of assay techniques and highlights some of the applications that can benefit from the selection of a range of assay techniques. Of particular interest are the detection limit and accuracy of non-destructive assay methods. Care and attention to these two requirements are of the utmost importance. Furthermore, these parameters do not behave similarly and must be managed on their own. Methods that offer good (i.e. low) detection limit performance do not necessarily offer high accuracy by default and may be wildly inaccurate under unfavourable conditions.

Further, it is often the case that the diversity of sample characteristics and/or the range of parameters that have to be measured, favours the use of a complementary suite of assay techniques rather than a single, preferred system. As such, it may be the case that no single assay technique can be selected that best or adequately suits *all* of the

requirements. The complexity of the assay technique (e.g. with regard to the number of different types of calibration and whether or not particular measurement correction methods are required) depends upon the sample characteristics. For example, if more is known about the samples being measured or if there is greater uniformity in their properties, the measurement can be much simpler with only a single calibration required.

Considerable savings can sometimes be achieved by integrating various techniques within a single chamber. For example, passive neutron counting is often combined with active neutron counting within a single assay chamber. High resolution gamma-ray spectrometry is often combined with passive neutron counting for plutonium assay, providing the isotopic vector that allows the total Pu mass to be calculated from the ^{240}Pu effective mass measured by passive neutron counting. Any savings obtained by combining measurement functions in this way must however be balanced against the potential compromises in measurement performance and also the risk of the complete assay facility failing because of a fault with one component of the system.

A comprehensive review, covering most of the NDA techniques described in this guide, is given in [1].

3.1 Neutron Techniques

Neutrons may be emitted from nuclear material through many different production routes [2], for example (α, n), (n, 2n), spontaneous fission (SF) and induced fission. Neutrons are highly penetrating in high atomic number materials and therefore neutron detection techniques have advantages over gamma based techniques when the material is of bulk nature or is contained within metallic or other dense materials such as in filled waste drums or product cans.

Passive neutron techniques rely upon the detection of natural neutron emissions from the assay material (neutrons originating from the material itself) without relying on any external excitation of the sample. In plutonium, for example, the fertile isotopes (^{238}Pu, ^{240}Pu and ^{242}Pu) undergo spontaneous fission releasing bursts of "coincident" neutrons emitted practically at the same moment in time. Additionally, most plutonium isotopes and other radionuclides (such as ^{241}Am, occurring as a decay product of ^{241}Pu), decay through alpha particle emission thereby producing single "random" neutrons through (α, n) interactions in certain light elements that are in intimate contact with the α-emitting radionuclide (e.g. PuF$_4$).

Active neutron techniques may be employed when the isotopes in the sample being assayed do not emit sufficient spontaneous neutrons to permit precise measurements to be made within a realistic time period, or when a direct measurement of a fissile

isotope is required. It is possible to induce fission or other neutron producing reactions within these nuclides using an external excitation or interrogation source. Normally, a moderating assay chamber is used to moderate and thermalise the source neutrons, to build up a flux that interrogates the sample container. The ensuing prompt or delayed fission neutrons can then be counted using a moderating detection chamber, as for passive neutron detection techniques.

Neutron detection equipment for use in industrial facilities is generally based upon either ^{3}He or boron trifluoride (BF$_3$) gas-filled proportional counters, although alternative detector types including ^{4}He and H$_2$ gas-filled detectors, fission chambers and scintillators are also available. The ^{3}He-filled proportional counters are most widely used, and have superseded the BF$_3$ counters (mainly due to health and safety considerations). The alternative detectors (^{4}He, H$_2$, fission chambers and scintillators) are, however, of limited use for waste assay (apart from very specific applications) because of their much lower detection efficiencies. Further information on the configuration and operation of neutron detectors, and the benefits and limitations of each type of detector is given elsewhere [3,4].

Note that in recent years there has been a world-wide shortage in stocks of ^{3}He. This has been in part due to increased demand for ^{3}He for neutron detectors for use in national and border security applications and also because the number of facilities involved in the production of the gas has rapidly declined. ^{3}He does not occur naturally on earth in significant quantities. Since the late 1960s it has been readily available as a by-product of the tritium gas produced to support nuclear weapons programmes. Reductions in nuclear arsenals around the world and the subsequent closure of key facilities that produce the purified gas from the tritium decay products have led directly to this shortage, which at the time of publication shows no sign of being resolved.

Suppliers of neutron detectors and the systems in which ^{3}He has hitherto been used are therefore increasingly looking to alternative methods of detecting neutrons. These alternative detector types inevitably have different characteristics from ^{3}He-based detectors and so will have an impact on the properties/performance of the assay systems in which they are used. Since, to date, very few (if any) production assay systems have been manufactured that use alternative detector types, in the following discussion ^{3}He type neutron detectors are assumed.

3.1.1 Passive Neutron Counting Techniques

For the interpretation of passive neutron counting data four variables are usually considered:

        (i)   effective mass of plutonium (or other nuclear material), m_{eff},

(ii)   neutron leakage multiplication, M_L,

(iii)   efficiency of the counting system, ε,

(iv)   ratio of neutrons produced by (α,n) reactions to those produced by spontaneous fission, α.

In order to <u>fully</u> characterise the assay item, at least four measurable quantities are needed to determine these four unknowns, otherwise assumptions for some of these parameters must be made. The techniques listed below provide different means for this interpretation.

In general, the fewer sources of neutrons that a technique is sensitive to the more specific it is to particular groups of isotopes. Total neutron counting is sensitive to all neutron sources, rendering it least specific in terms of isotope identification.

For passive neutron counting, the assayed quantity is usually the ^{240}Pu effective mass, m_{eff} (sometimes also known as the ^{240}Pu *equivalent* mass). The assay response comprises contributions from the spontaneously fissionable isotopes of plutonium ($^{238, 240, 242}$Pu), as well as other species such as $^{242, 244}$Cm, ^{252}Cf, and (to a lesser extent) ^{238}U. As such, m_{eff} can be expressed as a linear combination of these isotopes [5]. The presence of ^{242}Cm and ^{244}Cm in particular (in irradiated fuel / residues), can give rise to gross over-estimation of the Pu mass if its presence is unknown, owing to the very high spontaneous fission branching ratio for these isotopes. The effect is even more extreme for ^{252}Cf, although this is not normally present. The magnitude of this effect is described, together with compensation strategies, in [6,7] In order for the ^{total}Pu mass to be calculated from the measured m_{eff}, an appropriate conversion factor or isotopic vector must be applied. This is often obtained from the analysis of a separate high resolution gamma-ray measurement using a suitable code for determining isotopic composition, such as MGA or other codes [8,9,10,11,12,13].

The passive neutron counting techniques described in this section may be supplemented by the use of various matrix correction and background reduction and correction techniques (described in Section 4) which can often be incorporated within the passive neutron system. Such correction techniques can be used to optimise the performance for a specific measurement scenario.

3.1.1.1   Passive Total Neutron Counting (TNC)

In TNC measurements, all neutrons are counted identically with no attempt made to distinguish between them [14]. As plutonium mass is typically the desired output, knowledge of the isotopic composition, the α-ratio, leakage multiplication factor, M_L, and the measurement system detection efficiency, ε, must be well known. This generally restricts TNC measurements to well-characterised materials (for example where one particular source of neutrons dominates the overall neutron emission, e.g. plutonium metal (SF) or PuF$_4$ (α, n)), or where the sample is well-characterised such

that the specific (α, n) and spontaneous fission emission rates (per gram of material) are known. For instance, TNC methods may be used in the assay of uranium hexafluoride (UF_6) via its high specific (α, n) emission.

Generally, for fissile material contaminated waste, the presence, abundance and proximity of light elements is uncertain, and the use of total neutron counting is therefore limited. Total neutron counting may be used as a 'clearance' or threshold technique to prove the absence of, or give an upper limit for the mass of, fissile material where a high accuracy is not required. The generally superior statistical precision allows TNC to measure materials at very low thresholds, where the absence of neutrons from any source is used to prove that fissile material is not present. This clearance monitoring role is limited by the fact that alpha emitters within the assay item may be mistaken for fissile material (due to (α, n) production), and would need to be verified using another technique. For facilities that process fissile material, TNC can therefore be a very valuable screening tool.

The use of TNC is particularly prone to error when the neutron emission has a significant contribution from the variable quantities of contaminants such as alpha emitters in an (α, n) matrix or through other neutron emitters including curium isotopes. In these cases it may still be possible to use TNC by linking it to other techniques, e.g. determining the total neutron count rate from irradiated fuel and using mass spectrometry or fuel burn-up calculations to determine the plutonium/curium ratio, but the application of these techniques to wastes requires a very careful, and often impractical, assessment of the waste stream characteristics.

Benefits of TNC

- Simple and robust electronics and data processing
- Can use portable equipment to carry out assessments
- Readily available acquisition systems and standard data processing algorithms
- Generally superior statistical precision compared to other neutron counting techniques
- May be useful for threshold monitoring for alpha–bearing waste materials
- Low burden of operator training as hardware and software is generally very simple.

Limitations of TNC

- Requires knowledge of isotopic composition
- Other species that emit neutrons will cause a potential overestimate of plutonium content
- If the sample characteristics are not well known, TNC can result in a large over or under estimate of the plutonium mass

- Requires knowledge of chemical composition (affects α) and/or matrix materials
- Requires knowledge or determination of sample leakage multiplication (M_L) to provide accurate plutonium mass
- Sensitive to background fluctuations
- Sensitive to changes in efficiency (ε) (due to moderators/absorbers in chamber)
- Highly dependent upon the system calibration which should ideally be carried out under conditions that are as realistic as practical.

3.1.1.2   Passive Neutron Coincidence Counting (PNCC)

Passive neutron coincidence counting [15,16], discriminates between the coincident (time-correlated) neutrons produced in bursts (from spontaneous fission events) and those random neutrons produced in (α, n) and other reactions.   The ability to discriminate in this manner permits PNCC measurements to be made even in the presence of (α, n) or ambient background conditions. For plutonium, PNCC is sensitive to spontaneous fission events in all species including [238]Pu, [240]Pu, [242]Pu, but also other spontaneously fissionable isotopes such as [238]U (albeit small), [242]Cm, [244]Cm and [252]Cf, which can result in interference. For the low plutonium masses typical of waste, the induced fissions generated by multiplication in fissile species such as [239]Pu and [241]Pu will also be detected by PNCC, but this usually represents a small increase in the assay response. As the mass of fissile material within the item increases, the neutron leakage multiplication increases [17,18,19,20,21,22,23,24], resulting in overestimation of the fissile mass, unless a self-multiplication correction is applied (see section 4).

The most commonly used form of PNCC counting system is the shift register hardware based coincidence analyser [15,16]. More recently, time correlated analysis (TCA) type PNCC systems (which record the time at which each individual neutron detection event occurs) have become more commonly used [25,26]. Such systems can use software based processing of the detected neutron pulse stream to perform the coincidence analysis and may also operate in list mode providing a record of the neutron pulse stream for later re-analysis.

PNCC is often combined with TNC to permit two of the unknown parameters (from m_{eff}, ε, α and M_L) to be determined. In coincidence systems, both the TNC response ("Totals") and the PNCC response (genuine or "Reals" coincidence count) are normally available. For example, systems are sometimes configured so that the plutonium mass and multiplication are solved using a known detection efficiency and an assumed α ratio. However, the common "Bohnel single point model" [18,19] upon which this is founded, requires several assumptions that must be fully understood before the technique is implemented.

Benefits of PNCC

- Requires no knowledge of chemical composition, although high (α, n) levels in low atomic number materials (e.g. fluorides) may cause problems due to pile up of accidental coincidences and subsequent poor statistical precision

- Relatively insensitive to background neutron radiation conditions (though due consideration to shielding and location of the assay system is needed) as the coincidence neutron background is usually much smaller than the total neutron background

- Readily available acquisition systems and standard data processing algorithms

- Knowledge required of the waste/stream parameters is generally less than TNC

- Generally more accurate than TNC

- Modest burden of operator training.

Limitations of PNCC

- Requires knowledge of isotopic composition

- Other species that emit coincident neutrons can cause a potential overestimate of plutonium content

- Poorer precision than TNC – requires higher efficiency systems to achieve comparable precision to TNC (more detectors)

- Requires knowledge of and correction for leakage multiplication

- Higher sensitivity to changes in efficiency due to neutron absorbers/moderators or variation of the position of the plutonium within chamber (varies with ε^2). This limitation is minimised in the design of most systems, as well-designed measurement chambers will have flattened "empty" response profiles

- Highly reliant upon the performance of an appropriate system characterisation/calibration

- The high sensitivity chambers used for low level mass assays require substantial amounts of background shielding (this provides an interaction path for cosmic ray interactions, particularly if large amounts of lead are included to shield the system from gamma radiation).

3.1.1.3   Passive Neutron Multiplicity Counting (PNMC)

The neutron multiplicity technique [27,28] is able to differentiate between coincident neutron events involving different numbers of neutrons. The acquisition is used to determine the number of times singles (totals), double coincidences (reals), triple coincidences (triplets) are observed. In practical assay systems counting precision is generally too poor for quantitative assays with higher order multiplicities (greater than three neutrons). However, information regarding the higher orders is still valuable as cosmic ray events may give rise to very high multiplicity bursts and this technique provides an excellent method of filtering them out.

PNMC permits three of the unknown parameters (from m_{eff}, ε, α and M_L) to be determined, requiring only one of the parameters to be known (or assumed). Again, the common "Bohnel single point model" [18,19] upon which this is founded, requires several assumptions that must be fully understood before the technique is implemented. Careful interpretation of PNMC results based on this approach is therefore necessary to avoid misleading results.

For safeguards applications, where the sample geometry (small measurement chambers) and material characteristics (little matrix effect) are better controlled, a PNMC technique assuming known detection efficiency is well established, but is more problematic when applied to the assay of waste, where the count rates may be quite low and the distribution of the plutonium within the waste matrix and the associated α value may not be well known.

PNMC can also be used to determine an effective detection efficiency which best fits the assay data for a distribution of sources, but complications can occur. For example, it is possible that the observed signal from a Pu 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.

Recent experience with waste assay has, however, shown that PNMC can give improved performance [29,30] under some conditions. Furthermore, PNMC can become valuable when the range of conditions can be bounded. For example where the plutonium content is known to be low, neutron leakage multiplication M_L, will be negligible.

The Maximum Likelihood approach [31] permits multiplicity analyses to be made when definite values for at least one of the unknown parameters cannot be assumed. Upper and lower bounds are allocated for each parameter with an assumed probability distribution. The multiplicity equations are iterated with varying perturbations applied to each of the terms (within the assumed bounds) until best agreement between predicted and measured data is obtained.

Benefits of PNMC

- Chemical composition, leakage multiplication or system calibration may be considered variables (depending upon the specific analysis approach taken)
- Impure samples such as mixed oxide wastes can be assayed
- Cosmic spallation effects can be rejected
- Can determine 3 of the 4 unknown quantities (ε, M_L, α, m_{eff}) using the three measured quantities (Totals, Doubles and Triples). However, this requires careful

interpretation. In this sense PNMC can, under certain conditions, provide more accurate results than PNCC.

Limitations of PNMC

- Requires knowledge of isotopic composition
- Other species that emit neutrons can cause a potential overestimate of plutonium content
- Sensitive to high neutron backgrounds due to use of totals in the multiplicity algorithms
- More complex data acquisition and data processing techniques
- Lower precision than total or coincident neutron counting – requires very high efficiency and constant efficiency systems to achieve the required performance (many detectors) with good background shielding and long count times
- Even higher sensitivity to changes in efficiency due to the presence of absorbers/moderators or the sample position within the chamber (varies with ε^2 for doubles and ε^3 for triples). In PNMC, the chamber detection efficiency is usually maintained as a variable in the system algorithms
- Still requires knowledge of at least one parameter (i.e. ε, α, or M_L)
- Dead-time corrections have much higher importance and can be an accuracy limiting factor at high count rates
- High burden on the operator with regard to training, as data often requires very careful interpretation. Expert consultation can be beneficial.

3.1.2 Active Neutron Counting Techniques

3.1.2.1 Source-driven Active Neutron Coincidence and Multiplicity Counting (ANCC and ANMC)

Fissions can be induced in fissile nuclides such as ^{239}Pu and ^{235}U by using an external neutron or gamma ray interrogation source. If the interrogation source is random in nature in contrast to the coincidence response of the sample (i.e. an (α, n) source such as ^{241}Am-Li, or a (γ, n) source such as ^{124}Sb-Be) then the coincidence technique is able to discriminate between the fission neutrons and the interrogating radiations [32,33,34]. In practice, the same coincidence or multiplicity electronics as used for PNCC or PNMC systems, is suitable. The use of low energy neutron sources such as ^{241}Am-Li also limits the interrogation response to the fissile isotopes only, as the interrogating neutron energy is below the threshold for fission in isotopes such as ^{238}U.

Active Neutron Coincidence Counting can be extended with multiplicity counting [1] analogous to the passive neutron multiplicity counting to allow Active Neutron Multiplicity Counting (ANMC).

Generally, active neutron chamber designs are very similar to those for PNCCs. In fact, ANCC/ANMC chambers are essentially PNCC/PNMC chambers that have been modified to allow insertion of random neutron sources.

ANCC systems are not generally used for plutonium measurements due to the limited benefits provided. Such systems more typically find use for the measurement of uranium product wastes, or for the radiological characterisation of unirradiated fuel elements or assemblies (fuel collars). Additionally, ANCC measurements do not prove very useful when there is a high passive coincidence signal, such as in high plutonium mass samples or 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.

Benefits of ANCC/ANMC

- Direct measurement of fissile isotopes (as long as there is no interference from fertile nuclides)
- Useful for ^{235}U measurements where the interrogating source type (^{241}Am-Li) permits discrimination between the thermally induced fissions in ^{235}U and the fast fission of ^{238}U
- Long half-life sources (^{241}Am based) may be used
- Can utilise conventional multiplicity acquisition electronics to gain further sample characterisation data
- Burden of operator training is modest for ANCC, since this technique is the simplest type of active neutron counting.

Limitations of ANCC

- Lumps of fissile material can cause underestimation of the true fissile mass, due to self-shielding as a result of the low penetrability of interrogating thermal neutrons
- Accidental coincidence rates may be high from installed neutron sources – increases the statistical uncertainty
- Raised dose to operators from constantly exposed sources, if design does not take this risk into account
- Careful attention to dead-time correction is required in order to avoid uncertainties, especially for the higher order multiplicities
- Difficulty in obtaining purely random neutron sources.

3.1.2.2   Differential Die-Away (DDA)

Differential die-away [35,36,37,38,39] is based upon the build up and decay of a thermal neutron flux in the walls of a moderating assay chamber, following the

injection of a short pulse of fast neutrons. Induced fission events then follow the decay of the interrogating thermal neutron flux over time. Prompt, fast fission neutrons are detected in a set of dedicated fast neutron detector packages (usually ^{3}He counters embedded in a polyethylene jacket), which are rendered insensitive to the thermal neutron flux by external cadmium cladding.

The name "Differential Die-Away" refers to the large difference in die-away times between the chamber cavity (typically of the order of several 100 μs depending upon the materials and the chamber/assay item size) and the fast neutron detector packages (typically of the order of 10 μs). The difference in response between an empty chamber and a chamber with fissile material is directly proportional to the quantity of fissile material within the chamber. Neutron interrogation is typically performed using high intensity 14 MeV neutrons from "D-T" pulsed neutron generators along with a chamber design optimised to quickly thermalise the interrogating neutron flux in order to optimise the fission yield.

The DDA technique is used as a powerful screening tool, capable of detecting fissile material at milligram levels. It is often used for screening the total fissile content of samples, for applications such as criticality control. 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

- Direct measure of fissile isotopes (total fissile mass, the quantity of most interest for nuclear criticality safety measurements)
- Useful for ^{235}U and ^{239}Pu measurements ("total fissile") in mixed streams
- The relative response of the technique to ^{235}U and ^{239}Pu is well matched to their relative importance for criticality control
- Highest sensitivity measurement (milligram quantities of ^{239}Pu or ^{235}U) due to large signal -to-background ratio
- High neutron interrogating flux (the 14 MeV neutron output can be adjusted) permits measurements in high passive backgrounds (e.g. spent fuels with Pu/Cm content)
- Precision can be increased by higher intensity sources
- Large interrogation flux permits good penetration of matrix and measurement of matrix properties
- For fissile masses well above the LLD, the large signal allows sophisticated data processing, e.g. imaging, to reduce geometrical uncertainties
- Can tailor the energy spectrum of the interrogating flux (using materials around the source) for optimised performance in specific sample / matrix types

- Systems can be designed for a wide variety of container types/sizes and waste forms.

Limitations of DDA

- Lumps of fissile material can cause severe underestimation of the true fissile mass [40], due to self-shielding as a result of the low penetrability of the interrogating thermal neutron flux
- Potentially large matrix effects requiring a complex correction technique when applied to waste assay
- Extensive calibration required (usually requiring much more time than for a passive neutron system), depending upon the application
- Cannot discriminate between plutonium and uranium in mixed streams
- Interrogating neutron pulses may paralyse electronics and therefore cannot be used for the Add-a-Source (AAS) matrix correction technique (see Section 4.2.2.1)
- High cost and maintenance burden of pulsed neutron generator based systems
- Complex control systems and algorithms
- Personnel operating the equipment must be trained to a greater degree of competence than for simpler systems. Further, it is advantageous to develop experienced maintenance staff.

3.1.2.3   **Californium Shuffler**

The Californium Shuffler technique [1] uses a high intensity ^{252}Cf neutron source mounted on a source transfer mechanism to interrogate the assay sample. The source is transferred rapidly from a shielded enclosure to irradiate the sample being assayed for a period of typically a few seconds. The neutrons from the source are thermalised in the chamber and produce fissions within any fissile material present. After this period the interrogation source is rapidly returned to its shielded location. Examples of shuffler installations in use at UK sites are given in [41,42].

The time taken to return the source to its shield (typically 0.5 to 1 second) is too long to allow prompt neutron detection (as in DDA) but still provides a useful, although much less abundant, "delayed" neutron signal from the neutron-rich, short half-life fission products formed. The delayed neutrons produced are counted in the TNC mode for a few seconds following the return of the source to its shield in order to determine the response of the system and thereby the amount of fissile material in the chamber.

Shufflers are used for assay applications at higher fissile masses (lower limits of detection of typically fractions of a gram of fissile material) where the high sensitivity of DDA is not needed. The lower limit of detection is inversely proportional to the

^{252}Cf neutron strength. Shuffler chambers are often lined with cadmium for all or part of a measurement to prevent thermalised neutrons from entering the container. This increases the mean energy of the interrogating neutrons and reduces potential self-shielding problems (see 4.3.1.5), allowing more accurate assays at higher masses than is achievable with conventional DDA. However, this increase in accuracy is obtained at the expense of a significant reduction in sensitivity.

Benefits of californium shuffler

- Direct measure of fissile isotopes, but incapable of directly discriminating between them
- Useful for ^{235}U and ^{239}Pu measurements ("total fissile") in mixed streams
- Widely used for measurements of "uranium–only" streams [43]
- High sensitivity measurement particularly for repeated interrogation/measurement cycles (typically tens – hundreds of mg of fissile material) but not as sensitive as DDA
- Large interrogation flux permits good penetration of matrix and measurement of matrix properties
- For fissile masses well above the LLD, the large signal allows sophisticated data processing, e.g. imaging, to reduce geometrical uncertainties
- Capability to "tailor" the energy of the interrogating spectrum for optimised performance in specific sample/matrix types
- Can be designed to measure a wide variety and size of container types and waste forms.

Limitations of californium shuffler

- Affected by self-shielding in lumps of fissile material [44,45,46], slightly less effect than DDA. Using a cadmium liner with a shuffler greatly reduces the effect of self shielding
- Potentially large matrix effects requiring a complex correction technique when applied to waste assay
- Extensive calibration required, depending upon the application (usually requiring much more time than for a passive neutron system) [47]
- Elevated passive background will worsen the detection limit performance (extremely difficult to perform shuffler measurements in the presence of curium)
- Cost and maintenance burden of source transfer and shielding systems
- High costs involved in maintaining and renewing the ^{252}Cf source (due to the relatively short half-life), although this is application specific and can be much lower than an equivalent DDA system
- Dose uptake to operators when replacing ^{252}Cf sources
- Complex control systems and algorithms

- Personnel operating the equipment must be trained to a greater degree of competence than for simpler systems. Further, it is advantageous to develop experienced maintenance staff.

3.1.3 Combined Active-Passive Neutron Counting Techniques

Neutron counting systems are often operated as combined passive plus active systems, allowing both passive and active measurements in a single, combined assay cycle, with no operator intervention required. This allows measurement of both spontaneously fissionable (passive) and fissile (active) isotopes present. The passive response includes contributions principally from the even mass number plutonium isotopes ($^{238, 240, 242}$Pu), and is generally expressed as the ^{240}Pu effective mass. The active response includes contributions principally from the odd mass number fissile isotopes ($^{233, 235}$U, $^{239, 241}$Pu) and is generally expressed as a ^{235}U equivalent or ^{239}Pu equivalent mass. Provided that there is reliable information on the plutonium isotopics, the $^{239, 241}$Pu isotopic masses can be calculated from the measured effective or equivalent mass, allowing the $^{233, 235}$U isotopic masses to be obtained by subtraction from the active assay result. A combination of passive and active assays, analysed in this way, allows a complete measurement of the individual plutonium and uranium isotopic masses.

A further benefit of performing both passive and active assays is that the assay results for both measurements can be compared for consistency. For example, for assay of waste streams containing plutonium, the active and passive plutonium results will be subject to a different (although sometimes related) set of uncertainties and bias. Inconsistency between the passive and active assay results can sometimes be attributed to the presence of self-shielding (giving rise to an underestimate in the active response) or self multiplication (giving rise to an overestimation in the passive response). This can provide valuable diagnostic information.

Neutron assay chambers are commonly designed to incorporate combined passive and active assay functionality. However, because the chamber materials/geometry and detector disposition optimisations are generally different for passive and active assay, the detailed chamber design must be optimised for either the passive or active assay performance. The detailed physics involved in these optimisations are a matter for the instrument designer. Nevertheless, the user should recognise that, as a result of the trade-offs and compromises involved in the design, a chamber optimised for one mode of assay (passive or active) will not necessarily also operate with optimum physics performance for the other mode.

These trade-offs are subtly different for the different types of active assay equipment and depend on the detailed performance requirements for the assay application as a

whole. Instrument designers will take these performance requirements into account when designing combined assay systems.

Although the details of the geometry/detector disposition optimisations are different, the hardware requirements for active and passive neutron counting chambers are very similar. It is often possible to readily convert a passive neutron counting chamber to an active system by merely keeping the same assay chamber materials, geometry and counter array, and adding the further features associated with the source and control systems. For example, a PNCC chamber may be readily converted into an ANCC chamber by slightly modifying the design of the end plugs to allow insertion of the [241]Am-Li sources (and ensuring that there is sufficient shielding present). A PNCC could be converted to a combined PNCC/shuffler system by adding the source and transfer / control mechanisms.

3.2   Gamma Techniques

Gamma-based non-destructive assay instrumentation can be used to quantify those radionuclides that emit gamma-rays. Although the vast majority of radionuclides produce gamma-rays, not all of them produce them in sufficient numbers or of an energy that allows them to be detected outside of the sample container after they have passed through the sample matrix.

Typically, all of the gamma-based techniques that are in common use for non-destructive assay are passive measurements, in that they rely on the fact that the radionuclides of interest produce gamma-rays naturally. Some of them make use of external sources, but these are used simply to measure and correct for the effects of the sample container and the matrix on the gamma-rays that are produced in the sample.

Gamma-emitting waste can be measured either by counting the total number of gamma-rays being produced by a sample or by counting the number of gamma-rays being produced as a function of gamma-ray energy. Non-energy specific, gross gamma counting instrumentation can be used to measure material where only a single gamma-emitting radionuclide (or a predictable and simple mixture of radionuclides) is present. Techniques that measure and make use of the energy spectrum of the gamma-rays being produced, so-called spectrometric methods, can be further subdivided into Low Resolution Gamma Spectrometry (LRGS) and High Resolution Gamma Spectrometry (HRGS).

The choice of technique depends on the application for which it is to be used. As a rule of thumb, the less that is known (or can be assumed) about the material that is to be measured, the more complex and sophisticated the measurement system

that is required. Depending upon the detector technology that is used, the degree to which the energies of different gamma-rays can be resolved determines the application for which a particular technique can be used. In some applications the techniques may be used in combination (e.g. with an instrument using one technique being used to pre-sort material for measurement by an instrument that uses a different technique).

The following sections describe the different instrument types that are in common use.

3.2.1 Gross Gamma Counting

Instrumentation using the gross gamma counting technique typically consists of an enclosure with walls made from a plastic scintillator material into which the sample is placed for measurement. Gamma-rays emitted from the sample interact in the scintillator material forming the walls of the enclosure where the pulse of light produced by each event detected is measured by a photomultiplier tube (PMT). The total count rate (of pulses above a certain energy threshold) produced in all of the walls of the enclosure is then used to determine the total activity of gamma-emitting radionuclides present within the sample. Since there is no energy information (or energy information is present but is not used), the calibration of this type of device relies on the mix of radionuclides (sometimes referred to as a fingerprint or isotopic vector) being reasonably constant and any attenuation of the gamma-rays being produced in the sample also being similar for all of the material measured. Its main advantage is its combination of high intrinsic detection efficiency and high sample-detector solid angle. However, the sensitivity of the technique is limited by its lack of energy resolution so that events cannot be unambiguously separated from the background.

The high detection efficiency is gained by sacrificing confirmation of the radionuclide fingerprint, which has to be assumed; if the sample differs significantly from what is expected, the result (both activity and attribution) will be incorrect.

Another example of a detector type that may be used for gross gamma counting is a Geiger-Muller tube.

Benefits of Gross Gamma Counting

- Plastic scintillator is inexpensive and can be shaped e.g. to form the walls of a measurement chamber
- High detection efficiency counters can be built
- Less complex and faster counting electronics can be used

- Low maintenance
- No special conditions (e.g. liquid nitrogen cooling) required to operate the detectors
- Low burden of operator training.

Limitations of Gross Gamma Counting

- No (or limited) energy specific measurement possible and so the mix of radionuclides present must already be known
- Samples with a different radionuclide mix are liable to incorrect interpretation.
- Unexpected radionuclides not resolvable – therefore they cannot be identified (although the gamma-rays they produce may be counted and mis-attributed)
- Effect of gamma-ray attenuation in the sample matrix must be taken into account in the sample calibration (to a greater extent than for other methods)
- Output of counter can be affected by temperature instability, especially for low energy gamma rays
- Lack of specific background discrimination
- Background shielding is often structurally heavy.

Note that other, energy resolving, types of detector (e.g. sodium iodide, as described in the following section) may be operated in a gross gamma counting mode.

3.2.2 Low Resolution Gamma Spectrometry (LRGS)

Gamma-rays produced by different radionuclides have different characteristic energies. Therefore the measurement of the energy of each gamma-ray as it is counted by the detection system allows the activity of different radionuclides to be determined separately. The interaction of gamma-rays with the surrounding material, especially that within the sample itself (the matrix), affects the energy spectrum of the gamma-rays that reach the detector and this effect must be taken into account in the instrument calibration. Since the attenuating effect of the surrounding material depends upon the energy of the gamma-ray, knowledge of the gamma-ray energy can be used to good effect when correcting for matrix attenuation.

A common example of a LRGS system is a thallium-activated sodium iodide, NaI(Tl), scintillator crystal coupled to a PMT in order to convert the light produced by each incident gamma-ray into an electronic pulse whose amplitude depends upon the energy deposited in the crystal. Sodium iodide detectors typically have an energy resolution (full width at half maximum, FWHM) of ~40-60keV at the ^{137}Cs gamma-

ray energy of 662 keV (see Figure 1), and are suitable for use when the gamma-spectrum is relatively simple, e.g. by virtue of it containing one or a few well-spaced photo-peaks or by its having a broad energy region dominated by one isotope. However, they are cheaper and require less maintenance than their high resolution counterparts (which are described in the next section) and they do not require special operating conditions (e.g. liquid nitrogen cooling). For an equivalent detection volume, LRGS detectors provide a cheaper solution compared with their HRGS counterparts.
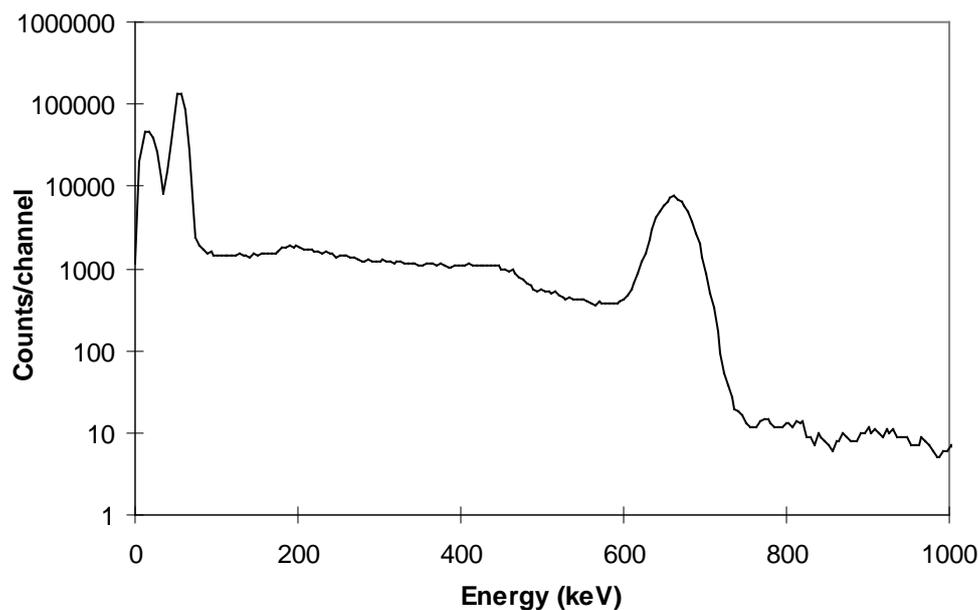
Figure 1   Typical LRGS Spectrum of ^{137}Cs

The choice of detector size and scintillator material used will depend upon the measurement application; larger detectors are used when a high counting efficiency is required and/or when the gamma-rays of interest are of high energy and unlikely to deposit all of their energy in a smaller crystal. Caesium iodide (CsI(Tl)) and bismuth germanate (BGO) are alternative materials that, because of the higher atomic number of their constituent materials, offer a better stopping power than sodium iodide for a given crystal size, although they have the drawback of poorer light transmission. Other factors, such as their robustness to mechanical damage or sensitivity to temperature (sodium iodide crystals will crack if there is a sudden large change in temperature) may also affect the choice of materials used. BGO is also particularly useful for gamma-ray measurements in high ambient neutron backgrounds as the material is relatively insensitive to neutron capture, compared with sodium iodide.

The output from scintillator detectors, especially those using PMTs, is strongly dependent upon temperature. For this reason, a spectrum stabilisation technique is usually employed that alters the detection system gain (usually by controlling the high voltage to the PMT) to compensate for changes in the location of a known photo-peak

that is always present in the spectrum (or present at pre-set times when a source is exposed). A special purpose stabilisation source or radionuclide doped scintillator material may be used for this purpose. More recently, pulsed LEDs have been successfully employed for this purpose where it is impractical to deploy a radioactive source. Irrespective of whether stabilisation techniques are used, it is always good practice to allow scintillation detectors to temperature stabilise before measurements are made. It should be noted that the PMT output can also be affected by magnetic fields (see Section 4.1.4.2).

A 3 inch diameter, 3 inch long, cylindrical NaI(Tl) crystal gives good all round performance in detecting gamma-rays in the energy range 30 keV to 2000 keV. Crystals of different shape and size are also in common use. Large bar-shaped crystals of NaI(Tl) of up to a metre in length are often used where high detection efficiency is required over a large volume (e.g. a 200 litre drum).

Benefits of LRGS

- Allows different gamma-emitting radionuclides to be measured separately
- Energy dependent matrix attenuation correction can be applied to the measurements
- Relatively inexpensive detection system
- High detection efficiency counters can be built
- Detectors and nucleonics are relatively low maintenance
- No special conditions (e.g. liquid nitrogen cooling) required to operate the detectors
- Lends itself to smaller and lightweight systems more easily employed in portable or mobile applications than HRGS systems
- Modest burden of operator training.

Limitations of LRGS

- Limited energy specific measurement means that only simple or predictable gamma emitting radionuclide mixtures can be measured
- More complex mixtures of radionuclides (with overlapping gamma-ray energies) may be measured incorrectly
- Unexpected radionuclides are not always detectable and may be missed or misinterpreted
- Scintillator material may be fragile and may be damaged by mis-handling or large temperature changes
- Detection systems may be affected by electrical noise, thermal drift or magnetic fields.

3.2.3 High Resolution Gamma Spectrometry (HRGS)

As is the case for LRGS, high resolution gamma spectrometry based systems work by measuring the energies of the gamma-rays reaching the detector. The techniques employed for radionuclide identification and quantification, system calibration and correction for matrix attenuation correction are identical or broadly similar to those used in LRGS systems. The improved energy resolution brings two main benefits:

- it allows more complex mixtures of radionuclides to be measured
- it improves the signal to noise ratio of the counts in the photo-peak relative to the ambient Compton continuum.

The most common form of detector technology used for HRGS is based on crystals of high purity germanium (HPGe) semiconductor material. An HPGe detector (usually single crystal p- or n-type coaxial intrinsic germanium) acts as a reverse biased diode which, for all practical purposes, does not produce any current when a high voltage bias is applied to it. A pulse of charge is produced when a gamma-ray interacts with the detector; the amplitude of the pulse produced is proportional to the energy deposited in the detector by the gamma-ray. HPGe detectors typically have energy resolutions (FWHM) of between ~0.6-1.2 keV at 122 keV (^{57}Co) and ~1.7-2.3 keV at 1332 keV (^{60}Co). They are required when the gamma-ray spectrum to be measured is complex, containing many gamma-ray lines from several radionuclides as in the example shown in Figure 2.
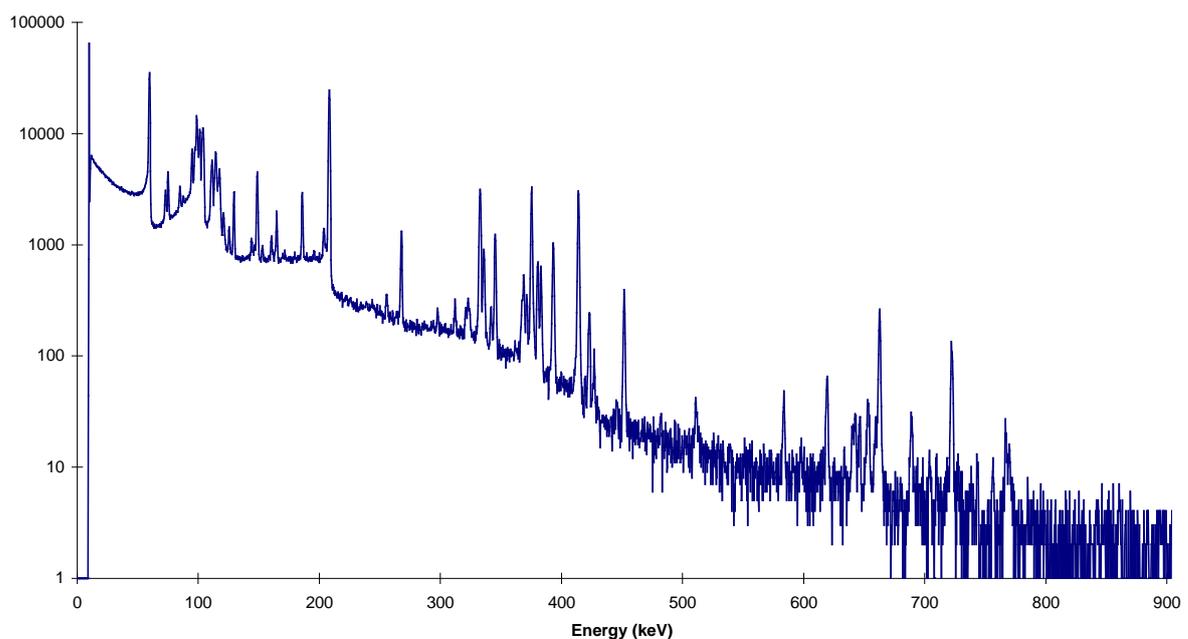
Figure 2   Typical HRGS Spectrum of Pu

The choice of shape and size of the detector crystal for a particular application is very important. Large crystals are used for applications where high detection efficiency is required or where high-energy gamma-rays are to be measured; smaller volume crystals can be used where a lower efficiency is required or where the gamma-rays being produced by the sample are of lower energy. Typical, cylindrical coaxial germanium crystals are 50-90 mm in diameter and 30-110 mm in length. For a 1.33 MeV point source at 25 cm, such crystals will have detection efficiencies, measured according to [48], in the range 15-150% relative to the efficiency for a 3 inch x 3 inch NaI crystal in the same geometry. It is worth noting that two HPGe detectors with different length:diameter ratios may have the same relative efficiency. In a real application, the actual efficiency depends strongly on gamma-ray energy and the geometry of both the crystal and sample. Clearly, relative efficiency is not the only parameter from which to infer detector performance.

With germanium detectors, different electrode geometries can be used to give better performance in different applications. Planar germanium crystals, typically having a thickness less than 20mm, provide more efficient charge collection, thereby giving better energy resolution, albeit the crystal thickness limits their practical application to energies of less than a few hundred keV. More recently, broad energy germanium detectors have become available, combining high resolution comparable to a traditional planar detector at low energy with a larger crystal size that maintains reasonable detection efficiency at higher energies.

Additionally, n-type germanium detectors have a much thinner layer of 'dead' material where the electrode is attached than p-type detectors, and are useful when very low energy gamma-rays (less than 40 keV) need to be measured. N-type germanium is also less sensitive to neutron damage and so is often used for measurements on samples that generate a significant neutron flux.

All HPGe detectors have to be cooled to a temperature below 100 K (-173°C) in order for them to operate. This is because intrinsic germanium has a small semiconductor band gap which means that at higher temperatures electrons are promoted into the conduction band and the material stops behaving as a semiconductor. Traditionally they are cooled using liquid nitrogen. The detector crystal is built into a cryostat assembly consisting of a vacuum insulated enclosure with the crystal connected via a high thermal conductivity anti-microphonic coupling to a cold finger that is immersed in a dewar of liquid nitrogen. Although various dewar and cryostat types and configurations are available that give liquid nitrogen holding times from 1 to 2 hours up to 2 to 3 weeks, the requirement for regular refilling means that such detectors have to be used in applications where they are in accessible locations and close to a reliable supply of liquid nitrogen. Remote filling systems (using a remote storage dewar and thermally insulated filling lines) can however be used where the detector

must be positioned in a location that is not normally manually accessible. The use of electro-mechanical cooling for HPGe detectors has been available for many years with an obvious attraction for applications where regular liquid nitrogen filling is problematic. However it is only in recent years that the technology has matured sufficiently to provide a reliable and cost-effective alternative to liquid nitrogen.

As in the case of LRGS, the output of a HRGS detector and its associated counting electronics is affected by thermal drift (although to a much smaller extent). Spectrum stabilisation techniques may again be employed, if a particular radionuclide is always present, to adjust the pulse amplifier gain or offset in order to lock the position of a particular photo-peak into a specific channel of the measured spectrum.

Benefits of HRGS

- Allows measurement of complex mixtures of gamma-emitters
- May be used to detect unexpected radionuclides or to interpret unknown radionuclide mixtures
- Energy dependent matrix attenuation correction can be applied to the measurements
- Medium detection efficiency counters can be built
- Enables direct measurement of, say, plutonium isotopic and/or uranium enrichment vectors.

Limitations of HRGS

- Detectors, unless electro-mechanically cooled, must be operated at liquid nitrogen temperatures adding an additional overhead requiring a high degree of maintenance
- Detectors and nucleonics may require high levels of maintenance in a plant environment
- HRGS systems are complex and thus may be more susceptible to problems
- Significantly more expensive than a LRGS system of equivalent efficiency
- Cool-down time (typically several hours) from room temperature precludes immediate use unless detector is routinely maintained at operating temperature
- Higher burden of operator training than LRGS systems, because both the hardware and the data interpretation are often considerably more complex.

3.2.4 Intermediate Resolution Gamma Spectrometry

The semiconductor material CdZnTe (CZT) has received considerable interest and development effort in recent years. The main benefits of CZT over HPGe are:

- CZT has a higher effective atomic number giving it a greater probability of collection of all the charge from an individual gamma-ray, and therefore higher detection efficiency for a given detector size
- CZT has a higher band gap energy enabling it to be operated at room temperature.

The drawback of a high band gap is that the energy required to create an ion pair is inherently higher and this translates into a lower number of ion pairs being produced and hence a poorer resolution. However, the resolution is still a factor of about 5 times better than that of the best NaI(Tl) detector, conferring sufficient advantage to make it attractive for the measurement of reasonably complex spectra that NaI(Tl) detectors would be unable to resolve effectively.

Unfortunately, it is now accepted that CZT crystals with volumes greater than $1cm^3$ and with acceptable charge transport characteristics, cannot be grown. However this has not precluded the construction of larger CZT detection volumes using segmented arrays of single crystals, using the following techniques;

- 3-D position-sensitive detector technology. In this method, the detector is electronically segmented into a large number of voxels. Signals from each voxel are corrected for carrier trapping effects. Coincident summing of multi-site interactions is used in this case to maintain the photopeak efficiency of the full-volume detector. This method requires complex signal processing electronics, and requires significant power to operate making it unsuitable for mobile systems.
- The coplanar-grid (CPG) technique. Coplanar grid detectors use a novel electrode structure to make spatially uniform measurements of the charge produced by gamma-ray interactions within the detector, requiring only very simple electronics to operate, making it possible to produce compact, low-power detector systems This results in good pulse-height resolution at high energy, typically better than 15-20 keV (FWHM) for a gamma-ray energy of 662 keV (^{137}Cs) and efficiency of ~8% for a detector with an effective volume of $2cm^3$.

Provided that the limitations associated with detector volume (and hence detection efficiency) do not preclude their use, CZT detectors are suited to applications that demand better resolution than can be provided by LRGS and for which the cooling requirements of HRGS systems render them impractical. This is frequently the case for portable gamma-ray spectrometry or hand held monitoring applications [49].

Even with the modest crystal sizes available today incomplete charge collection results in low energy tailing that complicates the spectral analysis. This is currently being addressed on four fronts:

- Surface contouring and passivation techniques are developing to reduce surface current leakage
- Two-stage Peltier cooling is often built into the detector head to reduce the crystal temperature to circa 30°C below ambient. This improves charge collection and significantly reduces the tailing
- Improved electrode designs are evolving in order to enhance charge collection particularly from deep trapping centres
- Existing HRGS spectral analysis codes are being adapted for use with CZT, particularly for complex incident gamma spectra and high count rates.

Benefits of CZT

- Better energy resolution than NaI(Tl) and other LRGS detector types
- Allows different gamma-emitting radionuclides to be measured separately
- Energy dependent matrix attenuation correction can be applied to the measurements
- Relatively inexpensive detector technology
- Low maintenance
- May be operated at room temperature (or at close to room temperature)
- Better temperature stability than that of NaI(Tl).

Limitations of CZT

- Detectors are small in size - high efficiency detectors are not available
- Energy resolution is not as good as HRGS and spectral artefacts can mean that data analysis (especially of overlapping gamma-ray energies) may be problematic
- Less operational experience in NDA systems compared to LRGS / HRGS systems.

3.2.5 Pulse Processing Electronics

The fundamental principles of the formulation and processing of detector pulses are described in [4]. It should be noted there have been significant advances in the processing and control of detector pulses away from Nuclear Instrumentation Module (NIM) electronics, where individual units for each area of the nucleonics are controlled at individual front panels to stand alone single units containing all

the nucleonics settings controlled by a PC. These PC controlled units have the facility to auto set up the system optimizing the pole zero, pulse shape etc. to manufacturers' set criteria.

3.2.6 Assay Systems

In different types of assay system, different strategies are used in order for the detection system (whether it uses gross gamma counting or spectrometry) to view the entire sample. These range from one shot measurement systems in which a single detector or array of detectors is positioned in order to view the entire sample over the full counting period to scanning systems where the detector or sample are moved relative to one another in order to allow the entire sample to be measured in stages.

3.2.6.1   One Shot

These are mechanically the least complex types of assay system and are typically used in applications where the matrix and activity distribution within the detector's field of view of the sample is known or can be assumed. The simplest type of one-shot system is a single gamma-ray detector which views the entire volume of a sample. An example of a multi-detector one-shot assay approach is a gross gamma counting system consisting of an enclosure into which a sample is placed. The detector-clad walls and door allow the gamma-emissions of the entire sample to be measured simultaneously.

To carry out measurements on large containers an array of detectors may be used, with each detector viewing separate volumes of the sample. Because all of the detectors collect data simultaneously, the time required to measure a sample to a given precision is minimised, however the requirement to cover the entire sample volume may mean that a large number of detectors and independent counting chains is required. In practice a balance is struck between system complexity and the spatial resolution afforded by using multiple detectors. Typically such systems use between 3 and 6 detectors to cover the height of a 200 litre drum.

Benefits of One Shot Counting

- Measurement time may be reduced allowing high sample throughput
- No complex scanning mechanisms required; mechanically simple to produce and maintain
- Minimal burden of operator training.

Limitations of One Shot Counting

- For measurement solutions requiring spectrometry there are associated cost and complexity issues
- Requires a priori knowledge / assumptions regarding matrix properties and activity distribution within detector field of view
- If assumptions regarding sample characteristics are incorrect then there is the potential for significant measurement bias.

3.2.6.2 Segmented Gamma Scanner (SGS)

The segmented gamma scanner [50,51,52] is the most common of the scanning type. The SGS is an axial drum scanner that uses one or more collimated HPGe or NaI(Tl) detectors to measure the emission of gamma-rays from different horizontal slices (or segments*)* of the sample, within which the matrix and activity are assumed to be uniform and homogeneous (typically between 4 and 20 segments are used for a 200 litre drum). The detector is treated as being perfectly collimated, that is, its response is uniform over its field of view, and the field of view is considered to be a narrow slice across the sample. The gamma ray attenuation characteristics of each segment may be measured using a transmission source positioned diametrically opposite the detector on the other side of the sample (see the discussion in section 4.2.4 for matrix attenuation correction). During the measurements the detector (and transmission source if present) is scanned vertically past the sample (or the sample is scanned past the detector/transmission source) and the gamma-ray spectrum of the container (with and without the transmission source exposed, if present) is collected while the sample is rotated to reduce bias caused by heterogeneity in the activity distribution and matrix materials.

Usually each of the separate segment measurements is made with the detector/sample at fixed height. On some systems the measurement is performed while the scan is taking place and with the vertical scan speed linked to the turntable rotation speed. This so-called spiral scan method allows the entire vertical surface of the sample to be viewed systematically and is often used during a pre-scan to identify hot-spots of activity.

SGS assays are best suited to low-density matrices but may also be used for high-density matrices provided the assumptions of the SGS analysis remain valid. Usually, for waste assay measurements, this can be assured by the availability of acceptable knowledge about the origin and character of the waste.
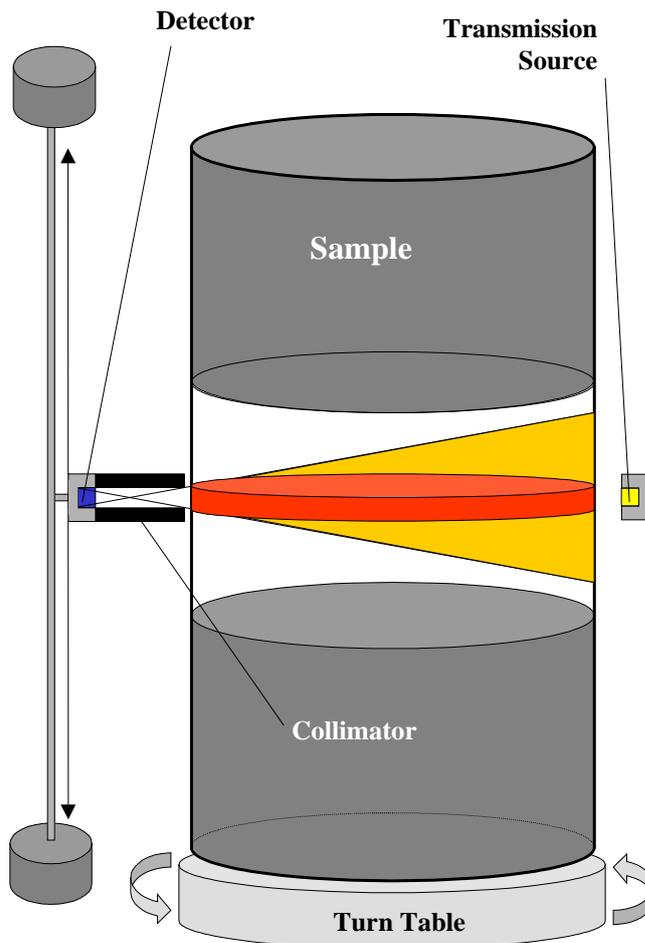
Figure 3   Principal Features of a Segmented Gamma Scanner

Benefits of SGS

- Improved measurement accuracy for samples with vertical heterogeneity
- Only a single detector and/or transmission source required (although throughput may be increased by using multiple detectors)
- Well-established and well-understood 'work horse' for the measurement of nuclear waste in drums.

Limitations of SGS

- Longer measurement times required to provide good sample coverage (although these may be reduced if additional detectors are used)
- Scanning mechanisms of intermediate complexity and high cost required

- Limited capability for detection and correction for the effects of radial heterogeneity within a segment (both of the matrix density and activity distribution)

- Maintenance intensive

- Requirement to purchase and manage a transmission source and transmission source exposure/movement mechanism (if present)

- Higher burden of operator training than one shot systems

- Care is required to avoid microphonic interference induced by vibration (and consequent loss of energy resolution) due to the movement of the detector-scanning platform.

3.2.6.3   Tomographic Gamma Scanner (TGS)

For a conventional SGS, in which the sample is rotated continuously with a fixed field of view per segment for the detector, the data will not support alternatives to the assumption that the sample gamma-activity is uniform within a given segment. Often this assumption is not representative of the material to be measured. The restrictions of the SGS analysis algorithm assumptions can be largely overcome in principle by using tomographic imaging techniques.

The tomographic gamma scanner [1,53] has the same basic components as the SGS, however it has more stringent detector collimation, incremental turntable rotation and an additional mechanical motion that allows the detector to move in the direction transverse to the axis of the sample. This allows the TGS to acquire three-dimensional transmission and emission tomographic projection data from which spatial emission and spatial attenuation maps can be determined. This spatial information provides a detailed correction strategy allowing sample heterogeneity to be taken into account. The penalty of acquiring data with the finer spatial resolution (typically 1600 volume elements for a 200 litre drum) is that data acquisition periods must be extended to achieve a given detection limit. Such a resolution may by necessary when assessing complex matrices in 200 litre drums, improving the information on matrix attenuation and distribution of activity substantially over a typical 8 to 16 segment SGS measurement.

Benefits of TGS

- Typically improved measurement accuracy for samples with vertical and radial heterogeneity

- Only a single detector and/or transmission source required, but throughput may be increased if more detectors are used

- Obtains a 3-D map and 3-D matrix compensation based on the actual source distribution as observed.

Limitations of TGS

- Long measurement times (at least a factor of two greater than for an equivalent SGS)
- Scanning mechanisms add to the mechanical complexity and therefore the maintenance requirements
- Complex data analysis algorithms may give rise to 'phantom' activity if care is not taken in establishing a response matrix for the collimator set-up
- Highly specialised and costly, with significant training burden.

3.3   Calorimetry

Calorimetry is the quantitative measurement of heat. There are a number of types of calorimeter and various applications [54]. The discussion here is limited specifically to radiometric calorimeters, whereby the heat generated by certain radioactive decay processes is used to quantify the amount of radioactivity.

Plutonium and tritium have high enough specific activities (due to their relatively short half lives compared with uranium) to generate sufficient heat for most practical measurement purposes. Essentially all of the energy produced in the decay process is released with the α-particles in the case of plutonium (the energy released by spontaneous fission is an insignificant contributor due to the much longer half life of this decay mode compared with that for α-decay) and β-particles in the case of tritium. These are slowed down over a very short distance within the material so that essentially all the thermal power generated by the decay is deposited within the sample. If the thermal power generated by the sample is measured and the relationship between power generated and mass of material is known, then the amount of material in the sample can be determined.

Calorimetry is potentially the most accurate non-destructive assay technique. The sample's thermal power can be related directly to fundamental physical quantities (i.e. voltage and current) and is immune to the matrix effects that affect neutron and γ-ray techniques. Moreover, corrections are not required for sample inhomogeneity or chemical form. The main disadvantage of calorimetry is the length of time necessary to achieve thermal equilibrium in the sample measurement chamber. Depending on the sample size and matrix, this can take many hours.

3.3.1 Fundamental Operating Principles

The basic components of a calorimeter comprise a measurement chamber (to house the heat-generating sample), a thermal barrier (or thermal semi-conductor) and a surrounding heat sink. Usually the sample is introduced in a closely-fitting container, that is close-coupled to the measurement chamber. The measurement chamber is

made of high thermal conductivity material. The term 'thermal element' collectively describes the complete assembly of these three main components. The thermal barrier is made of a low thermal conductivity material and separates the measurement chamber from the heat sink. It enables a measurable temperature gradient to build up between the measurement chamber and the heat sink. The surrounding heat sink is made of a high thermal conductivity material.

There are two different types of calorimeter, namely single chamber true isothermal calorimeters and heat flow calorimeters (with single and twin chamber variants).

3.3.2 Single Chamber True Isothermal Calorimeter

The single chamber, true isothermal (air-bath) calorimeter is the most common type of instrument currently in use for the measurement of plutonium and tritium. The measurement involves precisely balancing the measured electrical power with the thermal power produced by the radioactive decay of the sample. Electrical power is applied to the surface of the measurement chamber in order to maintain a fixed temperature.
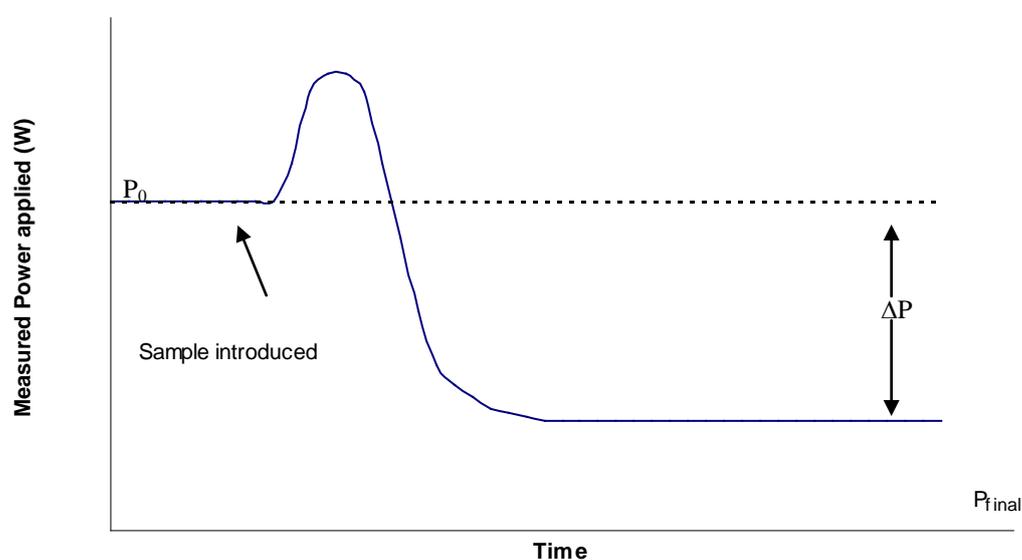
Figure 4   Isothermal Measurement

With reference to Figure 4, with no sample present and the calorimeter at equilibrium, the electrical power applied to keep the inner cylinder at a constant temperature (i.e. the equilibrium value of the chamber and the heat sink) has a value P_o, termed the base power. When a heat-producing material is introduced the applied electrical power is reduced to a value P_{final} in order to balance the increase from the sample. This mode of operation is called the Power Replacement or Power Difference method.

Both the measurement chamber (MC) and the heat sink (HS) are isothermal – they are each maintained at fixed temperatures (T) such that $T_{MC} > T_{HS}$ and there is heat-transfer between them.

In the case of true isothermal calorimeter measurements only, pre-heating of the sample to approximately the internal temperature of the thermal element can be used to reduce the measurement time and improve the equilibrium end-point prediction. To have any benefit several samples must be pre-heated together at the same time. This may not be possible in the case of plutonium due to criticality safety considerations. The disadvantages of pre-heating are that the instrument has a larger foot-print (required to accommodate the pre-heater cylinder and control equipment), the instrument is more expensive, and the pre-heating can increase the time to reach equilibrium if the sample is pre-heated to the wrong temperature.

In order to reduce measurement times isothermal calorimeters can utilise an equilibrium power prediction algorithm in which the decay of the applied electrical power is treated as an exponential function comprising a number of different time constants. After the short-lived components have died away a single exponential term remains. The calorimeter software detects the point at which a single exponential can be used to describe the decay of the applied electrical power. At this point the system fits an exponential function to the power decay data. Using the fit an extrapolation is employed to determine the final equilibrium electrical power, and hence the sample power. This algorithm can therefore be used to reduce the required measurement time. However, where measurement time is not a major consideration, or in the case of lumped and heterogeneous samples, operators are normally recommended to use the true equilibrium end-point rather than the predicted fit.

Benefits of isothermal calorimeters

- Simple technique in principle
- Direct measurement of sample power output gives high assay accuracy
- Low operator training requirement.

Limitations of isothermal calorimeters

- Long assay time required to reach thermal equilibrium (several hours)
- Less portable than other NDA techniques
- Not yet practically proven for routine bulk waste assay

3.3.3 Heat Flow Calorimeter with Isothermal Heat Sink

The Heat Flow calorimeter relies on a measurement of the temperature rise in the measurement chamber with respect to the heat sink after a sample is introduced. The temperature (or voltage) difference is related to the sample power by a calibration curve. The mass of the sample material is then determined, as for isothermal

calorimeters, from the measured power and knowledge of the sample effective specific power.

In the twin chamber variant of the heat flow calorimeter method two identical measurement chambers are connected 'back-to-back'. One chamber contains the sample to be measured while the other chamber is empty. The empty (reference) chamber monitors environmental variations (such as heat flow into or out of the measurement cell, electrical noise and interference, temperature), allowing a correction for heat capacity differences and short-term drift in temperature etc. Such a device is capable of measuring small samples or low power samples. This mode of operation is historically associated with water bath calorimeters (in which the water bath represents the isothermal heat sink).

- Benefits of heat flow calorimeters

- Twin chamber variants allow direct monitoring of environmental drifts in temperature etc, allowing measurements to lowest levels
- Very accurate assay of heat producing material
- Low operator training requirement.

- Limitations of heat flow calorimeters

- More expensive and complex technology
- Separate calibration with heat standard is required, since a direct measurement of temperature (through electrical measurements) is performed
- Long assay time required to reach thermal equilibrium (several hours)
- Less portable than other NDA techniques
- Not yet practically proven for routine bulk waste assay.

3.3.4 Application to plutonium assay

Calorimetry has specific application in the area of nuclear materials accountancy and safeguards. When combined with knowledge of the plutonium isotopic vector (usually determined by high resolution gamma-ray spectrometry), calorimetry provides a convenient, accurate and non-destructive measure of the total plutonium mass of the samples. The technique has advantages over other measurement methods such as PNCC and destructive analysis in that it does not suffer from matrix or neutron multiplication effects and is not biased by inhomogeneity or the presence of moisture in the sample.

For plutonium essentially all of the energy liberated in the decay process is absorbed in the sample or associated container and less than 0.01% of the total decay energy (resulting from neutrons and γ-rays) is lost to the calorimetry measurement process

[55]. The thermal power of plutonium samples generally ranges from below 2 W/kg up to almost 20 W/kg depending on the isotopic vector.

In most practical applications the overall assay accuracy is dominated by the uncertainties associated with the plutonium isotopic vector (including any americium and/or curium contribution).

Calorimetry is not normally applied to plutonium in solution, especially if there are significant quantities of heat generating fission products present.

3.3.5 Application to tritium assay

The mean energy of the β-particles emitted in the decay of tritium is about 6 keV. With a half life of 12.3 years, the effective specific power of tritium is ~324 mW g^{-1}, making calorimetry an appropriate method for measuring tritium. The limiting factors are a combination of power density and volume of the sample.

Calorimeters are often used to measure tritium-contaminated waste. Tritium exists in waste as a variety of species (e.g. as tritiated water, metal tritides or gas adsorbed onto plastic and metallic waste) and the measurement is unaffected by the chemical or physical form. In addition, calorimeters are used to measure tritium product in gaseous and concentrated form.

Calorimetry can be used to directly measure tritium absorbed on uranium in storage/transport containers. The uranium has a negligible effect on the assay due to its much lower specific power output.

3.3.6 Calibration issues

As the calibration of a calorimeter is different by nature to that of a typical neutron or gamma based non-destructive system it is discussed here, separately from the Section 5 description of Characterisation and Calibration. This section covers additional points that are specific to the calibration of calorimeters.

Either a heat standard containing a known amount of (typically) ^{238}Pu or an electric heater with accurately measured voltage and current is normally used for calibration of a calorimeter. Electric heater samples are common and convenient because they can be used to confirm the correct operation of the calorimeter without the need for a radioactive source.

In isothermal calorimeters, an absolute measurement is made since the power of the sample is measured directly and hence there is no requirement for a calibration in this mode of operation. However, in practice a small bias correction is normally necessary (relating the sample power to the measured power, typically by a straight line fit).

The mass of the sample material is determined from the measured power and knowledge of the sample effective specific power (see later discussion).

Heat flow calorimeters measure the heat flow through the calorimeter, which requires a fundamental calibration relating the electrical signal to the sample power. The temperature difference delta T (or voltage change or resistance change) is related to the sample power by a calibration curve (see Figure 5). Again, the mass of the sample material is determined from the power and knowledge of the sample effective specific power.

Individual components, such as thermopiles, are calibrated separately but this calibration does not necessarily take into account the operating environment the component will experience in the final system. Therefore the complete system should be calibrated during commissioning.
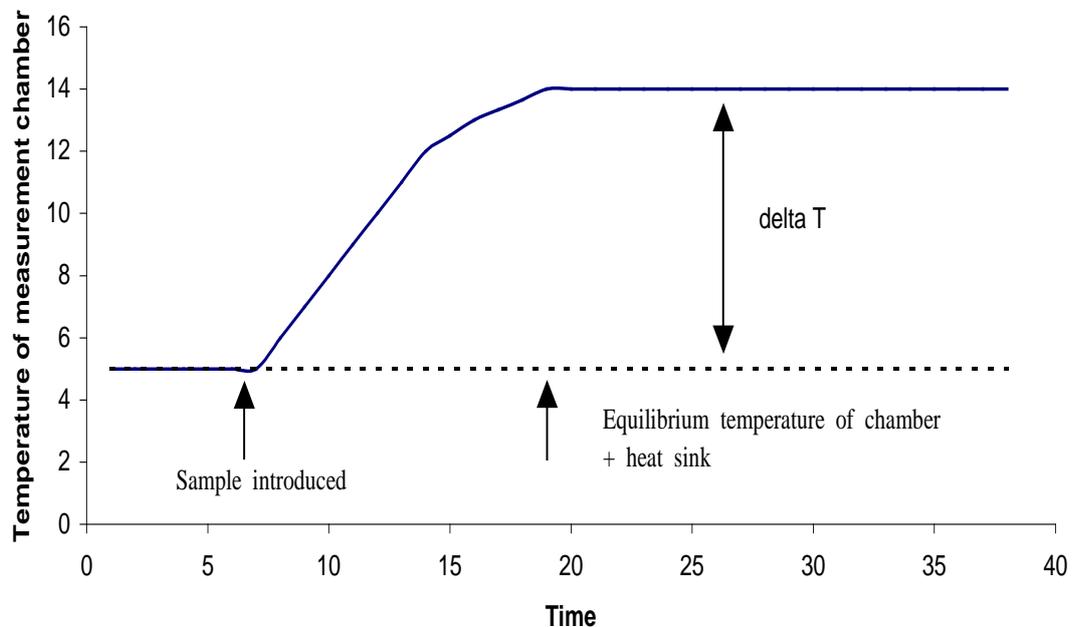
Figure 5   Heat flow measurement scheme

A variety of means are employed to confirm correct operation of a calorimeter and to validate the calibration. Periodically a check should be performed in which a sequence of electrical powers is injected into a measurement region and the applied power compared with the measured power. This verifies correct operation of all the instrumentation, although it does not take into consideration heat distribution errors and thermal leakage associated with samples which are located at different positions in the measurement chamber.

The calibration aspects of the calorimeter are straightforward. The electrical power is usually determined using a digital voltmeter (DVM) for measuring voltage, and a

combination of the DVM and a calibrated precision resistor to measure current. The DVM and calibrated precision resistor should be calibrated with traceability to national standards.

A realistic calibration of the calorimeter usually involves the use of an electrical sample (resistor chains) that accurately represents both the physical geometry and the thermal characteristics of the samples to be measured. In the case of nuclear standards, samples should be made using similar materials to those that will be measured. The electrical calibration sample is connected by wires to the calibration power supply. Errors associated with either heat leaking through the wires that pass into the sample or with heat generation in the wires have to be taken into consideration in the design of the calibration system.

The calibration measurements are conducted using procedures which mirror the actual plutonium or tritium measurement process. The calibration involves applying a known pre-selected power and then measuring the power that the sample evolves in the measurement chamber, as one would do in a normal measurement. The calorimeter can thus be calibrated in this way over its entire dynamic range. Calibration using heat standards such as ^{238}Pu, requires the use of a number of standards to cover the dynamic range of the calorimeter.

3.4 Other Techniques

3.4.1 X-ray Fluorescence

The X-ray fluorescence (XRF) technique uses an intense X-ray or gamma-ray source to excite fluorescence X-rays from the material under study. The fluorescence X-rays are characteristic of a given element, at well-defined energies, and their intensity is a function of the amount of element present. The energies of the X-rays produced are typically in the range 1 -100 keV, depending upon the element to be measured and the particular set of X-rays (e.g. K- or L-shell) generated by the element.

The instrumentation used in XRF measurement systems consists of a suitable source of X-rays or gamma-rays and a detector, positioned in such a way as to optimise the detection of fluorescence X-rays produced in the sample relative to those produced by the excitation source. The detection of fluorescence X-rays is no different from that of gamma-rays of similar energy and the detector types used are identical to those described in Section 3.2 (Gamma Techniques). However, the requirement to differentiate between fluorescence X-rays produced by different elements and to separate them from scattered X-rays or gamma-rays from the source means that high resolution spectrometry is usually employed in most applications of interest to this guide. For example, a planar HPGe detector is typically used in XRF systems that

measure the K X-rays (90 – 110 keV) from uranium and/or plutonium. The pulse processing and spectrum acquisition hardware and the issues affecting its use are as described in Section 3.2.

The nature of the XRF technique means that the fluorescence signal produced by the sample depends upon the energy and intensity of the excitation source, how well the source illuminates the sample and how many of the X-rays produced reach the detection system relative to those scattered from the source. This gives the XRF technique a number of unique features and limits its application:

- reproducible positioning of the source/sample/detector is very important
- a high degree of collimation and shielding is required between source, sample and detector
- high or variable attenuation of the excitation and fluorescence photons within the sample means that the applications for which the XRF technique is best suited are those where the sample does not severely attenuate the X-ray energies of interest (e.g. solutions or thin coatings) or where the bulk material can be characterised by a measurement of a limited surface layer
- absolute calibration is highly problematic and so XRF is usually employed to accurately measure *ratios* of elements, which produce X-rays at similar energies. In this case, all or most of the factors affecting the intensity of the measured signal are the same and so the ratio of the X-ray intensities is directly related to the amount of each element present.

- Typical applications are:

- measurement of the concentration of heavy elements in solution in tanks and pipework (e.g. in a reprocessing plant)
- measurement of the concentration of heavy elements in bulk materials (e.g. uranium/plutonium in MOX fuel pellets or fuel rods).

3.4.2   Laser-Induced Breakdown Spectroscopy (LIBS)

Laser-Induced Breakdown Spectroscopy (LIBS) is a form of optical emission spectroscopy (OES) in which a pulsed laser is used as the excitation source [56,57,58,59,60].   As with other OES techniques such as electric spark and inductively-coupled plasma (ICP), LIBS is strictly an elemental analysis technique although under certain conditions it can be sensitive to different isotopes and can be used to infer the presence of a chemical compound. The basic principle is illustrated in the following schematic diagram.

Laser-Induced Breakdown Spectoscopy
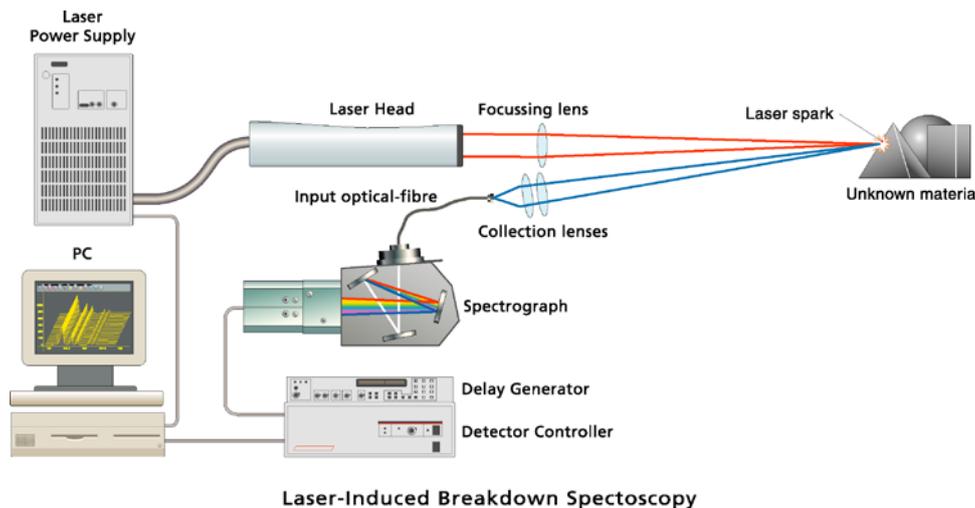
The output of a pulsed, low-energy laser (typically 10s to 100s of mJ) is focussed on to the surface of a sample of material. If the power density is sufficiently high (typically $1 – 10$ GW cm^{-2}), a small amount of material (typically tens of nanograms per laser pulse) is ablated from the surface of the sample and a short-lived but highly luminous microplasma with instantaneous temperatures exceeding 10,000 K is formed at the surface of the sample. At the end of the laser pulse, which is typically 10 nanoseconds, the plasma quickly cools as it expands outwards at supersonic speeds. Some of the light emitted by the plasma may be collected using an optical telescope and fed to a spectrograph to yield a digitised spectrum which is characteristic of the excited atoms and ions within the plasma. It is normal to use a time-gated detector with the spectrograph so that spectra can be recorded some time after the laser pulse (typically 1 microsecond). This time delay allows the plasma to expand and cool sufficiently to reduce the intensity of the background continuum caused by Bremsstrahlung radiation. Without time gating, the recorded spectra will usually be dominated by a broad continuum which obscures the atomic and ionic emission lines needed for analysis.

Benefits of LIBS

- Relatively simple technique which for some applications requires only moderate cost components
- Essentially an all-optical technique and so can be used remotely (100 metres has been demonstrated) using either a telescope system or a fibre-optic system for in-situ elemental characterisation of materials
- Used as a remote technique, LIBS is essentially immune to the effects of ionising radiation – has been used to analyse materials in a radiation field of 2000 Sv / hr (High-Level Waste)
- Can be deployed underwater (eg. in-situ material characterisation in spent-fuel storage ponds)

- Rapid technique – an analysis can be performed in a fraction of a second. LIBS can be a very effective in-situ rapid screening tool for certain applications
- Can be used to analyse the elemental composition of virtually any material and in any physical state (solid, liquid, gas)
- Can be used to remove and/or depth profile surface coatings, due to the ability of the laser to "drill" into these coatings (eg. detection of heavy metals in paint layers)
- Quasi-nondestructive technique – only a minute amount of material is consumed during a LIBS analysis
- Qualitative and, with appropriate calibration, quantitative measurements are possible.
- Good sensitivity (down to ppm level for some elements) and good precision (typically 1 – 5 % RSD)
- Isotope discrimination possible if an optical spectrograph of suitably high wavelength dispersion is used.

Limitations of LIBS

- Requires optical access to the material being analysed
- Surface analysis technique, not a bulk analysis technique
- Leaves a tiny crater on surface of sample (ie. LIBS is not a true NDT technique)
- Quantitative measurements require careful calibration of the LIBS instrument using matrix-matched calibration standards and precise control of the measurement conditions
- Quantitative measurements can be difficult to achieve when conducting remote analysis (due to difficulties in controlling the measurement conditions)
- Requires the use of a Class 4 laser, hence safety implications have to be considered.

Applications

- In-situ and remote elemental characterisation of radioactive materials within hot cells, gloveboxes, etc.
- "Through shield window" deployment of a telescopic Stand-Off LIBS (ST-LIBS) instrument for non-invasive elemental characterisation of materials inside a hot cell
- Fibre-optic LIBS probe for in-situ elemental characterisation of materials located within a hot cell, radwaste silo, nuclear reactor, spent-fuel storage pond, etc.

- Rapid screening of items during decommissioning / post operational cleanout operations
- On-line, real-time elemental characterisation of liquids and slurries.

Measurement Limitations and Factors Affecting Performance

4

IN THIS CHAPTER

- Environmental and Systems Effects
- Matrix and Homogeneity Effects
- Sample specific properties
- Statistical Constraints
- Operational Constraints

4.1   Environmental and Systems Effects

4.1.1   Background Effects

Background measurement and subtraction issues are discussed in detail in Appendix 2, in the context of operational conditions and lower limits of detection. The key technical aspects are also discussed here.

Conventionally, a measurement of background is made and its contribution is automatically subtracted from subsequent measurements. However, problems can arise when the background is changing or is affected by the presence of the sample on the monitor (the sample may shield the detector from the source of the background). Such effects can lead to measurement inaccuracies, but these can be minimised by ensuring that background measurements are carried out regularly and preferably with an (inactive) item that has the same physical characteristics (e.g. density) as the samples being measured in the instrument.

4.1.1.1   Neutron techniques

Neutron radiation arising from sources other than the item being measured may prove problematic. Many instrument systems are located within operational plants and the high penetration of neutrons means that background levels are generally elevated. The effects of background may be minimised through adequate shielding design of the instrument measurement chamber (borated polyethylene, etc.). Additionally, neutron coincidence counting systems are generally much less prone to background radiation fluctuations than total neutron counters, as ambient background neutrons tend to be random in nature.

Gamma radiation (from fission products or other radionuclides) poses a potential problem to neutron systems as high levels of gamma field may cause pile-up of photon-induced pulses that appear to the counting system as neutron events. These effects can be minimised by careful selection of the detector and associated nucleonics as well as the operating high voltage. Typically, detectors will not discriminate between neutrons and piled-up gamma pulses at gamma dose rates in excess of 1mSv/hr and may cause overestimates of the fissile mass, or underestimates due to unforeseen dead-time effects. Such dose rates are not uncommon, particularly in operational and redundant plutonium processing facilities where non-destructive assay equipment is widely used.

The use of large amounts of high atomic number (Z) materials for gamma shielding (or other high Z materials in the container itself) presents additional problems because these will significantly increase the likelihood of generating cosmic ray events that

give rise to high multiplicity bursts of neutrons, thereby increasing the effective neutron coincidence background. Neutron coincidence counting may be configured to perform multiple measurements of short duration and carrying out a statistical analysis to reject spurious bursts but, in this case, multiplicity techniques are advantageous as they will detect a high order burst (greater than 10) and enable direct rejection of these from the measurement. These problems become worse at high altitudes due to the greater abundance of cosmic rays.

The movement of neutron sources past assay systems may unduly affect the results produced due to the variation in background levels. Good shielding and choice of the appropriate technique will minimise this effect but in some cases it may be necessary to designate an "exclusion zone" around the monitor to prevent source movements through this zone while the instrument is in use (background assessment or assay). Real-time data analysis techniques can be applied (in software) to detect and correct for a limited number of fluctuations in background occurring during an assay (the process simply requires the counting interval to be split into a number of sub–intervals, and a statistical analysis performed). These techniques can also provide a warning if there are source movements in the vicinity of the assay chamber.

Even when the neutron background can be accurately measured and corrected for, the effect of the normal statistical fluctuations in the number of background and sample related counts about their respective means, imposes a residual uncertainty which has the effect of setting a lower limit of detection (LLD) or minimum detectable quantity (MDQ), below which the instrument cannot reliably detect or measure a particular radionuclide. The LLD/MDQ depend upon:

- background levels (count rates) relative to sample signal
- background and sample measurement times
- system sensitivity or efficiency (which is usually affected by variations in the sample matrix and source geometry)

4.1.1.2   Gamma techniques

Measurements using gamma-based instrumentation are always made in the presence of a gamma-ray background due to natural sources of radioactivity (e.g. naturally occurring uranium and ^{40}K in building materials and cosmic rays) or arising from interactions in the shielding around the detector. There may also be a significant contribution from other radioactive materials that may be stored nearby or awaiting measurement or from the build-up of contamination within the instrument.

The detector elements of the instrument are traditionally designed with built in shielding (e.g. as part of a collimator that restricts the view of the detector to the sample only) to reduce the level of the measured background but in many cases there

is a practical limit to the amount of shielding that can be provided. The positioning and orientation of the instrument in relation to other sources of background gamma-radiation can play a large part in reducing the level of the measured background.

The LLD/MDQ depend upon:

- background levels relative to sample signal
- interference from other radionuclides which may be present in the sample, especially those emitting higher energy gamma rays
- background and sample measurement times
- gamma-ray energy, specific activity (Bq per gram) and emission yield (gammas per disintegration) of the radionuclide of interest
- full energy peak detection efficiency at this energy
- attenuation effects due to the waste matrix and self-shielding within the source.

4.1.1.3   Calorimetry

The calorimeter cannot distinguish heat produced by radioactive decay from that produced by other mechanisms, such as chemical reactions. This is usually not a problem for two reasons:

- production of heat from long-lived isotopes normally found in nuclear material is constant
- chemical reactions by their very nature tend to have relatively short half lives and the time dependence of the heat output can be used to detect the presence of such reactions.

In the special case of pyrochemical materials where reactive species might be present, it is estimated that the heat of reaction would have a negligible effect on the overall heat measurement. In most cases, the use of hermetically sealed containers excluding air and moisture will mitigate against this problem. Procedural controls can also prevent such effects from adversely affecting measurements.

4.1.2   Radiation Scattering Effects from the Environment

Certain materials that are commonly found in operational plants have widely differing neutronic scattering properties. A measurement system positioned adjacent to a concrete wall will experience a high degree of neutron reflection compared to one positioned in the centre of a large room. This can significantly affect the instrument calibration in the sense that the detection efficiency for neutrons born inside the chamber, can be significantly altered. Consideration should be given to the design and location of assay equipment to minimise such effects. Also a review of system configuration and/or calibration should be performed if the monitor is moved or

surrounding areas of plant modified. This is of particular relevance to portable equipment.

Gamma ray assay systems are less susceptible to effects due to the environmental location of the equipment. The surrounding materials can affect the background radiation levels but this does not generally have a significant effect on the instrument calibration, only on the achievable LLD/MDQ. The composition of any local shielding/collimation in the immediate vicinity of the detector can substantially affect the detector response by way of scattering, and should be carefully considered during the design of the instrument.

Calorimetry-based non-destructive assay equipment is generally insensitive to background radiation.

4.1.3   Measurement Geometry

The particular detector/sample geometry employed in an instrument plays an important part in how well the measurement can be performed. The specification of the assay geometry must be carefully optimised for a given measurement application. Section 7.2.6 also discusses this issue in the context of portable measurement systems.

If containers of different sizes are being assayed, then the geometrical response variations within the different containers will vary. Calibrations need to take this into account so that the assay uncertainty can be determined correctly.

4.1.3.1   Neutron techniques

Neutron counting systems are generally constructed as fixed geometry measurement chambers, within which the assay item is placed. As such, geometric variations in detection efficiency can be minimised through good design with adequately placed detectors, moderator and neutron reflectors. A well-designed measurement chamber may have efficiency variations of only a few percent within the empty well. Turntables may be employed to ensure that the measurement chamber response is symmetrical about the centre of the (rotated) waste item.

In other cases the detectors may be located around an item in-situ, such as for pre-commencement assay of a glovebox for decommissioning. In these situations the efficiency variations may be significant and must be taken into account during assays. This variation in measurement geometry is particularly important for portable or modular equipment where defined geometries cannot always be guaranteed. In these cases the set-up testing and operation should include additional rigour to ensure that all potential uncertainties are included or "calibrated out" of the system.

4.1.3.2 Gamma techniques

A number of viewing/scanning strategies have been discussed along with their benefits and drawbacks. The effect that the detector size and shape has on the measurement in a given application has also been described in Section 3.2. In general, a detector is built into the instrument to view the sample in a particular way and may use shielding and collimation to confine its field of view to the sample or a particular region of the sample. The detector size, position and collimation defines the probability of detection of a gamma-ray of a given energy produced in the sample (the detection efficiency). The instrument design attempts to ensure that the efficiency of detection is reasonably independent of source position or the degree of dispersion within the viewed region of the sample is minimised (as described earlier, a turntable may be used to reduce the effects of positional variability). However, position response is not independent of matrix except for very light matrices.

The instrument is calibrated to determine the response (counts per second versus radionuclide activity) and this calibration normally assumes that the radioactive material is distributed in the sample in a particular way (usually it is assumed that it is uniformly distributed). The assumed distribution should be justified based on any available evidence or operational experience and, in any case, the impact of the assumed distribution being incorrect should be considered within the treatment of the measurement uncertainties. A separate calibration may be required for each measured radionuclide. For a spectrometry based system the calibration may be performed as a function of gamma-ray energy as well as matrix type/density. Calibration takes place by measuring samples of known activity (usually these are sealed sources produced by an accredited facility against a defined standard) in such a way as to simulate the activity distribution of the unknown samples. It can be a complex and time consuming activity to carry out a full set of calibration measurements covering all of the source and matrix combinations and distributions that are typical of real samples. Increasingly, computer modelling is used where it is impractical to cover the complete range of sample conditions, with the model being normalised by reference to a limited set of actual benchmark measurements.

An instrument may use a variable geometry. For example, in order to cope with a range of samples of widely ranging activities, a variable collimator aperture, absorber thickness or a means of varying the sample to detector distance may be implemented to maintain the optimum counting rate and so prevent overloading of the counting system or enable operation at very low count rates so that long counting times are avoided. Each collimator/absorber setting or detector position requires a separate calibration.

The choice of materials used in any detector collimator/shield also plays an important part. Typically high density and high Z materials (e.g. lead or tungsten) are used because of their good shielding properties but greater thicknesses of lower density materials that have better engineering properties (e.g. steel) may also be used. Given their proximity to the detector the quality of the materials used in the collimator/shield is important; any radioactive impurities present may have the effect of increasing the gamma-ray background in the detector to unacceptable levels. The interaction of gamma-rays from the sample in the shield may also produce secondary X-ray emissions, which in a spectrometry system may interfere with lower energy gamma-rays or X-rays that are to be measured. For this reason, collimators/shields are often lined with thin layers of materials (e.g. copper, cadmium or tin) to attenuate these secondary low energy X-rays.

Changes to a detector (e.g. following repair) or any other significant changes to the detector/ sample geometry usually mean that the instrument requires recalibration. To minimise the changes to the detector/collimator/sample geometry following maintenance, the detector mount and collimator should be designed so that its re-assembly is reproducible.

4.1.3.3   Calorimetry

The details of the measurement geometry are far less important for calorimetry than for neutron or gamma based assay systems. As already discussed, the assay accuracy is independent of the contents of waste containers with a particular, fixed geometry. However, if various shape/size containers are being assayed by calorimetry then the geometrical variation in the thermal properties of the chamber can become important with regard to the amount of time required to reach thermal equilibrium. This places demands on the instrument designer to ensure uniformity in the thermal properties. Similarly, the spatial distribution of heat-producing material inside the sample container can become important with respect to the varying thermal properties of the matrix and chamber material in the vicinity.

The size of the sample chamber affects the time required to reach thermal equilibrium since the larger the chamber the further from the chamber wall the heat producing sample is located. The sample size and thermal conductivity are the major factors affecting measurement time. The type of material to be measured, and hence the thermal conductivity, is defined by the particular application and cannot be altered. In principle, the sample size could be optimised to achieve a balance between the conflicting requirements of throughput and measurement accuracy.

4.1.4   Environmental and Electromagnetic Effects

Active control of the environment (air conditioning, temperature regulation, etc) can be used to reduce temperature dependency. Control of humidity within air

conditioning systems is important as levels that are too low can increase the potential for static electricity build-up.

Other noise sources, including microphonic pick-up and earth loop interference, can be prevented by good design procedures, utilising anti-vibration mounts or matting, common point earthing, insulation of detectors from mountings, short cable lengths from detectors to preamplifiers, etc. Testing can be carried out to ensure compliance with the relevant electrical immunity standards, depending on the circumstances of each application.

4.1.4.1   Neutron techniques

Neutron detectors tend to be less prone to temperature fluctuations than other non-destructive assay equipment but care should be taken to ensure that the systems are operating on the plateau area of the high voltage bias curve to minimise any fluctuations in system gain due to temperature shifts.

The high voltage bias supplies used (which can be as high as 4000 V for BF_3 detectors), coupled with high humidity levels can cause operational problems. Complete failure of the system would be rare but discharges in the cables or connectors can appear as neutron events, often in coincidence or at higher multiplicities. Similarly, static electricity discharges can also cause spurious counts to appear in neutron detection systems. In high humidity environments, sensitive electronics boards may be sprayed with a moisture sealant to reduce the possibility of flash-over interference.

Detectors produce very small charges for each neutron event that occurs. This makes them susceptible to induced currents caused by radiofrequency or other sources of electrical noise. Systems should be constructed using screened electronic modules and cables wherever possible to reduce the extent of any noise pickup. Mains-borne spikes or interference can be prevented using mains filters or line interactive uninterruptible power supplies. Due to the larger amount of energy liberated per neutron interaction, neutron pulses in BF_3 detectors are significantly larger than in ^{3}He counters. This means that BF_3 detectors offer better discrimination against gamma and RF noise events.

4.1.4.2   Gamma techniques (including XRF)

The temperature of the operating environment can influence the measurement. It has already been stated that some of the scintillator materials (e.g. NaI) used in LRGS systems may be damaged by sudden changes in temperature; a crack in the scintillator severely affects the light collection probability. Smaller changes in temperature can also have an effect on the output of a photomultiplier tube; they may also affect the gain and the resolution of the amplification and pulse height analysis electronics in

LRGS and HRGS spectrometry systems. Spectrum stabilisation techniques can be used to help minimise the effects of small changes, but these will not be effective if extreme changes are encountered. The best approach is therefore to control the environmental conditions.

The effect of electromagnetic interference may range from a complete loss of the detector output signal down to changes in the pulse height output (due to false interference 'pulses' summing with true detector output pulses). These effects can occur at any stage in the pulse processing chain, but the most vulnerable stage is the detector/preamplifier where the electrical signals are much smaller. It is for this reason that the preamplifier is normally positioned close to the detector and is often built into a single capsule assembly. The introduction of Electromagnetic Compatibility (EMC) standards has reduced the likelihood of problems due to interference, by defining an EMC environment in which the equipment will operate and to which components are tested by the manufacturers of both detection equipment and any electrical equipment that might be the source of such interference.

The output of photomultiplier tubes (used in LRGS detectors) is affected by the presence of strong or variable magnetic fields. Even a change in the orientation of the detector relative to the Earth's magnetic field or relative to the steel framework of a building has been seen to affect detector output. PM tubes should be shielded against magnetic interference wherever possible using mu-metal screens. The positioning of LRGS detectors in the proximity of equipment generating strong magnetic fields, e.g. electrical motors, should be avoided.

4.1.4.3 Calorimetry

For radiometric calorimeters, the thermal properties of the chamber are the single most important consideration. The impact of temperature fluctuations should be considered as part of the calorimeter design with the aim of rendering it insensitive to localised temperature variations.

4.2 Matrix and Homogeneity Effects

4.2.1 Neutron Techniques

Sample characteristics can become important for neutron assay systems and some means for overcoming the effects of these often-unknown parameters must be considered. Representative calibration drums/canisters should be used to investigate the effects of typical matrices during setting-up of the equipment. Characteristics that must be considered include:

- (α, n) contamination

The most commonly encountered (α, n) emitters in plutonium and uranium measurements include beryllium, boron, oxygen and fluorine (either chemically combined or in intimate contact with it). The presence of these elements within the sample will increase the overall neutron emission rate. The (α, n) emission yields from relatively low quantities of plutonium fluoride, for example, are sufficiently high to cause high accidental rates in PNCC systems. This gives rise to poor PNCC precision in the derived reals rate and an increased LLD.

- Moderators and Absorbers

Low Z elements within the sample matrix can moderate the neutron energy spectrum of the emitted neutrons, resulting in a large change in the system response. The effect of these elements can be to increase the detection efficiency of the system at low concentrations but to reduce the system response at higher concentrations due to absorption. Certain elements (including cadmium, gadolinium and boron) and materials (such as polythene, rubber and proprietary neutron shielding material) have a high thermal neutron capture cross-section and can significantly affect the neutron emission from the sample container. If these elements are likely to be present in significant quantities then neutron counting may not be the most applicable measurement technique. For active neutron assay systems, moderators and absorbers also affect the intensity and spectrum of the interrogating neutron flux, adding further complexity to the overall effects of the sample matrix on the assay response. These effects need to be considered either through an appropriate calibration, or a matrix compensation procedure.

- Source Position

Heterogeneity in the matrix and distribution of nuclear material within the item being measured can provide the dominant sources of measurement uncertainty usually encountered by non-destructive assay systems. Many systems are calibrated to assume uniform distribution of material and matrix and therefore any deviations from this assumption may result in large assay bias. The high penetrability of neutrons coupled with compensation techniques (as detailed in Section 4.2.2) helps to overcome such limitations, but matrix effects coupled with source position variability still form the main component of uncertainty. The effect of potential variations in matrix types and source positions/distributions should therefore be evaluated and appropriately accounted for in the determination of the total measurement uncertainty.

4.2.2 Matrix Correction Methods for Neutron Techniques

In order to account for the effects of the often-unknown matrix within the sample a number of compensation/correction techniques have been developed.

4.2.2.1 Add-a-Source (AAS)

The "add-a-source" (sometimes known as a "Matrix Interrogation Source" (MIS)) technique [61] involves the introduction of a well-characterised neutron source

(typically ^{252}Cf) at one or more locations around the waste item. The detection system is first characterised by measuring the system response at each source position with no waste item present, and then determining the effect on system response due to different calibration matrices.

The advantages of this technique include the fact that a measure of absorption within the matrix is obtained automatically, which is not possible using the count rate ratio techniques listed below. This technique will permit the effect of pure absorbers to be corrected for in addition to any moderating materials present, although there is a limit to how much correction is possible. A further technical advantage of AAS over other techniques is that the measurement is sensitive to matrix perturbations on the system die-away time and hence the fraction of coincidence events which is observable within the fixed coincidence gate (the so-called coincidence gate utilisation factor). Additionally, a sample container can be interrogated by the AAS at several positions, allowing a degree of compensation to be applied for spatial inhomogeneities in the matrix composition.

An important benefit of the AAS technique is that it can be used where the passive neutron emissions from the sample are low due to either low specific neutron emission values or due to low fissile mass within the sample. The correction provided is therefore independent of the mass of nuclear material such that a matrix correction is always available, with a similar statistical precision as higher mass assays. This contrasts with the flux probe approach (see below) that relies on the presence of a significant amount of fissile material in order to obtain a viable signal to obtain quantitative matrix compensation. The AAS response can be used to provide a warning flag when the matrix moderation / absorption properties are too severe for the system to function normally. This helps to reduce the risk of unidentified large assay errors.

The add-a-source technique sometimes suffers from a disadvantage in that multiple measurements of the sample and source may need to be performed (for example in the case of large crates etc.). This can extend the assay time for such measurements, but the use of high emission sources may mitigate some of these effects. Also, as the source positions are outside the sample, neutrons from the add-a-source will not have followed the same path through the sample as intrinsic emissions. In certain cases (e.g. point sources in severely inhomogeneous waste drums) this may result in an add-a-source correction factor that is significantly different to the actual matrix effect but this can be addressed through careful calibration.

Limitations of this technique include:
- need to shield the source
- need to account for the source under the radioactive substances regulations

- replacement of source at regular intervals (approximately every 5 –10 years for ^{252}Cf)
- deterioration in AAS counting statistics if sample neutron output is significantly higher than interrogating source.

The additional mechanical complexities in moving and shielding the source and the corresponding costs may not always be warranted.

The AAS technique is widely used in conjunction with PNCC but can also be coupled with DDA and californium shufflers. In this latter case, the californium shuffler source can be used to also perform the AAS functions simultaneously with the main shuffler measurement, so that no additional source, hardware or assay time is required.

4.2.2.2 Flux Probes

Fast neutrons emitted during spontaneous fission events within the sample are moderated within the walls of the neutron measurement chamber and interact within detectors installed in the walls of the chamber. As the moderating properties of the sample increase (due to greater quantities of low atomic number elements) the neutrons become thermalised within the sample rather than the chamber walls, and are detected by a separate set of bare "flux probe detectors" within the chamber itself (separated by an absorber layer such that they are neutronically isolated from the main detectors). The ratio of the total neutron count rate within the flux probe detectors to that within the main bank of detectors gives a measure of the effect of any moderator present and may be used to correct the assay results.

The flux probe technique has advantages over the add-a-source method in that the measurement needs only to be performed once (simultaneously with the sample assay) thereby minimising the overall assay period. Also, since the sample material emits the neutrons, they will pass through the matrix en-route to the detectors (unlike a source positioned outside the sample) and so the response is automatically sensitive to the spatial distribution of plutonium within the sample.

A major disadvantage of the flux probe technique is that the flux probe ratio differs for different types of absorbing matrix materials i.e. a plot of flux probe ratio versus moderator content for polythene will differ from that for PVC (as chlorine acts as an absorber). This means that the system must assume one matrix type (often a worst-case) and vary the amount of worst-case moderator within the algorithms. Additionally, the presence of pure absorbers will not be identified and the system may default to a low fissile mass. As the fissile mass within the sample decreases, the statistical precision on the correction will worsen due to the reduced neutron count

rates. Unlike the add-a-source technique, these conditions are not always identified using flux probes.

As with all matrix correction methods that rely on the use of the inherent radiation from the sample itself, a disadvantage of the flux probe technique is that a viable signal is not obtained for very low masses close to the lower limit of detection. The statistical quality of the signal improves as the quantity of fissile material increases.

Flux probes can be used with PNCC. They are also widely used with DDA and californium shuffler active neutron assays, providing a further compensation for the effects of the matrix on the interrogating flux. Typically in DDA / shuffler systems, bare ^{3}He counters are placed in close contact with the outer surface of the container, and shielded from thermal neutrons that have not interacted in the container by partial cadmium wrapping. In DDA, the time distribution of thermal neutron events can be used to provide additional information on the matrix properties. DDA and AAS measurements may be combined to determine the effect of matrix absorption and moderation respectively in order to derive an overall correction factor for matrix effects [35].

4.2.2.3 Ring Ratio

The ring ratio technique employs two or more concentric rings of detectors positioned so that the inner ring of detectors is under-moderated and the outer ring over-moderated. With no matrix material in the chamber the ratio of counts in the inner ring to that in the outer ring is determined.

As the matrix moderator begins to thermalise the neutrons from the sample the count rate in the inner ring increases while the count rate in the outer ring falls. The change in the ratio of inner ring to outer ring count rates can therefore provide a measure of the thermalisation within the sample matrix. Unlike the flux probe method, the ring ratio method is insensitive to matrix absorbers as both sets of detectors are on the same side of the cadmium absorber liner (if a cadmium liner is employed). The ring ratio technique is often used to correct for the effects of variable sample moisture content [62] for high accuracy measurements (e.g. safeguards measurements of PuO_2 pellets).

4.2.2.4 Multiplicity Technique

The system calibration factor is often treated as a variable within the multiplicity algorithms such that changes in the system detection efficiency caused by moderating or absorbing materials can be varied as the system tries to obtain the best fit to the data available. Again, this technique has limitations when large quantities of neutron absorbers are present or when multiple sources are present. When using PNMC, it should be borne in mind that a single point model is often assumed and this can lead

to difficulties with the data interpretation, particularly for inhomogeneous matrices or plutonium distributions.

4.2.2.5 Imaging Algorithms

An emerging technique that may be useful to provide more refined matrix correction or selection of the actual detection efficiency relies on determining the spatial distribution of sources within the assay item [63,64,65,66]. This source distribution map can be coupled with other matrix correction techniques (e.g. add-a-source) to provide a more refined compensation.

Other imaging techniques that are in wider use often rely on correlations between ratios of count rates in different detectors to determine the sample position(s). Although not providing an "image" in the true sense, these methods can significantly improve assay accuracy by providing a global compensation for inhomogeneities in the plutonium/uranium spatial distribution.

In practice, it is beneficial to couple information on the spatial distribution of fissile material with information on the spatial variability of the matrix composition. In this sense the sample spatial distribution and the matrix homogeneity are connected and hence should be discussed together. This potentially allows a matrix correction appropriate to the matrix in the vicinity of the fissile material to be applied, thus improving the overall assay accuracy. This correction technology is not routinely available; solutions are bespoke to the assay system and the specific application.

4.2.2.6 Real Time Radiography (RTR)

RTR matrix corrections are best suited to gamma measurement systems due to their sensitivity to high atomic number materials, but the technique is also suited to sorting of waste streams to identify and reject prohibited items such as free bulk liquids. RTR may also assist in selecting a calibration factor based upon the actual fill levels within the sample (or drum) rather than assuming that material is present in the whole volume of the sample. In this respect RTR also has potential use for neutron assay systems in identifying partially filled containers for which an alternative calibration may be required.

Developments in image recognition and computer power may enable an increase in the use of RTR coupled with more standard forms of non-destructive assay instrumentation.

4.2.2.7 Operator Selected Calibration

Although not a matrix correction technique as such, it is often feasible to determine the possible combinations of materials that are likely to be present in the assay item and to use this knowledge to determine appropriate alternative calibrations for the expected matrix types. Most facilities have conditions for acceptance of samples or

wastes that provide a list of materials that may be present within the assay item, and many require declaration of the physical contents of the package. Monte Carlo modelling techniques or test measurements with mock-up matrices can be used to assess the effects of different combinations of materials on the calibration, and derive "composite matrix" calibrations if required. In many cases the best calibration is obtained by ensuring a high degree of knowledge of the matrix composition for real waste. This allows an "operator selected calibration" to give an accurate result. If the assumed matrix composition is known to be representative of real waste then this technique is potentially more accurate than any of the technically challenging matrix compensation techniques described above.

The use of operator selected calibration data removes complexity from the assay system hardware and also simplifies algorithms and software. For facilities with well-characterised waste streams there may be little benefit in the added complexity of an automatic matrix compensation technique.

4.2.2.8 Cadmium Liners

A thermal neutron absorbing sample cavity liner (most commonly cadmium) is often used in passive neutron assay chambers. Neutrons thermalised in the polyethylene walls of the chamber that would normally be reflected back towards the sample and cause induced fission events, are absorbed by the cadmium. This reduces the effects of neutron multiplication (which results in an overestimation of the plutonium mass if not corrected for). It should be noted that the introduction of the cadmium liner greatly reduces the overall efficiency and so raises the lower limit of detection. At high fissile material masses where multiplication is a problem, a cadmium liner will benefit the result.

The liner also reduces the low energy gamma-ray flux when measuring Pu samples that contain a high proportion of Am-241, thereby reducing the potential for gamma-ray induced counting events (as discussed in section 3.1.1.1).

4.2.3   Gamma Techniques

The gamma-rays produced by radionuclides present in the sample are scattered and absorbed as they pass through the material of which the sample is made (the matrix). This attenuation of the gamma-ray signal would lead to an underestimation of the activity present if no correction were made for it. The amount of attenuation suffered by gamma-rays as they pass through the matrix depends upon:

- the energy of the gamma-ray
- the density of the matrix
- the matrix type (most importantly the atomic number of elements).

The effect of the matrix also depends on the measurement technique. As gamma-rays pass through the matrix they will interact in different ways. Individual gamma-rays may be completely absorbed in a single interaction or they may be initially scattered, and as they are scattered they lose energy until they are finally absorbed. Spectrometric measurement techniques identify a given radionuclide by the energy of the gamma-ray; only those gamma-rays that have passed through the matrix and have kept all of their original energy contribute to the measurement signal for that radionuclide. Alternatively, a gross gamma detector system may have greater measurement efficiency than a spectrometry based instrument because it includes both scattered and unscattered gamma-rays in its count.

For some samples or radionuclides the effect of the matrix may be so extreme that the gamma-ray signal emanating from the sample is coming only from a layer of material near its surface. This may not be a problem if the sample is sufficiently homogeneous that a measurement of the sample surface represents the entire contents. In this case the calibration of the instrument takes into account that the inner region of the sample is effectively invisible to the detector (often referred to as the infinite thickness method). However, there is always the risk that a particular sample is non-uniform and this would go undetected.

4.2.4   Matrix Correction Methods for Gamma Techniques

In many simple types of gamma-based instrumentation the effect of matrix attenuation on the gamma-ray signal is taken into account in the instrument calibration. Obviously this relies on the degree of attenuation being reasonably constant from one sample to another.

Where such an approach is not possible, one of three alternative techniques may be used to correct for the effects of matrix attenuation.

- The sample weight is used to calculate the mean density of the material in the sample and a correction is applied to the measurement based on the relationship between matrix density and gamma-ray attenuation (this is known as the weight or mean density method). Note that this method may result in significant error if no account is taken of the fill height of the material in the sample container when the density is calculated or if there are large variations in the density of the materials present.
- The difference in attenuation suffered by gamma-rays of different energies from the same radionuclide is used to infer the mean matrix density and used to correct for matrix attenuation (the so called differential attenuation method).
- A transmission source may be used to measure directly the effect of matrix attenuation. An external gamma radiation source (e.g. ^{152}Eu) that produces gamma-rays at one or more energies covering the range of interest is beamed across the

diameter of the sample. The reduction of the count rates in the transmission source peaks is used to calculate the mean matrix density (as a function of gamma-ray energy) in that region of the sample. The transmission factor is determined, allowing for source decay, by measuring the ratios between the transmitted net peak areas with those obtained through an empty sample container. The matrix attenuation correction factor applied to a measured gamma-ray line is obtained from the transmission source data using a semi-empirical relationship.

4.2.5   Calorimetry

The measurement result is unaffected by the matrix. However, the thermal conductivity of the material which separates the heat generating components of the sample from the sample container wall (the sample packaging or matrix) will affect the time required for the flow of heat to reach thermal equilibrium and hence the overall measurement time.

4.2.6   Muon detection

A major component of the cosmic ray flux at sea level is high energy muons which are easily detectable using commercially available plastic scintillators. This highly penetrating form of natural radiation can be used as a tool to assist in the characterization of the waste material.

As the muons pass through any material they can be scattered or absorbed by interactions within the matrix. The resulting attenuation can therefore be used as a measure of the density of the matrix material. The highly penetrating nature of these particles allows application of this technique where the density for the material is too high for traditional RTR and gamma transmission techniques or for density measurements of large volume waste items.

The interactions of the muons and other cosmic ray particles within the matrix or the measurement device itself can also give rise to secondary radiation. These signals will often add to the measured background and therefore affect the detection limits of a system. Background correction techniques that account for the elevated background signal by correlating the induced background component with the incident cosmic rays can be applied to reduce this effect.

4.3   Sample specific properties

The physical and chemical form of the material being assayed may require special consideration within assay equipment design. Neutron techniques are less prone to errors caused by sample density changes, container types, etc. although corrections are often applied to overcome the effects of container material compositions if this

data is available. Calibration of the systems should attempt to include the effects of different container designs explicitly.

4.3.1 Neutron Techniques

4.3.1.1 Chemical form

The chemical composition of the assay material, or its proximity to low Z materials, has a great effect on the neutron emission from the sample. Alpha-emitters close-coupled with certain materials, particularly fluorine, boron or beryllium, can create such a high (α, n) neutron emission component that PNCC techniques can become swamped by accidental coincidence rates and assay bias can occur if these effects are not adequately taken into account by the calibration. In these cases PNMC may be more applicable. Again, the design and calibration of the equipment should consider these effects.

4.3.1.2 Other neutron emitting radionuclides

Other radionuclides in the assay sample may affect the results from non-destructive assay equipment. Nuclides of particular concern to neutron-based assay include californium and curium; the latter being particularly problematic in the higher burn-up fuel materials that are becoming more common. In certain cases the interference from these intense neutron emitters will mask the emission from plutonium (and almost certainly always mask uranium neutron emission) and compensation techniques or fingerprinting based upon the curium content must be employed.

4.3.1.3 Effect of source distribution

Assay chambers are designed to minimise the inherent spatial variability of response, so that the overall assay uncertainty as a result of an unknown spatial distribution of source material, will be minimised. However, for dense moderating matrices the spatial variability of detection efficiency within the sample container can be large, such that material at the central regions is effectively hidden from view. For active neutron systems the spatial variability of the interrogating flux adds further complexity, such that the spatial variability of response can be very large.

Since the topics of matrix and source inhomogeneity are closely related, the various correction techniques have already been generally described in Section 4.2.2.

4.3.1.4 Effect of neutron self-multiplication

Neutrons from either spontaneous fission or (α,n) reactions can induce fissions in a sample, giving rise to what is termed a multiplication event [17,18,19,20,21,22,23,24]. The definition of the sample multiplication, M, is the total number of neutrons that exist in the sample divided by the number of neutrons that were created. M depends on the neutron energy spectrum, the sample composition and the sample density. The effect of neutron self-multiplication is an excess of emitted neutrons above that

calculated from a knowledge of the sample mass, the spontaneous fission rate and α. If uncorrected a self-multiplying sample will therefore produce an assay result that is biased high. M can be estimated using PNCC from the reals to totals ratio if α is known, or it may be solved for using PNMC.

4.3.1.5   Self-shielding effects

Neutron attenuation can occur when the fissile material being measured by active neutron interrogation is present in a form that causes attenuation of the interrogating neutron flux. This so-called self-shielding [40,45] effect can occur, for example, when measuring highly enriched uranium (HEU) that is present in lumps or aggregates in waste material assayed by ANCC, DDA or californium shuffler. The mean free path for interrogating thermal neutrons in HEU metal is only a fraction of a mm, so that only the outer regions of lumps with dimensions of the order of mm or more, are interrogated. This means that severe underestimation can occur, if the calibration does not allow for the presence of lumped fissile material. The potential underestimation increases as the density and enrichment of the fissile material increases.

The effect of self-shielding can be allowed for in the calibration of active neutron assay systems if fissile calibration standards are used that have a shape, size, density, enrichment and chemical form that is typical of the material present in the samples to be measured. The extent of the potential self-shielding problem can be minimised by increasing the mean energy of the interrogating neutron flux so the penetration into lumps of fissile material is improved. Cadmium liners around the sample container are widely used in californium shuffler active neutron assay systems to eliminate the thermal component of the interrogating flux.

As fission cross-sections for ^{239}Pu and ^{235}U (i.e. the typical fissile nuclides) are much lower for a harder (higher energy) flux, the induced fission response is reduced but the potential self-shielding and consequent underestimation of fissile mass in lumps is greatly reduced. By taking two measurements, with and without the cadmium liner in place, a degree of self-shielding compensation can be achieved under certain conditions. An additional benefit of a cadmium liner is for dispersed fissile material where the Cd:no-Cd ratio can be used [41] to provide information on the matrix moderation.

Alternative techniques of self-shielding compensation have been investigated for DDA, using different time windows that correspond to different energies for the interrogating flux. However, all self-shielding compensation techniques break down when even small quantities of moderating material are present in the sample, because the thermal flux is rapidly regenerated.

4.3.2   Gamma Techniques

4.3.2.1   Effect of source distribution

One of the most important contributions to inaccuracy in measurements made by gamma based instrumentation on large samples is the effect of non-uniform source distribution. The magnitude of the effect of source radial non-uniformity is dependent on gamma energy and matrix density. Significant underestimation of gamma activity occurs when an instrument has been calibrated assuming a uniform activity distribution and all the activity is actually present as a single point at the centre of the container. In this case, the gamma-rays from the sample must pass through the maximum amount of the sample matrix and so suffer the maximum attenuation. Depending upon the size of the container and the density of the sample matrix, the underestimation may range from a few percent to several orders of magnitude.

In many applications there are arguments that can be made for why this theoretical worst case is not a realistic possibility; for example the probability of all of the activity being present in a single location may be very small. If the activity is present in the form of even a small number of randomly positioned point sources, the likelihood is that one or more of these point sources will be near the surface of the container. In this situation the underestimation caused by even the most severely attenuating sample matrix reduces significantly (from several orders of magnitude to factors of 2 or 3). For each case this effect has to be evaluated and appropriately accounted for in the determination of the total measurement uncertainty.

4.3.2.2   Self-Shielding (self-attenuation) effects

A special case of gamma-ray attenuation can occur when the radionuclide being measured is present in a form that itself causes severe attenuation of the gamma-ray signal. This self-attenuation effect can occur, for example, when measuring uranium that is present in lumps. The soft 185 keV gamma-ray produced by ^{235}U coupled with the high density of uranium in its metallic or its oxide form means that only 10% of the gamma-ray signal from ^{235}U in a one-gram sphere of uranium metal emerges to be measured.

The effect of self-attenuation can be allowed for in the instrument calibration if the calibration is carried out with standards that have a size and chemical form that is typical of the material present in the samples to be measured. The differential attenuation of gamma-rays of different energies from the same radionuclide can also be used (as in the corresponding matrix attenuation correction method – see Section 4.2.4) if suitable multi-energy gamma-ray emitters are present. However, the technique soon saturates; in effect the lump behaves as if it were infinitely thick so that only a fixed thickness of its outer skin contributes to the emergent gamma-ray intensity. The situation is further complicated if there is a wide range of lump sizes in

the same container. In practice, lump correction algorithms based on differential peak analysis have limited applicability for waste assay applications.

4.3.2.3 Non-gamma emitters / weak gamma emitters

Gamma-based techniques can be used only for measuring radionuclides that emit gamma-rays at a sufficient rate and energy for detection. In some cases, the combination of specific activity (Bq per gram of the isotope) and gamma-ray branching fraction (number of gamma-rays produced per disintegration) associated with a radionuclide of interest, is too low for it to be of practical use in the assay of the radionuclide at the required levels. Some radionuclides may also produce only low energy gamma-rays; low energy gamma-rays (less than 50 keV) are severely attenuated when passing through the sample matrix or container walls (see the discussion in the section on matrix effects) and when large amounts of high density materials are present (e.g. materials of density 0.5 g/cm^3 or more) gamma-rays with energies less than 200 - 300 keV may also have severe difficulty escaping.

When radionuclides cannot be directly measured, the amount present may be inferred by a known relationship with a radionuclide that is measurable. A plant-supplied fingerprint of ratios between all radionuclides that are expected to be present in the sample is then used to calculate the activity of radionuclides that cannot be measured from those that are readily observable.

Note that the art of radionuclide fingerprinting invariably involves destructive sampling and is outside the scope of this guide. Although the generic methods for combining uncertainties (see Section 6) for inferred nuclides can be applied, no guidance can be given regarding the derivation of those components. Users should be cautious when employing supplied generic fingerprints; any fingerprint should be verified against the process for which it is being used.

4.3.3 Calorimetry

The physical nature of the sample and the distribution of heat generating material within it (e.g. metal billet, high density oxide powder, scrap or heterogeneous item with large voidage) will affect the overall measurement time. Moreover, the measurement precision as a function of measurement time will also be affected. Typically, 1 kg of plutonium oxide can be measured successfully in under four hours, while the equivalent weight of pyrochemical salt (containing 100 to 200 g plutonium) can take up to 20 hours to measure. The measurement time is also highly dependent upon sample packing.

Calorimetry is most precise for materials with relatively high plutonium concentrations. Consequently calorimeters are normally used for measuring bulk solids such as plutonium oxide, plutonium metal (billets, buttons) and pyrochemical

materials containing significant quantities of metal 'shot' or species such as plutonium trichloride. Because of the restricted size for optimum performance of the calorimeter sample well, measurement tends to be on the packet scale with a normal upper limit of the order of 30 litres capacity.

4.4   Statistical Constraints

Any non-destructive assay technique comprises a measurement of the ambient background radiation level and an assay of the waste item or sample. In order to maintain the operation of particular plant these measurements have to be performed within a specific time period, dictated by many constraints – a very significant one being plant throughput. Generally, a balance has to be made between the need for plant throughput and the need to obtain measurements with an appropriate precision. This is discussed further in Sections 6, 7, and Appendix 2.

4.5   Operational Constraints

Non-destructive assay systems should be designed with consideration of the use of the equipment in its intended environment, which may be laboratory, or industrial. Assay techniques developed under laboratory conditions are seldom suitable for direct installation into an industrial facility due to the sensitivity of equipment, complexity of operation and setting up, and the difficulty in the interpretation of results. If the system operation is overly complex, the plant operator may not be able to use the system in the intended manner or produce consistent results.

Reliability and maintainability of the equipment is another consideration to meet throughput and plant production targets. Instrument designs should consider the reliability and future availability of components used within the instrument and also the ease of repair or replacement of such components and ease of testing. This is covered in detail in Section 7.

Tamper prevention and data back-up devices should be considered to prevent the accidental or malicious corruption of system parameters and measurement data.

Characterisation and Calibration

5

IN THIS CHAPTER

- Calibration Requirements
- Calibration Procedure
- Reference Standards
- Working Standards
- Uncertainty

Calibration is the link between the result of a measurement and a reference standard. It is an essential part of any radiometric measurement and is achieved by deriving the quantitative relationship between the response of the measurement system and a given quantity of the substance, in a representative form and containment. The term "calibration" is often abused and taken to have different meanings. Quite often, measurements taken to verify correct operation or to confirm that existing measurement relationships are correct are referred to as calibration. These other measurements are more appropriately termed "check measurements", "calibration checks" or "verification of calibration" (discussed in Sections 7.3 and 7.4), and should not in themselves be confused with calibration. However, the term calibration will continue to be used routinely and it is therefore important that the scope and meaning of such activities are clearly defined prior to the activity taking place.

It is very important to recognise the difference between "calibration" and "calibration check" measurements. The term "calibration" refers to determination of the primary relationship between the measured parameter and the quantity of interest (mass or activity). In contrast, the term "calibration check" refers to measurements performed to demonstrate continued validity of an existing calibration.

The terms "calibration" and "characterisation" are both often used to describe the process of determining the response of a non-destructive assay system, which is the ratio of the observed reading of an instrument to the true value of the quantity producing that reading. To this end reference standards are commonly used in a measurement system to establish the relationship between the basic instrument response and the attribute(s) of interest.

Radiometric characterisation is the process of determining the fundamental response of a detector to a defined source (which may be a reference standard), and may include the effect of intervening matrix and/or shielding materials. As necessary, a series of individual characterisations can be combined to provide an overall characterised response. Such an approach would be conducted for example to formulate the response of a particular detector to a shielded, extended (area or volume) source given the availability of only an effective point source for the characterisation process.

Calibration is the procedure to establish the quantitative relationship between the response of an instrument and the quantity to be measured. It is essentially the outcome of one or more (appropriately combined) experimental characterisations to establish the required response factor. In that circumstance, where the calibration is the result of a single characterisation, then the source used is entirely representative of the assay geometry (i.e. source-detector arrangement) of interest and the associated overall uncertainty in the calibration can be derived from the uncertainties in individual source attributes. This particular circumstance is very much the norm in destructive analysis (DA) where samples are prepared identically to the source used

for the single characterisation (i.e. calibration) needed to allow a straightforward ratio approach to the determination of activity.

In a large number of assay applications – particularly those where there is no reference standard counterpart to the source under assay (i.e. the actual source geometry itself is unknown but can be spatially bounded from other evidence) - the approach to calibration is through the aggregation of a number of individual characterisations for example using measurements with a point source or standard placed at different positions in turn. In such circumstances the total assay uncertainty, including that from the calibration process, is usually dominated by factors related to the range of response corrections (essentially those arising from matrix and geometrical effects) needed to synthesise the required response and to acknowledge that the source geometry can only be bounded. The total assay uncertainty is therefore largely independent of the reference standard; in such cases the requirement to use or possess well-characterised reference standard(s) for calibration purposes may not bring about any distinct improvement in the resultant assay uncertainty. At this point neither is it detrimental to interchange the terms "calibration" and "characterisation"; they have the same meaning.

Notwithstanding the differences between calibration and characterisation activities, as described above, the term "calibration" is often used to denote a variety of activities. Furthermore, calibration and/or characterisation activities may be performed at various stages in the setting to work of an assay system or methodology. Section 7 describes the various stages in the setting to work of a new system. The term "commissioning" is often used to denote the essential work program that is carried out so that the system can be authorised for routine operation with real samples. Calibration and characterisation will form a key part of this work.

It is common practice for characterisation measurements to be performed using inactive, surrogate waste drums. This may be performed at the supplier's factory prior to shipping the system to the end-use facility and this may be preferable because logistically it may be simpler to perform such measurements at the factory. This is sufficient to characterise the system, noting that aspects such as the matrix variability and positional sensitivity are functions of the physics design of the system and the detector geometry and as such will not change when the system is moved to the final location. Once the system has been installed at the end-use facility, subtle changes may have occurred and for this reason calibration is normally conducted so that the calibration parameters reflect the final environmental conditions. This "final calibration" may take the form of measuring the final system detection efficiency, with reference to a limited number of measurements with a relevant transfer standard. The system parameters can then be adjusted using correction factors.

5.1 Calibration Requirements

The initial characterisation and calibration of a NDA instrument should be performed only after correct operation has been guaranteed. This may not be practical for those systems where calibration is carried out by the manufacturer and confirmed during the setting-to-work tests. In the case of the initial calibration, this will be assured through the setting-to-work tests (see Section 7.2.2). An assessment of the anticipated uncertainty should be carried out beforehand.

For fixed, installed systems, the calibration should normally be checked or repeated at regular intervals of defined and justified length according to an established programme. Such an approach is often convenient with regard to scheduling of activities to ensure minimum disruption to normal operations. A calibration check (performed, for example, annually) is often performed on the same occasion (immediately following) as the planned maintenance. It is good practice to carry out such calibration check measurement regularly, for ease of demonstration of continued validity of the existing calibration. However, it is important to note that the important issue is ensuring that control and calibration check procedures are in place to verify continued validity of the system calibration. For installed systems which are used infrequently or mobile assay systems which are deployed only when necessary for irregular campaigns of measurements, "test before first use" calibration or calibration check measurements are often performed instead and this is considered wholly appropriate.

Calibration check measurements are required only to verify continued validity of the existing full system calibration. For example, measurement with a single working standard at the centre of a PNCC measurement cavity would suffice to check whether the system detection efficiency is the same as that at the time of the original, full system calibration. Provided that no other conditions can have changed (such as the detector geometry and chamber wall construction) then the matrix and geometry effects cannot have changed so there is no need to conduct measurements with sources at different positions in different matrices. In fact, it is often (but not always) the case that the regular QA check measurements will fulfil this purpose.

The requirement to recalibrate a system should be assessed by a competent technical authority on a case by case basis. This assessment should consider the changes to the system, end-use application range and/or environmental conditions, in order to decide whether recalibration is necessary and if appropriate, what extent of recalibration is necessary.

Consider the example of moving a PNCC system to a new location in which at least one of the chamber walls would then be located adjacent to a concrete wall. In this

case the neutron reflection from the concrete wall could have a significant effect on the system's detection efficiency (including the relative responses of the different detector banks) and therefore a complete recalibration and characterisation is likely to be necessary. The extent of the effort required may include a new determination of the relationship between the system count rate and Pu mass. This may include self-multiplication because of the changed albedo reflection effect, therefore necessitating measurements with different mass Pu standards or an assessment with the aid of Monte Carlo modelling or alternative means. Further, if the reflection introduces a degree of asymmetry to the system response, it may be necessary to perform new calibration measurements to determine the variation of response with Pu position inside the chamber. The reflection may also introduce a perturbation to the effects of variable waste matrices, possibly necessitating additional test measurements or calculations.

A similar example would be consideration of the validity of continued operation of a PNCC system if one or several of the detector counting channels had failed or there was loss of ^{3}He gas pressure by some other mechanism (such a condition might be diagnosed by observation of a deterioration of performance on the quality control chart). A new efficiency calibration is likely to be necessary and the asymmetry introduced may affect the positional response. However, the variability of response with matrix type may not be affected significantly, so that additional matrix measurements are not necessary. Similarly, gradual deterioration may be observed (in the resolution of a reference peak) in the control chart readings for a HRGS detector, due to growth of the dead layer (attention to the maintenance regime can minimise the risks of this). A technical authority may deem that a new calibration of the detector is necessary.

Similar consideration should be given to changes in the operational envelope, for example the proposed operation of a system with a new waste drum matrix type. It may be that the existing calibration is still valid, because the matrix compensation techniques will function adequately with the new matrix type. Alternatively, new calibration measurements may be required and the results of these would be critical in deciding whether the existing system can be authorised for operation with the new matrix type, when considerations such as maintaining the required detection limit and total measurement uncertainty performance are taken into account.

Other changes may occur with regard to the environmental conditions, for example a changing plant operations schedule might adversely affect the gamma background in the vicinity of a segmented gamma scanner. In this case the calibration is unaffected because all that is affected is the statistical quality of the results and detection limits; there is no change to the relationship between the detected "full energy" peak count rate and the nuclide activity. However, a similar level of assessment is recommended

as for the above examples, because it may still be very important to implement control measures to avoid the detection limits being elevated to unacceptably high levels and to avoid undue bias in drum activity results.

Equipment should be checked or re-calibrated using a working standard (see Section 5.4) following any adjustment, modification, or any change in the surroundings or in the operating envelope, for which it has been shown (following an appropriate assessment as described above) to have a potentially significant effect on the calibration. It is recommended that checks be made routinely to ensure that conditions have not changed significantly. For example, continuous recording of system backgrounds and quality control measurement results can provide a useful early warning of changing conditions or impending system failure. A record should be maintained of the results of stability checks in such a way as to enable the ready identification of trends away from stability (see Section 7.3.2). Re-calibration of the equipment should be carried out only following a visual inspection of the system and rigorous functional testing of the system operation (see Section 7.4.2).

Implicit in the above discussion is the assumption that modification control measures are implemented (see Section 9). Recalibration will involve changes to some or all of the system/software parameters, and should be considered as a system change. As such, the modification control records should include records of recalibrations and the assessments made to determine when they should be performed.

The calibration and calibration check measurements do not always require a high level of specialist physics expertise; the calibration measurements can often be performed by field analysts or operators following defined procedures. The resultant data is then analysed/checked by specialist physics staff and/or technical authority. The underlying physics of the calibration method must be sound. The calibration should, where possible, be covered by a written procedure describing the method employed, reference standards, other equipment used and the quantification and treatment of sources of uncertainty. The procedure should cover the specified operating envelope of the equipment. The specialist technical authority should check and approve the calibration and this is considered a necessary step in authorising the system for routine operation.

Where practicable sources that are traceable to national standards should be used; note that the traceable parameter may not be the radiation emission. The uncertainty of the standard should be commensurate with the application. Note that, for simplicity, the term reference standard is used in the following discussion irrespective of the extent of its traceability.

5.2   Calibration Procedure

Guidance for the calibration of non-destructive assay equipment is available from a number of sources such as [67] and [68]. In essence two calibration techniques may be used. The first is the recording of the reading of the equipment for an appropriate reference standard. The other (analytical) technique is the calculation of the response for a standard from known or calculated values of the relevant parameters that comprise the theoretical or semi-empirical formula describing the response for that material. In practice, as standards rarely match the physical/chemical form of the substance to be assayed, calibration is usually a combination of both techniques. The response of the equipment to a standard is measured and this is related to the response for the quantity to be measured by calculating the relevant conversion or correction factors.

5.2.1   Calibration Function

The reading (R) may be represented by a mathematical function of the form:

$$R \; = f \, (\, A, \, k_1, \, k_2, \, \ldots\ldots k_{i.}\ldots \, k_n \,) \; + \; B \tag{1}$$

$$\begin{aligned}
\text{where } A = \quad & \text{activity or mass being determined,} \\
k_i = \quad & \text{parameter with influence on response, independent of } A, \\
B = \quad & \text{response to background.}
\end{aligned}$$

The calibration of an instrument should span the whole range of A and the dynamic ranges of all the parameters k_i.

The parameters influencing calibration could include the following examples in the case of fixed installed assay systems:

k1   position of the material within its container (geometry)
k2   dimensions, materials and construction of the container (configuration)
k3   detector geometry relative to the container
k4   physical characteristics of the matrix (material type, density)
k5   physical characteristics of the fissile material
k6   chemical composition of the matrix
k7   chemical composition of the fissile material
k8   self-shielding effects
k9   self-multiplication effects
k10   environmental conditions (temperature, humidity, etc)

For mobile, reconfigurable assay equipment, other significant factors are:

k11   geometry-specific efficiency calibration
k12   impact of local background at the time / location of the measurements

In some situations the experimental determination of instrument response across the entire range of k_i values may be impractical or impossible to achieve. In such situations the use of Monte Carlo or other modelling techniques may offer an acceptable alternative provided that the approach is validated by sufficient benchmarking of the model, performed against achievable experimental measurements. Calculated detector calibration parameters are becoming more commonly accepted, for example there are software-based modelling codes which construct a geometrical model of the sample – detector geometry and calculate the detection efficiency for a given source geometry, thereby removing the need for radioisotope test sources. As such, a detector may be supplied which is pre-characterised with the aid of computer models which are suitably benchmarked against reference measurements. This characterisation then allows a mathematical model to be developed for an item to be measured (for example a waste drum), thus developing the system calibration. Further discussion on the topic of modelling tools, is given later (see Section 7.2.5).

In practice, the k_i values are often inter-related, for example the positional response variability depends on the waste drum matrix. Furthermore, the background count rate, B, may be highly dependent upon the k_i values, for example the shielding afforded by the waste drum matrix means that the gamma background is dependent upon the drum mass and appropriate compensation may be necessary to avoid under-reporting the measured activity (which would be the case if a background was measured with an empty drum).

In many cases, for any given set of values of the k_i-parameters, the background-corrected reading can be confidently expected to be proportional to the quantity *A* being measured and to be zero for a zero amount of *A*. However this is not always the case. Self-shielding can mean that the curve "flattens off" at higher fissile masses whilst self-multiplication can have the opposite effect with the response increasing more rapidly as the mass increases. The linearity may be investigated by measuring the response for at least four suitable values of *A*; one value should be greater than or equal to the maximum that is expected to be encountered in practice, and another should be less than or equal to the minimum; however, it is impractical to attempt to measure below the LLD. A weighted least squares fit is then made to the data, normally using a quadratic relationship. The inverse squares of the random uncertainties are used for the weighting. If the second order term is insignificant a linear relationship may be assumed. If the fit indicates a significant constant term (an offset) this should be investigated and, where warranted, be included in the response determination. It should be noted that it is not always possible, due to a lack of appropriate standards, to perform calibration measurements over the desired range. However, it may be sufficient to use fewer standards, or even a single standard,

provided that the method is substantiated with the aid of separate assessment (often involving additional measurements/modelling). Central to these considerations are the properties of the calibration standards in relation to the real fissile material/activity to be assayed. For example, if it is known or assumed that any fissile material present within a waste package will be dispersed throughout the volume such that self-shielding and self-multiplication are negligible, then calibration with a single standard (carefully constructed such that the self-shielding and self-multiplication are negligible) is likely to be valid. The assumption of a linear response function must then hold because of these assumptions and it would not be appropriate to use additional, available higher mass standards for which the physical and chemical properties are different (because the self-shielding and self-multiplication properties are likely to be different). On the other hand, calibration of a system measuring hold-up of liquor or solid residues in a process vessel, may be critically dependent upon the use of mass standards covering the complete dynamic range. As the mass or activity of the liquor increases, the volume and shape of the active liquor are constrained by the shape and size of the vessel, in a systematic manner which will determine the self-attenuation and / or self-multiplication.

The reading may then be expressed in the form:

$$R \; = \; K A \; f \, (\, k_1, \, k_2, \dots \, k_i \, \dots \, k_n \,) \; + \; B \tag{2}$$

where K is the calibration constant or response for a reference set of values of parameters k_i. For this set, the function $f(k_i)$ would usually be set to unity. The constant K and its uncertainty are derived from the weighted least squares fit.

The response should be measured for the anticipated ranges of values of the parameters k_i. For each parameter, a mathematical relationship with response may be assumed provided that the function is known to be monotonic for the range of values over which it is applied. Expert guidance should be sought on which type of relationship is suitable; polynomial or power law formulae are commonly used. The number of terms should be realistic and not arbitrarily large.

The number of measured responses (for a given k_i) should be at least two greater than the number of terms in the formula. One value of the parameter being varied should be greater than or equal to the maximum that will be encountered in practice and another should be less than or equal to the minimum. The fit should yield values of the coefficients of all terms in the formula and their uncertainties. Using the derived coefficients to extrapolate beyond the minimum and maximum values is not recommended.

For some systems, calibration may be developed based on databases or "look-up-tables" which define the functional relationships $f(k_i)$. In some cases, the calibrations

are handled by system software which uses internal algorithms such as "least squares fit" techniques to establish the nearest match k_i values so that the calibration co-efficient can be matched from the database or look-up table. Interpolation is sometimes performed when the measured k_i value for a measured sample (for example when it is based on a matrix compensation algorithm), lies between the corresponding values for two calibration points. In other cases, the system application software may be configured with calibrations for several pre-defined matrices. This approach is often better and more robust for simple applications with well segregated, discrete waste forms, so that the operator simply selects the waste type from a menu screen. In these cases, it is necessary to consider the implications of variabilities in the actual matrix properties for real waste, from those of the calibration matrices, as part of the assessment of the total measurement uncertainty.

5.2.2 Positional Dependency

The positional dependency is usually accounted for by assessment of the response at a set of discrete points that provide good coverage of the measurement volume. This approach usually offers a more practical solution to the problem of simulating a dispersed source, due to lack of availability of dispersed reference standards. However, for specific applications, and where appropriate dispersed standards are available, the latter approach is reasonable. The response at these points may be determined through carefully-staged, point-like source calibration/characterisation measurements, modelling, or a combination of both, applying corrections for self-shielding, etc. A smooth curve that fits these points is usually applied to generally describe the response over the entire reference measurement volume. A volume-weighted average response, representing the material uniformly distributed in a matrix throughout the whole measurement volume is usually constructed. Each point may be selected to represent a predefined volume element within the measurement volume. The response (often expressed as "counts per second per gram of fissile material" or "counts per second per Becquerel of activity") from each position is then assigned a weight proportional to the volume of the region represented by that measurement point. Such an approach yields vital information regarding the applicability of the assay technique since the measurement geometries must be consistent with radiation penetration. The number of measurement points should be carefully selected to adequately quantify the changing response function through the sample volume. For example, a low density soft waste matrix affording little gamma attenuation may only require a few measurement positions. However for a drum of grouted waste measured on a PNCC system the response is subject to very large radial variability. In this case, a large number of measurement points may be required. This activity is usually performed during commissioning and also significantly contributes to the performance demonstration (see Section 7.2).

The volume-weighted average approach is appropriate for situations where the radioactive material has an equal probability of being located anywhere within the item to be assayed. Wastes typically exhibit this probability distribution. In such circumstances, by using a volume-weighted average approach, long-term inventory bias is avoided. However, there may be situations where a different distribution is more likely. For streams of small packages placed in the bottom of a large measurement chamber, the calibration should be weighted towards the response nearer the bottom of the chamber to avoid any such bias. Additionally, there may also be a small number of situations where the need for accuracy is overshadowed by a need to not underestimate. In these special situations, it may be prudent to weight the calibration towards the areas of minimum response and accept a bias in return for avoiding the possibility of underestimation.

Historically, this area may not have consistently received appropriate attention. It has been common practice to select calibration points based on criteria such as convenience of source placement (e.g. centre of measurement volume), worst-case scenario, etc. In order to improve accuracy and minimise total measurement uncertainty, the calibration should be weighted towards the distribution of highest probability (e.g. volume-weighted average for most waste streams) unless there is another overriding factor being considered. This does not mean that convenient calibration points cannot be used in future. Such measurement points (e.g. centre of measurement volume) usually have advantages regarding reproducibility. The response from these points may be adjusted based on the probability weighting factors in order to develop the calibration curve when the response profile and probability distribution are known. Such an approach can be used to advantage to reduce the number of measurements with fissile standards. For example, a Pu source calibration may be performed with multiple sources at a convenient position inside a reference drum (for example at the centre of an empty drum). The spatial response profile may then be measured using a ^{252}Cf spontaneous fission neutron source, to determine the correction factors to convert from the drum centre response to the volume-weighted average response. It should also be noted that the positional response profile is matrix-dependent and would most likely have to be separately determined for each matrix.

5.2.3 Documentation

The calibration of an instrument for an assay application should be documented in sufficient detail to permit independent validation of the derived response. The result of a calibration should be expressed in terms of values for K (or its reciprocal) derived from the fitting of reference values of the parameters k_i. The calculations should be independently checked by a second competent person. The staff carrying out the calibration and checking the results should be identified.

The calibration documentation should contain;

- identification of the equipment including its location
- date of calibration/characterisation
- name of organisation/person(s) carrying out calibration/characterisation
- reason for calibration/characterisation (commissioning, breakdown, routine, etc)
- description or identification of method used
- calibration/characterisation values (graphs may be used if appropriate)
- uncertainties
- details of standard used
- details of any relevant conditions under which the calibration/characterisation was made
- details of any special equipment employed
- Reference to any procedures which were followed when performing the work.

It should be noted that the confidence level used to express uncertainties is simply a matter of convention. Different applications may require different confidence levels to be quoted. For example, certain applications may frequently require uncertainties to be quoted at 3 standard deviations (> 99% confidence). In any case, calculation of the expanded total measurement uncertainty requires (see Section 6) each term to be evaluated at one standard deviation. Whilst expression of the total measurement uncertainty at one standard deviation is equally as valid as two standard deviations, it should be noted that the confidence level for the former is only 68 %, as compared with 95 % for the latter. Thus it is imperative that the calibration documentation states the confidence level associated with the declared calibration uncertainty.

It is good practice to compare the calibration values obtained with those from previous occasions and to investigate any trends and inconsistencies.

The requirement for equipment to bear a calibration status label, showing the date of calibration and the date when the next calibration is due, should be considered. In practice, this is more applicable to calibration check measurements (see Section 7.4.2). The label is simply a suggested means of reminding the management and operations team of when calibration (checks) are due. Of course, alternative means of management control to ensure that these checks are performed at the prescribed intervals, are equally valid.

It is recommended that, where possible, an equivalent level of documentation be adopted, for "calibration check" measurements (see Section 7.4.2).

5.3   Reference Standards

Calibration should be performed using reference standards (sometimes referred to as calibration standards) based on material with the same chemical and physical composition as the material being assayed. In cases for which this is not practical, suitable alternatives may be employed provided that the radiation emissions are representative of the actual material for the type of assay system under consideration. For a plutonium-measuring neutron coincidence counting system that cannot be calibrated using a sufficiently high mass of plutonium it may, for example, be possible to use ^{252}Cf instead. This is acceptable provided that adequate consideration is given to the potential impact of self-multiplication in dense aggregates of Pu (applying correction factors as appropriate). Modelling tools can be employed to study such effects and apply appropriate correction factors; it is important to study the uncertainties associated with these factors. The lack of general availability of representative Pu standards means that increasing reliance is being placed on computer modelling and alternative sources such as ^{252}Cf and these tools are now mature for specific applications.

Assurance should be provided to show that the standard is appropriate for the material to be assayed. Care must be taken to ensure that conversion factors are accurately known and the additional uncertainty is taken into account. This ensures that self-multiplication and / or self-shielding, is representative of the real material to be measured. A typical sequence of events in fabricating standards for non-destructive assay is given in [67].

Where practical the reference material should be packaged and contained in an identical, equivalent or similar representative configuration to that of the material being measured.

Reference standards should be uniquely identified and securely held; protected from damage, deterioration and misuse; and be periodically inspected to check physical integrity.

Reference standards should ideally be calibrated with traceability to national or international standards. They should therefore have a certificate or a formal record stating the quantity of the material in the standard and its uncertainty at a stated level of confidence, on a specified date. Other details that may be relevant depending on the end-use are the physical and chemical form of the standard and the properties of its encapsulation. It should also be noted that the use of alternative reference materials originating from a laboratory or operational facility is an acceptable alternative, provided that there is sufficient confidence in the knowledge of the source properties and that adequate equivalence of the material to the real material to be assayed, can be

shown. An example is the use of process material at an operational site to construct fissile reference standards, for use in subsequent calibration of assay systems at the same site. This approach has the advantage that the standards are known to comprise feed material of the same origins as the material to be measured in the assay systems.

The anticipated end-use of the samples is an important consideration. For example, a set of reference PuO_2 standards manufactured for the purpose of measurement (by high resolution gamma spectrometry) of the relative isotopic composition, may be selected for Pu mass calibration of a PNCC system but the level of information provided on the calibration certificates may not be appropriate (in this case only the relative Pu isotopics might be declared but the total Pu mass might not be declared). In the case of ^{252}Cf neutron sources used for calibration of neutron assay systems, it is important to determine the concentrations of any impurities (that is, isotopes other than ^{252}Cf). As such, it should be noted that ^{250}Cf is a prolific spontaneous fission neutron emitter with a half-life which is much longer than that of ^{252}Cf so that for aged sources the neutron emission from ^{250}Cf can become significant such that the effective half-life is increased slightly from the normal 2.65 years. Failure to ensure that the ^{250}Cf content of a source is specified, can therefore give rise to an unacceptably large bias in the neutron emission rate compared to the certificate value. In practice the periodic measurement of the neutron output from long lived ^{252}Cf sources may be necessary to overcome such impurity issues.

Ideally, reference standards should not be used for any purpose other than calibration. It is recommended that working standards (see next section) be used for the frequent checks of stability instead of reference standards.

Standards should ideally have sufficient provenance regarding their radioisotope content, chemical form and physical properties, that their radiative properties can be predicted simply by using radioactive decay correction procedures. The sources must be of sufficient activity to result in a calibration factor with suitable uncertainty for the application. Where insufficient information is available (for example known but unquantified levels of contaminants, as in the case of ^{252}Cf discussed earlier) standards should either be recalibrated or replaced every five years or five half-lives (whichever is the shorter), or the impact of not doing so assessed in the context of the measurement application. Provided standards are well designed, manufactured and maintained such that the only variable is radioactive decay these time consuming and costly activities may be avoided.

5.4 Working Standards

Working standards should be used for routine checks (or "calibration checks") of the stability of NDA systems. They may also be used for determining the relative

response of parameters that are then cross-referenced to absolute measurements made with a reference standard.

The calibration of a working standard does not need to be absolute or traceable to national standards. It is however essential for a working standard to have proven stability so that the radiation emissions are predictable with time. Working standards should be stored, maintained and documented in a similar manner to that specified above for reference standards.

The reproducibility of the measurements using working standards must be good enough so as not to mask significant trends in the calibration of the equipment. The level of inconsistency at which investigation or remedial action are required should be specified.

5.5   Uncertainty

The overall uncertainty associated with the calibration of an assay application shall be evaluated. This evaluation must be derived by an accepted method (see Section 6) by combining the individual components of uncertainty involved in the calibration. These values should be justified in the calibration documentation. The value of the overall uncertainty must be expressed in a manner that enables the total uncertainty of the assay to be determined.

The main sources of uncertainty arise from the quantities shown in expression (2), Section 5.2.1.

In the case of nuclear materials safeguards applications, precise and accurate assays are required, which may necessitate extended count times to achieve high statistical precision on the calibration parameters and therefore achieve accurate calibration curves. However, for some waste assay applications a higher uncertainty may be acceptable for the calibration parameters because the uncertainties associated with the measurement of real waste items will be much greater. The uncertainties on the final assay results relate to departures from the ideal calibration conditions; these are discussed in Section 6.

- **Reference standard**

The uncertainty of the activity or mass for the reference standard is normally that given by its certificate. However, if the information on the certificate is inadequate, further calibration of the standard may be required.

- **Observed reading(s) of instrument to the calibration standard and background**

The uncertainty of the equipment reading is basically that due to the counting statistics of the detection of ionising radiation combined with other random effects, perhaps of unknown origin. This uncertainty is generally random and may be determined by a series of repeated measurements of the same quantity under unchanged conditions (see Section 6.2). It may be possible to theoretically calculate the uncertainty due to counting statistics and thereby investigate the presence of any other random effects.

The influence of this uncertainty is controllable by varying the number of measurements or the length of the measurement interval. It is recommended that, where practical, this uncertainty be reduced to a relatively low level compared with the other uncertainties; 2% or less would be a reasonable target.

In the case of a series of measurements where the activity or mass of the reference material in the standard is varied, the uncertainty for the value of the coefficient $(R - B)/K$ is obtained from the weighted linear, quadratic or other fit to the results (a linear fit is used most often). A weighted non-linear fit may be required for other parameters.

- **Parameters with influence on response (k_i)**

These uncertainties arise from the uncertainties in the values of the parameters and in environmental conditions (e.g. physical and chemical properties, geometry, temperature). See Section 6 for further details. They are derived by employing the measured variation of response with changes in those parameters, for example, from the polynomial fits described above.

Uncertainties arising from differences between calibration and assay assumptions require evaluation. These uncertainties may be considered as uncertainties of the assay, but they are frequently evaluated as part of the calibration process.

For illustration, a typical characterisation/calibration program for a waste drum PNCC assay system would usually include the following steps.

- Identification of representative Pu standards
- Evaluation of correction factors to account for self- multiplication
- Evaluation of mass and Pu isotopic composition of standards
- Evaluation of uncertainties in correction factors and isotopics
- Identification of representative calibration drum matrices (considering matrix type, fill height, homogeneity)

- Measurement of standards at different positions inside each matrix calibration drum to determine volume weighted average (VWA) calibration parameters
- Determine spatial variability of response due to unknown source position inside each calibration drum
- Determine uncertainty due to unknown matrix/matrix compensation process, by studying the deviations expected (for real waste forms) from the VWA calibration conditions.

One may consider that the last two components form part of the initial characterisation phase. They deal with evaluation of the expected measurement uncertainties for real waste measurements, but are derived from measurements/calculations using the calibration standards/drums. Thus, careful selection of the calibration standards/drums, ensuring that they span the full range of the expected properties of the real material to be assayed, is essential.

Treatment of Uncertainties

6

IN THIS CHAPTER

- Scope
- What is measurement uncertainty?
- Steps in Estimating Measurement Uncertainty
- Worked Example

6.1 Scope

The purpose of this section is to provide guidance in areas where questions commonly arise when applying the concepts presented in standard reference material (see, for example [69] and [70]) on uncertainty determination and treatment applied, in this instance, to NDA systems. Further reading may be found in [71,72].

6.2 What is measurement uncertainty?

The objective of any measurement is to establish the value of some parameter of interest. Given the statistical nature of radioactive decay, the measurement result can only ever be an *estimate* of the value of that parameter; hence, the measurement result is incomplete without a statement of *uncertainty*.

It is important not to confuse uncertainty with error. For any given measurement, error is the difference between the true value and the measured value, whereas uncertainty is a measure of the dispersion of the values that could reasonably be attributed to the quantity being measured. We seek to identify causes of error and, where possible, eliminate them from NDA measurements; as a minimum, we seek to establish the size of the error.

Unfortunately, for most practical non-destructive assay systems, it is not possible to establish the size of the error for every measurement we make because we need to use the same system, having the same configuration, to make measurements of samples that possess (often quite) different characteristics. Under these conditions, there will always be some uncertainty about the size of the error on any given measurement. We need to quantify this to give our measurement any meaning.

When considering measurement uncertainty it is reasonable to ask:

1) What the range of possible errors is;
2) Whether there is any chance the actual error could lie outside of the range.

Hence, it is apparent that two statements are needed to quantify measurement uncertainty. The first of these provides the range of values within which we believe the actual error lies. The second (known as 'confidence') provides a statement of how sure we are that the 'actual' measurement error lies within the defined range.

It is important to understand that the error on an individual measurement may, by chance, be negligible; however, the uncertainty may still be fairly large, simply because we can never be sure exactly how big the error is. For this reason, the uncertainty should never be interpreted as representing the error itself; simply as 'bounding' the range in which it may be expected to lie, with the required level of confidence.

For NDA systems, measurement uncertainty is a fact of life. However, accepting a measurement has an uncertainty does not imply doubt about the validity of the measurement; on the contrary, knowledge of the uncertainty implies increased confidence in the validity of the measurement result.

6.3   Steps in Estimating Measurement Uncertainty

For simple systems, where the contributing factors are few, a practical approach to establishing the measurement uncertainty may be to make a number of measurements of known sources in known configurations and geometries. The measurement uncertainty can then be estimated from a statistical analysis performed on the results.

This approach is often appropriate where the measurement geometry and source location are well defined and where the parameter of interest is that which is being measured, or at least there is a fixed relationship between the parameter of interest and the parameter that is being measured.

However, many NDA systems are complex; the sources, locations and configurations are extremely variable and often the parameter of interest does not have a fixed relationship with the parameter being measured. Hence, many factors can influence the measurement uncertainty and this approach may not be practical because of the number of samples that would need to be prepared and the number of measurements that would need to be made.

An alternative (and often used) approach is to calculate (or estimate) an uncertainty budget. The following paragraphs outline a typical methodology (or model) used to estimate measurement uncertainty; the model is consistent with the concepts defined in [69] and [70].

This does not preclude the use of other models or methodologies.

- **Step 1 – Understand how the measurement works**

In practice, measurement uncertainty comprises contributions from a number of sources. Estimation of measurement uncertainty requires the analyst to assess the effects of all possible sources of uncertainty. This can be a complex task.

The first stage is to understand how the measurement works. A clear and unambiguous definition should be constructed, which includes not only the behaviour of the measurement system, but also the behaviour of the material being measured and the conditions and environment under which the measurement is being made. All stages and relationships required to produce the result should be considered.

- ## Step 2 - Identify sources of uncertainty

Once we understand how the measurement works, sources of uncertainty can be identified. It is a fundamental assumption that the instrument is operated within its specified operating envelope.

For many practical applications, a preliminary study should quickly identify the major contributors; the overall measurement uncertainty is almost entirely controlled by the major contributions. It is suggested that a 'reasonable' estimate of uncertainty can be made by concentrating effort on the contributions from these sources. This guide suggests the effort expended should be commensurate with the purpose of the measurement.

Sources of uncertainty typically considered when assembling uncertainty budgets for non-destructive assay systems are:

- Number of radiation induced events detected;
- Background Radiation;
- Calibration;
- Measurement Geometry;
- Activity Distribution;
- Matrix Effects;
- Isotopic Composition;
- Interference from Other Species;
- Dead Time Effects;
- Instrumental Drift;
- Algorithms;
- Physical Constants;
- Environmental Conditions and Surroundings.
- Sample properties – self-shielding or self-multiplication

These are discussed further in Appendix 1.

Some effects are caused by irregular events that, although they are potential sources of measurement error, are not generally considered when estimating uncertainty. Instead, we seek to minimize (or ideally avoid) their occurrence by the adoption of good practice, and by monitoring their presence. Generally, as soon as an error is noticed, the recently-measured results should be examined and re-evaluated. These 'irregular events' may typically include:

Operator Error

The possibility of operator error can be reduced by implementing a suitable, regulated training programme, with routine reviews and, where necessary, refresher courses. There should be adequate safeguards in place to prevent operation by personnel who are untrained and unauthorised to carry out the task in question unless they are properly supervised.

Equipment must be operated according to written, established procedures that are adequately maintained and properly controlled. These procedures should specify (as a minimum) the scope of operation of the equipment. Equipment design should include a consideration of human factors and ergonomics to minimise complexity and consequently the likelihood of operators incorrectly using the instrument or misinterpreting the results produced.

Instrumental Malfunction due to Environmental Conditions

Equipment must be designed so that effects due to adverse or extreme environmental conditions of, for example, temperature and humidity, are minimised.

Where there is potential for environmental conditions to cause malfunction, the ranges of relevant environmental parameters must be measured at the location in which the measurements are to be performed and the equipment must be ascertained to function acceptably within those ranges.

If it is necessary to operate outside the specified ranges (working out of doors, for example) then adequate controls must be put in place to ensure the accuracy is not compromised.

Equipment must be designed so that the effect of voltage fluctuations and noise spikes in the electricity supply (for example) are minimised. Instruments should be properly earthed and screened from sources of electrical noise. They should, as far as is reasonably possible, be located away from electrically-noisy machinery.

Manufacturers should indicate the limits of the EMC (see Section 7.1.3) environment

in which measurement performance is guaranteed. Measurements should not be performed outside of this envelope.

Software Errors and Faults

Software should be produced against established QA standards to facilitate auditing and identification of faults and bugs. Software should be purchased preferably from suppliers who have demonstrated compliance against such QA standards.

Step 3 - Estimate the contribution from each identified source

Conventional treatment of uncertainties is to estimate the effect on the measurement result from the variation (or dispersion) that may be seen for each individual uncertainty component, with every other component being considered at its assumed value (i.e. by the method of partial derivative). The individual contributions can then be combined to establish the overall (or total) uncertainty.

The data available to us will vary in provenance. In some instances we may have a complete data set available, such that we can apply conventional statistical techniques. In other instances we will have significantly less data; for instance, we may only know (or be able to establish) the mean (or expected value) and the extremes of variation. In this instance, standard statistical techniques cannot be used and there is a need to follow the uncertainty treatment approaches given in [69] and [70].

Both of these texts refer to Type A uncertainty components for which conventional statistical techniques can be applied to derive the standard deviation of a set of measurement data. Uncertainty components for which we only know the mean and the extremes of variation are referred to as Type B uncertainty components. Ref [69] and [70] provide a methodology for estimating the dispersion of these components. The methodology advocates fitting a standard shape of 'distribution' to the available data. Standard texts provide a set of values to be used when calculating dispersion for Type B uncertainty components considered to be distributed according to Gaussian (Normal), rectangular, triangular and 'U shaped' distributions. The methodology does not preclude the use of other forms where these can be justified; however it does not provide values for other forms of distribution, so these will need to be determined.

The methodology described within both [69] and [70] allows us to consider the standard deviation of the Type A uncertainty component(s) and the dispersion associated with the Type B uncertainty components(s) to be analogous, using the collective term 'standard uncertainty'.

Step 4 - Calculate the combined standard uncertainty

The information obtained in step 3 will consist of a number of quantified contributions to the overall measurement uncertainty. All such Type A and Type B uncertainty components can then be combined (and combined together) using standard statistical techniques.

There is an implicit assumption that the overall uncertainty distribution will be Gaussian (or Normal), centred on the mean value of the parameter, having a standard deviation σ.

Step 5 – Determine the measurement uncertainty and the confidence

Mathematically, confidence equates to the proportion of the area beneath the probability density function (pdf) that lies within a defined range. It follows that confidence is the probability that the true value will lie within the defined range. Conventionally, the range is declared in multiples of σ; the greater the range, the higher the confidence that it will contain the true value. Given the implicit assumption that measurement uncertainties will be distributed according to a Gaussian (Normal) distribution, standard tables can be used to equate multiples of σ to confidence.

Both [69] and [70] call the co-efficient of σ the 'coverage factor' (k). The coverage factor (k) should be chosen to be commensurate with the level of confidence required. Standard references quote the following:

> k = 1 gives a double sided confidence level of approximately 68%
> k = 2 gives a double sided confidence level of 95%
> k = 3 gives a double sided confidence level of 99.7%

It must be noted that these coefficients are for a double sided confidence interval. If a single sided confidence interval is required, the coefficients must be derived using single sided tables.

Hence, measurement uncertainty is quoted as:

- Having a certain range;
- At a certain level of confidence (quoted in multiples of σ, or as a percentage).

For example, if the combined standard uncertainty σ is calculated to be 10% and the measurement uncertainty needs to be determined with a 95% confidence:

The total measurement uncertainty would be quoted as:

Having a range of ±20%, at a 95% (corresponding to a 2σ) level of confidence.

The range is also often commonly referred to as the 'expanded uncertainty'.

For assay systems in certain applications, we are required to declare only an upper value of uncertainty (the lower value is irrelevant in this application). By way of illustration, if the requirement were to establish the upper value of uncertainty, with a 95% level of confidence (i.e. a single sided confidence interval), again using σ = 10% the total measurement uncertainty would be quoted as:

Having an upper value of +16.45%, at a 95% (corresponding to a 1.645σ) level of confidence.

In this instance, k = 1.645. As illustrated within the example, the relationship between k and % confidence differs between single sided and double sided confidence intervals.

6.4 Worked Example

It is considered useful to provide an example of how an uncertainty budget may be developed using the afore-mentioned Type A and Type B methodology. This is provided in Appendix 1.

Certification, Performance Demonstration, Routine and Maintenance Testing

7

IN THIS CHAPTER

- ■ Certification
- ■ Performance Demonstration
- ■ Routine Testing and Quality Control
- ■ Scheduled Maintenance

7.1 Certification

Certification to relevant national and international standards is required for equipment and instruments used to perform non-destructive assay. Several generic manufacturing standards have been aggressively applied for many years, and more focused standards are also applicable. In particular, some European compliance (CE marking) standards are mandatory, e.g. the EMC directive does not just apply to products which are sold to a customer, it also applies to specialist equipment for 'own use' by the manufacturer. In this section, an attempt has been made to capture the main standards for which certification apply regarding NDA equipment and applications.

It must be stressed that for many of these standards, certification is relevant in two parts. The manufacture of the component or system is only one part. The installation and application of the equipment is equally important when one is concerned with certification. After all, the best-designed and manufactured equipment may fail to deliver its intended function if it is incorrectly installed or used. Section 7.1.1 describes the first part, or manufacture certification, and Section 7.2 describes the second part, or application certification and performance demonstration.

7.1.1 Quality of Manufacture

Many reputable manufacturers of instrumentation are regularly audited by accreditation bodies and are capable of self-certifying their equipment against relevant standards. For those that are not, there exist organisations that can inspect and provide independent certification.

The main series of standards currently relevant for the quality management of manufacture is BS EN ISO 9001:2008. Certification of a manufacturer's facilities and production/testing programmes can be achieved through a number of independent accreditation bodies.

It is important to note that a general supplier certification to one of these standards does not guarantee the same degree of testing and documentation for all products delivered. Individual products may be offered with varying degrees of testing, commensurate with the needs of the customer. It is important for a customer to know his requirements and ensure that they are appropriately satisfied.

7.1.2 General Safety

There are several sets of regulations that in some way apply to the manufacture and use of NDA equipment. It is important that everyone involved understands

and satisfies his responsibilities under these regulations. The following is a list of the main United Kingdom and European legislation that generally apply:

- The Health and Safety at Work Act 1974
- Electricity (Factories Act) Special Regulations 1908-1943
- 92/58/EEC European Safety Signs Directive
- The Electricity Safety, Quality and Continuity Regulations, 2002
- The Electricity at Work, Regulations 1989
- Ionising Radiation Regulations, 1999
- 2006/42/EC European Machinery Directive
- 2006/95/EC European Low Voltage Directive
- 2004/108/EC European Electromagnetic Compatibility Directive
- CDM 2007 Construction (Design and Management) Regulations 2007
- BS 7671 Requirements for electrical installations lEE Wiring Regulations 17th edition
- BS EN 61010-1:2001 Safety requirements for electrical equipment for measurement, control and laboratory use – Part 1: General requirements.
- BS EN 60950-1:2006 Information technology equipment – Safety – Part 1: General requirements

7.1.3   Electromagnetic Compatibility (EMC)

This requirement is satisfied if the equipment is tested and found to be compliant with the following two standards:

- BS EN 61000-6-3:2007 Electromagnetic compatibility (EMC) – Part 6-3: Generic standards – Emission standard for residential, commercial and light-industrial environments
- BS EN 61000-6-1:2007 Electromagnetic compatibility (EMC) – Part 6-1: Generic standards – Immunity for residential, commercial and light-industrial environments

It should be stressed that a system made up of components that individually satisfy these two standards does not necessarily automatically satisfy the requirements. The act of connecting components together can change the EMC properties of the system. Thus, it is important that the design of the complete system is assessed as compliant or it is tested after final assembly.

Another very important consideration, and possibly the most limiting, is the electromagnetic environment at the final location of use. Only newer facilities will be constructed with power supplies and other operating equipment compliant with these standards. For older facilities, this will almost certainly not be the case.

It is important to consider and be aware of the electromagnetic environment in the facility and to ensure that any newly installed non-destructive assay equipment is designed and installed to ensure EMC compliance. The power supply and earth for sensitive instrumentation should be as independent as possible from that supplying other 'noisier' equipment.

For installed systems near to other electrical or electronic equipment, consideration should be given to performing an electromagnetic environment survey after final installation. The results of a survey would provide verification that the electromagnetic environment does not adversely impact on the assay system and that the assay system does not impact on any other system that meets the required EMC standards. At the very least, the commissioning tests should be performed while other equipment in the local area is operating (and running up or down during test measurements) to be fully aware of the possible effects of any interference. If this interference ultimately cannot be avoided, such tests define the extent of such interference and appropriate management controls implemented to ensure that individual measurements are not affected.

7.2   Performance Demonstration

Stated simply, performance demonstration is the collection of tests that show that the system is fit for purpose, i.e. it satisfies the user requirement or specification. Depending on the complexity of the instrument or system and the specific application, this can include any or all of the following: manufacturer production testing, customer acceptance tests, site acceptance tests and on-site commissioning. In almost every instance, the latter will be required.

It is important to recognise that instruments cannot be validated independently from the application. In most cases, it is unlikely that generic instrument tests will be sufficient to adequately demonstrate fitness for purpose for a given application. Therefore, if an already validated system is to be used for a different purpose, some additional testing specific to this new application will almost certainly be required.

Performance testing of the instrument and the given measurement application will establish:

- a realistic and defensible value for the measurement uncertainty for each type of measurement for all allowable measurement conditions will have been determined (refer to Section 6)
- boundaries of operating conditions for which results are valid will be defined
- conditions under which the system may be vulnerable will be known
- credible failure modes will be known and indications of failure understood.

7.2.1 Manufacturer Production Testing

For mainstream applications, commercial off-the-shelf products may be available. These systems have been developed to satisfy applications that are common to a wide area of the industry. It is possible that considerable testing has already been performed on these products. For reputable manufacturers, it can generally be assumed that the testing has taken place under controlled conditions and the data will be of sufficient quality. However, before relying on any of this information, the quality of this test data should be confirmed.

Due to the fact that these products are designed to perform a generic function, it is highly unlikely that rigorous testing of every perceived application has been performed. Manufacturer testing will have been designed to demonstrate generic capabilities as well as to satisfy manufacturing quality control requirements. Once an application has been selected, a full evaluation of the equipment and the method under actual measurement conditions is required to determine the measurement uncertainty described in Section 6. It is unlikely that any generic testing performed by the manufacturer will satisfy all of these requirements. However, some part of these requirements may have been satisfied and some tests may not necessarily require being repeated by the operator. For any of these original tests to be valid, the conditions under which they were performed must be consistent with the intended measurement conditions, including aspects such as time, geometry, environmental conditions, materials being measured, etc. If any of these differ significantly, then it may be more efficient to repeat the test rather than try and extrapolate results from manufacturer test data.

7.2.2 Setting to Work Tests

Complete testing during setting to work is essential in order to maintain control of the measurement process. It is this testing which effectively satisfies almost all of the requirements for performance demonstration.

For more complex custom-installed systems, this may involve one or more of the following stages:

- Works Acceptance Testing (at the supplier facility)
- Customer Acceptance Testing (at the supplier facility)
- Site Acceptance Testing (at the end user facility)
- Commissioning (as part of the process at the end user facility)

For reasons described below, the final stage cannot be avoided, but under some circumstances may be combined into the previous stage. For simpler systems, the final stage may be considered on its own.

It is worth noting that it has been common practice for test plans to be produced as part of the purchase contract by the supplier, at least for all but the last stage of on site commissioning. To get the most value out of these tests, and indeed the instrument itself, it is absolutely critical that the statement of user requirements, or specification, is accurate and complete. It is equally important that both the supplier and customer are in agreement on how much of the performance demonstration testing is to be co-ordinated by the supplier. While it is the aim of the customer to have a fully tested and qualified installed system, it may be in the supplier's interests to only perform the minimum amount of testing that is sufficient to satisfy the contract. Unless the additional testing required to fully satisfy the performance demonstration has been specified and included in the contract, there is a strong risk that little more than basic performance tests will be included in any supplier generated test plans.

There is an additional risk to be aware of if one is relying on the supplier to generate all of the performance demonstration test plans. Suppliers tend to do business with many different customers, each having different requirements. It is therefore crucial that the end-user independently determines the scope and extent of testing to demonstrate the required performance. This allows the validity of any pre-existing performance data gathered by the supplier to be judged in the correct context.

7.2.2.1 Works Acceptance Testing

Works acceptance testing is generally applied as a contract control tool. Most often its purpose is to define a key milestone to allow a contract stage payment. It is an opportunity to prove the system to the customer in order to provide confidence that the contract may continue. Test conditions may involve a partially assembled instrument tested under a very limited range of conditions such that extrapolation estimates may be made. Judgements can then be made as to whether the final system will work as expected under actual operating conditions. There is no default list of aspects that must be tested at this stage. The testing is designed to test whatever the customer and supplier agree is necessary to demonstrate that the milestone has been reached.

The data from this stage usually offers little towards performance demonstration. Very often the manufacturer is limited in the testing materials available or permitted at his facility. Additionally, the test conditions are unlikely to represent all of the expected operational conditions. The instrument may be only partly assembled and often only parts of the process are tested, or different stages are tested independently. Any critical tests would be repeated under operational conditions at a later stage.

This stage of testing is worthwhile because each stage of testing increases the probability that any errors present will be detected and corrected prior to the instrument going into active service. It is also a very necessary part of contract

project management for large projects. As this is usually the first chance a customer has to view the concept unfolding from the supplier's viewpoint, an additional benefit of this stage is the opportunity to capture misunderstandings regarding the user requirements or specification.

7.2.2.2 Customer Acceptance Testing

Customer acceptance testing is usually scheduled after all of the work that can be undertaken at the supplier's facility has been completed. This stage precedes site installation and its satisfactory completion gives the customer confidence that the on-site installation and post-installation operation of the instrument are likely to be satisfactory. The amount of testing necessary to provide this confidence is agreed between the customer and supplier.

As this stage of testing usually takes place at the supplier's facility, it is unlikely that an accurate simulation of true operational conditions can be made. There are likely to be constraints that limit how closely the testing conditions can match the true operational conditions at the customer's site, such as:

- the range of radioactive material that can be used at the supplier's facility may be small or not typical of those on the customer's site

- the supplier's facility may be newer, and cleaner from an EMC standpoint

- the testing environment may be more like a controlled test facility rather than a busy industrial environment, etc.

As for works acceptance testing, the data from this test are unlikely to be directly applicable to supporting performance demonstration unless it can be demonstrated that the above points have been addressed. However, it is another stage of testing that increases the probability of any errors present being detected and corrected prior to the instrument being put into active service. Calibration can be achieved at this stage only under special circumstances including the availability of suitable radioactive materials and matrix simulants.

The primary purpose of this stage is to demonstrate that the design and build work is indeed complete and the system is ready for site installation. It is a key opportunity for catching any remaining minor discrepancies with respect to the user requirements. Any problems up to this stage may still be remedied at the supplier's facility. Any problems that persist or are discovered after this stage will likely need to be rectified on-site at greater inconvenience to both the customer and supplier. This stage is an easily identifiable milestone and is often used to award a contract stage payment.

7.2.2.3 Site Acceptance Testing

This is usually the last stage of contractually motivated testing. The prime purpose is usually to demonstrate that the supplier has indeed satisfied the user requirements and, barring any warrantee considerations, the contract is complete. The required level of testing to demonstrate this must be agreed between the supplier and customer.

The testing is performed following installation at the end user facility. All major functions of the instrument are normally tested under conditions close to those anticipated during normal operations. For tests performed under realistic operating conditions, the results will be directly applicable to satisfying the performance demonstration.

The tests are usually designed to demonstrate that the installed equipment is fully operational and every programmed function works as specified in the user requirements.   It is not necessary that all permutations of all possible varying conditions be tested in order to demonstrate this. Thus, it is unlikely that this level of testing will result in a complete performance demonstration, unless previously agreed to do so with the supplier. However, these tests will contribute to the performance demonstration and will not require repeating under the next phase. A partial or full calibration may be performed during this stage. A full calibration must have been carried out prior to the system being made available for use.

7.2.2.4 Commissioning

This stage of testing picks up where the site acceptance testing left off. In this stage, all remaining tests to satisfy performance demonstration are carried out (see Section 7.2.3).

The tests are performed under normal operating conditions and will cover all of the necessary permutations of varying measurement conditions to be encountered during normal operations.   The tests will also simulate abnormal conditions to assess potential vulnerabilities.   In effect, everything is tested to highlight the limits of performance, and to know the reaction of the system under all foreseeable operational conditions. Once all of this information is known and understood, management procedures can be confidently developed that will constrain operations to within acceptable bounds of performance and avoid areas of vulnerability.

7.2.3 Scope of and Important Considerations for Setting to Work Tests

Although it is impossible to generically specify what tests are required for a complete performance demonstration, this section should provide a useful starting point regarding any individual instrument and application. The following key areas should be addressed in order to fully understand the system and application:

The instrument should, wherever possible, be tested under normal operating conditions for its intended application and under the entire range of anticipated environmental conditions. For example, if motorised equipment is close by that could conceivably be operated during a measurement, the instrument should be tested with this equipment operating. Note that, for this specific example, the most vulnerable situation occurs when the motor starts, so the test should be designed to capture the effects of the motor starting during a measurement cycle. If it is discovered that the system is vulnerable to other equipment in the area, then management controls to prevent their operation during measurements should be used.

In general, enough should be known about the physics behind each process to develop a crude prediction of the expected results for each test. It is not enough to blindly accept the data and assume that they must be correct simply because they have been generated by a sophisticated instrument. If the data do not agree with the prediction then either the instrument has not been set up properly or the physical process is not understood well enough. Neither situation is acceptable and further investigation is required.

A geometrical response profile should be generated where appropriate. This is necessary to calculate any positional uncertainty effects. Measurements should be taken in order to enable mapping of the entire measurement geometry, e.g. for box or well counters, the response at all points within the box or well must be known. For this test, the use of symmetry can aid in reducing the total number of measurements required to synthesise a response profile.

The effects of all expected matrices must be known. It is important to note that for all but the most sophisticated imaging instruments the effects of matrix and position cannot be easily separated. For most applications it will be more convenient to measure and assess their combined effects by developing a measurement profile for each matrix type. If automatic matrix determination/compensation is enabled then it should be tested across the full range of expected matrices.

The effects of the physical and chemical properties of the species to be measured must be assessed. This is especially important when considering the effects of differing lump sizes of fissile material on the detected response.

If the calibration is based on any assumptions, then these assumptions must be tested or otherwise confirmed to be true. For example, if it is assumed that the activity will be concentrated in one location, but it also possible for it to be dispersed, then the effect of such variable source geometry must be determined.

Each hardware and software function should be tested unless it has been disabled. Functional tests should test for negative as well as positive response. For example, if a function is triggered at a threshold value, it should be tested both just above and just below the threshold to demonstrate that it is initiated appropriately.

The possible impact on results of known credible fault conditions should be assessed. The indications of revealed faults should be understood, as should the impact of potential unrevealed faults.

It should be noted that, as long as the instrument and application remain unchanged, this level of detailed system characterisation is required only once and will not have to be repeated.

7.2.4 Commissioning for a Different Application

When an existing system is to be used for a different application, then there is no guarantee that the original performance demonstration will be valid for this new application. This may be equally true if the instrument is to be modified in any way. If either situation is encountered, then a detailed assessment of the deviation between the conditions of the original performance assessment and the new conditions must be performed. It is only necessary to repeat testing for any differences from the original performance demonstration.

Example 1: If a new package size is to be introduced, but the assay chamber configuration remains unchanged, then only the effects of the spatial/matrix changes will need to be re-determined.

Example 2: If the well configuration is to be modified, then all of the geometrical calibration and testing will require repeating. This is typical of mobile neutron slab counter systems that are positioned around in-situ objects. For a new geometry many parameters will change requiring significant reassessment. Some of these parameters subject to change are efficiency, die-away time, reflection, geometrical response profile, etc. Significant reassessment will be required to re-evaluate measurement uncertainties.

7.2.5 Computer Modelling

Complex modelling codes are becoming more widely used as design and calibration tools for NDA equipment, to assess performance and to predict the effects under extremes of conditions. It is now possible to model some conditions for which it is virtually impossible or highly impractical to take physical measurements under controlled conditions. As computing power improves, modelling becomes more attractive.

Modelling, by definition, mimics a real process using a mathematical model. It is therefore not perfect and is limited by the validity of the assumptions and the appropriateness of the model. The quality of any model can and should be assessed prior to relying on any modelling information. This process of benchmarking the model is achieved by comparing actual measurements with the results of the model considering identical conditions. Without this benchmarking information, it is impossible to know the accuracy of any of the modelling information. UK national and international benchmarking exercises have already been performed for some codes in wide use and more are planned for the future [73,74,75,76,77,78,79,80,81,82].

A comprehensive introduction into modelling codes is given in ref [83].

7.2.6   Special Considerations for assay with mobile equipment

Much of the above discussion concentrates on the various staged processes which are relevant for testing, characterisation, calibration and setting to work, of major new installed assay systems.   These testing phases may be implemented over a considerable timescale.

Many measurements are often undertaken using mobile, reconfigurable assay equipment. Whilst these measurements often involve simpler assay equipment and less sophisticated measurement techniques, this is not always the case.   PNCC measurements are often performed with mobile, reconfigurable neutron counting slabs, for example. Similarly, in-situ gamma spectrometry is often performed using a mobile detector for applications where the sample to be measured cannot be moved. Such applications often involve special considerations and it is recommended that an assay procedure is prepared to include equipment set up, checking and field operation to provide confidence in the final assay data. Relevant factors include:

- The calibration should reflect the known or assumed sample-to-detector geometry (including matrix and/or other shielding effects) to allow a realistic determination of the response factor and its associated uncertainty.
- Instrument performance checking regime that provides confidence in equipment functionality during the in-field assay process.
- The effect of plant background conditions which may affect the system performance and / or require erection of temporary shielding
- The effect of other local environmental conditions on the assay performance

Modelling codes are often used for calibration of reconfigurable measurement systems. It is recommended that separate method substantiation is performed on a case-by case basis.

7.3 Routine Testing and Quality Control

Once an instrument has been calibrated, deployed, tested and commissioned, routine testing and maintenance are generally required to provide confidence that the instrument continues to operate properly and perform the functions for which it was supplied.

7.3.1 Types of Test

A number of terms are used within the industry to describe different tests that can be performed. The terms used within this guide are "Standardisation Test", "Functional Test" and "Calibration Check"; each of these types of test is discussed further below.

7.3.1.1 Standardisation Test

The objective of the standardisation test is to demonstrate that the counting chain (from detector through to analyser) is working correctly. For example, in a gamma spectrometry based assay system, it is normal good practice to test that the centroid of one or more peaks appear in the expected channel(s) and that the number of counts in each peak is as expected (allowing for source decay). Sometimes, practitioners will also check that the FWHM (Full Width at Half Maximum) is as expected.

The test should be performed as often as deemed necessary (as a minimum this should be in accordance with manufacturers recommendations), using a characterised source. The source need not be a formally traceable.

Under very controlled (laboratory) conditions, the standardisation test may be used to demonstrate that the system can provide the expected result for the measurement of a known source. For most practical assay systems, however, this is not the case; the test simply demonstrates the system counting chain is functioning correctly. In general, this type of test is relatively quick and simple to perform. For this reason it is the test performed with the greatest frequency and is used to augment other types of test (described below) as part of an operations and maintenance strategy.

7.3.1.2 Functional Test

It is good practice to routinely perform measurements of known quantities of active material, under reproducible conditions to provide confidence that the system continues to function and respond in a normal fashion. Some of the commonly used terms for such measurements are "operational checks", "functional checks", "daily check measurements", and "quality control check measurements".

Although it is not necessary that functional test measurements resemble regular assay measurements, a close resemblance to normal measurement conditions does contribute to the overall picture of 'normality'. A measurement closely resembling

normal operations is simpler and may contribute to greater confidence that the system is behaving normally.

Generally speaking, the more functions that can be tested the better. For example, if automatic matrix correction is employed, then the test should be designed to exercise that component of the system. If the system has both active and passive functions, then both should be tested as part of the functional test. Similarly, if the system makes gamma and neutron measurements, the results of which are combined to give the final assay result, the test should be designed to exercise this functionality.

Functional tests for systems that are to be used to simultaneously assay and report inventories for several different isotopes/elements should involve test material that mimics the expected mix.

Dialogue with the supplier will be invaluable to help understand the workings of each function and the failure modes that can be routinely tested for.

A well-designed testing programme is simple, easy to use and understand, and will clearly identify partial or total failure of any critical component/function or excessive abnormal operation.

7.3.1.3   Calibration Check

It is good practice to routinely check that the system is still within calibration and that the calibration remains valid for the materials the system is required to assay.

The calibration check involves testing of the measurement system response using a known working reference standard/source (or sources). These need not be traceable to national standards. In effect this check perpetuates the validity of the original calibrated response of the system.

For some systems (such as fixed geometry neutron detection systems) a calibration check comprising a measurement made at a single point, but under strictly controlled conditions, may be appropriate to check the calibration.

Ordinarily the calibration check measurement is significantly less onerous and time consuming than the original calibration of the measurement system, but it serves to verify the latter. Indeed this is the intent, since repeating the full calibration at intervals would often be impractical and inefficient.

The calibration check should be appropriately documented.

7.3.2 Frequency of Testing

The frequency of testing is governed entirely by the tolerance to the system failing to perform in the required manner. This statement is equally applicable to all types of testing.

If a test has not been performed with the required periodicity or the result of a test has fallen outside of acceptable performance criteria, it may be necessary to question the validity of all measurements made since the last successful test measurement.

7.3.3 Portable Instrumentation

Following set-up, testing should be performed to demonstrate that the system delivers the functionality and performance for which it will be used. A further test should be performed as soon as reasonably practical following the field measurements to provide confidence that it was operating correctly whilst on plant.

7.3.4 Performance Criteria

Careful attention should be given to selecting appropriate pass/fail criteria in support of test measurement schemes. If the criteria are unnecessarily restrictive, then too many perfectly normal responses will be measured as failure. This leads to an unnecessary loss of confidence concerning the quality of the instrumentation that may prove very difficult to regain. A large incidence of 'false failures' may also contribute to complacency, whereby a 'real failure' may be missed.

Test measurement records should be kept as simple as possible: too much information may lead to confusion. Simple pass or fail criteria for each test measurement will greatly contribute to the success of the programme. Adoption of too many pass/fail criteria for each individual test measurement should be avoided.

It is common practice (and, indeed, encouraged) to investigate more complex parameters, for instance whether too many successive test measurements lie on the same side of the mean. However, such parameters are better investigated by recording the test measurement data at the time of the test, and allowing instrument specialists to examine it periodically to look for evidence of trends.

7.3.5 Actions to be Taken in Case of 'Failure'

Since any measurement of radioactivity is statistical in nature, it is to be expected that some test measurements will 'fail', by chance. It is considered good practice that, should any test measurement 'fail', operations should cease until an investigation has identified whether the 'failure' is a matter of chance, or symptomatic of a genuine issue.

It is good practice to assess whether the cause of the 'failure' could have affected measurements made prior to 'failure' and since the last successful test.

Once the cause is understood, it may be possible to resume operations under increased supervision if the instrument is only partially impaired, and it is possible to control the quality and robustness of measurements until remedial action is taken. Otherwise, measurements should not resume until the problem has been adequately remedied and tested.

Should a single measurement appear to fail, it should still be recorded and not replaced by the results of an immediate re-measure that so happens to fall within acceptable limits. For statistical analysis to be effective, the results of every test measurement are important. Some operators may form the opinion that the success of the test is all important and may believe that it is acceptable to keep taking further operational check measurements following a failed measurement and recording the first of such repeats that pass. This practice must be discouraged as it destroys the randomness of the data generation process that is critical to the success of any statistical analysis technique and may mask a developing problem.

7.4   Scheduled Maintenance

It is good practice to perform a rigorous inspection and testing of the system periodically. This has two main purposes: to confirm that the existing calibration and instrument performance in its entirety continues to be valid and to discover developing hardware degradation so that remedial action may be taken prior to actual component failure. As such, it is considered good practice to perform standardisation tests, functional tests and calibration checks at this time.

The frequency and extent of scheduled maintenance activities is generally application specific and should be commensurate with the requirements and emphasis placed on the instrument. Guidance on the frequency and extent of such activities may be available from the system supplier.

7.4.1   Hardware Degradation

The maintenance inspection should include an overall visual inspection of all components. Any visible damage should be repaired. Any other maintenance actions that are recommended by the supplier should also be carried out. This may involve checking the operation of switches, LEDs, protection circuits, etc. More complex mechanical systems will have additional maintenance requirements, such as inspecting and changing belts, lubricating chains or other moving parts, etc. The physical integrity of the detector chains should also be checked. For example, plateau measurements help to confirm integrity of sealed proportional neutron tubes. An

irregular-shaped plateau is one sign of degradation either within the detector itself or its amplifier.

Sufficient testing should be performed such that all credible fault conditions are examined. This may include additional testing to confirm continued software integrity. Manufacturer guidance will be invaluable in setting up appropriate hardware inspection/maintenance schedules.

7.4.2 Testing Following Maintenance

One of the most vulnerable times for an instrument is immediately following maintenance. There is always a risk that another component may have failed during maintenance or some aspect of the maintenance may have been carried out incorrectly. Extra care should be taken to test the equipment thoroughly prior to return to service (usually by successful completion of a functional test or calibration check) to ensure that either of these two situations have not occurred. It is important to perform tests on all components that have been serviced and further tests on any other aspect that could conceivably have been affected. For example, following a detector failure, the efficiency of the newly installed detector should be checked and a correctly completed measurement cycle confirmed.

Competence and Responsibility

8

IN THIS CHAPTER

■ Competence

■ Responsibilities

8.1   Competence

Personnel involved in specifying, delivering, testing, operating and maintaining NDA equipment should be demonstrated to be competent to discharge those responsibilities. An assessment of competence should be based on formal training, previous experience and qualifications; these should be relevant and appropriate to both the nature and duty of the equipment, and to the role being undertaken.

8.1.1   Training

Training may include, but not be limited to:

- Theory,
- Practice,
- On the job training.

Training should address:

- Knowledge and implementation of standards, guides, procedures and instructions,
- Record keeping,
- Production of documentation,
- Consequences of failing to discharge the role in the prescribed manner.

Personnel undergoing training should be formally assessed; results of the assessment should be recorded.

The training received must be reviewed periodically to ascertain its continued relevance to the tasks being performed. The need for, and frequency of, refresher training should be considered.

Individual training records should be stored for future reference. Records should be maintained, stating which personnel are trained, competent and qualified to perform specific tasks and, where appropriate, should state the systems (or types of system) for which they are deemed competent.

Personnel that have not completed the formal training requirements may perform duties only under supervision from a competent person.

8.1.2   Qualifications and Experience

The qualifications required by personnel involved in specifying, delivering, testing, operating and maintaining NDA equipment should be relevant to their role(s) and to

the activities to be performed. The level of previous experience and its relevance should be closely matched to the level of competence required. In general the level of competence should be allied to the complexity of the task being performed. It is generally considered that a higher level of competence is required for calibration, operation and maintenance of portable assay instruments, as compared to their fixed counterparts, due to the inherently variable measurement conditions with which they contend.

8.2   Responsibilities

The roles involved in specifying, delivering, testing, operating and maintaining NDA systems should be defined. Personnel fulfilling those roles must be made aware of their responsibilities through appropriate training and job definition.

Personnel with overall responsibility should be clearly identified. It is their responsibility to ensure that tasks associated with specification, delivery, testing, operation and maintenance of assay systems are performed on schedule, via a defined reporting interface, and that all work is carried out in accordance with the appropriate standards, guidance, procedures and instructions. They must ensure that any deviation is recorded and justified.

It is the responsibility of the personnel performing any individual activities to notify the person with overall responsibility of any instances where it may be necessary to deviate from standard practices and procedures or where results are not as expected.

Modification Control

9

IN THIS CHAPTER

- Prevention of Unauthorised Modification

- Configuration Control

- Obsolescence Tracking

Incorrect modification of a non-destructive assay system can affect the performance of the system and may have major, detrimental effects.

For this reason, any proposed modification needs to be carefully considered before implementation. In this context, modification includes changes to the way the system is configured and operated, as well as changes to the system itself.

If a modification is to proceed, the implications of the modification need to be fully assessed and the modification needs to be implemented with an appropriate degree of rigour.

9.1   Prevention of Unauthorised Modification

Measurement systems often include signal processing hardware that can be re-configured using switches and dials on the hardware modules. Such units should be installed in such a way as to prevent accidental or unauthorised changes to system settings.

Many NDA systems incorporate software. This often has facilities to select different measurement options and change configuration settings or calibration factors. Such facilities should be managed to prevent the system being used with incorrect settings. Access to application code and operating system commands should be restricted to authorised, competent personnel.

9.2   Configuration Control

Any changes to the system, including configuration parameters and values, must be recorded and the appropriate documentation updated, such that it reflects the current status of the system.

It is good practice to periodically check the system configuration to ensure that actual settings agree with the recorded settings.

9.3   Obsolescence Tracking

A NDA system normally includes several proprietary electronics modules. This modular format allows downtime for breakdown maintenance to be minimised by simple module replacement. However, this type of module will often become obsolete over the lifetime of the instrument. It is prudent to check on a regular basis the continued availability of replacement modules and to plan system upgrades (or increased spares holding) should a component become obsolete.

Appendix 1 – Uncertainty in NDA Measurements

10

IN THIS CHAPTER

- ■ Typical sources of uncertainty in NDA measurements

- ■ Worked example

This appendix supplements Section 6. As such, Section 6 should be read ahead of the material contained herein.

This appendix discusses sources of uncertainty that are typically considered as contributing to the overall (or total) measurement uncertainty and provides a simple worked example of an uncertainty budget. The purpose of the example is not to provide a template, but rather to illustrate the use of the Type A, Type B methodology (as described in [69] and [70]) in the context of non-destructive assay measurements.

This section is included for guidance; it is ultimately incumbent on the analyst to ensure that all significant sources of uncertainty have been considered and to justify the ranges of uncertainty assigned to each component.

A1.1   Typical sources of uncertainty in NDA measurements

Number of radiation induced events detected

If we consider a collection of unstable atoms, we can be certain that all will eventually decay. However, we can never know exactly when any single atom within the collection will decay; it follows that we can never know how many atoms will decay within the measurement period. In fact it is almost inevitable that, if we were to make a number of measurements of the same sample, under the same conditions, the number of counts detected would be different for each measurement; yet each one would be a valid estimate.

Statistical theory dictates that (for such randomly occurring events) as the number of events detected increases, so the relative uncertainty decreases. This fundamental uncertainty underlies all measurements of radioactivity.

Sometimes we obtain a single count; sometimes we obtain a series of counts (which can then be combined; sometimes additional information can be gained by making the measurements in this way). In either case, this is a Type A uncertainty component because the standard deviation on the measurement result may be estimated by applying standard statistical techniques to the measurement result(s).

Background Radiation

In reality, the number of radiation events detected is a composite of those events that originated from the sample and those that originated from the background such that instrument reading = (sample + background).

It is normal practice for the measurement result to be corrected to remove the effects of background events.

For neutron measurement systems, the background contribution is usually estimated from measurements made either directly before and/or directly after the sample measurement, or from a series of measurements without the sample present.

This technique can be applied to gamma systems and there may be reasons why this is the most appropriate method. However, an alternative approach, which is used when the application permits, is to determine the background from the count rates in the channels either side of the peak(s) of interest.

In any event, there will be an uncertainty about the result, due to the random, varying nature of radioactive decay. Again, the uncertainty is a Type A uncertainty component; the standard deviation may be estimated by applying standard statistical techniques to the measurement result(s).

Background Corrected Result

As there is uncertainty on both the sample measurement result and the background measurement result, it follows that the background corrected result will also have an uncertainty. It is generally this uncertainty (rather than the uncertainty on any of the contributing measurements) that is used within the uncertainty budget.

The uncertainty on the background corrected measurement is established by combining the contributing uncertainty components, using conventional techniques. (This calculation can be quite complex, particularly if events emanating from sample and background cannot be considered to be independent).

The uncertainty is most usefully expressed as a percentage of the background corrected value (and is considered to be a Type A uncertainty since it was derived by application of standard statistical techniques).

Calibration

The uncertainty in the calibration of non-destructive assay equipment is discussed in Section 5.5. In general we know the expected value and extremes, but the shape of the distribution is not always quoted. Hence, this uncertainty is a Type B uncertainty. It is therefore necessary to establish the shape of the distribution.

Experience or additional knowledge may enable the analyst to justify a specific form of distribution. Where no further information is available, it is normal

practice to assume a rectangular distribution. (This is considered the limiting case, because it provides the widest range of uncertainty).

Both [69] and [70] provide factors that can be applied to symmetrical Gaussian, rectangular, triangular and U-shaped distributions to derive the standard uncertainty. Clearly, other forms of distribution exist and, in some circumstances, may be more appropriate; however, no guidance is provided as to how to derive the standard uncertainty for other forms. This does not exclude the use of other forms of distribution, if appropriate; however, in these instances, the standard uncertainty will need to be derived and justified by the analyst.

Measurement Geometry, Activity Distribution and Matrix Effects
Practically, assay measurements may need to be made on samples of variable size and shape, where the activity distribution and matrix effects are variable. To accommodate these effects calibration is performed by positioning a known source (or set of sources) at known locations relative to the detector in the defined matrices.

If the response of the system is sensitive to fluctuations in measurement geometry, activity distribution and matrix effects, each of these will produce a component of uncertainty. Potential for the radioactive material to exist in lumps can contribute further to the uncertainty.

It is possible to consider the effects of each uncertainty component separately. However, for most practical systems the measurement geometry, activity distribution and matrix effects cannot be considered to be independent of one another. This can make combination of uncertainty components complex because co-variance needs to be quantified and considered.

A pragmatic approach to overcome issues of co-variance is to develop a combined uncertainty that is an aggregation of geometry, activity distribution and matrix effects. For instance, the range of uncertainty may be estimated based on measurements made using:

- a point source, at the most advantageous position for measurement, within the most advantageous matrix,

- a lump at the most disadvantageous position within the most disadvantageous matrix,

and where the calibration case would be a distributed source within the average matrix.

In this instance the uncertainty component would be Type B, because we have only established the mean and extreme values. However, we may have some difficulty in establishing the precise form of the distribution. Under such circumstances it is acceptable to apply the limiting rectangular form and use the factors provided within the standard references to calculate the standard uncertainty.

Measurement geometry, activity distribution and matrix effects are normally the dominant sources of uncertainty in NDA measurements; hence, the overall (total) measurement uncertainty is extremely sensitive to their value. Clearly, the combinations of source distribution, matrix and geometry given in the example above provide the widest possible variability in measurement conditions; hence they will yield the greatest range of measurement uncertainty. It may be that this extreme range of measurement conditions is not credible and other, less extreme combinations can be justified.

Although, it is notionally possible to perform a more comprehensive set of measurements in order to establish the form of the uncertainty distribution, the number of combinations necessary to construct it quickly becomes punitive. Indeed, obtaining and handling the necessary sources and constructing the range of matrices necessary may well be impractical.

Mathematical modeling provides an alternative approach to assess the form of the uncertainty distribution, with the modeled results being used to generate the standard deviation (i.e. the uncertainty component can be treated as Type A).

It is normal practice to benchmark the mathematical model by performing a limited number of measurements of source/distribution/matrix combinations.

Although modeling can lead to a better understanding of uncertainty, it can be a complex and time consuming activity. The effort expended should be commensurate with the purpose of the measurement.

Isotopic Composition
Thus far we have considered factors that have related directly to estimating the quantity of radioactive decay taking place during the measurement period. Sometimes this can be the parameter of interest but often this is not the case, and it is necessary to apply a conversion factor to obtain the required units of measurement.

One common problem is that the conversion factor does not have a simple, fixed relationship with the number of events detected, across the population of samples

to be measured. For instance, the relative contributions from uranium and plutonium vary with fuel type, enrichment, irradiation history and cooling time.

This uncertainty component is normally Type B.

Interference from Other Species
Interference from radionuclide species present within the sample, other than those of interest, can be a problem for all techniques.

An estimate of the uncertainty requires an estimate of the amount of species that are likely to be present and knowledge of their effect on the system response.

This uncertainty component is normally Type B.

Dead Time Effects
It is necessary to estimate the extent of dead-time and pile-up effects in detector electronics (single-channel and multi-channel analysers, for example). Different situations should be examined individually and the limits assessed for the activities encountered. Most modern systems allow for the input of dead-time correction parameters.

The manufacturers of loss-free counting systems incorporating pile-up rejection or live-time correction should provide information on the uncertainties for given count rates and gamma energies.

This uncertainty component is normally Type B.

Instrument Drift
The conditions producing drift should be controlled as specified by the manufacturer. The manufacturer's specification should include information on the drift to be expected under normal operating conditions over a defined period of time. It is expected there will be procedures in place to minimize instrument drift, but where this is not possible its effect should be considered.

This uncertainty component is normally Type B.

Algorithms
A sensitivity analysis should be performed on any algorithms employed within the measurement system. The effect of assumptions made in an algorithm should be tested at the limits of those assumptions.

The effect on the uncertainties of the parameters used in an algorithm should be evaluated by using the algorithm with limiting values. The results calculated at the

limits should be taken as indicating the uncertainties arising from use of the algorithm.

This uncertainty component is normally Type B.

Physical Constants
Evaluated, universally-adopted values from accepted literature sources should be used for any physical constants that are employed. These sources should also give evaluated uncertainties. The sources used should be documented.

This uncertainty component is normally Type B.

Environmental Conditions and Surroundings
The uncertainties due to fluctuations in environmental conditions (such as temperature) can be estimated based on knowledge of the variability in the relevant parameter.

The uncertainties due to changes in the surroundings are, generally, a matter of judgment. Where possible, fluctuations in environmental conditions should be minimized.

This uncertainty component is normally Type B.

A1.2   Worked example

The following worked example is for a system that estimates total plutonium mass by combining the background corrected reals count rate, measured using PNCC techniques and the measured $\%^{240}Pu_{eff}$, measured using HRGS techniques.

It should be noted that the expressions used within this example have been selected because they are considered to be basic expressions that exist within many standard texts and can be readily derived from first principles. This guide acknowledges that alternative expressions exist; these are no less valid and it is not the intent of this guide to preclude their use.

For the purposes of our example, let us consider that the best estimate of the plutonium mass is obtained using the following equation: -

$$Pu = \frac{R1(net)}{K} x \frac{100}{\%^{240}Pu_{eff}}$$
(equation A1.1)

where: $R_1(net)$ =   Background corrected "reals" count rate

K =   Response factor to convert "reals" count to $^{240}Pu_{eff}$ mass

for a reference matrix.

%^{240}Pu$_{eff}$ = % isotopic ratio as measured by the HRGS system.

The example has been selected because this measurement and technique are in common use within the industry and, as such, the principles should be well understood by most readers.

For the purposes of this example, we will assume that we can estimate the Total Measurement Uncertainty (TMU) using the following equation:

$$\left(\frac{\sigma Pu}{Pu}\right)^2 = \left(\frac{\sigma R}{R}\right)^2 + \left(\frac{\sigma K}{K}\right)^2 + \left(\frac{\sigma\% \, ^{240}Pu_{eff}}{\% \, ^{240}Pu_{eff}}\right)^2 \qquad \text{(equation A1.2)}$$

The three major components included are: -

- Standard uncertainty of the background corrected neutron reals count rate (R);

  - Standard uncertainty of the response factor (*K*);

  - Standard uncertainty of the %^{240}Pu$_{eff}$ measurement.

(All other potential sources of uncertainty having been demonstrated to have a negligible contribution to the TMU).

Standard uncertainty of the background corrected neutron reals count rate (σR);

The gross equipment reading is the number of detected events, N, which is a combination of genuine events due to the presence of fissile material and background events. Generally, the background is subtracted from the total number of events detected to obtain the net reading, which is the desired quantity. As it is not possible to subtract the true background events from a measurement value, a previously-measured reference background value, B, is used instead.

According to Poisson statistics, the uncertainty (σ) on the estimate can be evaluated as the square root of the number of detected events. This is true of both the sample measurement and the background measurement. Following uncertainty propagation rules, the combined standard uncertainty on the net reading, expressed as a percentage of the net reading, is calculated as:

$$\left(\frac{\sigma N_{net}}{N_{net}}\right) = \frac{\sqrt{N + B}}{N - B} x100 \qquad \text{(equation A1.3)}$$

Note that this calculation uses number of detected events, *not* count rate.

In this instance,

$$N = (R + A)_{sam} - A_{sam} \qquad \text{(equation A1.4)}$$

and

$$B = (R + A)_{bkg} - A_{bkg} \qquad \text{(equation A1.5)}$$

where (R+A) is the measured reals plus accidentals count and A is the measured accidentals count obtained during the sample (*sam*) and background (*bkg*) data acquisitions.

In this instance, the combined standard uncertainty on the net "Reals" count is calculated as:

$$(\frac{\sigma R_{net}}{R_{net}}) = \frac{\sqrt{[(R + A)_{sam} - A_{sam}] + [(R + A)_{bkg} - A_{bkg}]}}{[(R + A)_{sam} - A_{sam}] - [(R + A)_{bkg} - A_{bkg}]} x100 \qquad \text{(equation A1.6)}$$

It is common practice to use segmented counting, coupled with segment rejection, rather than to make a single count; this allows segments that contain neutron activity originating from noise or cosmic ray interactions to be identified and rejected to prevent the measurement result from becoming biased. For systems of this type, it is normal to estimate the standard deviation using only the set of "Reals" results (in practice, this tends to yield a slightly larger uncertainty) than using equation A1.6.

The duration of the measurement and the number of segments obtained will affect the uncertainty on the neutron system count rates. Segment acquisition time is usually chosen to give optimum performance of the neutron segment rejection algorithm.

A fairly typical value for the standard uncertainty on the "Reals" count for systems of this type would be 5%; though it is important to note that this depends on the Pu mass and hence neutron signal, low Pu masses giving higher uncertainties than high masses. Recourse to equation A1.1 demonstrates this will contribute a standard uncertainty of 5% to the plutonium mass result.

Therefore:

$$\frac{\sigma R}{R} = 5 \%$$

Standard uncertainty of the response factor (σK);

The uncertainty on the response factor includes both matrix and source position effects. This uncertainty can be comprehensively investigated using modelling techniques, in which case the standard deviation can be determined using statistical techniques. For the purposes of this example we will assume we have data, obtained from efficiency measurements made at the locations of average, highest and lowest efficiency and these are distributed symmetrically about the mean.

In this instance, we consider the uncertainty component to be a Type B uncertainty component.

For the purposes of this example consider:

K(average) = 15 reals per second per gramme of $^{240}Pu_{eff}$
K(min) = 12 reals per second per gramme of $^{240}Pu_{eff}$
K(max) = 18 reals per second per gramme of $^{240}Pu_{eff}$

For the purposes of this example, we shall assume we are able to put forward a justification (possibly based on experience) that a triangular distribution is appropriate to this uncertainty component. With reference to [69] and [70], the standard uncertainty (σ) for a triangular distribution can be estimated by dividing the semi range by $\sqrt{6}$.

Hence:

$$\sigma(triangle) = \frac{(range)}{2} x \frac{1}{\sqrt{6}} \qquad \text{(equation A1.7)}$$

Therefore:

$$\sigma(K) = \frac{(18-12)}{2x\sqrt{6}} \qquad \text{(equation A1.8)}$$

Therefore:

$$\frac{\sigma K}{K_{average}} = 8\ \%$$

Standard uncertainty on the %$^{240}Pu_{eff}$ determination ($\sigma^{240}Pu_{eff}$)

Many practical systems determine the %$^{240}Pu_{eff}$ using data provided by some form of an analysis code. For the purposes of this example we will assume that we know the relative uncertainty associated with the %$^{240}Pu_{eff}$ result to be not worse than $\pm 10\%$, but we have no further knowledge.

In the absence of any further information, and with reference to [69] and [70], it is normal practice to fit a (limiting case) rectangular distribution between the extremes.

The situation can be represented as follows:

<div align="center">-10%   % ^{240}Pu$_{eff}$ result   +10%</div>
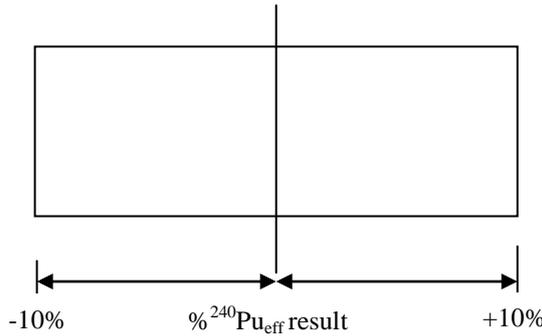
the standard uncertainty (σ) for a rectangular distribution can be estimated by dividing the semi range by $\sqrt{3}$.

In this instance, the semi range is simply 10%.

Therefore:

$$\frac{\sigma(\%^{240}Pu_{eff})}{\%^{240}Pu_{eff}} = \frac{10}{\sqrt{3}} \approx 6\% \qquad \text{(equation A1.9)}$$

Combining the individual uncertainty components

Refs [69] and [70] recommend that these uncertainty components be combined according to equation A1.2:

$$\left(\frac{\sigma Pu}{Pu}\right)^2 = \left(\frac{\sigma R}{R}\right)^2 + \left(\frac{\sigma K}{K}\right)^2 + \left(\frac{\sigma\%^{240}Pu_{eff}}{\%^{240}Pu_{eff}}\right)^2$$

Hence:

$$\frac{\sigma Pu}{Pu} = \sqrt{5^2 + 8^2 + 6^2}$$

Therefore:

$$\frac{\sigma Pu}{Pu} = 11\%$$

Determination of confidence interval

Refs [69] and [70] state that the combined uncertainty distribution will be Gaussian (Normal) and that the calculated combined standard uncertainty may be equated to 1 standard deviation (σ) for a Gaussian (Normal) distribution.

In this instance the confidence interval can be established by taking multiples of σ. The appropriate multiplier, for the required level of confidence (or probability) may be taken directly from standard tables (single or double sided as appropriate).

For this example we will assume that a 95% double sided confidence interval is required, so it is necessary to multiply the combined standard uncertainty by a factor of 2.

Hence, the confidence interval is declared as;

$$\pm 22\% \text{ (95\% or } 2\sigma \text{ confidence)}$$

If a 95% single sided confidence interval were required, it would be necessary to multiply the combined standard uncertainty by 1.645.

In this instance, the confidence interval would be declared as

$$+18\% \text{ (95\% or } 1.645\sigma \text{ confidence)}$$

or

$$-18\% \text{ (95\% or } 1.645\sigma \text{ confidence)}$$

as appropriate.

Note:

This example has been selected to demonstrate how the Type A and Type B methodology may be applied to:

- an uncertainty component that may be expected to have a different value for every measurement, (i.e. the random error on the "Reals" count)

- an uncertainty component that may be expected to vary from sample to sample, but which may be expected to remain constant for repeated measurement of the same sample (i.e. the error on the calibration factor is systematic for repeated

measurements of the same sample, but random across measurements of the sample set)

- an uncertainty component whose value is not known precisely, but may be expected to remain constant across all measurements, for all samples (i.e. the relative error on the $\%^{240}Pu_{eff}$ is systematic)

Appendix 2 – Treatment of Background

11

IN THIS CHAPTER

- Measurement of Background

- General Considerations

- Influence of Background on Measurement

- Uncertainty and Detection Limit

Most modern NDA systems provide a net result that incorporates some method for subtracting background radiation from the measured response. This operation is necessary to remove any potential bias in the results due to other sources of radiation present in the signal that are not due to the object being assayed. There are several different ways of performing background measurements. There are also various operational parameters including, for example, limits of detection and total measurement uncertainty, that require careful attention to the background subtraction. This appendix covers the relevant issues and experience regarding background subtraction in assay methods.

A2.1 Measurement of Background

A2.1.1 Simultaneous Background Measurement

If an additional detector or set of detectors is available outside of the measurement chamber, then the background may be measured simultaneously with the package measurement. This ensures that the background being subtracted is directly correlated to the background conditions during the measurement. However, short of reproducing an entire identical second measurement chamber specifically for the purposes of background measurement, it is difficult to directly compare the background from the "background following detectors" to that in the measurement chamber detectors. Although this method appears to have advantages in theory, in practice it is very expensive and not very practical. It is rarely, if ever, employed.

A2.1.2 Representative Background Measurement

Periodic measurements may be taken with no known active material inside the measurement chamber to represent typical background conditions over a period of time. It is assumed that this background measurement is typical of the background conditions for measurements taken at a later time. This background is then subtracted from future measurement results to provide a net result for each measurement. This method is simple and relatively effective. It is this method that is almost exclusively used.

A2.1.3 Background Subtraction in Gamma Spectrometry Systems

Some gamma spectrometry systems have multiple methods of background subtraction. These may be organised into two main categories: Compton continuum subtraction and spectrum subtraction. Compton subtraction deals with subtraction of events under a peak that are not due to the nuclide being measured, but due to scattering effects from other incident gamma photons of higher energy. These scattering events tend to smear across the spectrum, and the real peak effectively sits above this continuum. The net peak area should be calculated by subtracting the

Compton contribution that lies below the peak. This subtraction contributes to measurement uncertainty in the usual way. Thus, the larger the Compton continuum, the higher the statistical uncertainty.

Spectrum subtraction accounts for measurable peaks in the background spectrum. This is important if the background spectrum shows the visible presence of nuclides that are also in the sample to be measured. If these are not subtracted properly the sample measurement will overestimate these nuclides by the amount present as background. It is important to note that this spectrum subtraction also includes a Compton subtraction component, so the calculation of this effect on the statistical measurement uncertainty is not trivial.

A2.1.4 Background Subtraction in Neutron Coincidence Counting

The Reals rate cannot be directly measured, but is calculated by subtraction of the Accidentals rate from the (Reals+Accidentals) rate, both of which are measured directly. This involves subtraction (as defined in Equation A1.6 of Appendix 1) of two relatively large quantities which affects the statistical uncertainty on the final Reals rate. This is specifically important with active well coincidence counters because the Accidental count rate from the ever-present random neutron source is usually very high and likewise affects the limits of detection.

A2.2   General Considerations

A2.2.1   Stability of Background

If the background is truly stable, in the sense that there is known to be no drift in the average value with time, then it need only ever be measured once. However, since in practice the background is not truly stable it should be updated regularly with the most recently measured representative background always becoming the reference for subtraction.

Ideally, the background should be updated whenever ambient background conditions are known to have changed. This can be whenever packages known to contain active material have been moved around an instrument, or the results of any measurement indicate a problem with the reference background. One such example is a large negative net result for a given measurement. This indicates that the current background conditions are significantly less than when the reference background was measured. If measurements are allowed to continue with this uncorrected, significant bias in the measurement results is introduced. In practice, the background update period should be commensurate with the time scales upon which the background is known to vary.

The frequency of update should be chosen based on an optimisation of many criteria: desired production throughput, relative stability of the background, sensitivity of the instrument to the background, the level of accuracy desired at the lower measurement range, etc. It should not be too impractical to perform an update at least once per day for frequently used systems (assuming that the background is stable on an average daily cycle and has been proven to be so), or before each use for infrequently used systems.

Although it has been historically common for systems to convert all negative net results to zero, this is certainly contrary to best practice. If a system is working properly, packages that contain zero active material will have net measurement results that fluctuate close to, but equally probably on both sides of zero. If there is a large negative result for such a package it is a clear indication that the background has changed and requires updating. If the system were to arbitrarily convert the large negative result to zero, the operator would not be alerted to the change in conditions. Viewing negative results is not itself completely effective in detecting changes to background conditions, but it is one obvious indicator that should be taken advantage of. If there is a significant risk that background conditions can change frequently, or even during measurements, then there are statistical tools that can be employed in software that are capable of detecting such changes. In neutron coincidence counting, for example, cosmic rays can produce high multiplicity bursts of coincident neutrons, and the time variation of the cosmic ray flux can therefore give rise to a significant non-Poisson component in the coincidence neutron count rate. Statistical software tools can help to filter out occasional large bursts, but these tools also have their inherent limitations.

A2.2.2 Representativeness of Background

The background measurement conditions should ideally be identical to those employed in a normal measurement. For example, if a heavy package is to be measured, the reference background measurement should be taken with a similar heavy inactive package within the measurement chamber. This is important because the measurement package itself may change the background conditions. If this effect is not accounted for, then errors in the background subtraction may result. In the example described above, the heavy package may be suppressing the background due to its shielding properties. In this particular case, the result is a larger than true background value being subtracted from the measurement and an underestimate of the radioactive contents.

It is recognised that for high-throughput systems measuring packages of varying properties, it is often impractical to have multiple reference backgrounds taken using multiple reference packages. This is likely to have unreasonable throughput as well

as storage and management implications. It is therefore not unreasonable to default to a single reference background measurement, as appropriate. However, if this approach is chosen, it must be recognised that there will be a corresponding penalty in terms of an increased measurement uncertainty. An assessment should therefore be carried out and accounted for in the estimates for the limits of detection and total measurement uncertainty.

Alternatively, it is possible to adjust the background based on detected or selected known properties of the measured package, but this is an exercise left for instrumentation developers to consider. An example of this is correlating the Add-A-Source matrix correction parameters in PNCC, with the sample matrix-dependent system background.

Some systems continuously monitor and update background conditions. However, these are not without their limitations. At times the background should not be automatically updated, such as during movement of active material in the vicinity of the equipment. These approaches require methodologies to ensure that the continuously-updated background remains representative.

A2.2.3 Non Subtraction of Background

In some special situations it may be advantageous to consider not subtracting background. Such situations can include certain safety or threshold counting applications where the need for accuracy is overshadowed by a need to avoid any possibility of underestimation, however slight. If background is not subtracted, then any slight fluctuation under the true value due to the counting statistics attributable to background subtraction is avoided.

This method should be used with caution, as at least two negative side effects will require careful management. Firstly, the overall trend will be a positive bias above the true value, with an average value directly related to the magnitude of the background. This may not be of extreme consequence when concerned with individual measurement results, but if any longer term trending or summing for inventory purposes is being performed, the increasing bias may not be trivial. Secondly, if the decision threshold is low, the bias in individual measurements may lead to the occurrence of false positives, which may generate unnecessary concern or a decrease in confidence in the measurement process.

A2.3   Influence of Background on Measurement Uncertainty and Detection Limit

A2.3.1 Assessment of Background contribution to Measurement Uncertainty

The contribution to the overall uncertainty due to the background [69,70] should be calculated from the overall background variation and not from the background count rate alone. When the standard deviation of the background is assumed to be the square root of the total background counts collected, (i.e. strictly based on the magnitude of the background) it is implicitly assumed that all of the background variation is due to random counting statistics. As there are other contributing factors, this results in an overly optimistic estimate of the background variation that in some cases can be grossly optimistic. The effects of increased background variation are most severe when counting times are long and the background is updated infrequently.

The most practical method of accurately assessing the total background variation is empirical measurement during the commissioning period. For example, if the background is to be measured (during routine operations) only once per day, several additional measurements may be taken throughout a day to simulate normal measurement conditions. The difference between these later measurements and the initial reference background is representative of the average total background variation. For situations where counting times are, by necessity, very long, the potential variation of the background during the measurement itself may no longer be insignificant and will require a similar assessment.

A2.3.2 Limits of Detection

The effect of the random fluctuations in the instrument readings for the sample measurement and for the background measurement produces a random uncertainty in the net background-corrected count rate. This has the effect of setting values for the quantities variously termed limits of detection and minimum detectable quantities [84,85,86].

Three quantities are recommended for use in NDA measurements -

- **Critical level, L_c**

This is the net count rate that must be exceeded in order to conclude that the sample contains any measurable radioactive material above background. The critical level should not be used as a practical quantity for reporting minimum detectable amounts.

The critical level may be defined as

$$L_c \;=\; k\,\sigma_o$$

where k is the *one-sided* coverage factor and σ_o is the standard deviation of a zero net signal.

For reporting at a confidence level of 95%, $k = 1.65$. This equates to a 5% probability that a true mean count rate of zero will be falsely recorded as a positive value, i.e. a false positive declaration.

- **Detection limit, L_d**

This is the minimum amount of radioactive material that can be quantified with a specified degree of confidence.

The detection limit may be defined as

$$L_d \;=\; L_c + \; k\, \sigma_d$$

where k is the one-sided coverage factor and σ_d is the standard deviation of a net count rate corresponding to L_d. If k is chosen to be 1.65, then L_d is a practical detection limit which you are 95% confident of detecting. Equally there will be a 5% probability that genuine activity is incorrectly attributed to background, i.e. a false negative declaration.

- **Less-Than Level, L_t**

Nothing has been said about the quantity of radioactive material which could be present even though it is not detected; that is, the measured count rate is less than L_c. In general, neither L_c nor L_d satisfies the requirement for the Less-Than Level *(L_t)*. The Less-Than Level *(L_t)* is defined as the maximum true count rate which a sample could have based on the measured count rate (which is less than L_c);

$$L_t = R_s + k\, \sigma_s$$

where R_s is the measured net count rate and σ_s is its corresponding standard deviation.

- **Reporting Considerations when Aggregating Results**

If values at or below the detection limit are simply summed it must be recognised that the total value may be an overestimate of the true value, hence due consideration must be made when comparing this value to overall limits. Although there is a range of guidance available which considers aggregation of results, currently there is a lack of consistency in approach.

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IN THIS CHAPTER

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Additional Reading

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- ■ Additional Reading

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