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Locating and Characterizing Disused Sealed Radioactive Sources in Historical Waste



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LOCATING AND CHARACTERIZING
DISUSED SEALED
RADIOACTIVE SOURCES
IN HISTORICAL WASTE

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LOCATING AND CHARACTERIZING
DISUSED SEALED
RADIOACTIVE SOURCES
IN HISTORICAL WASTE

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 2008

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Marketing and Sales Unit, Publishing Section
International Atomic Energy Agency
Vienna International Centre
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FOREWORD

Disposal and long term storage of radioactive waste were practised by many Member States from the beginning of the use of radioactive material in various fields such as research, medicine, industry and nuclear power generation. This waste included radioactive material permanently sealed in capsules or closely bonded and in a solid form that are called 'sealed radioactive sources'. Despite their predominantly small physical size, many sources contain very high concentrations of radionuclides, which make them potentially dangerous to human health and the environment and could give rise to unacceptable radiation doses during maintenance or in the event of human intrusion.

Many of the existing storage and disposal facilities were developed and began operations long before current regulatory requirements took effect, and before technological advances, safety assessment methodologies and quality management systems became available. Changes in national laws, regulations and disposal methods have evolved and improved with time. Various Member States have ongoing programmes both to upgrade these facilities and/or to develop new near surface disposal facilities.

Recently, a binding international regime for radioactive waste management was established through Article 12 of the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management. This article states that, "each contracting party shall in due course take the appropriate steps to review the results of past practices in order to determine whether any intervention is needed for reasons of radiation protection". It is anticipated that the Joint Convention will result in safety reassessments of certain repositories and that corrective actions will be pursued based on these assessments. Disused sealed radioactive sources dominate the radioactive content of some storage and disposal facilities and are, because of their high activity and/or long half-lives, critical factors in the safety assessment of these facilities.

The corrective actions may include adoption of new waste acceptance criteria and container specifications, building additional engineered barriers, installing hydrogeological cut-off walls, improving cover systems, waste retrieval and other measures. Retrieval of radioactive waste may be a corrective action in cases where sealed sources containing long lived or high activity radionuclides or other problematic wastes have been placed in shallow boreholes or other near surface repositories. Location of disused sealed sources in retrieved waste followed by source characterization may be included as part of the retrieval process prior to re-storage or re-disposal. Waste location and characterization may (i) provide valuable information for updating safety assessments to improve predictions of future facility performance, or (ii) detect the presence of disused sealed radioactive sources within the containers that would not meet the acceptance criteria for final disposal.

Sealed radioactive sources in historical waste pose a significant challenge from a characterization point of view. In some cases the sources are heavily shielded, making characterization difficult. In other cases sealed sources were conditioned to encapsulate sources for theft prevention or to improve shielding, or for other reasons. A general approach to characterization of radioactive waste, retrieval operations and restoration of waste historical data has already been provided by the IAEA. However, characterization methods for sealed radioactive sources in waste have not yet been adequately addressed by the IAEA.

Recognizing the need of many Member States without well developed nuclear programmes to properly locate, identify and characterize sealed radioactive sources present in historical waste, the IAEA has decided to issue this publication. It provides information and guidance on the methodologies and techniques which could be used in these activities. For each of the methods described the applicability to the characterization of sealed radioactive sources in historical waste is discussed and the advantages and disadvantages are highlighted.

The IAEA wishes to express its thanks to all those who took part in the preparation of this publication. The IAEA officer responsible for this publication was A. Kahraman of the Division of Nuclear Fuel Cycle and Waste Technology.

EDITORIAL NOTE

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CONTENTS

SUMMARY	1
1. INTRODUCTION	1
1.1. Background	1
1.2. Objective	3
1.3. Scope	3
1.4. Structure	3
2. CHARACTERIZATION DRIVERS	3
2.1. Need for remedial actions	3
2.2. Characterization drivers	4
2.2.1. Radiation protection	4
2.2.2. Licensing of storage and disposal facilities	5
2.2.3. Operational and post-closure safety	5
2.2.4. Categorization of sealed radioactive sources	6
2.2.5. National inventory of sources	6
2.2.6. Segregation of sources for selection of a management option	7
2.2.7. Emergency preparedness and response	7
2.2.8. Criticality and nuclear material accountancy	7
2.2.9. Compliance with waste acceptance criteria	7
2.2.10. Transportation	8
3. CHARACTERISTICS OF SEALED RADIOACTIVE SOURCES	8
3.1. Equipment containing sealed radioactive sources	8
3.1.1. Medical applications	8
3.1.2. Industrial applications	10
3.1.3. Sources in research	14
3.2. Source design	14
3.2.1. Source shape	15
3.2.2. Source dimensions	16
3.3. Characteristics of radionuclides in disused sealed radioactive sources	17
3.4. Grouping of sealed radioactive sources for guiding their location and characterization	17
3.4.1. Gamma measurements	17
3.4.2. Neutron measurements	19
3.4.3. Laboratory analysis	19
4. LOCATION AND CHARACTERIZATION METHODS	20
4.1. Visual inspection	21
4.2. Basic radiation detection systems	22
4.3. Location of sealed radioactive sources by gamma camera	23
4.4. Dose rate methods	23
4.5. Gamma assay methods	24
4.5.1. General considerations	24
4.5.2. Rotational scanning	25
4.5.3. Segmented gamma scanning (SGS)	27
4.5.4. SGS and swivel scanning (SGS/SW)	27
4.5.5. SGS and angular scanning (SGS/AS)	27

4.5.6.	Transmission corrected SGS (TC-SGS)	28
4.5.7.	Transmission corrected SGS/SW (TC-SGS/SW)	28
4.5.8.	Transmission corrected SGS/AS (TC-SGS/AS).....	29
4.5.9.	Transmission and emission tomography	29
4.6.	Imaging techniques.....	29
4.6.1.	Radiography	29
4.6.2.	Autoradiography	32
4.6.3.	Tomography	33
4.7.	Neutron counting methods	34
4.7.1.	General considerations	34
4.7.2.	Passive neutron techniques.....	36
4.7.3.	Active neutron techniques	39
4.8.	Computer simulations and modelling	40
5.	LOCATION AND CHARACTERIZATION SYSTEM FOR SEALED RADIOACTIVE SOURCES	40
5.1.	Specification of the system	41
5.2.	Retrieval of historical waste data	42
5.3.	Preliminary identification	43
5.4.	Detection of sealed radioactive sources inside waste.....	44
5.5.	Characterization	44
5.6.	Instrumentation	45
5.6.1.	Pocket size instruments.....	45
5.6.2.	Hand-held and portable instruments.....	45
5.6.3.	Mobile systems	46
5.6.4.	Specialized instruments.....	46
5.7.	Quality management	46
6.	CONCLUSIONS	47
	REFERENCES	49
	CONTRIBUTERS TO DRAFTING AND REVIEW	51
	STRUCTURE OF THE IAEA NUCLEAR ENERGY SERIES.....	52

SUMMARY

The objective of this publication is to provide information and guidance on the methodologies, which could be used to locate and characterize disused sealed radioactive sources in historical wastes.

Although disused sealed radioactive sources currently tend to be handled separately from general radioactive waste, this was previously not always the case. In some cases the presence of disused sealed radioactive sources is indicated in existing inventories of historical storage and disposal facilities. In others, it has been conservatively assumed that sealed radioactive sources have been mixed with the waste, at least in some of the disposal facilities.

The various drivers for characterization of sealed radioactive sources are discussed in the report. The main drivers include confirmation of the existing radionuclide inventory; operational and post-closure safety; sorting and segregation; compliance with downstream waste acceptance requirements and transport regulations. The principal driver for retrieval of sealed radioactive sources from a repository is normally the post-closure safety case. This will contain limiting scenarios for exposure of various groups of people (members of the public) to some radionuclides in the waste and will identify what types (if any) of disused sealed radioactive sources and/or waste need to be removed and what specific additional precautions are needed to improve the repository safety. This will form the basis of the sealed radioactive source retrieval and characterization plans.

A sealed radioactive source is usually a low volume, high activity material. It is important that sealed radioactive sources and radioactive waste inventory be compiled and validated.

It is inevitable that a publication of this nature also addresses general radioactive waste management issues, including waste characterization. Sealed radioactive source characterization generally focuses on recognizing and determining the activity of individual radionuclides, while waste characterization is more general. However, sealed radioactive source characterization requires more advanced methods and sophisticated techniques compared to those for other waste items.

Recently, a binding international regime for radioactive waste management was established through Article 12 of the Joint Convention on the Safety of Spent Fuel Management and on the Safety of Radioactive Waste Management [1]. This article states that, “each contracting party shall in due course take the appropriate steps to review the results of past practices in order to determine whether any intervention is needed for reasons of radiation protection”. It is anticipated that the Joint Convention will result in safety reassessments of certain repositories and that corrective actions will be pursued based on these assessments. Disused sealed radioactive sources dominate the radioactive content of some storage and disposal facilities and are, because of their high activity and/or long half-lives, critical factors in the safety assessment of these facilities.

This publication considers a range of location and characterization techniques for disused sealed radioactive sources from simple to advanced. It is stressed that the methods should be selected according to the different characterization drivers, fit the purpose and be developed within a quality management system.

The main non-destructive characterization methods discussed in the report are visual inspections, dose rate methods, gamma assay methods, radiography, tomography, neutron radiography and neutron assay methods. The advantages and disadvantages of each method are identified and the issues, which should be addressed when formulating and implementing the sealed radioactive source characterization system, are discussed.

1. INTRODUCTION

1.1. BACKGROUND

Many countries (e.g. the USA, Russian Federation, UK, the former USSR republics, Eastern European States, China) currently have historical radioactive waste inside storage or disposal facilities designed and constructed a long time ago. Some disposal facilities have never been licensed; those that have been licensed

were often non-compliant with internationally accepted principles, and in most cases there were no environmental impact statements for the facilities. Some of these facilities are not compliant with modern standards and some form of waste recovery and facility remediation is recognized to be necessary. Waste to be retrieved from storage facilities or repositories may or may not be conditioned. Information regarding the waste inventory is generally incomplete, inconsistent with quality management system requirements and inadequate for assessing the long term safety of the facility.

Leakage from the vaults or pits into the environment has been detected in some disposal facilities and in others there is a recognition that this might eventually occur. Some disposal facilities have already been remediated. The waste has been retrieved from the original facility, characterized, reconditioned and transported to new or upgraded disposal facilities. However most of the problematic disposal facilities are still awaiting a solution.

Some of the storage facilities are also experiencing problems. In some cases the facility did not perform as planned. Unfavourable conditions inside some stores have caused corrosion and degradation of waste packages. In many cases, inadequate storage conditions have resulted in disappearance of original markings, labels, and signs that could help identify the origin and characteristics of the waste.

When the waste is eventually recovered, it will undergo a variety of processes to allow it to be disposed of at authorized facilities or sent for interim storage pending disposal. The processes of waste sorting, segregation and characterization are essential and often represent difficult steps towards achieving these objectives.

For the purposes of this document, the determination of the presence and identification of the position of disused radioactive sources in waste is defined as 'location'.

Characterization is defined by the IAEA in Ref. [2] as the "determination of the physical, chemical and radiological properties of the waste to establish the need for further adjustment, treatment, conditioning or its suitability for further handling, processing, storage or disposal".

Disused sealed radioactive sources in historical waste pose a significant challenge from the points of view of location and characterization. In some cases the sources are heavily shielded, making location and characterization difficult. In other cases sealed radioactive sources were encapsulated for theft prevention or to improve shielding, or for other reasons. Such sources can be found inside cemented waste drums where cement provides additional shielding.

It is often difficult and sometimes impossible to consider sealed radioactive sources and other radioactive waste separately since they were often mixed and will therefore be retrieved together. Customized solutions are frequently required, e.g. removing the shielding may be necessary to improve the characterization of sealed radioactive sources. Many retrieval projects have run into difficulties because insufficient attention has been paid to waste characterization. One common failure in the selection of the characterization method(s) is that inadequate attention has been paid to the characterization drivers.

Recovery of data on such waste may be possible in some cases and the IAEA has published specific guidance on retrieval of historical waste inventory data [3]. Nevertheless, if data are not retrievable and/or a reliable information source does not exist, the waste could be re-characterized and then re-conditioned. Retrieval of waste followed by its conditioning is the subject of another IAEA publication [4]. While this publication focuses on the waste containing sealed radioactive sources, more general aspects of waste characterization are provided in Ref. [5]. However, specific characterization methods for sealed radioactive sources in waste have not yet been adequately addressed by the IAEA.

Guidelines, procedures and methods need to be developed to allow those Member States planning or implementing historical repository remediation programmes to evaluate and characterize their radioactive waste, including disused sealed radioactive sources. Characterization of waste could be essential in situations where no data exist and where verification of historical data is not possible.

Although this publication is a stand-alone report, the reader should view the characterization of sealed radioactive sources in the broader context of waste characterization. The methods used to characterize sealed radioactive sources are the same or similar to those for characterizing other radioactive waste. Since the characterization process requires significant resources, process definition, equipment specification and procurement, all aspects of the waste characterization process should be considered. Furthermore, the whole process should be planned and implemented according to modern quality management system requirements. Moreover, personnel exposure should be as low as reasonably achievable (ALARA).

1.2. OBJECTIVE

The objective of this publication is to provide information and guidance on the methodologies and techniques which could be used to locate and characterize disused sealed radioactive sources in historical radioactive waste.

The publication is addressed to Member States which do not have a fully developed nuclear fuel cycle (front end to back end) or do not have sufficient experience in both the management of sealed radioactive sources and historical waste.

1.3. SCOPE

The scope of this publication is restricted to the location of disused sealed radioactive sources in historical waste and their radiological characterization (e.g. radionuclide, activity). Special attention has been paid to identification of those characterization drivers which significantly influence the selection of characterization methods.

General radiological safety aspects and particular safety aspects associated with the presence of non-radiological material in the waste are not addressed [6]. Characterization of nuclear fuel cycle waste and mixed radioactive waste is in the scope of this publication.

1.4. STRUCTURE

This publication describes a broad range of techniques and methods for the location and radiological characterization of sealed radioactive sources in historical waste. It is stressed that the selected methodologies should be commensurate with the radiation safety risks involved. Section 2 discusses the main waste characterization drivers. Section 3 provides background information on the types of equipment containing sealed radioactive sources and on the main mechanical/physical characteristics of sealed radioactive sources. The most commonly used radionuclides in sealed radioactive sources are discussed and grouped according to the methods used for their characterization. Section 4 presents a survey of the principal non-destructive methods that can be used for the location and characterization of sealed radioactive sources. Section 5 provides guidance on the content of the entire characterization system. Section 6 presents the conclusions.

2. CHARACTERIZATION DRIVERS

2.1. NEED FOR REMEDIAL ACTIONS

The need for remedial actions in historical waste storage or old disposal facilities arises for a number of reasons, including those indicated below.

- The quantities of radionuclides in the storage/disposal facilities due to the presence of high activity disused sealed radioactive sources are too large, and this results in unsafe conditions for the workers. For example, the dose rate at the top of the disposal vault at Tammiku, Estonia was $\sim 150 \mu\text{Sv/h}$ [7].
- There is evidence of uncontrolled release of radioactivity from the storage vaults or wells. For example, sampling and analysis of the water from monitoring wells situated near the storage/disposal site has indicated the presence of elevated levels of tritium in the Kiev Radon facility [7].
- Problems have occurred in wells (boreholes) dedicated to the storage of sealed sources. For example, in Romania [8] well blockages have occurred, local contamination has been found which is indicative of leaking sources and water has been found inside the wells.

- The regulations have changed in such a way that the facility no longer conforms to modern safety standards. For example, the permissible doses to the public have been reduced since these types of storage/disposal facilities were constructed and commissioned. The safety assessment of the Püspökszilágy storage/disposal facility in Hungary [9] highlighted concerns about the long term safety of the facility. Some scenarios, notably those related to inadvertent human intrusion, were predicted to give rise to substantial doses to members of the exposed population. These studies highlighted the need for intervention to improve the long term performance of the facility, particularly by removing some or all of the long lived and high activity sealed radioactive sources.
- There is evidence of procedural and design non-conformance. For example, in many repositories unconditioned waste and conditioned and unconditioned sealed radioactive sources have been disposed of in the same vaults. The unconditioned sealed radioactive sources may need to be removed and conditioned. These conditioned sealed radioactive sources may need to be disposed of elsewhere to improve the long term safety of the repository.
- Stakeholders' perceptions need to be addressed to ensure that they have confidence in the operational facilities. For example, the views of stakeholders are being sought before remediation is carried out at the Püspökszilágy storage/disposal facility in Hungary [9].

2.2. CHARACTERIZATION DRIVERS

It is often the case that waste management facility designers experience difficulties in identification of appropriate characterisation methods and equipment. This can lead to mis-specification of the equipment, resulting in assay methods that are not fit for their purpose. It is therefore important to identify the characterization drivers at an early stage in the project. The key drivers for characterization of disused sealed radioactive sources in historical waste are discussed below.

2.2.1. Radiation protection

One of the main goals of characterization of sealed radioactive sources in historical waste should be to detect high activity sealed radioactive sources in order to ensure the radiation protection and safety for workers, and then to isolate these sealed radioactive sources from the waste. Thus a first level of detection and location would be to identify the presence of a sealed radioactive sources of high activity. Examples of storage of disused sealed radioactive sources mixed with other radioactive waste are shown in Fig. 1 [7].



FIG. 1. Storage of historical miscellaneous waste, including disused sealed radioactive sources [7].

2.2.2. Licensing of storage and disposal facilities

Operation of any storage or disposal facility must be authorized by a relevant regulatory body. The legal person responsible for a storage/disposal facility should apply to the regulatory body for an authorization which may take the form of a licence. A proposal for an authorization should, in a written application to the regulatory body, specify the quantities, types and characteristics of the sealed radioactive sources to be stored or disposed of. This authorization will be needed for new storage/disposal facilities if it is decided to retrieve sealed radioactive sources from old facilities and place them in new or upgraded ones.

2.2.3. Operational and post-closure safety

Knowledge of the quantities and characteristics of radioactive waste to be stored or disposed of is essential for demonstration of the safety of storage/disposal methods. It is internationally agreed that the demonstration of safety is performed by the safety case. A safety case is defined in Ref. [2] as “An integrated collection of arguments and evidence to demonstrate the safety of a facility. This will normally include a safety assessment, but could also typically include information (including supporting evidence and reasoning) on the robustness and reliability of the safety assessment and the assumptions made therein”.

A safety assessment is defined in Ref. [2] as “An analysis to evaluate the performance of an overall system and its impact, where the performance measure is radiological impact or some other global measure of impact on safety”.

An operational safety case normally needs to be prepared before operations (e.g. handling of radioactive waste and sealed radioactive sources) can start at a storage or disposal facility. The designers/operators of these facilities must ensure that the risk of release of radionuclides to the environment will be as low as reasonably achievable (ALARA). This principle is defined [10] as “the process of determining what level of protection and safety makes exposures, and the probability and magnitude of potential exposures, ‘as low as reasonably achievable’, economic and social factors being taken into account”.

The operational safety case will contain limiting scenarios, which must be avoided or managed to ensure that they do not cause any harm. Knowledge of the inventory is therefore necessary to assist in identifying limiting scenarios and managing various hazards. It will also contain measures necessary to avoid criticality. In many facilities, including low and intermediate level waste repositories, there will probably be insufficient fissile material present to initiate criticality. If this is not the case, it must be demonstrated that the fissile material inventory will not form a critical mass.

The post-closure safety case for a repository will contain limiting scenarios related to the exposure of various groups of people to some radionuclides in the waste. These critical radionuclides are often long lived, alpha and pure beta emitters. Knowledge of the activities of these isotopes is therefore necessary to produce a credible post-closure safety case. The post-closure safety case will identify what types (if any) of sealed radioactive source need to be removed to improve repository safety. This will be a major driver for repository remediation and form the basis of the remediation and characterization plans.

Data on waste will be needed from several sources, with levels of detail and uncertainty estimates that depend on the objective of the particular safety assessment. The following data are typically required:

- (a) Waste characteristics (radionuclide composition as a function of time, total inventory, physical and chemical characteristics, including gas generation rates, and mass transfer parameters under disposal conditions);
- (b) Container characteristics (mechanical and chemical performance under disposal conditions).

The initial source of information on historical waste will normally be the inventory of the radiological (and sometimes chemical and potentially toxic) content of the waste. If little or no inventory data exist, the characterization measurements will provide a new basis for the inventory. If radioactive inventory data already exist, additional characterization measurements should generate confidence in the historical waste inventory and provide additional data. It should be noted that characterization of the chemical and toxic content and the associated potential health hazards is far more difficult to tackle.

2.2.4. Categorization of sealed radioactive sources

Categorization is an approach which is used mainly when the quantity of elements (i.e. objects or ideas) considered is large, to ease management of the elements by reducing their number. Categorization is realized by selecting the main features (criteria) and by structuring these criteria.

Categorization of sealed radioactive sources may be helpful at any stage from its original use through to its final disposal. It will assist in developing sealed radioactive source management strategies, in optimizing decisions regarding the national registry of sources, organizing the handling operations, planning and designing the conditioning and storage facilities, giving a broad indication of the potential hazards involved with the various types of sealed radioactive source and in record keeping.

Some countries have created their sealed radioactive source categorization systems at different times according to their needs. These systems are different in nature and frequently do not cover the features of sealed radioactive sources important for the management of disused sealed radioactive sources as radioactive waste. The IAEA and ISO have developed internationally acceptable categorization systems [11, 12] that can be recommended to those countries that do not have a sealed radioactive source categorization system or have decided to improve it. It is obvious that without knowledge of the radiological characteristics of sealed radioactive sources it would not be possible to define the category to which the source belongs.

2.2.5. National inventory of sources

The use of sealed radioactive sources is expected to be controlled under the radiation protection programme. A specific concern of waste management is to ensure that sealed sources are properly controlled after they are no longer used, and eventually are disposed of. This concern requires that any new source is tracked throughout its life, that commitments for its disposition be made prior to its import, and that a plan is established and implemented for safe management of the source after it has become disused/redundant.

Every state should establish a national inventory of radioactive sources. The responsibility for the national inventory system normally lies with the regulatory body. This inventory should, as a minimum, include the most active sources (categories 1 and 2) as described in Ref. [11].

The national inventory system should be based on the registry systems established and maintained by the sealed radioactive source users. It should keep track of a sealed radioactive source from the time of import into the country until the source has become disused and is to either be returned to the manufacturer or permanently disposed of. The registry system should contain essential information on the source. The system should enable data to be readily accessible and retrievable, while being resistant to tampering. An acceptable registry system can range from a manual system (such as card files) to a computerized database.

A source registry system should typically contain the following information:

- (a) Type of source;
- (b) Identification numbers (source and container);
- (c) Radionuclide identification;
- (d) Activity and date of determination;
- (e) Supplier/manufacturer;
- (f) Certificate of conformity;
- (g) Former user (authorized institution and responsible person);
- (h) Place of storage (authorized institution and responsible person);
- (i) Other information as may be considered necessary.

It might be the case that no information is available on the source or device. In this case characterization methods should be applied with the use of suitable instrumentation to obtain information on the following:

- (1) Nuclide identification;
- (2) Activity estimation;

- (3) External contamination;
- (4) Leakage status;
- (5) Physical properties of the source and/or device.

Acquiring these data is considered to be the absolute minimum requirement for handling, conditioning and storage of sealed radioactive sources.

2.2.6. Segregation of sources for selection of a management option

Preliminary characterization of sources may be necessary to allow them to be segregated into manageable types. For example, segregation of sealed radioactive sources from other waste, segregation of low activity sources and those containing short lived radionuclides and removal of the sealed radioactive source from its cement encapsulation matrix. Leaking sources must be segregated from other sources and separately collected to avoid cross contamination.

It may be possible to carry out an initial segregation of waste according to the history of the facility, the existing recorded waste data and existing measurements. Thus, depending on the completeness and robustness of existing data, only a simplified characterization programme for verification and validation could be implemented.

2.2.7. Emergency preparedness and response

Characterization of sources is needed to ensure that emergency preparedness plans and responses to accidents are commensurate with the risks associated with the sources.

2.2.8. Criticality and nuclear material accountancy

Characterization may be required to meet criticality control, even if it is not a very frequent or crucial issue in most Member States, and/or nuclear material accountancy requirements. This would be the case if the waste contains fissile and fertile material.

2.2.9. Compliance with waste acceptance criteria

It must be ensured that the waste packages conform to the waste acceptance criteria for processing, storage and disposal. Characterization should be performed to ensure that the waste packages containing sealed radioactive sources conform to these acceptance criteria.

Some characteristics of disused sealed sources result in difficulties in accepting them for disposal in near surface repositories. For example:

- (a) High total activity — The basic property of category 1 and 2 sources [11] is their residual high activity. This high activity can preclude disposal in a near surface repository. For example, the high activity of a category 1 source is responsible for elevated local temperatures and may result in radiological damage to the source vicinity. Gas generation due to radiolysis, decay products and corrosion must be considered.
- (b) High specific activity — A considerable number of higher activity sources exceed the specific activity limits for a waste package to be accepted in near surface disposal facilities because that could give rise to unacceptable radiation doses in the event of inadvertent human intrusion. The source could maintain its structural integrity far beyond the institutional control period.
- (c) Unfavourable physical and chemical characteristics — Although robust construction is employed in the manufacture of sealed sources they are not indestructible. The chemical form of the radioactive contents of a sealed source will determine the degree of dispersability and radionuclide migration within the repository.

Only after a careful safety assessment, which will include among others the above factors, can a decision be made on the acceptability of a specific sealed source for disposal in a near surface repository.

2.2.10. Transportation

On- and off-site transport of radioactive waste needs to be compliant with the relevant transport regulations. The IAEA Regulations for the Safe Transport of Radioactive Material [13] specify the packaging requirements for transport by all modes, and for the in-transit storage of all types and quantities of radioactive material. These regulations require either qualitative or quantitative criteria to be met, depending on the level of hazard posed by the quantity and form of the radioactive contents of the waste.

Sealed radioactive sources are generally encapsulated, fulfilling the requirement for 'special form radioactive material' when they are manufactured. They are then delivered to customers either in a type A or type B package, depending on the activity. Knowledge of the radionuclide activity is therefore necessary to demonstrate compliance with relevant on- and off-site transport requirements.

3. CHARACTERISTICS OF SEALED RADIOACTIVE SOURCES

Sealed radioactive sources have been widely used throughout the developed and developing world for a variety of medical, industrial and research applications since the discovery of radium by Marie and Pierre Curie in 1898. Since the 1950s the choice of radionuclides available for various nuclear applications has increased with the advent of particle accelerators and nuclear reactors for production of artificial radionuclides.

This section addresses the types of devices containing disused sealed radioactive sources, some design features of sealed radioactive sources, and the properties of the most commonly used radionuclides. A more complete list of radionuclides used in sealed sources can be found in Refs [11, 14]. The sealed radioactive sources are sorted into groups, which can contribute to the selection of the techniques and methods that can be applied for their characterization [11].

3.1. EQUIPMENT CONTAINING SEALED RADIOACTIVE SOURCES

The equipment which may contain sources of interest in this publication varies widely in construction and application. Descriptions of some of the main types of equipment are provided below.

3.1.1. Medical applications

Most known applications of sealed radioactive sources in medicine relate to manual or afterloading brachytherapy and teletherapy.

3.1.1.1. Manual and afterloading brachytherapy

Brachytherapy (therapy at a short distance) is a term that is used to describe the interstitial or intracavitary application of radioactive sources by placing them directly in the tumour (e.g. breast, prostate), in moulds (e.g. skin, rectum) or in special applicators (e.g. vagina, cervix).

Originally, brachytherapy techniques involved the use of individual needles or manual afterloading. Relatively low activity sources were used in these applications. Historically, ^{226}Ra encapsulated in platinum in either needles or tubes of a few mm in diameter and up to 5 cm in length was used (Fig. 2). Emission of alpha

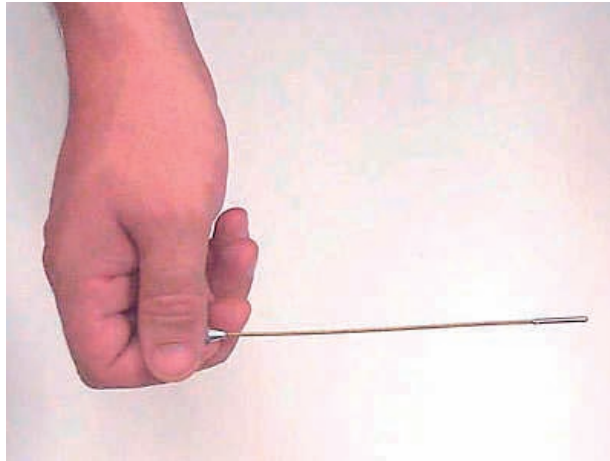


FIG. 2. ^{226}Ra applicator.

particles (helium nucleus) leads to pressure buildup in a sealed capsule. A buildup of pressure may eventually damage the encapsulation, resulting in a release of radioactivity.

Remote afterloading techniques were developed in the 1970s. ^{226}Ra was replaced mainly by ^{137}Cs and ^{60}Co and more recently by ^{192}Ir and ^{252}Cf . These techniques involve the use of machines which can contain a large number of relatively low activity sources, but which taken together represent a significant inventory stored in a single, relatively transportable container. An example of brachytherapy equipment is shown in Fig. 3. This type of equipment typically contains activities of up to 185 GBq (usually of ^{137}Cs). Remote afterloading equipment is used to arrange sources into an appropriate configuration and to transfer them either pneumatically or on the end of a cable into the patient applicator.

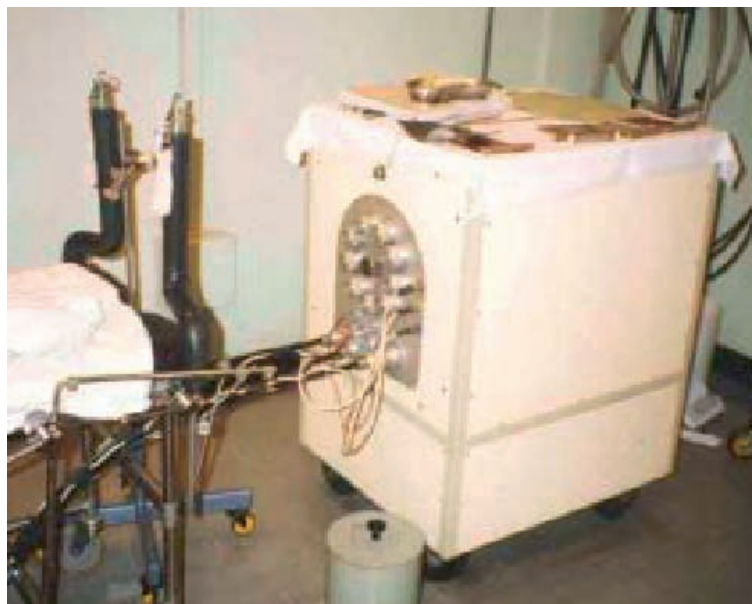


FIG. 3. Typical brachytherapy machine

3.1.1.2. Teletherapy

The other principal medical application of sealed sources is teletherapy, where a large source (typically ^{60}Co but possibly ^{137}Cs) of several hundred TBq is used, external to the body, to irradiate a tumour. These sources are invariably mounted in heavily shielded housings made from steel, lead or depleted uranium ('teletherapy head') and used in shielded enclosures, normally of brick or concrete of about one meter thickness. The head incorporates a shutter mechanism and beam collimating system. The shield material may be lead, tungsten or depleted uranium in a steel shell. ^{60}Co teletherapy heads can contain up to 500 TBq of activity.

Teletherapy equipment became available about 1951, and initially used ^{137}Cs . ^{137}Cs continued to be used through the 1960s but was gradually superseded by ^{60}Co . Because ^{137}Cs was replaced by ^{60}Co it became necessary, due to the shorter half-life, to replace sources at regular intervals, usually every 5–7 years. Teletherapy equipment was therefore designed to allow the sources to be removed from the head and transferred to shielded transport containers in situ rather than in a shielded cell. An example of a ^{60}Co unit is shown in Fig. 4.

3.1.1.3. Other medical applications

Sealed radioactive sources are also used in medicine for bone densitometry (^{241}Am , ^{153}Gd and ^{125}I), for whole blood irradiation (^{137}Co , ^{60}Co) and as gamma radiosurgery knives (^{60}Co).

3.1.2. Industrial applications

In heavy industries such as steel foundries or fabrication, portable, mobile or fixed radiographic equipment incorporating various radionuclides may be installed in purpose built enclosures. Mobile or fixed installations incorporate heavier shielding than portable source housing.



FIG. 4. Typical teletherapy machine.

Typical industrial applications with their main isotopes are shown below:

- (a) Industrial radiography: ^{60}Co , ^{192}Ir , ^{75}Se , ^{170}Tm , ^{169}Yb , ^{137}Cs (historical); $^{241}\text{Am/Be}$, ^{252}Cf (neutron radiography);
- (b) Moisture detectors: $^{241}\text{Am/Be}$, ^{137}Cs , $^{226}\text{Ra/Be}$, ^{252}Cf ;
- (c) Well logging: $^{241}\text{Am/Be}$, ^{137}Cs ;
- (d) Gauges: ^{137}Cs , ^{60}Co , ^{241}Am , ^{85}Kr , $^{90}\text{Sr}(+^{90}\text{Y})$, ^{32}P , ^{147}Pm ;
- (e) Static eliminators: ^{241}Am , ^{210}Po , ^{226}Ra ;
- (f) Lightning rods: ^{241}Am , ^{85}Kr , ^{226}Ra (historical);
- (g) Dredgers: ^{60}Co
- (h) X ray fluorescence analysis: ^{55}Fe , ^{109}Cd , ^{238}Pu , ^{241}Am , ^{57}Co ;
- (i) Calibration: ^{60}Co , ^{137}Cs ;
- (j) Smoke detectors: ^{241}Am , ^{239}Pu .

3.1.2.1. Radiography equipment and gauges

Industrial radiography sources have been widely used for the inspection of pipework and tank welds, etc. Most industrial radiography equipment consists of a source holder, which may contain depleted uranium shielding, and one or more sealed sources (Fig. 5). The source holder is normally also a transport container within the meaning of the international regulations for the safe transport of radioactive material. While the majority of industrial radiography source holders are relatively small, typically 20–30 kg, there are a number of mobile designs which weigh several hundred kilograms and may contain high activity sources. Source activities of a few TBq are common in radiography.

The construction, for example, of petrochemical installations can involve the use of portable industrial radiographic sources of up to 5 TBq of ^{192}Ir for testing of welds in pipes and tanks. ^{137}Cs sources have been used in the past, but these have been replaced by ^{192}Ir and ^{60}Co . ^{169}Yb and ^{170}Tm are also used. The portable containers (afterloaders) for these sources can contain up to several tens of kilograms of shielding material, such as depleted uranium (which has a low specific activity and is a fertile nuclear material), lead or tungsten.

Neutron and gamma sources are used for the determination of density, material levels, porosity and moisture and the hydrocarbon content of geological structures, soils or building materials in oil exploration and in the mining and construction industries. Source capsules are held in source holders, typically 200 mm in length \times 50 mm in diameter. The source holders are transported in type A or B neutron shielded containers.

The most common neutron sources are $^{241}\text{Am/Be}$ (typically up to 800 GBq per source) but some use has been made of $^{239}\text{Pu/Be}$, $^{244}\text{Cm/Be}$, ^{252}Cf and $^{226}\text{Ra/Be}$. The dimensions of storage and transport containers for industrial neutron sources were and still are typically several tens of centimetres (e.g. 45 cm diameter, 50 cm high); the bulk of the shielding will normally be polyethylene or paraffin wax. The activity of older gamma sources was between 50 and 100 GBq ^{137}Cs . The shielding for these gamma sources was normally made of lead or depleted uranium.

In many industries it is necessary to measure the thickness, density or moisture content of a material while it is being manufactured. Application of gauges using radiation from radioactive sources enables non-contact



FIG. 5. Examples of gamma radiography afterloaders containing ^{192}Ir or ^{137}Cs sources.

measurement. A considerable number of different sources may be used. For instance, beta sources (e.g. ^{85}Kr and ^{90}Sr) are used for measuring the thickness of paper, plastics and thin light metals, whilst gamma sources (e.g. ^{60}Co) may be used in industrial process control (e.g. steel and electronics industry) or for measuring the density of coal, rock and oil.

3.1.2.2. *Static electricity eliminators*

In many industries the generation of static electricity during manufacturing creates problems leading to attraction of dust or a possible fire hazard. In order to minimize these problems, static electricity eliminators incorporating an alpha source (e.g. ^{241}Am , ^{226}Ra and ^{210}Po) have been used. These vary in size from hand held devices of a few centimetres to fixed installations up to several metres long. Since the eliminators utilize the emitted alpha particles, the source holder is fragile and will not withstand physical abuse or fire, either of which may result in the spread of contamination.

3.1.2.3. *Smoke detectors*

Historically, radionuclides such as ^{239}Pu , ^{226}Ra and ^{241}Am have been used in smoke detectors. The radioactive material were sealed inside a single envelope (metal foil) or deposited on a ceramic support and vitrified. A partially dismantled smoke detector containing ^{239}Pu manufactured in the former USSR is shown in Fig. 6.

3.1.2.4. *Industrial irradiators*

High energy gamma irradiators are used world-wide for a number of purposes, including sterilization of medical supplies and food irradiation. These involve the use of arrays of very high activity sources (i.e. the average total activity per sterilization unit is about 40 PBq ^{60}Co). Large volumes of material are passed through the source beam. These will typically be presented on pallets carried past the source beam on a conveyor belt (Fig. 7).



FIG. 6. *Partially dismantled smoke detector containing ^{239}Pu (Soviet design).*

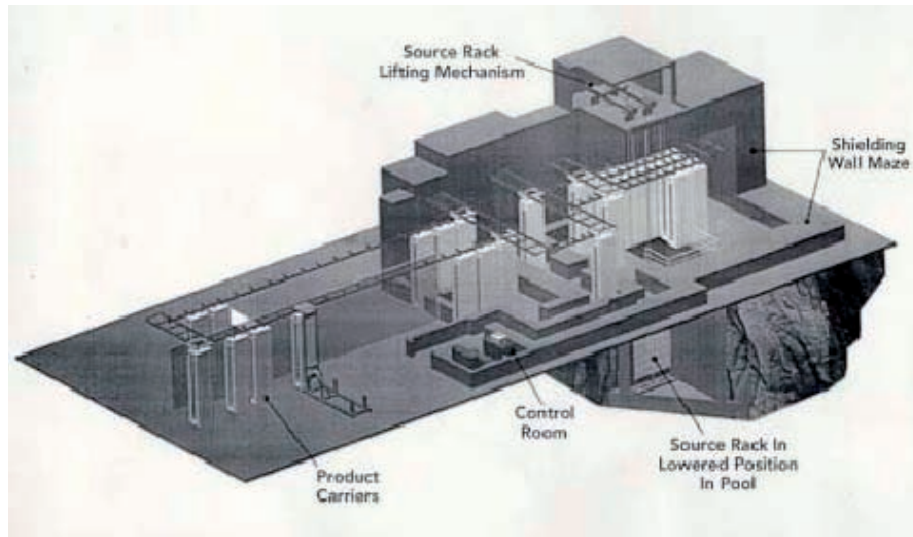


FIG. 7. Layout of a sterilization plant.

3.1.2.5. Radioisotopic thermoelectric generators

Radioisotopic thermoelectric generators (RTGs) are devices that use the decay heat of a radioisotope to produce electricity. The two radionuclides that have been most frequently used are ^{90}Sr (330 TBq – $2.5 \times 10^4\text{ TBq}$) and ^{238}Pu (1 – 10 TBq). The power that is typically generated can vary from a few watts to tens of kW, depending on the activity and radioisotope. There are no moving parts in these devices and, since they are designed to operate unattended for tens of years, they are ideal for supplying power to equipment in remote areas (Fig. 8). Hence they have been deployed fairly extensively in the Arctic regions and in space. Many of the devices were originally put in position by the military forces of the USA and the former USSR for remote monitoring or for navigation purposes. It should be noted that it is very unlikely to find a radioisotopic thermoelectric generator in historical waste.



a)



b)

FIG. 8. Radioisotopic thermoelectric generators: a) from the former USSR, b) from the USA.

3.1.3. Sources in research

Applications of radioactive sources in research are very varied. Almost any radionuclide can find a use in some research work. Almost all alpha, beta, gamma or neutron emitting radionuclides may be required for research. The range of activity is large and the sources can be transportable or fixed in installations.

Use of radium sources for calibration purposes has been extensive, and may still continue. $^{226}\text{Ra}/\text{Be}$ and $^{238}\text{Pu}/\text{Be}$ sources have been used in university training programmes. Research work is often carried out in project form, e.g. thesis work and under contract. Equipment, including radioactive sources, may have been obtained specifically for the project. When the work is completed, there may be no further use for the sources, which then become redundant and disused.

Research irradiators have been produced for both gamma and neutron irradiation. There is no standard design principle for irradiators used for research purposes, due to the range of applications. In many cases, research establishments have produced irradiators based on their own designs. There are, however, two main design principles; those in which the source position is fixed and those in which the source is moved to an exposed position.

Irradiators in which the source is moved out of its shielded position into an exposed position are normally used for the irradiation of large samples. These are typically located inside a shielded room or cell of considerable volume, into which samples are placed. With the doors locked, the source is then brought out of its shielding by a remote mechanism. A system similar to that used in many industrial radiography applications (e.g. teleflex) is often used with these irradiators.

An alternative approach is to hold the source in a fixed position and move the sample close to it. From the early 1960s there was a considerable market for fixed source irradiators. A number of companies developed a range of products based on similar principles, and some of these products are still sold today. A typical fixed source irradiator will have one or more sources positioned in the centre of a large, shielded source holder, weighing perhaps a few tonnes. A sample is placed in a sample chamber and the sample chamber may then be moved into position close to the source. There may be a number of sample locations, giving a range of radiation dose rates. Examples of the mobile irradiators used in the former Soviet Union (^{137}Cs) are shown in Fig. 9. Research irradiators of this generic type may contain 100–200 TBq of activity.

3.2. SOURCE DESIGN

The capsule material of sealed radioactive sources is made generally, with very few exceptions, of stainless steel or inert metals like titanium, and more rarely platinum.

Since the 1990s, in order to comply with international standards, the sealed radioactive sources should be marked for identification with an engraved serial number, and for sources with sufficiently large dimensions, the



FIG. 9. Sources used in mobile irradiators in the former Soviet Union.

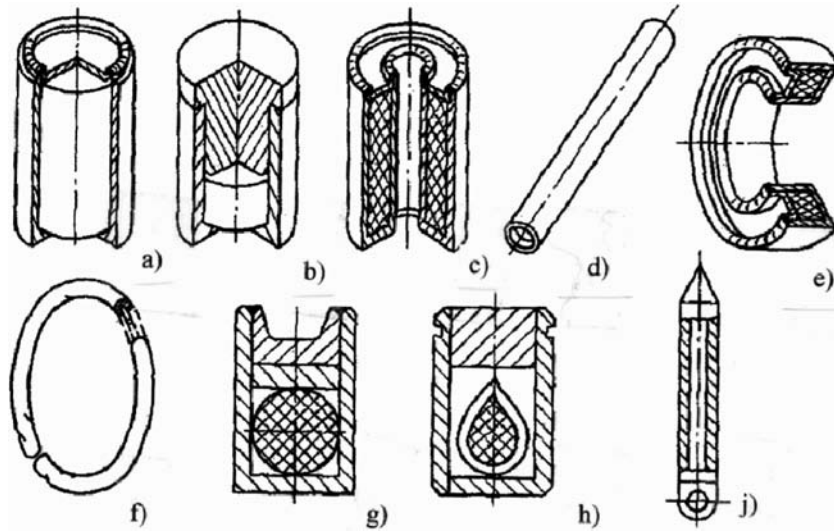


FIG. 10. Radioactive sources with the volume activity. a) ^{60}Co ; b) ^{153}Gd ; c) ^{124}Sb ; d) ^{60}Co ; e) ^{241}Am ; f) ^{241}Am , ^{109}Cd , ^{57}Co ; g) ^{137}Cs ; h) ^{169}Yb ; j) ^{137}Cs .

radionuclide, activity and date may also be given. In some cases, however, the small size of the source prevents marking but the activity of such sources is usually low. The code is also provided in the source certificate.

3.2.1. Source shape

The active part of the radioactive source may have various forms. For β - γ radiation the active parts may be in the form of cylinders, disks, granules, plates and wires (Fig. 10).

For α sources, some β sources and low energy γ sources the radioactive material is fixed as a thin layer on the surface of the plate (Fig. 11). Source holders and the source itself used in medical teletherapy units are presented in Fig. 12.

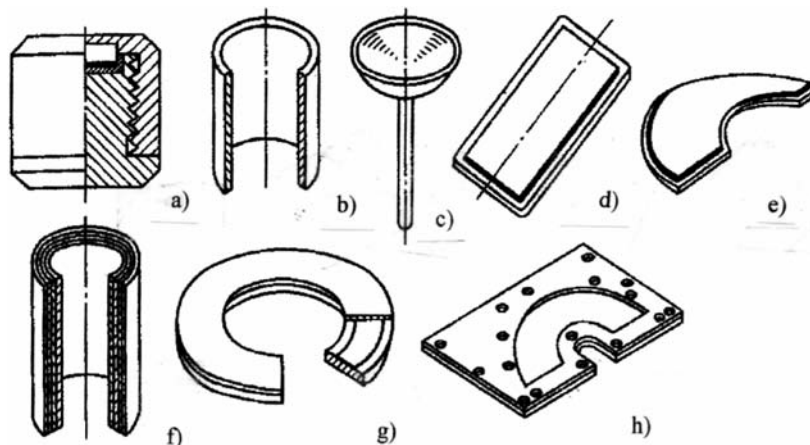


FIG. 11. Surface radioactive sources. a) β sources with $^{90}\text{Sr} + ^{90}\text{Y}$, $^{144}\text{Ce} + ^{144}\text{Pr}$, $^{106}\text{Ru} + ^{106}\text{Rh}$; b) α source with Pu ; c) β source with $^{90}\text{Sr} + ^{90}\text{Y}$; d) α source with Pu ; e) β source with ^{147}Pm ; f) β source with $^{90}\text{Sr} + ^{90}\text{Y}$; g) X ray source with ^{55}Fe ; h) β sources with $^{90}\text{Sr} + ^{90}\text{Y}$, $^{144}\text{Ce} + ^{144}\text{Pr}$.



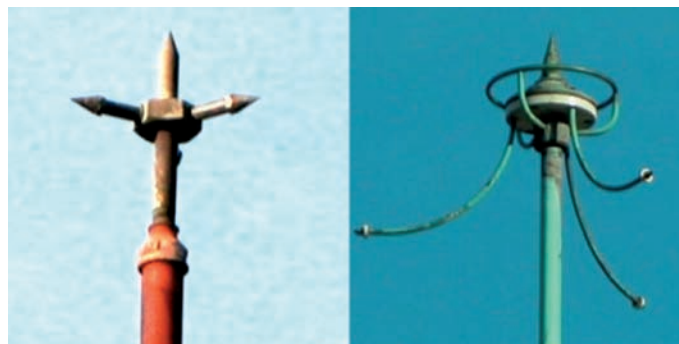
FIG. 12. Source holders and the source (bottom right) used in medical teletherapy units.

3.2.2. Source dimensions

Geometrical parameters of sources are important when choosing available techniques for conditioning and storage. In addition to the typical cases described above, there are particular applications where sources are very large. For example, there are lightning rods greater than 1 meter in length containing ^{226}Ra or ^{241}Am (Fig. 13). The geometrical form of some of these devices is complex, with radioactive source material distributed on the surface of the structure. Many of these devices were manufactured many years ago and little consideration was given to the problems that would be encountered during decommissioning.

Typical dimensions of high energy gamma sources (irradiators):

- Outer diameter (\varnothing) of the capsule: 10.5–11.5 mm;
- Length (l) of the capsules: 80–452 mm;
- Wall thickness of the capsules: 0.5–1 mm.



^{241}Am

$^{226}\text{Ra}/^{241}\text{Am}$

FIG. 13. Lightning rods.

The form of the active material (nickel plated) can be:

- Disc (\varnothing 6–7 mm, height 5–7 mm);
- Slug (\varnothing 6–7 mm, height 20–80 mm).

Typical dimensions of industrial sources (NDT, gauging techniques, etc.):

- Outer diameter of the sources: 3–6 mm;
- Length of the sources: 3–20 mm;
- Wall thickness of capsules: 0.5–1 mm.

The form of the active material (nickel plated) can be:

- Pellet (\varnothing 1–3 mm, height 0.3–1 mm);
- Slug (\varnothing 3–5 mm, height 3–5 mm);

The typical dimensions of sources for radiotherapy:

- Outer diameter of the sources: 20–25 mm;
- Length of the sources: 23–38 mm;
- Wall thickness of capsules: 0.5–1 mm.

The form of the active material (nickel plated) can be:

- Pellet (\varnothing 1 mm, height 0.5–1 mm);
- Disc (\varnothing 5–20 mm, height 1–2 mm).

The typical dimensions of sources for brachytherapy:

- Outer diameter of the sources: 1–1.5 mm;
- Length of the sources: 5–8 mm;
- Wall thickness of the capsules: 0.1–0.2mm.
- The most common form of the active material is wire (\varnothing 0.5–0.8 mm, length 1–6 mm).

3.3. CHARACTERISTICS OF RADIONUCLIDES IN DISUSED SEALED RADIOACTIVE SOURCES

Table 1 provides information on those radionuclides which need to be considered when locating sealed radioactive sources inside radioactive waste and characterizing them. It is noted that some of these isotopes have relatively short half-lives and will therefore have decayed to safe levels before they have been retrieved.

3.4. GROUPING OF SEALED RADIOACTIVE SOURCES FOR GUIDING THEIR LOCATION AND CHARACTERIZATION

For the purpose of selecting location and characterization methods, the sealed radioactive sources listed in Table 1 have been subdivided into different groups as given below. It is cautioned that the groupings are for guidance only and that specific expert advice should be sought on a case by case basis.

3.4.1. Gamma measurements

In relation to historical waste, the sealed radioactive sources containing the radionuclides listed below might, under specified conditions (e.g. activity level high enough), be located and characterized by gamma

TABLE 1. TABLE 1. CHARACTERISTICS OF RADIONUCLIDES IN DISUSED SEALED RADIOACTIVE SOURCES

Radionuclide in sealed radioactive sources	Use	Half-life (Years)	Emissions applicable for characterization				Applicable group * G, A, N (see, 3.4)
			α	β	γ	n	
³ H	Military, industrial applications; gaseous light emitting devices; targets for neutron tubes (high activities)	12.3		✓			A
¹⁴ C	Thickness measurement	5,700		✓			A
²² Na	Oil, mining and paper industries	2.6			✓		G
⁶⁰ Co	Industrial radiography; oil; mining paper-industries; teletherapy and brachytherapy; food irradiation	5.27			✓		G
⁶³ Ni	Electron capture detection	100.1		✓			A
⁸⁵ Kr	Process control in the paper and plastics industries; lightning rods	10.72			✓		G
⁹⁰ Sr	Thickness measurement; eye applicators; power generators	28.6		✓			A (Bremsstrahlung where applicable)
¹³³ Ba	Calibration sources	10.5			✓		G
¹³⁷ Cs	Thickness, level or density measurement; radiography, well logging, sterilization, teletherapy; food irradiators	30.17			✓		G
¹⁵² Eu	Oil, mining and paper industries	13.6			✓		G
²²⁶ Ra	Brachytherapy; lightning rods; smoke detectors	1600			✓		G
²²⁶ Ra/Be	Oil well logging; moisture detection	1600			✓	✓	G, N
²³⁸ Pu	High activity: ground and spatial RTGs (most hazardous sources); pacemakers. Low activity: smoke detectors; static electricity eliminators.	87.7	✓		✓	✓	G, N, A
²³⁸ Pu/Be	Oil well logging; industrial application; moisture detection	87.7			✓	✓	G, N
²³⁹ Pu	Smoke detectors; gas analysers	24,100	✓		✓		G, A
²³⁹ Pu/Be	Mining industry, oil well logging; moisture detection	24,100			✓	✓	G, N
²⁴¹ Am	Density measurement; static electricity elimination; smoke detectors; lightning rods; bone densitometry	432	✓		✓		G, A
²⁴¹ Am/Be	Mining industry; oil well logging; moisture detection; neutron activation	432			✓	✓	G, N
²⁴¹ Am/Li	Neutron activation	432			✓	✓	G, N

TABLE 1. TABLE 1. CHARACTERISTICS OF RADIONUCLIDES IN DISUSED SEALED RADIOACTIVE SOURCES (cont.)

Radionuclide in sealed radioactive sources	Use	Half-life (Years)	Emissions applicable for characterization				Applicable group * G, A, N (see, 3.4)
			α	β	γ	n	
²⁴⁴ Cm/Be	Mining industry; oil well logging, moisture detection; neutron activation	18.1				✓	N
²⁵² Cf	Cement industry; oil well logging; neutron activation; medical application	2.64				✓	N

* **G**: Gamma non-destructive measurements; **N**: Neutron non-destructive measurements; **A**: Laboratory analysis.

measurements. The radionuclides shown in bold have relatively very strong gamma energies, making them easy to measure. One possible complication is the presence of other gamma emitters and shielding effects if the sealed radioactive sources have been co-disposed of with other waste. This group is indicated in Table 1 by (G).

²²Na, ⁶⁰Co, ¹³³Ba, ¹³⁷Cs, ¹⁵²Eu, ²²⁶Ra, and, to a lesser extent: ²⁴¹Am, ²⁴¹Am/Li, ²⁴¹Am/Be, ²³⁸Pu/Be, ²³⁹Pu/Be, ²²⁶Ra/Be.

For non-destructive assay (NDA) using gamma techniques, the following gamma emitting nuclides can be in most cases ignored for the reasons provided below:

- Due to low energy gamma lines: ²⁴¹Am (except if scattered background from higher gamma energies is low enough);
- Due to low intensity: ²³⁸Pu, ²³⁹Pu.

As one of the main safety goals related to retrieval of historical waste is to be able to detect high activity sealed radioactive sources in order to ensure radiation protection for workers, the first level of detection would make it possible to identify sealed radioactive sources of safety relevant activity level, in order to recover and temporarily store the source and later to characterize it.

Some compound neutron sources like ²⁴¹Am/Li, ²³⁸Pu/Be, ²³⁹Pu/Be, ²⁴¹Am/Be, ²²⁶Ra/Be can also be identified by measuring the high energy gamma ray produced by the following reactions ¹H(n, γ)²H, ⁹Be(α , n γ)¹²C and ⁷Li(α , $\alpha\gamma$)⁷Li [15–18]. Moreover, high energy gamma rays produced by neutron activation reactions (n, γ) on housing materials (e.g. aluminium, iron, titanium) and neutron shielding materials (e.g. borated paraffin) may be used for detection of the neutron sources.

3.4.2. Neutron measurements

The sealed radioactive sources listed below can be detected by neutron measurements, passive and/or active methods. This group is indicated in Table 1 by (N).

²²⁶Ra/Be, ²³⁸Pu, ²³⁸Pu/Be, ²³⁹Pu, ²³⁹Pu/Be, ²⁴¹Am/Be, ²⁴¹Am/Li, ²⁵²Cf.

3.4.3. Laboratory analysis

In some cases, preliminary investigations of the upper layers of raw waste in storage or disposal vaults or in waste packages will not provide an indication of the presence of sealed radioactive sources in waste because:

- (a) Some sealed radioactive sources emit radiations which are difficult to detect (beta radiation, low energy gamma radiation);

- (b) The radiation from other waste may interfere with the radiation from the sealed radioactive sources and make it difficult to detect low activity gamma or neutron sealed radioactive sources;
- (c) The radiation from the sealed radioactive sources is difficult to detect due to its own shielding or the shielding provided by other waste items and/or waste matrix.

If under such conditions sealed radioactive sources cannot be characterized by 'in field' or 'in situ' non-destructive assay methods then they should be retrieved, sorted out, segregated and characterized. The sorting of waste (in a glove box or a hot cell, depending on the radiation hazard) will allow identification of sealed radioactive sources (or items which look like sealed radioactive sources) by visual inspection using the IAEA catalogue [19] or by means of basic radiation detection equipment (e.g. G-M detectors, beta probes, proportional counters, scintillation spectrometers).

The low energy levels of penetrating radiation from such sealed radioactive sources reduce the exposure risk during characterization, and in many cases the low activity and radiotoxicity reduce the risk further during disposal.

A pragmatic approach adopted in many countries is to characterize sealed radioactive sources by destructive methods only when it is absolutely necessary, in accordance with the characterization drivers (see Section 2.2). In the unlikely event that such sealed radioactive sources are sent to a laboratory for detailed analysis, the preparatory procedures and measurements would typically include the following:

- (1) Wiping the source holder, taking the activity into solution and measuring the activity using liquid scintillation counting (LSC) and gamma spectrometry;
- (2) Removal of the source from its holder, preparation of samples, sample analysis (e.g. by LSC and gamma spectrometry).

More details of sampling and analysis methods are given in Refs [5, 20].

If a sealed radioactive source is submitted to a laboratory for destructive analysis it would no longer be considered a sealed source and would require subsequent treatment and conditioning in conformance with relevant waste acceptance criteria.

The radionuclides of concern identified in Table 1 in group (A) are as follows.

Pure beta emitters: ^3H , ^{14}C , ^{63}Ni , ^{90}Sr : most of radiation energy would be absorbed in the encapsulation material.

Tritium is of particular concern as it was used in targets for neutron generators with very high tritium content. Indeed, most of the countries manufacturing such generators used to consider the neutron tube: glass tube containing the tritiated target, as a sealed source, because tritium was confined in the sealed glass tube. Identification of the potential presence of such tritium tubes in waste (on existing records, and by visual identification when retrieving waste) would be of high priority to avoid spreading of tritium in the environment and in the waste processing facilities.

Transuranic isotopes: ^{239}Pu and ^{241}Am : these isotopes have been used in smoke detectors and calibration sources.

4. LOCATION AND CHARACTERIZATION METHODS

This section gives an overview of different techniques and methods [21–31] that may be applied for the location, identification and characterization of disused sealed radioactive sources. Most of the described methods are applied for general radioactive waste characterization. However, the location and identification of disused sealed radioactive sources are more complex for the following reasons:

- (a) Highly localized and concentrated activity (hot spot);
- (b) Possible heavy shielding leading to strong attenuation of the emitted radiation (cold spot);

- (c) Interference between the radiation emitted by the sealed radioactive sources and that emitted by other waste inside.

Moreover, the strategy and methods for characterization of newly generated sealed radioactive sources from nuclear applications and those for characterization of sealed radioactive sources in historical waste may be different. Historical waste to be retrieved from storage or disposal facilities will often have the following characteristics:

- (1) The waste was mixed, so that the fingerprint method is difficult, even impossible to apply;
- (2) It will be difficult to rely on available information regarding retrieved waste packages, if these exist;
- (3) Retrieved waste packages (mainly drums) will often be in poor condition, so that their handling and further characterization will not be possible without repacking drums and/or reconditioning waste;
- (4) Sometimes old vaults contain only raw waste which needs to be retrieved, sorted out and repackaged before further characterization.

Thus, mainly for radiation protection purposes, location and retrieval of sealed radioactive sources will in many cases need to be carried out for comprehensive characterization. Visual inspection and dose rate cartography (gamma and neutron) of accessible waste should be carried out before any retrieval operations.

For each of the described methods the applicability to the location and characterization of sealed radioactive sources in historical waste is discussed and advantages and disadvantages are highlighted.

4.1. VISUAL INSPECTION

Visual inspection is a very useful technique for identification of a number of parameters, which will assist in the identification and characterization of sealed radioactive sources. These parameters include information on and the physical condition of the shielding/housing of the sealed radioactive sources (e.g. marking and labelling). Identification of sources is eased by the recognition of associated shielding and equipment. The shapes and sizes of sources depending on their purposes vary considerably; however, the specific design of devices or housings containing radioactive sources may assist in visual recognition of the sources. Data collected in the past of typical sealed radioactive sources encapsulating and holding structures could give an indication of which radionuclides might be present and their approximate activity content.

The IAEA has developed a comprehensive system of data on sealed radioactive sources. The system includes data on the actual sealed radioactive sources, as well as on the devices housing the sources. The package is in the form of a computer database and is called the International Catalogue of Sealed Radioactive Sources and Devices [19].

This catalogue provides vital information for a wide range of individuals and organizations on industrially manufactured radioactive sources and devices, designated as 'source models' and 'devices models'. This catalogue facilitates source identification based on limited information available (or 'found') for given radioactive sources or devices, and thereby assists in handling them safely.

The catalogue utilises a vast amount of data to provide information for a given unknown source or device for which only a few data are available. The information is managed through three main linked databases.

- (1) Source models: This database contains data on source models (i.e. radionuclides, activity, category, shape, size, manufacturing details, uses, manufacturer/distributor, etc.);
- (2) Device models: This database contains data on device models including transport containers (i.e. description, applications, manufacturer/distributor and identification of authorized sealed sources inside the device);
- (3) Suppliers: This database contains addresses of manufacturers and distributors of sealed radioactive sources and devices housing such sources.

The catalogue options include an identification system (browser), in which the inquirer fills in a form displayed on the screen. When the form is completed, the catalogue will deliver the identity of the most likely sources or devices.

Mirrors, magnifier glasses, binoculars, remote controlled video cameras and others can be used for visual inspections and are highly recommended for obtaining initial information subject to compliance with the ALARA principle. Visual observations can provide information, which can be cross-referenced to entries in the inventory.

Endoscopy, the use of small remotely operated cameras, is a simple technique which can be used when access to the source is difficult. It could be used for remote visual inspection of sources and containers. For example, it could be used for detection of markings/labelling of sealed radioactive sources, or evidence of corrosion and moisture inside the waste containment system. The data may be captured and stored using digital cameras and/or video recorders.

The main advantages of the visual inspection methods are:

- Fast, simple and easy to use;
- Tools are cheap.

The main disadvantages of these methods are:

- Installation and implementation may involve a high radiation risk for the operators;
- Limited to qualitative information;
- Accessibility to the waste may limit applicability.

4.2. BASIC RADIATION DETECTION SYSTEMS

Most spent sealed sources will have associated beta and/or gamma emissions. These can be detected with unsophisticated equipment such as an end window or a thin walled Geiger-Müller (G-M) tube monitor. Scintillation monitors, if available, are preferred because of their shorter response times. A very useful addition would be the fitting of the monitor with an audible output so that the operator would not need to watch the meter constantly.

This basic equipment is not applicable or not reliable if the source is an alpha or neutron emitter, or if a beta source is positioned in a shielding housing. Instruments sensitive to alpha or neutron radiation may be required if sources with such radionuclides are expected to be encountered.

From the detection point of view, shielded alpha or beta sources are the most difficult to detect. For beta sources, in the absence of measurable levels of Bremsstrahlung, an end window G-M tube monitor is probably best suited, provided safe opening of the shield can be arranged and the taking of measurements can start at several metres distance, to avoid saturation of the G-M counter.

The detection of neutron sources will not always require access to a neutron monitor. Some gamma or X ray emissions associated with $^{226}\text{Ra-Be}$ and $^{241}\text{Am-Be}$ neutron sources can be detected by using a G-M tube or thin crystal scintillation monitor. This indirect detection technique will, however, introduce uncertainties caused partly by the low energy of the photons ($^{241}\text{Am-Be}$) and partly by shielding itself. It may also lead to a significant underestimation of the risks involved in future management of the source.

Where several sources of widely different activities need to be detected, a collimated monitor with defined angular response may be particularly useful.

Detecting high activity sources such as teletherapy sources, known or suspected to be partly or wholly unshielded, requires special considerations. Because of the high radiation field, conventional or readily available equipment may saturate and resulting readings will be unreliable.

Alpha, beta and low energy X and gamma ray contamination monitors could be used for detecting source leakage by the wipe test method.

Inferential techniques based on key nuclides, which may be used to characterize well-qualified waste generation and conditioning processes, are not applicable for characterization of sealed radioactive sources.

4.3. LOCATION OF SEALED RADIOACTIVE SOURCES BY GAMMA CAMERA

A special instrument that may be used and recommended for location of sealed radioactive sources and hot spots in historical waste is the gamma camera. This device allows images to be made of the gamma ray intensity and its 2D distribution superposed on a picture or video image. This allows the position of the radioactivity to be identified and an initial qualitative estimation to be made of the source intensity. The images of the gamma rays are obtained by using a pin hole collimator and a detector array which allows the gamma rays to be detected and their direction and intensity determined. Dedicated software is used with these cameras to represent the gamma radiation as a false colour picture.

Gamma cameras are typically used in dismantling operations for pre-characterization of the facility, or to identify hold-up of nuclear materials in complex processes (See Fig. 14). Since the pin hole collimator strongly reduces the gamma ray flux that will impinge on the array detector, coded aperture collimators are currently under development to produce the images.

The main disadvantage of the gamma camera is its high cost. Nevertheless, its use should be recommended as often as possible because of its great ability to locate highly active sealed radioactive sources in historical waste at distance and good radiation protection conditions.

4.4. DOSE RATE METHODS

Dose rate measurements can provide an indication that a sealed radioactive source is present as a hot spot in the waste. Hot spots can be detected by slowly scanning the waste surface and observing the recorded dose rate and point by point measurements on a predefined grid. Where possible, the measurement should be performed automatically to minimize exposure to the operators and to provide better data traceability.

Gamma dose rate measurements give the primary information on the existence of gamma radiation and can under certain conditions (use of collimators) be used to determine the approximate location of the sealed radioactive sources and to assess their activity. The activity of the sealed radioactive sources can be quantified by using the measured dose rate, the dose to activity conversion factor and the nuclide composition of the sealed radioactive sources (if known).

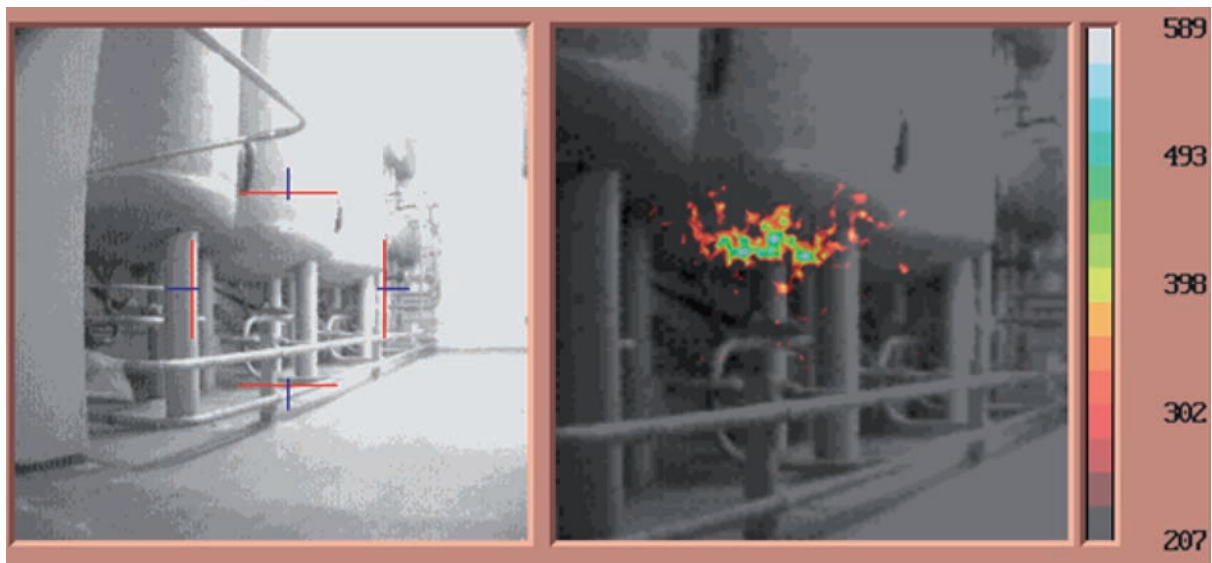


FIG. 14. Typical use of a gamma camera to identify hold-up of radioactivity in a tank. The picture at the right shows that radioactivity is concentrated at the bottom of the tank.

For a mono-nuclide point source the following equation can be used:

$$P = AK/r^2$$

where P = dose rate (mSv/h);

A = activity (GBq);

K = conversion factor (mSv·m²·h⁻¹·GBq⁻¹);

r = distance (m).

This approach relies on the ability to calculate the effect of shielding (e.g. all encapsulation structures) on the measured dose rate. It is advised to use the appropriate calculation models and build-up factors, and take account of the shielding structures and measurement geometry. Commercially available software programs are often used for this purpose [32].

Neutron dose rate measurements using appropriate neutron detectors and hand-held equipment may make it possible to detect neutron sealed radioactive sources or to confirm the presence of such sources. Usually, only neutron sealed radioactive sources with significant activities (>10 GBq ²⁴¹Am/Be for instance) could be detected in historical waste at short distances (a few meters).

The main advantages of these methods are:

- The measurement devices are not expensive;
- The instruments are easy to use;
- The measuring time is generally quite short.

The main disadvantages of these methods are:

- Dose rate measurements are insufficient to fully characterize the sealed radioactive sources because they require additional detailed information such as radionuclide composition, encapsulation materials, structure materials, matrix and shielding.
- The absence of hot spots does not necessarily mean that sealed radioactive sources are not present, for instance, if sealed radioactive sources are inside a shielded block or container.

It may generally be necessary to obtain additional information using other more advanced techniques (e.g. gamma assay methods, imaging techniques) performed on sources conditioned in packages.

4.5. GAMMA ASSAY METHODS

4.5.1. General considerations

Gamma assay techniques are mainly applicable in the following cases:

- (a) Waste packages retrieved from historical waste disposal or storage units contain sealed radioactive sources enclosed in a drum with concrete lining (homogeneous matrix) and withstanding handling without major reconditioning;
- (b) Historical waste (raw waste or damaged packages) repacked in containers (drums, metal overpacks, etc.) and characterized before sorting and reconditioning of waste.

These methods are applicable for sealed radioactive sources containing nuclides with gamma emission intensities and energies high enough to allow their detection. For instance, location and characterization of some ^{238}Pu sealed radioactive sources in waste packages would be difficult or even impossible.

Gamma assay techniques are based on gamma spectrometry. Most detectors used in gamma spectrometry use either NaI(Tl) scintillation (low resolution), HPGe (high purity germanium) semi-conductor (high resolution) and CdTe (medium resolution) detectors. Systems varying from hand-held devices to specially installed systems using these methods have been available for a number of years [21].

Low resolution systems require less maintenance and no cooling. A disadvantage is that only simple spectra (less than 10 well resolved peaks in an energy range from 35 keV to 1500 keV) can be collected and analysed. However, it can be used to identify sealed radioactive sources in historical waste if the activity level of sealed radioactive sources is high enough and taking into account that sealed radioactive sources are mainly 'mono' nuclide sources.

Medium resolution systems (CdTe) are a good compromise between low and high resolution systems. High resolution systems require cooling (liquid nitrogen or electro-cooling) and are normally expensive and less robust than room temperature devices. The advantage of a high resolution system is the ability to deal with a very high number of different gamma ray peaks.

Gamma spectrometry allows the identification of the different radionuclides and under certain conditions their activity. For quantitative analysis, two main parameters need to be known:

- (1) Gamma ray absorption in the waste matrix and/or shielding material of the sealed radioactive sources; this is mainly accomplished using a priori information (e.g. measurement geometry, density, matrix composition) or transmission measurements;
- (2) The spatial distribution of sealed radioactive sources in the waste.

The assessment of the gamma ray absorption by the waste and the spatial distribution of sealed radioactive sources can be obtained by dedicated gamma scanning techniques. Gamma scanning sequentially collects gamma spectra at different locations at the surface of a waste object. Complexity of gamma scanning methods can vary from simple manual measurements up to fully automated systems.

Different automated gamma scanning techniques have been developed for general waste assay purposes and may also be applicable for characterization of sealed radioactive sources in historical waste. An overview of typical scanning techniques for waste characterization is introduced in Fig. 15.

The choice of the suitable scanning technique depends on the required measurement accuracy, the homogeneity of the waste matrix and the distribution of sealed radioactive sources.

These techniques are currently mainly used for characterization of waste streams arising from operation of nuclear facilities, or for some specific radiological survey and expertise of waste packages. Such methods may be used efficiently to detect low and very low activity sealed radioactive sources, e.g. few tens of kBq. Higher activity sources may be detected by more simple methods.

4.5.2. Rotational scanning

Rotational scanning is technically the most simple gamma scanning technique in waste characterization. The detector is located at a fixed height and distance with respect to a continuously rotating waste package. The detector sees the complete waste package (open geometry). The aim of the rotation is to average matrix and source distribution effects (see Fig. 15).

The main advantages of this method:

- Identifies gamma emitting nuclides in the waste;
- Easy to perform.

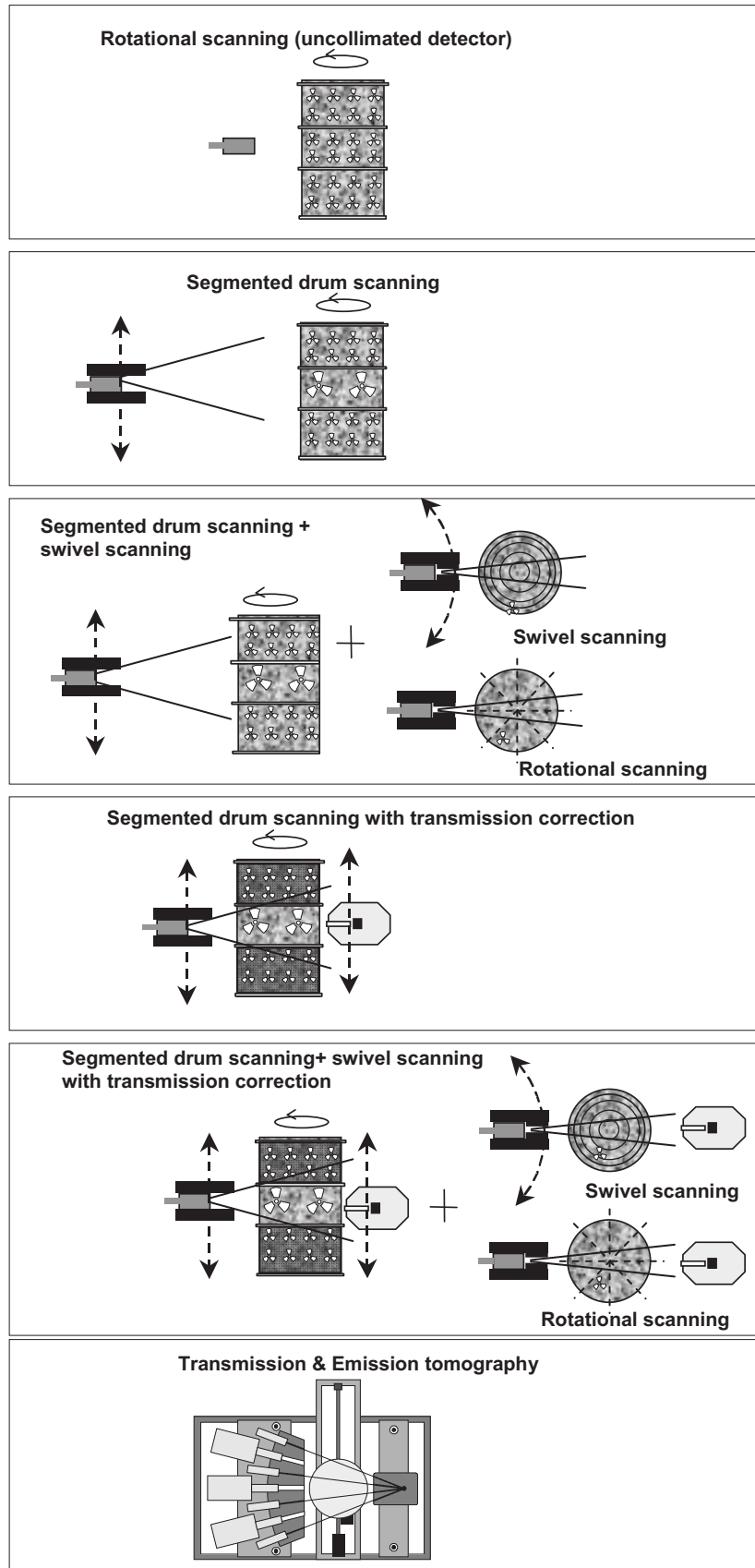


FIG. 15. Overview of available gamma scanning methods.

The main disadvantages of this method:

- This method is not applicable for location of sealed radioactive sources inside a waste package;
- In case a sealed radioactive source is suspected inside a waste package, it may provide limited information which is not sufficient for the full characterization of the sealed radioactive sources.

4.5.3. Segmented gamma scanning (SGS)

SGS is the most commonly applied gamma scanning technique in waste characterization. Gamma spectra are collected at different fixed vertical positions along the waste axes with a collimated detector at a fixed detector–waste distance. The waste package is continuously rotating. In this way a complete waste package is measured segment by segment (see Fig. 15).

The main advantage of this method:

- In the case of a homogeneous density distribution, vertical variations in activity distribution could give an indication of the presence of sealed radioactive sources.

The main disadvantages of this method:

- It cannot discriminate between radioactive waste and sealed radioactive sources per segment (identical to rotational scanning);
- Limited accuracy for determination of sealed radioactive source activity.

Guidance on the use of advanced methods for processing data from SGS and related techniques is provided in Ref. [33]. This publication contains reference iso-plots calculated by the MCNP-X transport code [34] to assist in the interpretation of processed data when investigating the possible presence of sealed radioactive sources inside waste packages (See Section 4.8).

4.5.4. SGS and swivel scanning (SGS/SW)

SGS/SW is an advanced gamma scanning technique allowing determination of the vertical and the radial distribution of the activity in the waste, assuming that the density distribution is homogeneous. The scanning mode is the same as for SGS with the additional feature that the detector swivels on a horizontal plane (see Fig. 15).

The main advantage of this method:

- In the case of a homogeneous density distribution, vertical and radial variations in activity distribution could give an indication of the presence and location of sealed radioactive sources.

The main disadvantages of this method:

- Limited radial resolution of the source distribution;
- Limited accuracy for determination of sealed radioactive source activity, better than segmented gamma scanning;
- Requires sophisticated software and equipment.

4.5.5. SGS and angular scanning (SGS/AS)

SGS/AS is an advanced gamma scanning technique which allows determination of the vertical, the radial and the angular distribution of the activity in the waste package, assuming that the density distribution is homogeneous. The scanning options are the same as those described above for SGS/SW, with an additional

option that the waste package is rotated stepwise. Angular scanning allows the radial and angular source distribution of a selected segment to be taken into account.

The main advantages of this method:

- In the case of a homogeneous density distribution, vertical, radial and angular variations in activity distribution could give an indication of the presence and location of sealed radioactive sources;
- Better accuracy for sealed radioactive source location and characterization than SGS/SW.

The main disadvantages of this method:

- The method is often limited to the determination of 3 to 4 hotspots per segment;
- Requires sophisticated software and equipment similar to SGS/SW.

4.5.6. Transmission corrected SGS (TC-SGS)

TC-SGS is an advanced gamma scanning technique similar to that described above for SGS, but with matrix corrections based on transmission measurements using an external gamma ray source (see Fig. 15).

The main advantages of this method:

- Gives an indication of the vertical variations in activity distribution, which could give an indication of the presence of sealed radioactive sources;
- Gives an indication of the variation of the waste density between segments, which could indicate the presence of shielding material of a sealed radioactive source.

The main disadvantages of this method:

- Cannot discriminate between radioactive waste and sealed radioactive sources in a segment;
- Limited accuracy for determination of sealed radioactive source activity, better than segmented gamma scanning;
- Licensing of external source required and continuous surveillance of the external source is necessary.

4.5.7. Transmission corrected SGS/SW (TC-SGS/SW)

TC-SGS/SW is an advanced gamma scanning technique similar to that described above for SGS/SW, with matrix corrections based on transmission measurements using an external gamma ray source (see Fig. 15).

The main advantages of this method:

- Gives an indication of the vertical and radial variations in activity distribution, which could indicate the presence and location of sealed radioactive sources, assuming a homogeneous density distribution per segment in a waste package;
- Gives an indication about the vertical and radial variations of the waste density between segments, which could indicate the presence of shielding material of a sealed radioactive source.

The main disadvantages of this method:

- Limited radial resolution of the source distribution, compared to TC-SGS/AS;
- Limited accuracy for determination of sealed radioactive source activity, compared to TC-SGS/AS;
- Requires sophisticated software and equipment similar to that for SGS/SW and TC-SGS;
- Licensing of external source required and continuous surveillance of the external source is necessary.

4.5.8. Transmission corrected SGS/AS (TC-SGS/AS)

TC-SGS/AS is an advanced gamma scanning technique similar to that described above for SGS/AS, with matrix corrections based on transmission measurements using an external gamma ray source.

The main advantages of this method:

- Gives an indication of the vertical, radial and angular variations in activity distribution which could indicate the presence and location of sealed radioactive sources, assuming a homogeneous density distribution per segment;
- Gives an indication of the vertical, radial and angular variations of the waste density between segments, which could indicate the presence of shielding material of a sealed radioactive source.

The main disadvantages of this method:

- The method is often limited to the determination of 3 to 4 hotspots per segment;
- Requires sophisticated software and equipment similar to that for SGS/SW and TC-SGS;
- Licensing of external source required and continuous surveillance of the external source is necessary.

4.5.9. Transmission and emission tomography

The combination of transmission computed tomography (TCT) with emission computed tomography (ECT) is the most advanced gamma scanning method and allows a very accurate determination of the density and source distribution in a waste package (see Fig. 15). More information on this imaging technique is given in Section 4.6.3.

4.6. IMAGING TECHNIQUES

Imaging techniques are used to identify any variation in the distribution of density and/or attenuation coefficients in waste, which may indicate the presence of sealed radioactive sources. As shown in Fig. 16, imaging techniques can be categorized according to the type of external radiation source (except autoradiography) used for the creation of the images (e.g. X rays, gamma rays and neutrons), measuring technique (e.g. film/converter, digital systems, gamma detectors) and processing of data (e.g. radiography, tomography). The principles of these photon and neutron imaging techniques are similar; however neutrons interact with the nucleus while photons interact with the electron clouds around the nucleus. The mass attenuation increases with increasing atomic number in case of photon imaging. However, there is no such simple rule for neutrons [26].

For implementation of these techniques, waste has to be conditioned in packages, preferably in 100 L or 200 L drums. Thus, raw waste or waste contained in damaged containers has to be reconditioned before characterization. Before reconditioning, the most active sealed radioactive sources might be identified using more simple techniques (e.g. dose rate measurement).

4.6.1. Radiography

4.6.1.1. Photon radiography

X ray imaging techniques are suitable for assay of low density waste. They work on the principle that X ray or gamma ray beams are directed at and attenuated in waste.

Radiography can identify high or low density regions within a waste matrix. Higher density regions could be indicative of lead shielding which may be associated with sealed radioactive sources. On the other hand, lower density regions may be indicative of the presence of shielding around neutron sources (e.g. hydrogenous materials) or of low atomic number sources.

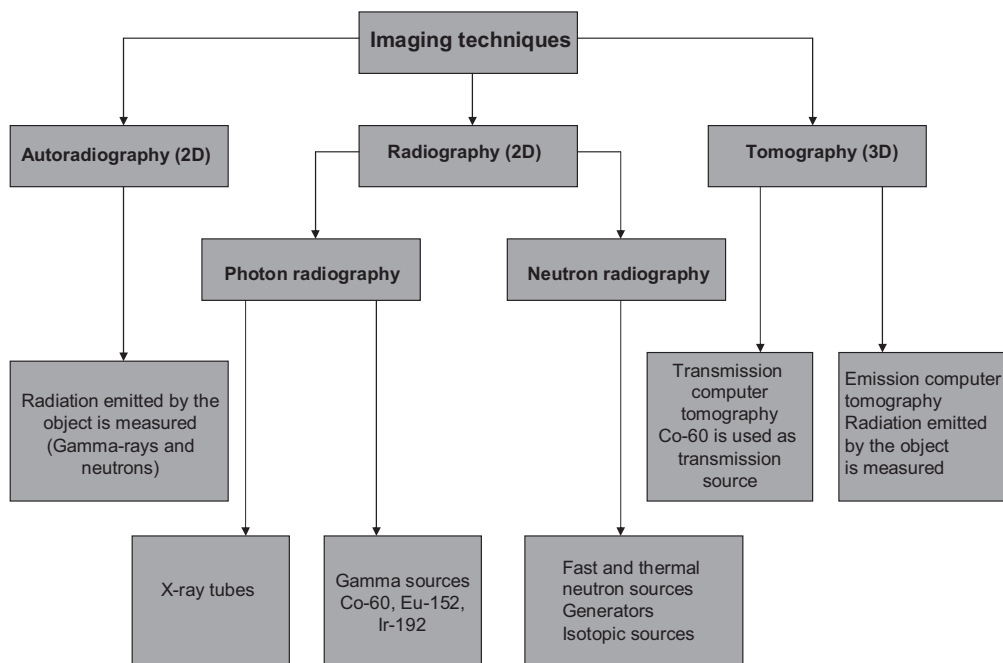


FIG. 16. Schematic description of commonly used imaging techniques. The radionuclides in the figure refer to the external sources.

High energy photon (gamma and X rays) imaging is applicable for the characterization of high density or large waste packages.

In film radiography the transmitted radiation is absorbed by photographic film and it provides qualitative information on the inspected item. In digital radiography the transmitted radiation is absorbed by a digital detection system. In addition the latter can provide quantitative information such as the attenuation correction factor for determining the absolute activities. In both cases, images represent attenuation along the different beam paths through the waste. Digital radiography with ^{60}Co as the external source is currently the most commonly used imaging technique for characterization of radioactive waste.

Imaging techniques can be operated in low or high resolution mode. The typical resolution of photon imaging for waste characterization is in the range of about two to ten millimetres. Low resolution is adequate for most applications in waste characterization, but may not be sufficient for identifying small unshielded sources.

The main advantages of these methods:

- Radiation sources and detection systems are readily available;
- The technology is mature and commercially available at a relatively low cost for 100 L and 200 L drums;
- The technique can be adapted for waste of varying shapes and densities;
- Digital radiography has a high throughput in comparison to other applicable imaging techniques.

The main disadvantages of this method:

- Detection of sealed radioactive sources is negatively affected by the presence of shielding materials;
- Discrimination between materials which have similar scattering and absorption properties is not possible;
- The equipment is expensive for large size packages.

Figure 17 illustrates radiographs of low and high density waste packages.

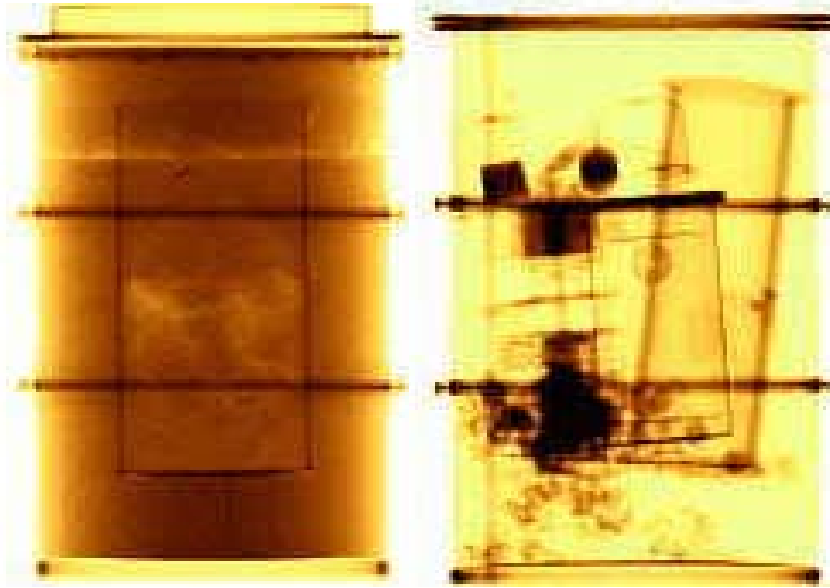


FIG. 17. Gamma ray digital radiographs (^{60}Co) of two different waste drums. The left image shows inactive concrete lining containing cemented evaporator concentrates in an inner canister. The right image shows a radiograph of a drum of raw waste.

4.6.1.2. Neutron radiography

Each atomic nucleus has a different attenuation coefficient, depending on its absorption and scattering cross-sections. Elements with adjacent atomic numbers can have widely different attenuation coefficients for neutrons, and this varies from element to element, even between isotopes of the same element [26]. In contrast to photons, neutrons are not strongly attenuated by materials of high atomic numbers like Pb, W and depleted uranium, which are commonly used as shielding materials. This can be exploited by the inspection of shielded sealed radioactive sources or identification of regions with similar atomic numbers as the matrix. In addition, neutrons are strongly attenuated in light density materials containing elements like H, Li, Be, B, which allow the identification of low atomic number regions inside a waste matrix.

Neutron radiography can be categorized according to the energy of the neutrons, i.e. as thermal and fast neutron imaging. The penetration depth of neutrons into materials depends on their energy. Fast neutrons have a higher penetration depth than slow neutrons and the penetration depth increases with increasing energy. Fast neutrons can be also used as an element sensitive imaging method. If the neutron energy matches a resonance corresponding to a specific element, the neutrons scattered by that element will give relatively more contrast than the surrounding matrix. In addition, prompt (n, γ) reactions can be used to identify the chemical composition of the waste contents.

Presently, neutron radiography is not commercialised for characterization of waste (because of adequacy of the technique, cost of equipment, high level of expertise needed). However, transportable neutron radiography systems using neutron generators and isotopic neutron sources can be considered as a complementary imaging method to the above photon imaging techniques. The resolution of neutron radiography depends strongly on the detection system. In order to increase the detection efficiency for such applications, the thickness of the neutron converter or scintillators can be increased, but this leads to poorer resolution. Digital neutron radiography cannot be applied to inspection of radioactive material since the emitted gamma radiation affects the quality of the neutron radiography images, causing white spots. In this case, nitrocellulose films, which are insensitive to gamma radiation, may be used [35]. These films are easily handled and can be processed in daylight.

The main advantages of this method:

- High attenuation for elements having low atomic numbers is obtained, which provides a good image contrast for the determination of the location of waste containing low atomic number elements;
- Despite low attenuation for elements having high atomic numbers, the technique allows determination of the location of shielded sealed radioactive sources in waste.

The main disadvantages of this method:

- The costs are high;
- The degree of complexity means that highly qualified personnel are required;
- At present it is mainly a research tool associated with high neutron flux facilities.

4.6.2. Autoradiography

Photons emitted by the waste itself could also be used as a radiography source. Autoradiography may give an impression of the photon flux distribution at the surface of the waste item [31]. Autoradiography uses spontaneously emitted radiation from a radioactive source or any radioactive material to form images on X ray film. The X ray film may be digitized afterwards. In this technique the radiation emitting material and the film are placed in close contact and left for a sufficient time to create the image. This technique can easily and quickly be applied to reveal the presence of sealed radioactive sources in packages of historical waste. Autoradiography can be used as a complementary or alternative method to the X or gamma radiography, since in some cases the penetration power of X or gamma rays does not allow creation of an image of the sealed radioactive sources in the waste drum or package.

The main advantages of this method are:

- Very fast method (in sufficiently high radiation levels) to determine the existence and location of the sealed radioactive sources or of hot spots (e.g. activated material) in a waste drum;
- The method is cheap and easy to apply.

The main disadvantages of this method are:

- Radiation hazard for workers during application of the method;
- Not selective between sealed radioactive sources and other hot spots in waste (such as activated material);
- Interior of the waste drum cannot be imaged/investigated;
- Low radiation level at the package surface cannot produce a recognizable indication on the film;
- Long exposure time for low activity sources is not compatible with industrial conditions of retrieval of waste from historical disposal or storage sites;
- Complicated placement/configuration of a number of sealed radioactive sources with shielding materials may cause insufficient discrimination;
- Interpretation of autoradiograms may be difficult, even impossible without sufficient supporting data.

Figure 18 shows a metal box containing ^{241}Am sources taken from lightning rods and their X ray radiography and autoradiography images. The radiogram taken with an X ray tube and Kodak AA400 film was obtained at 150 kV, 5 mA, 2 minutes exposure time and 700 mm film focus distance. The autoradiography image of the ^{241}Am sources was taken by 5 hours exposure time. As can be seen from the autoradiogram, some of the ^{241}Am sources have high activity, some have low.

The autoradiography technique can easily be applied to detect and locate the sealed radioactive sources in historical radioactive waste packages. Further characterization, such as type of radionuclide, activity, requires other methods. Duration of the process depends mainly on the radiation level at the outside surface of the waste drum.



(a)

(b)

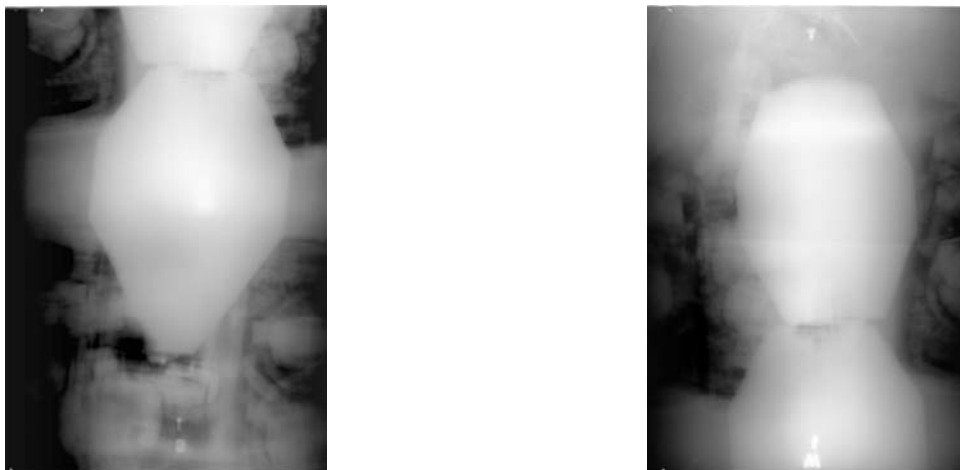
(c)

FIG. 18. (a) A metal box containing ^{241}Am sources dismantled from lightning conductors, (b) X ray radiography image of the box, (c) Autoradiography image of the box.

Fig. 19 shows the digitized ^{60}Co radiograms of a 100 L historical waste drum. It is seen that two similar big sealed radioactive source shieldings (approximately centralized) and several other small shielding had been embedded into cement mortar. Only ^{226}Ra radiations were detected from this historical waste drum by using a portable gamma spectrometer.

4.6.3. Tomography

Transmission computer tomography (TCT) is a technique in which multi-directional measurements using an external gamma ray source (generally ^{60}Co) are used. These provide data to reconstruct the density distribution of material within the waste by means of complex mathematical methods. In waste characterization,



(a)

(b)

FIG. 19. ^{60}Co radiograms of a cemented drum containing two similar and other types of shielding containers; (a) bottom part, (b) upper part of the drum

TCT is generally used in combination with emission computer tomography (ECT). Through multi-directional measurements of the gamma radiation emitted by the waste, ECT allows the reconstruction of the activity distribution of the detected radionuclides. For reconstruction of the ECT image, the local density distributions of material within the waste (obtained by TCT) are used. The techniques, including the density and activity imaging reconstruction software, are well developed, but implementation is complex and has to be supported by highly qualified personnel. The mathematical reprocessing of TCT and ECT data requires high accuracy and therefore good counting statistics, which is associated with long counting times. The high resolution mode of TCT allows determination of small spatial variations in the density distribution (around 5 mm) in waste and therefore determination of the location of small sources. The high resolution mode of ECT allows identification of small radioactive point sources (around 5 mm). The combination of TCT and ECT can be used to confirm that a density variation is a radioactive source and also to quantify the source activity.

At present, the TCT/ECT technique is selectively used for the characterization of historical waste with very complex matrices.

The main advantages of this method:

- Tomography systems are commercially available, namely for 100 L and 200 L drums;
- Systems can be adapted to various waste items of varying shapes and densities;
- Location of sealed radioactive sources inside packages containing shielding materials is possible;
- Accurate quantification of activity when TCT and ECT are combined is possible.

The main disadvantages of this method:

- System costs are high;
- The degree of complexity means that qualified personnel are required for operation, calibration and maintenance;
- Limited throughput in high resolution modes of TCT and ECT;
- Long time for measurement and processing is needed.

Figures 20 and 21 illustrate an application of waste characterization using TCT in high resolution mode.

4.7. NEUTRON COUNTING METHODS

4.7.1. General considerations

Neutron counting is the generic name that is used to indicate a number of techniques that rely on the detection of neutrons. Neutron counting is only applicable to sources that emit neutrons by, for example, one of the following processes [22–24]:

- (a) Neutrons emitted from (α , n) reactions, like in $^{241}\text{Am}/\text{Be}$, $^{244}\text{Cm}/\text{Be}$, $^{239}\text{Pu}/\text{Be}$ or $^{238}\text{Pu}/\text{Be}$ sources. The transuranic isotopes emit alpha particles and these interact with beryllium or other light elements to produce neutrons.
- (b) Neutrons emitted during the spontaneous fission of radionuclides like ^{238}Pu , ^{240}Pu , ^{242}Pu , ^{242}Cm , ^{244}Cm and ^{252}Cf . In the spontaneous fission reaction a group of neutrons, containing on average 2 to 3 neutrons (prompt neutrons), is simultaneously emitted.
- (c) Induced fission in fissile materials such as ^{239}Pu , will result in simultaneously emitted neutrons. These fission neutrons may also induce new fissions and hence lead to neutron multiplication.

The most commonly used neutron detector in non-destructive assay systems is the ^3He gas proportional counter. The ^3He counter is a thermal neutron detector and it is used for detection of neutrons by combining it with a moderator, e.g. high density polyethylene. Some types of hand-held neutron dose rate meters are equipped with BF_3 detectors that could allow detecting neutron sources by contact measurement around waste packages.

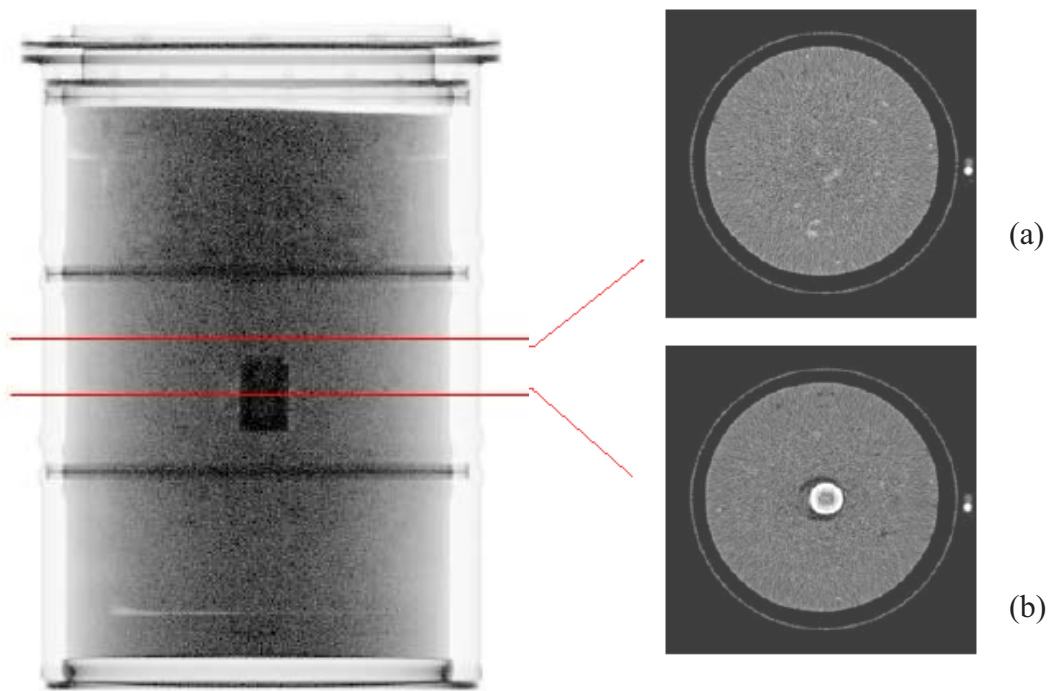


FIG. 20. The left diagram shows a digital radiograph of a waste containing a high density object. The right diagram shows tomograms obtained at two different drum heights, above (a) and through a dense object (b). The measurements were performed with an external ^{60}Co source.

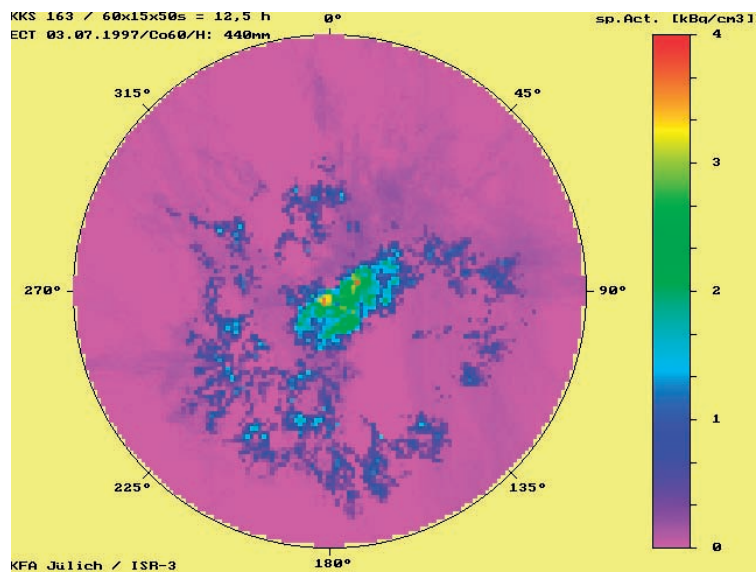


FIG. 21. ECT image showing the distribution of radioactivity (^{60}Co) in a segment of a cemented waste drum.

To obtain high detection efficiency, single detectors are combined in detector arrays. The detector configuration can either be variable (e.g. slab counters), or fixed (e.g. drum counters based on a measurement cell). Neutron counting techniques are generally subdivided into passive and active neutron counting techniques [24, 25, 27].

Passive neutron counting techniques measure the neutrons spontaneously emitted by the sealed radioactive sources. Passive neutron techniques are generally also subdivided according to the specific counting electronics used. Gross neutron counting or total neutron counting, coincidence neutron counting and neutron multiplicity counting are the techniques applied in non-destructive assay of waste. Passive neutron counting techniques have been successfully used to detect neutron sealed radioactive sources in waste packages.

Active neutron counting techniques use interrogation with an external source to trigger some reactions. For example when neutron interrogation is used, secondary neutrons are produced in the fissile or fissionable nuclides. Photo-fission may also be used to create secondary neutrons, although this technique is not commonly applied in waste assay, and not for detection of sealed radioactive sources. Active neutron techniques are complex, expensive and have limited applications in relation to sealed radioactive sources. They would allow detection of sealed radioactive sources containing ^{239}Pu inside a waste package if the activity of the neutron source is high enough.

It should be mentioned that beryllium contained in most of disused neutron radioactive sources is a highly toxic material that has to be considered in the inventory of waste accepted for disposal.

4.7.2. Passive neutron techniques

4.7.2.1. Gross neutron counting

Gross neutron counting is the simplest neutron counting technique from the instrumentation point of view. Every source of neutrons can in principle be measured by gross neutron counting if the activity is high enough. However, to perform quantitative analysis the following information needs to be known:

- (a) The relative contribution of all of the different neutron producing processes, e.g. (α , n)-neutron rate, fission neutron rate, neutron multiplication.
- (b) The isotopic composition of the source, e.g. a $^{239}\text{Pu}/\text{Be}$ source may actually also contain other isotopes of plutonium with different neutron production rates. The Pu isotopic composition may be determined by gamma spectrometry.
- (c) The absolute detection efficiency for the sealed radioactive sources in the waste that is measured. This generally requires a procedure to correct for matrix effects and source distribution effects.

Gross neutron counting can also be applied to determine the location of hot spots based on neutron count rate distributions [24]. A count rate distribution may be obtained from a passive neutron waste assay system that records the count rates in each or a group of detectors (see Fig. 22 below). Count rate distributions may also be obtained with a slab counter that is successively placed at different orientations relative to the waste.

The main advantages of this method:

- Easy to use.
- May be used as a method to determine the presence of neutron sources. This method may be combined with gamma scanning.
- Under certain conditions, gross counting may be used to provide a conservative upper limit of the source activity.

The main disadvantages of this method:

- High cost of the measurement system compared to the objectives to be achieved.
- Of limited use for quantitative assessment.

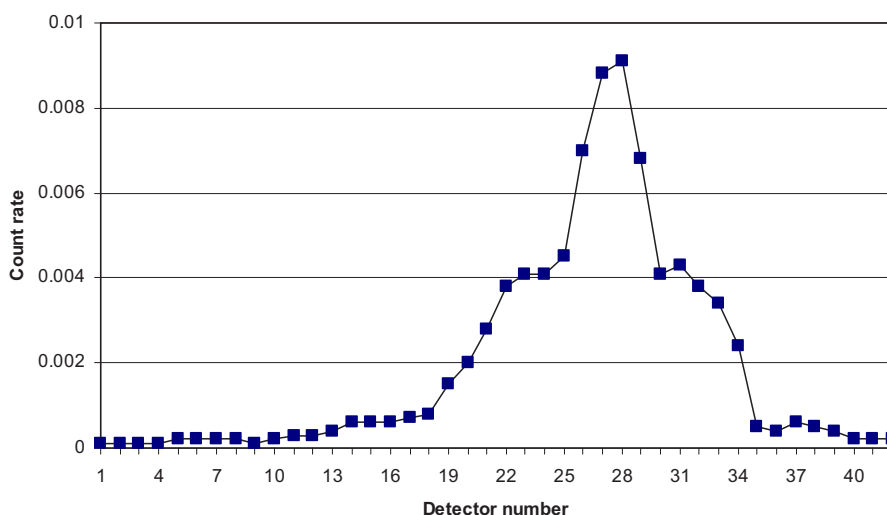


FIG. 22. Count rate distribution obtained with a point neutron source inside waste which was measured by a hexagonal detector configuration containing 42 detectors around the waste package.

Radionuclides in sealed radioactive sources which may be detected in non-alpha bearing waste are the following:

- (a) ^{252}Cf ;
- (b) $^{241}\text{Am}/\text{Be}$, $^{244}\text{Cm}/\text{Be}$, $^{239}\text{Pu}/\text{Be}$: above few MBq, depending on the system detection efficiency;
- (c) ^{238}Pu (difficult to detect, even pacemaker sources).

Gross neutron counting is not applicable for smoke detectors and calibration sources.

4.7.2.2. Passive neutron coincidence counting (PNCC)

Since the production of (α , n) neutrons depends on the chemical and physical composition of the source there is no constant correlation between the alpha activity and the neutron production. If the radionuclide also emits fission neutrons then the quantification can be based on selectively counting fission neutrons. This is achieved by coincidence counting that counts only those neutrons that are emitted simultaneously (within a specified time window). Generally, coincidence counting electronics also provide the gross neutron count, so that two independent measurement quantities are obtained.

For quantitative analysis the following parameters need to be known:

- (a) The isotopic composition of the neutron sources.
- (b) The different processes leading to coincident neutrons (e.g. fission neutrons from spontaneous or induced fission (when multiplication is present); for example (n , $2n$) interactions in Pu/Be sources ^9Be (n , $2n$) ^8Be will contribute to the coincidence count rate) and other related factors.
- (c) The absolute detection efficiency of the sealed radioactive sources in the waste, taking account of matrix effects and source distribution. The add a source (AAS) [25] technique uses an external neutron source to measure the matrix effects. However, the AAS technique may fail on waste containing shielded sealed radioactive sources.

The main advantages of this method:

- Discrimination between randomly emitted neutrons and simultaneously emitted neutrons, reducing the number of unknown parameters in the analysis;

- Comparing the gross count rate with the coincidence count rate may, under certain conditions, provide information on the radionuclide in the source.

The main disadvantages of this method:

- Coincidence count rate is proportional to the square of the detector efficiency; hence relatively high detection efficiency is required to apply this method;
- Applicability of coincidence counting may be limited by a high accidental coincidence count rate resulting in high measurement uncertainty;
- Needs matrix corrections (using for example ASS technique);
- Needs assumptions on the source distribution and isotopes.

The main radionuclides in sealed radioactive sources which could be detected in non-alpha bearing waste are the following:

- (1) ^{252}Cf ;
- (2) $^{244}\text{Cm/Be}$.

Passive neutron coincidence counting is not applicable for other neutron sources, smoke detectors and calibration sources.

4.7.2.3. Neutron multiplicity counting (NMC)

Neutron Multiplicity counting (NMC) [36] is a technique that determines the number of neutrons simultaneously detected (in practice within a small time frame) from a radioactive source.

Based on a physical model describing the neutron production and detection processes, the detected neutron multiplicities can be related to the emitted neutron multiplicities. The method allows determination of equivalent source strength without prior calibration and takes account of matrix effects and source position. The equivalent source strength is generally expressed in terms of a ^{240}Pu equivalent mass. However, when applied to neutron sources, the physical model should include the entire different neutron producing reactions, e.g. in Pu/Be sources, ^9Be (n, 2n) ^8Be should also be considered.

The main advantages of this method:

- Prior knowledge of the neutron detection efficiency is not required.

The main disadvantages of this method:

- Isotopic composition of the source needs to be known.
- Assumes that all neutrons are emitted from a single point. When two distinct sources are present in the waste, the analysis will be biased.
- Limited use at high count rates (i.e. $>1\text{E}5$ counts/s).
- Very sensitive to the effects of counting losses.
- Requires high detection efficiency.

The method is mainly applicable for high activity Pu/Be sealed radioactive sources. Passive neutron coincidence counting is not applicable for most other neutron sources, smoke detectors and calibration sources.

4.7.3. Active neutron techniques

4.7.3.1. General considerations

In active neutron counting techniques (neutron interrogation techniques) the sealed radioactive source is irradiated with neutrons produced by an external neutron source, e.g. a neutron generator or an isotopic source (e.g. ^{252}Cf) or high energy photon beams. The technique may be applicable to sealed radioactive sources that contain fissile material that produce secondary radiation (prompt and delayed neutrons) as a consequence of the induced fission reaction. To increase the interaction rate, thermal neutrons are generally used. For that purpose, those external source neutrons, which have much higher neutron energy, are first thermalized in the walls of the measurement cell containing the waste package. The production of secondary neutrons by induced fissions depends on the neutron flux produced in the waste. This flux depends on the matrix properties of the waste (e.g. moderation, absorption) and needs to be quantified in each measurement. For that purpose, flux monitors are commonly used. In neutron interrogation it is necessary to discriminate between the neutrons produced by the external source and the secondary neutrons produced by the sealed radioactive sources. This is generally achieved by pulsing the interrogation source.

The most widely used neutron interrogation technique is the californium shuffler, which is described below. There are some additional interrogation techniques which are not widely used, but for completeness are summarized below.

Fixed source irradiation systems use an external source (e.g. $^{241}\text{Am}/\text{Li}$ or $^{124}\text{Sb}/\text{Be}$) that is fixed in the measurement cell [21]. The external source continuously exposes the sample to neutrons. The detectors are mainly sensitive to the high energy fission neutrons while relatively insensitive to the low energy source neutrons. Moreover, the relative position of the interrogation source and detectors is optimized to reduce the detection of the interrogation neutrons.

The photon interrogation technique uses high energy photons (6 MeV) to cause photo fission, and the fission neutrons are then counted in this active neutron counting technique [24]. The high energy photons are created as Bremsstrahlung photons by high energy electron beam accelerators. The end point energy is chosen to be above the threshold for photon fission, but below the threshold for most other photon–neutron reactions. This technique is not commonly applied in waste assay. However, the use of high energy photons may circumvent the flux reduction problems from which the neutron interrogating techniques suffer when interrogating sealed radioactive sources in neutron shields.

4.7.3.2. Californium shuffler

Waste is exposed to neutron radiation from a californium source, which is mechanically shuffled back and forth between the irradiation position and the shielding position of the ^{252}Cf source. The relatively long time needed to withdraw the source prevents the detection of the prompt fission neutrons, therefore only the delayed neutrons can be measured.

The main advantages of this method:

- Relatively simple active interrogation method;
- Californium shuffler may be installed in a neutron passive system;
- Increased sensitivity over passive neutron methods for the main fissile isotopes.

The main disadvantages of this method:

- Strong absorption of the ^{252}Cf neutron flux occurs in sealed radioactive sources shielding;
- A safety justification and a licence are required to operate the californium source and shuffler.

The method is mainly applicable for high activity Pu and Pu/Be sealed radioactive sources. Californium shuffler is not applicable for smoke detectors and calibration sources.

4.8. COMPUTER SIMULATIONS AND MODELLING

Illustration of expected radiation flux deviations at the outline of a simulated waste package can be very helpful in assessing a real waste package which might have a complex inner structure. Several sealed radioactive sources can be modelled at different locations of a waste package with different nuclides and activities. Waste matrix can also have a uniform radioactivity and density for simulation purposes. Radiation doses on the surface of this simulated waste package can be calculated. Results of these calculations can be introduced in many different ways, e.g. two dimensional iso-plot coloured graphs. The same type of graph of the real waste package which is produced with real measurements can be compared to the graph produced for simulated waste. Trying different geometries of the content of the simulated waste package may result in close (or identical) graphs of the real one. This is not; however, proof that the simulated content is identical to the real waste content due to other factors that might be in effect; nevertheless this is valuable information for estimation and assessment of the real waste content.

A number of methods for the calculation of expected photon fluxes from a simulated waste package containing sealed radioactive sources are available. In Fig. 23, the results of the Monte Carlo neutron photon transport code (MCNP) are seen as iso-plots of the gamma flux changes as a function of the distance between the ^{60}Co sealed radioactive sources and the drum wall [33, 34]. The affect of the shielding container is also seen. The disturbance due to the uniform background on the sealed radioactive sources signal is clearly observed. It can be noticed that as the distance between the sealed radioactive sources and drum wall increases, the shape of the elevated energy flux (hot spots) due to the sealed radioactive sources changes from circular to elliptic and becomes less visible.

The main advantages of this method:

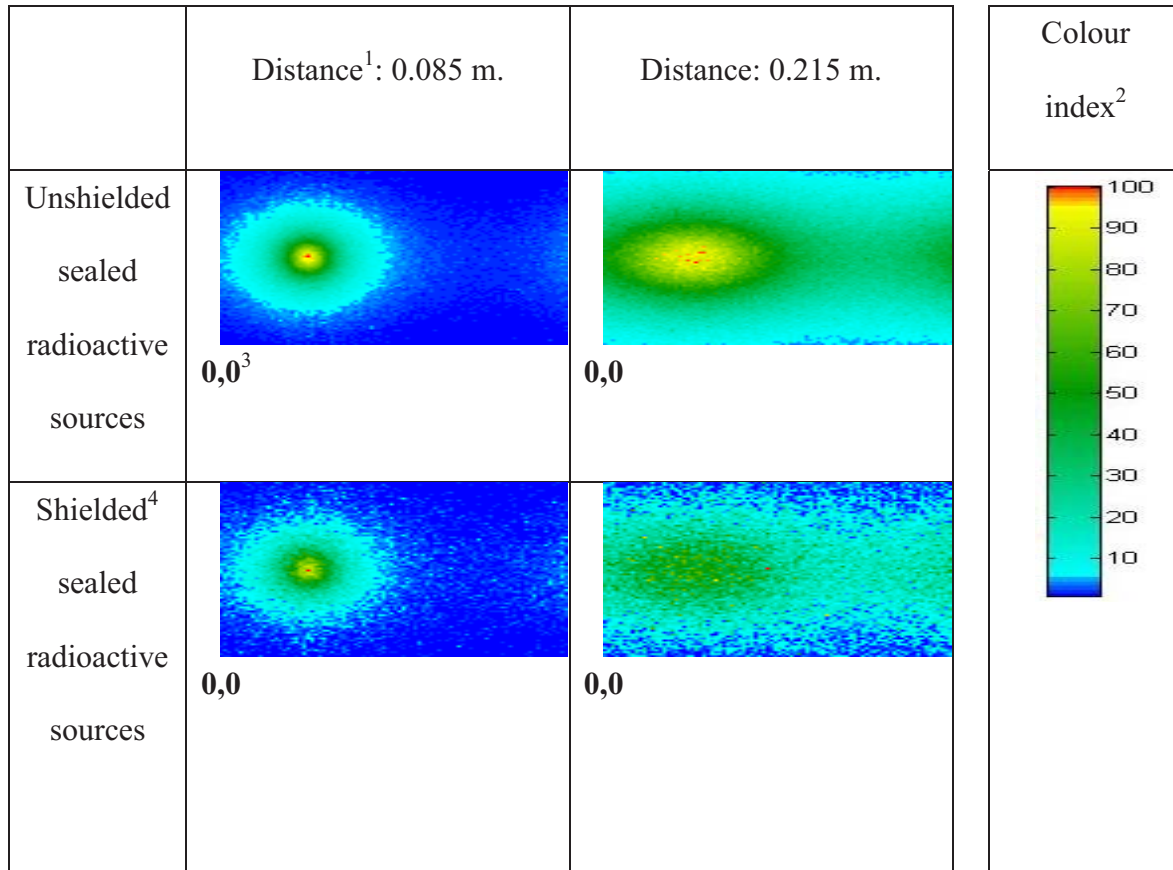
- Iso-plots provide a better understanding of photon fluxes that can indicate the presence of a sealed radioactive source and dense materials like a shielding container in a historic waste package;
- Complexity caused by inhomogeneous nature of historic waste containing immobilized sealed radioactive sources can be addressed to some extent.

The main disadvantages of this method:

- Requires sophisticated software and highly qualified staff.

5. LOCATION AND CHARACTERIZATION SYSTEM FOR SEALED RADIOACTIVE SOURCES

One of the main characterization drivers is to enhance the safety of disused sealed radioactive sources in storage or disposal and to protect the environment from radiation hazards associated with them. For disposal, it is necessary to study and guarantee post-closure safety as explained in Section 2. To do so, generation or completion of the sealed radioactive source inventory is an essential step. When sealed radioactive sources need to be characterized, a characterization system must be set up and performed according to an adequate management system. The sealed radioactive source characterization system must be managed, performed, assessed and controlled to provide adequate confidence that quality requirements and objectives are met. The management system should provide confidence that environmental, technical and characterization requirements are fulfilled.



¹ Distance between the location of the assumed point source and the drum surface where the flux is measured. Iso-plots for 0.085 m and 0.215 m distances are seen. The height of sealed radioactive sources placement from the drum bottom is 0.45 m for both studies.

² This colour bar shows the per cent of the net data values (NDV). Red indicates that NDV is above 95%. Yellow means that NDV is between 85 and 95 %. Green indicates 15 to 85 and light blue gives 5 to 15 % of NDVs.

³ The Y axis of each iso-plot corresponds to the height of a standard 220 L drum (h: 0.9 m, d: 0.6 m) and the X axis to the outline of the drum of 1.8 m. The seam of the drum is assumed to have the coordinates (0,0).

⁴ Shielding is assumed a spherical lead container which has a diameter of 0.15 m.

FIG. 23. Iso-plots of the calculated total energy flux at the outline of a simulated waste package. A 1 mBq ⁶⁰Co sealed radioactive source is placed in two different geometries, with and without shielding, to show the effect of distance and shielding. The MCNC-X transport code was used for calculations [34].

Completing and ensuring accuracy of the inventory is a step wise process which includes the organization of characterization tasks following the different processes of the remedial actions, such as:

- Collection and processing of existing data regarding historical waste and its sealed radioactive source content;
- Preliminary characterization of waste, including sealed radioactive sources before retrieval;
- Characterization of retrieved sealed radioactive sources.

5.1. SPECIFICATION OF THE SYSTEM

Before establishing a sealed radioactive source location and characterization system, it is necessary to provide a specification, which will identify the system requirements. This can be a detailed specification for a

particular piece of equipment, if the waste operator is confident that this equipment is required. Alternatively, a functional specification can be provided which describes the problem (including the tasks to be performed and the required outputs) and allows the supplier to use professional judgement on the choice of system. This may, for example, allow the supplier to propose integrated or single systems, depending on the problem.

The issues which need to be addressed at least by the waste operator include those listed below.

- (1) Details of waste and disused sealed radioactive sources, including their history, age, expected radionuclides and physical form of the waste and sealed radioactive sources, waste matrix including materials (e.g. neutron absorbers), which may interfere with the measurement techniques, dimensions of packaging;
- (2) Details of the overall process, including a description of the facility which will contain the characterization system(s), working environment of the equipment, constraints on the dimensions of the equipment, throughput requirements (e.g. drums/day), maximum times available for measurements;
- (3) Details of interfacing equipment (e.g. if the proposed new characterization equipment is to be used in conjunction with existing equipment);
- (4) The accuracy required, including acceptable precision and sensitivity;
- (5) The acceptance tests required at the factory and at the intended equipment location to demonstrate that the system (including the software) is fit for purpose;
- (6) Identification of any additional validation to be carried out by the purchaser;
- (7) Degree of expertise necessary and available to operate the equipment;
- (8) Statement on required maturity of the equipment, for example if innovative technologies are accepted;
- (9) Spares to be held, identification of times to repair malfunctioning equipment and maintenance frequency requirements;
- (10) Available finances (if this is a constraining issue) and an indication if the equipment is to be hired or purchased;
- (11) Provision of a compliance matrix showing where the proposed new equipment is (and is not) compliant with the above requirements;
- (12) A quality management system which should be documented into procedures, implemented, maintained and continually updated [37–39];
- (13) Provision of operational and training procedures.

The operator needs to provide evidence that all the above concerns are addressed.

5.2. RETRIEVAL OF HISTORICAL WASTE DATA

Data collection, recovery and processing are the first step in generation of the inventory of historical waste [1]. Therefore, assuring the completeness and accuracy of the inventory includes verifying the content and quality of all relevant information. This needs consideration of waste forms, waste characterization already performed and existing radioactive waste inventory, storage or disposal locations. The focus is on the type and radiological characteristics of the sealed radioactive sources in the waste storage and disposal facilities.

During data retrieval it may be necessary to consult former or retired employees who may be able to provide any relevant information. A list of data retrieval actions may include:

- (a) Identification of sealed radioactive sources in historical waste;
- (b) Collation of information on the relevant sealed radioactive sources including physical, chemical and radiological properties, manufacturer's name, drawings, serial number, date of production, users of sealed radioactive sources, leak test certification, measured dose rates, conditioning materials (if applicable);
- (c) Identification of the volumes, amount, mass, radioactivity and locations of all waste packages containing sealed radioactive sources;
- (d) Identification of the numbers of sealed radioactive sources which have been treated or untreated and those still inside their shielding structures;
- (e) For those sealed radioactive sources which have been conditioned, identification of those encapsulated in concrete, polyurethane foam, paraffin wax and/or other matrices;

- (f) Identification of the types of additional shielding used for the sealed radioactive sources (e.g. lead, cement, depleted uranium);
- (g) Identification of the packaging used for conditioned and unconditioned sealed radioactive sources (e.g. plastic bags, specific shielded transport containers, drums, metal and wooden boxes);
- (h) Identification of those sealed radioactive sources which have been inserted into vaults with no backfill, partial backfill and complete backfill;
- (i) Group the sealed radioactive sources (see Section 3);
- (j) Correct the activity of sealed radioactive sources due to decay.

On the basis of the inventory and an understanding of the requirements, a sealed radioactive source location and characterization plan should be produced which includes the justification of selected methodology and techniques (see Section 4). The plan should be in accordance with the waste acceptance criteria of the storage or disposal facility which will receive the reconditioned sources. Moreover, it should be drawn up according to the retrieval and conditioning strategy and techniques already defined.

5.3. PRELIMINARY IDENTIFICATION

Preliminary investigations may be conducted to gain a partial understanding of the location and condition of the sealed radioactive sources and waste in the storage or disposal facility. These preliminary investigations will provide a very useful indication of what to expect during full scale waste retrieval. Some investigations discussed below relate to waste as well as sealed radioactive sources, since these two are inextricably linked. The investigations may include inspections and measurements like those listed below.

- (a) Carry out visual inspections, e.g. by inserting video cameras, endoscopes, mirrors and lights into the waste storage location (where possible) to provide information on:
 - The presence of manufactured devices which could contain sealed radioactive sources inside shielding, increasing difficulty to detect and identify sources used in teletherapy, brachytherapy, industrial radiography or gauges, etc;
 - Identification of potential sealed radioactive sources (for instance metal cylinders), look for labelling and marking on the sealed radioactive source packaging or outer surfaces, which could provide information on the manufacturer, radionuclide activity and date of assessment and other information presented in Section 5.2;
 - Colour coded waste packages which could indicate their origins and possible contents;
 - The condition of the waste packages; for example if the sealed radioactive sources and waste packages are still intact, whether metal corrosion related to the sealed radioactive sources and waste packaging has taken place, and if self compaction of the waste has taken place.
- (b) Insert a dose rate meter into the vaults to provide information on the ambient and surface dose rates near waste items inside the vaults.
- (c) Insert a suitable gamma system to detect variations of gamma radiation inside the vaults.
- (d) Insert a suitable neutron detector to measure neutron dose rate or counting and detect neutron sources.
- (e) If available, use a gamma imaging system (such as Cartogam[®]) to locate hot spots in the accessible layers of waste. This could be used first to remove the first layer of waste/waste packages, then before working on the second layer, and so on.

A strategy regarding retrieval, pre-conditioning, on-site transport, storage, and other waste management steps might be finalized, using the preliminary identification results, complemented with additional health physics safety measurements and information from the waste inventory.

5.4. DETECTION OF SEALED RADIOACTIVE SOURCES INSIDE WASTE

In the case of raw waste inside disposal vaults, before any retrieval operations gamma and neutron dose rate measurements, visual inspections and, if available, gamma images would be performed. This would make it possible to locate the most active and hazardous sources in accessible parts and layers in order to determine how to retrieve them under safe conditions, before commencing retrieval operations.

A case by case approach should be adopted to retrieve these sources, to pack them into adequate shielded containers and to store them.

In the case of packaged waste, after retrieval from the waste storage or disposal facility, the waste package is investigated to determine whether sealed radioactive sources are present. This is one of the most challenging steps in the detection of sealed radioactive sources inside waste:

- (a) Weigh the waste package and record the measured weight:
 - If an unusually high weight is recorded (consider the size of the waste package and fill height) this may be indicative of heavily shielded sealed radioactive sources inside the waste package;
 - If the weight is as anticipated, small shielded and/or unshielded sealed radioactive sources may still be inside the waste.
- (b) Use a gamma dose rate meter to determine surface dose rates in several positions on the waste package:
 - If increased dose rate levels (hot spots) are recorded in localized areas, this could be indicative of shielded or unshielded sources;
 - If no significant variations in dose rate levels are recorded there may be no sealed radioactive sources or non-gamma emitting sealed radioactive sources inside the waste or, worst case, sealed radioactive sources located near the centre of the measured waste package;
 - Use a suitable neutron detector to determine the presence of any neutron emitting sealed radioactive sources in the waste package.
- (c) Use a suitable imaging/radiography technique to identify waste containing high or low density material:
 - If a density variation is observed, this may indicate the presence of shielding material (low or high density);
 - High density could be associated with gamma sources and low density with neutron sources.
- (d) Use a suitable gamma system to determine the possible locations of sealed radioactive sources in the waste and their radiological properties:
 - If a high gamma count rate is recorded in localized areas this might be indicative of unshielded sources or high heterogeneity of waste activity distribution;
 - If there is a large area backscatter region this could be indicative of shielded sources;
 - If specific high energy gamma rays are observed this could be indicative of a neutron sealed radioactive source.

Combine the outputs from the above measurements and derive conclusions on whether shielded or unshielded sealed radioactive sources are present in the waste.

5.5. CHARACTERIZATION

After location of a disused sealed radioactive source in a waste package or in a mixture of waste a characterization plan should be developed. If retrieval of historical data is not possible, all efforts should be undertaken to characterize the sources using the NDA methods specified in Section 4. If there is a lack of information on the sealed radioactive sources and/or non-destructive assay fails to give further information to characterize the sealed radioactive sources in waste, a step-by-step retrieval of the sealed radioactive sources from the waste packages and ultimately from the shielding structures may be considered. At each step of this process, the characterization of the sealed radioactive sources can be performed using basic radiation detection techniques.

During retrieval and characterization operations, all necessary precautions should be taken to ensure that the risk is maintained as low as reasonably achievable.

Priority should be given to retrieval of both gamma and neutron sources with high levels of radiation hazard. In this case sophisticated characterization techniques may not be needed.

Sealed radioactive sources which do not present a high radiation hazard but which are important for the safety of the disposal facility (compliance with waste acceptance criteria) should also be considered for retrieval. In this case, sophisticated characterization techniques may be needed.

The use of sophisticated and costly characterization methods is not justified for sealed radioactive sources which do not represent a radiation hazard and which are not important for the safety of the post-closure phase of the future disposal facility.

After characterization, the sealed radioactive sources need to be conditioned based on the waste acceptance criteria of storage or disposal facilities.

5.6. INSTRUMENTATION

A large variety of instruments are available on the market and even more are being employed by dedicated institutions for specific purposes.

5.6.1. Pocket size instruments

Pocket size instruments can be used for location of sealed radioactive sources in historical waste. The models of this type of instrument should be easy to use, even by non-specialized staff. The purpose is to allow a quick and qualitative assessment of suspect sources. They must be battery powered and shock and water resistant. There also should be minimum requirements for maintenance and calibration. Typically, these instruments are auto ranging and serve as alarm indicators by audio, visual or vibration alarm. The high sensitivity required to detect low activity sources usually requires solid state detectors and rules out classic Geiger-Müller tubes. Even neutron sensitive small devices are available.

5.6.2. Hand-held and portable instruments

Hand-held and portable instruments are used also for location and identification (Fig. 24). They are generally more bulky than pocket sized instruments and exhibit more features. With the requirement for increased sensitivity, more complex evaluation procedures are required, leading to the inclusion of spectroscopic evaluation techniques that yield information on the type of radioisotope (radionuclide identifiers). However, the use of such instruments is more demanding and requires a higher degree of training. Under the guidelines of the response strategy, such instruments are typically employed after initial detection and more detailed characterization of the material detected is desirable, specifically when detailed location or identification is required.



FIG. 24. Typical hand-held instrument used for location and identification of disused sealed radioactive sources.

Some survey meters operate together with telescopic detectors, which allow operators to stay away from radiation sources during measurements. A number of commercially available instruments are available.

5.6.3. Mobile systems

Mobile systems are more complex and bulky but provide better sensitivity than hand-held instruments. Their area of application comprises, for example, investigation of large areas and measurement of waste packages (drums, boxes). Their capabilities are usually tailored to meet the needs of the specific situation, for example with respect to sensitivity, type of radiation detected, package size and cost. Similarly, various methods of data processing and transmission are employed either locally or remotely.

Figure 25 below shows a mobile passive neutron counting system with two slab counters.

5.6.4. Specialized instruments

According to the complexity of a specific case, more elaborate equipment may be employed and more specialized personnel involved in locating and characterizing disused sources in historical waste (See Fig. 26). Though some field laboratory type capabilities have been introduced with recent instruments, allowing complex in situ identification techniques (e.g. high resolution gamma spectroscopy using high purity Ge detectors), certain problems will still need to be referred to laboratories and their highly specialized staff and equipment.

5.7. QUALITY MANAGEMENT

Characterization of sealed radioactive sources must be carried out under an adequate quality management system which should include such elements as organization and instructions, document control, control of purchased materials, process control, non-conformance control and corrective actions, record keeping, audits and training of personnel [37–39].

An important aspect of the quality management system for characterization of sealed radioactive sources is the qualification of the assay method. This would typically include validation studies, calibration (using reference materials), operator training/qualification and routine verification measurements.

Measurement tools and instruments should be checked before their use and periodically calibrated. Calibration should be performed using the certified equipment with reference to recognized standards (or to other databases where no standards exist). The accuracy of the reference materials should be known. This requirement applies to gases, reactants and radioactive sources which are used in analytical applied programmes.



FIG. 25. Neutron slab counter.

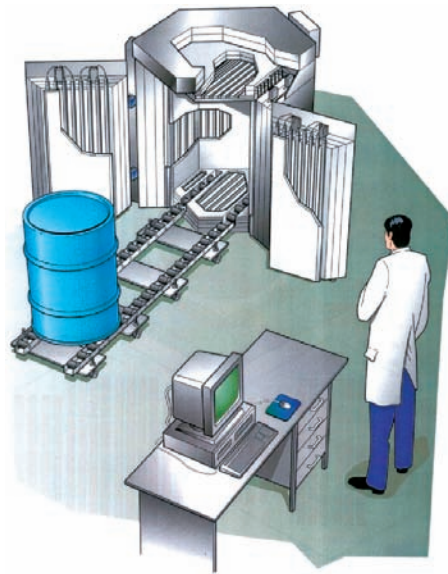


FIG. 26. Generic layout of a passive neutron counting system (showing the ^3He counters) for assay of waste packages.

6. CONCLUSIONS

The main non-destructive methods for location and characterization of sealed radioactive sources in historical waste are introduced. The advantages and disadvantages of each method are identified and the issues, which should be addressed when formulating and implementing the sealed radioactive source characterization system, are discussed. In order to establish and implement a sealed radioactive source location and characterization programme in historical waste, the following recommendations should be considered:

- (a) The reasons and objectives of the work (characterization drivers) should be defined clearly.
- (b) A work plan must be established. This plan is based on the characterization drivers and available (and/or adaptable) technology options considering site specific conditions. The plan should include the radiation protection of workers involved in the location of sealed radioactive sources, retrieval and characterization operations as well as protection of the environment.
- (c) A quality management system must be developed in parallel with the work plan. All activities associated with the characterization of sealed radioactive sources in historical waste must be planned under this system. The system should cover the technical procedures and instructions of each planned activity. This will ensure the reliability of acquired information.
- (d) Operational areas should be checked in view of the planned activities. Equipment and instruments for location and characterization of sealed radioactive sources should be specified and procured.
- (e) A preliminary assessment using simple methods such as dose rate measurement, visual inspections and, if available, gamma imaging (use of gamma cameras), should be performed to locate the most active and hazardous sources in accessible parts and layers of waste and waste packages to be retrieved, prior to performing waste retrieval operations.
- (f) Non-destructive assay techniques should preferably be used. Gamma spectrometry and neutron counting techniques are widely used. Highly active gamma or neutron sealed radioactive sources may not require sophisticated assay instruments.

- (g) Radiography and tomography techniques may be suitable for location of sealed radioactive sources in historical waste packages and in support of the decision to retrieve sealed radioactive sources from unconditioned or conditioned waste packages.
- (h) Destructive sampling and analysis of sealed radioactive sources may be considered when NDA techniques are not applicable or adequate to obtain the required information [40].
- (i) Specific guidance from technical experts on characterization issues should always be sought on a case by case basis.

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CONTRIBUTERS TO DRAFTING AND REVIEW

Bastürk-Tatlısu, M.	Atominstytut der Österreichischen Universitäten, Vienna
Bruggeman, M.	SCK/CEN, Belgian Nuclear Research Centre, Belgium
Ekinci, Ş.	Turkish Atomic Energy Authority, Turkey
Green, T.H.	NUKEM, United Kingdom
Hordijk, L.	South African Nuclear Energy Corporation (Necsa), South Africa
Kahraman, A.	International Atomic Energy Agency
Kupca, S.	Canadian Nuclear Safety Commission (CNSC), Canada
Liu, Xinhue	National Nuclear Corporation (CNNC), China
Mauerhofer, E.	Institut für Sicherheitsforschung und Reaktortechnik (ISR), Germany
Neubauer, J.	Consultant, Austria
Pillette-Cousin, L.	AREVA TA, France
Poluektov, P.	Russian Scientific Research Institute of Inorganic Materials, Russian Federation
Rodrigues, F.	Ontario Power Generation, Canada
Simenon, R.A.C.	Association Vinçotte Nucleaire, Belgium
Tsyplenkov, V.	Consultant, Russian Federation
Velzen, L.P.M.	NRG Arnhem, Department of Radiation and Environment, Netherlands

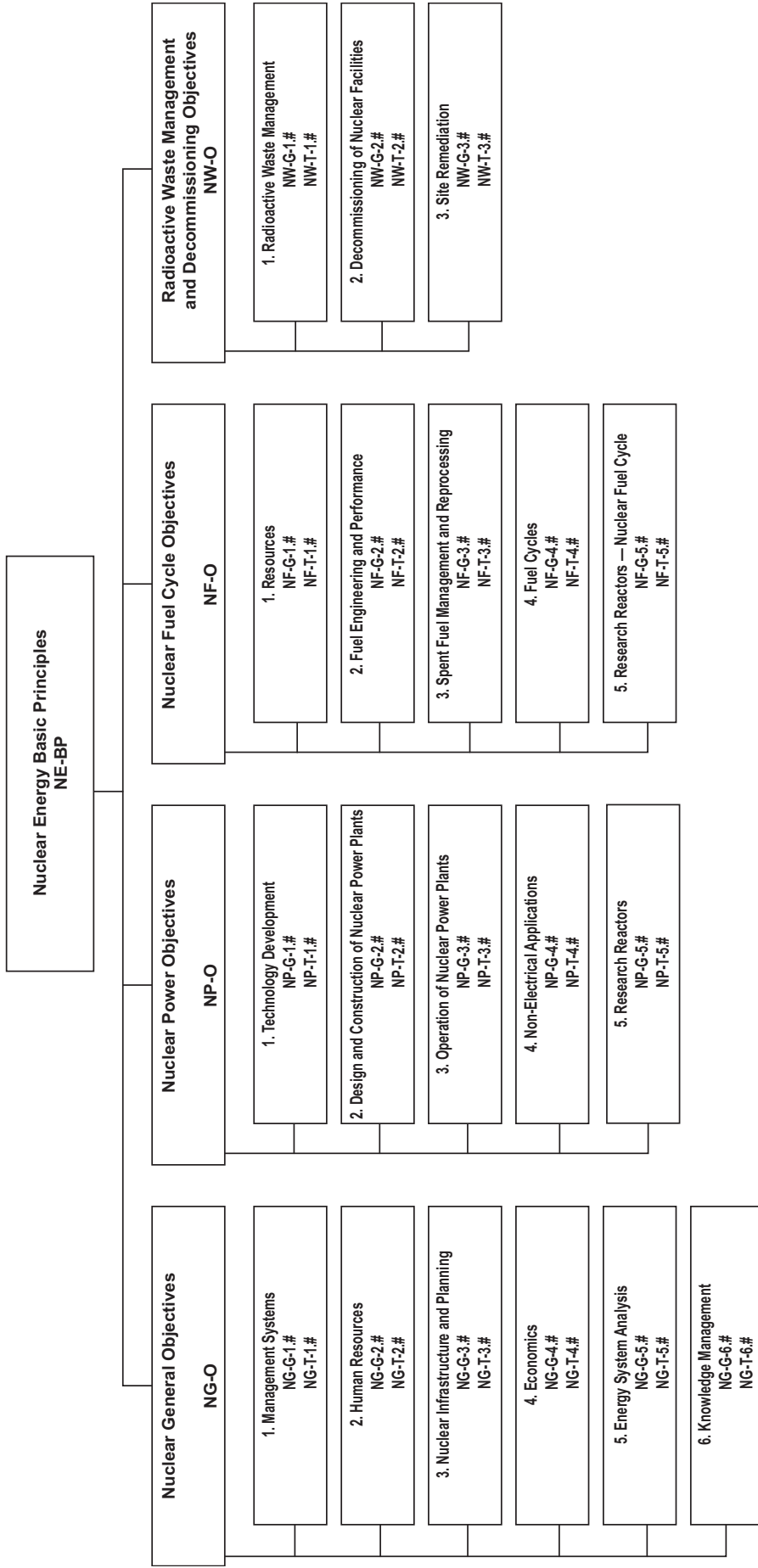
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