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## **CHARACTERISATION OF LONG-LIVED LOW AND INTERMEDIATE-LEVEL RADIOACTIVE WASTES IN THE NORDIC COUNTRIES**

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### **Abstract**

The present report is a final report from a study on characterisation of radioactive wastes in the Nordic countries. The study has mainly been focused on long-lived low and intermediate level radioactive waste. Methods to measure or estimate the activity content and the general composition are discussed. Recommendations are given regarding characterisation of waste under treatment and characterisation of already produced waste packages.

### **Sammanfattning**

Föreliggande rapport är en slutrapport från en studie om karakterisering av radioaktivt avfall i den nordiska länderna. Studien har huvudsakligen fokuserats på lång-livat låg- och medelaktivt avfall. Metoder för att mäta eller uppskatta aktivitetshåll och kemiskt innehåll diskuteras. Rekommendationer ges beträffande karakterisering av avfall under behandling och karakterisering av redan producerade avfallskollin.

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## 1 Introduction

The fifth four-year NKS program for the period 1994-1997 includes one waste project, two reactor safety projects and a total of six projects about radioecology, emergency preparedness and information. The waste project (NKS/AFA-1) is focused on safety in the final disposal of radioactive waste. It is divided into three sub-projects dealing with [NKS 96]

- Waste characterisation (NKS/AFA-1.1)
- Performance analysis (NKS/AFA-1.2)
- Environmental impact statement (NKS/AFA-1.3)

The project was originally directed towards disposal of long-lived low and intermediate level radioactive waste. It was however extended based on different conditions in the countries. Long-lived low and intermediate level radioactive wastes are e. g. in many cases handled together with short-lived radioactive waste and environmental impact assessment processes are in some cases focused more on disposal of spent fuel.

The present report is the second (and final) report for the subproject on waste characterisation. The first report [Brodén 95] is written in Swedish, Danish and Norwegian.

The original object of the subproject was to collect information about experiences regarding waste characterisation for long-lived low and intermediate level radioactive waste in the Nordic countries and also to study and possibly also develop new waste characterisation methods. The project was however somewhat extended so that it no longer was limited to long-lived waste and adjusted to different conditions in the Nordic countries:

- Denmark has facilities for waste handling at Risø. Long-lived low and intermediate level radioactive waste is handled together with short-lived low and intermediate level radioactive waste in the facilities.
- In Finland the nuclear power plants are the main producers of long-lived low and intermediate level radioactive waste. Long-lived low and intermediate level radioactive waste is handled together with short-lived low and intermediate level radioactive waste at the power plants.
- Iceland has only small quantities of radioactive waste from hospitals, research and industry.

- Norway has facilities for waste handling at Kjeller. Long-lived low and intermediate level radioactive waste is handled together with short-lived low and intermediate level radioactive waste in the facilities.
- In Sweden the long-lived low and intermediate level waste comes from the nuclear power plants and Studsvik. The waste from Studsvik has however most in common with the waste in most of the other Nordic countries.

## 1.1 References

[Brodén 95]

K Brodén, S Carugati, K Brodersen, T Carlsson, P Viitanen, T Walderhaug, S E Pålsson, S Backe and A Sörlie. "Waste characterisation of long-lived low and intermediate level waste. Intermediate report for the AFA-1.1 Project." (In Swedish, Danish and Norwegian.) Nordic nuclear safety research 1995, Report NKS/AFA-1(95)1.

[Brodén 96]

K Brodén. "Nordic nuclear safety research, AFA-1. Safety at waste disposal. Annual report 1996. Plans for 1997." Nordic nuclear safety research 1996, Report NKS/AFA-1(96)14.

## **2 Destructive and non-destructive measurements**

The wide range of methods available for measurement of activity or content of specific isotopes in radioactive waste is conveniently separated into methods depending on sampling of the waste material and methods where the information is obtained from measurements of the external radiation or other knowledge about the waste.

Sampling and analyses of waste streams can be carried out routinely and combined with other information about the conditioning process to give information about activity contents in the resulting waste units.

Non-destructive measurement methods are of special importance for the characterisation of the finished conditioned waste units where interference with the physical integrity of the unit should be avoided. However, destructive sampling from a small number of waste units may also be used for quality checking of information obtained in other manners. Such sampled waste units have to be reconditioned.

### **2.1 Sampling methods and representativity**

Radioactive waste materials may range from the completely homogeneous to single point sources distributed at unknown positions in solid materials of variable type and density.

Sampling of homogeneous waste streams (solutions, ion exchange resins, incineration ashes, metals homogenised by melting) is fairly easy with sampling frequency and representativity of the analytical results determined by the variability of the composition of the waste stream.

Destructive sampling of homogeneous waste units (bituminized or cemented evaporator concentrates, ion exchange resins, etc.) is in principle also straight forward, although practical sampling can be difficult and may require special equipment such as diamond drills for coring in cementitious materials, other types of drills for bituminized materials, etc. Analyses of a single sample of the conditioned waste material is representative for a homogeneous waste unit and can be converted to total content provided the amount of waste in the unit is known.

Destructive sampling of inhomogeneous waste units (compressed solid waste, cemented metallic scrap, closed sources with or without shielding, etc.) is on the other hand difficult to carry out in a meaningful manner. The problem is the representativity of samples obtained by core drilling in such units. Combination with advanced imaging technique (see Section 3.5) for localisation of relevant sampling positions may improve the credibility of contents based on analyses of drill samples.

## **2.2 Non-destructive measurements and representativity**

Besides the non-destructive manner of the measurement the principal advantage of determining isotope contents in waste units by measurement of external  $\gamma$ - or neutron radiation is that the methods in principle are integrating and combine contributions from all point sources of the measured radioisotope within the unit. However, effects of geometry and uneven distribution of shielding within the units will influence the representativity of such measurements. In addition it is only a minor number of the long-lived radioisotopes that is possible to measure by non-destructive methods.



### 3 Measurement methods

This section contains a short presentation of the various activity measurement principles used in management of radioactive waste.

#### 3.1 Alpha analysis

Some of the long-lived radioisotopes in radioactive waste are  $\alpha$ -emitters ( $^{226}\text{Ra}$ ,  $^{238}\text{U}$ ,  $^{227}\text{Np}$ ,  $^{229}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Am}$ , etc.).

Methods used for contamination control, etc. during handling of uranium, plutonium and other actinides can also be used for estimating contents of  $\alpha$ -emitters in radioactive waste.

Alpha radiation has a very short range of penetration and measurements can therefore only be carried out on unconditioned waste or waste samples after suitable chemical destruction.

Direct measurements of  $\alpha$ -emitters is restricted to surface contamination, either directly by mapping activity distribution on accessible surfaces by use of a suitable proportional counter or indirectly by measurements on filter paper or similar materials used for sweep tests on contaminated surfaces. Only figures for total  $\alpha$ -activity are obtained.

Measurement of concentrations of  $\alpha$ -emitters in samples from waste streams or from conditioned waste must be carried out after chemical destruction and dissolution of the material. Radiochemical methods based on ion-exchange, complexation and manipulation of redox conditions are used for purification and separation of the various radioelements. The radioisotopes are thereafter precipitated by electroplating onto stainless steel discs and counted in vacuum using silicon solid state detectors in an  $\alpha$  spectrometer. The method is extremely sensitive and gives the distribution of the  $\alpha$ -activity on the various radioisotopes according to the discrete energies of the  $\alpha$ -particles.

The principal disadvantage is that the radiochemical separations are complicated and time consuming making  $\alpha$  spectroscopy a rather costly method.

#### 3.2 Beta analysis

Some important long-lived radioisotopes in radioactive waste are pure  $\beta$ -emitters ( $^3\text{H}$ ,  $^{14}\text{C}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$ , etc.) However, many isotopes with short or intermediate half-life are also  $\beta$ -emitters and this, in combination with the limited possibilities of instrumental methods for discrimination between  $\beta$ -radiation from the various radioisotopes, make simple  $\beta$ -counting likely to overestimate the contents of long-lived radioisotopes in

waste materials. An exception is when administrative information shows that the contamination is due to a single isotope.

Beta radiation has a somewhat longer penetration range than  $\alpha$ -particles, but also the direct measurement of  $\beta$ -emitters is mainly restricted to surface contamination. Measurement of contamination using proportional counters directly or on sweep test filter paper may give a rough idea about the gross  $\beta$ -contents.

Real quantitative determination of the concentration of specific  $\beta$ -emitters can only be carried out after chemical destruction and dissolution of samples of the waste material. Extensive purification for other radioisotopes must be carried out using radiochemical methods. Analytical methods well established in radioecology studies for environmental samples may be employed e.g. for  $^{90}\text{Sr}$ , but methods aimed directly at determination of long-lived  $\beta$ -emitters in radioactive waste have also been developed in later years [Müller 97].

After separation the investigated  $\beta$ -emitter is precipitated as a thin layer sample and the activity determined by use of a proportional counter. Alternatively the activity can be measured using scintillation counting. A special type is liquid scintillation where the  $\beta$ -emitter is dispersed in a fluorescence liquid converting the  $\beta$ -particles to light which is detected by a photomultiplier.

### 3.3 Gamma spectroscopy

Measurement of external  $\gamma$ -radiation by a Geiger-Müller counter can be valuable as a simple method for determination of activity of a known radioisotope regarded as an unshielded point source.

Gamma radiation is emitted at discrete energy levels characteristic of the various isotopes and this is utilised in  $\gamma$  spectrometry for identification of the isotopes. The energy range of interest is from about 40 to 2000 keV.

Relatively few of the long-lived radioisotopes of interest in radioactive waste management are  $\gamma$ -emitters and some of these have only a low yield of photons per decay. An example is the 1001 keV peak from  $^{238}\text{U}$  which is only emitted in 0.59 % of the decays.

Sometimes the content of a long-lived  $\alpha$ - and  $\beta$ -emitting isotope can be estimated from equilibrium with short lived  $\gamma$ -emitting daughters. An example is  $^{226}\text{Ra}$  which can be measured as the daughter product  $^{214}\text{Bi}$  provided the material has been stored for some weeks in such a manner that escape of the in-between gas-formed daughter product  $^{222}\text{Rn}$  is prevented.

The principal advantage of  $\gamma$ -radiation is that it can be detected on the outside of the investigated sample or even on a finished conditioned waste unit. However, the ability

to penetrate surrounding material is dependent on the energy of the  $\gamma$ -rays and the density and thickness of the material separating the source and the detector.

In practice it is mainly radiation from  $^{60}\text{Co}$  (1173 and 1332 keV),  $^{137}\text{Cs}$  (661.6 keV) and sometimes  $^{154}\text{Eu}$  (723, 1274 keV and other peaks) that can be detected by  $\gamma$ -scanning of finished waste units. Commercial equipment for performance of such measurements is available, see Section 6. The signal from the drum is often obtained while the waste drum is rotated and moved in vertical direction past a collimator used to diminish the  $\gamma$ -signal from the drum. Other methods to integrate the signal from the waste unit are also available, but the principal uncertainty in such measurements is always due to lack of knowledge of the geometrical distribution of the radioisotope and the shielding inside the unit. This is not a problem if the material inside the drum is known to be reasonably homogeneous, if not, a higher uncertainty must be accepted. Improvements are possible ranging from relatively simple variations of the so-called "add a source" principle, where external radiation from a  $^{60}\text{Co}$  source is used to estimate density variations of the waste material, to very complex imaging techniques as described in Section 3.5.

The detectors used in  $\gamma$ -scanning are normally of the Ge-type. They give high resolution of the  $\gamma$ -peaks and that is an advantage when weak signals have to be evaluated or if the waste is fresh and contains short-lived isotopes. Detectors of the NaI type have low resolution but are cheaper and easier to operate. They may be used on old waste if the primary need is to obtain a measurement of the  $^{137}\text{Cs}$  content.

For samples obtained from waste streams or by destructive sampling of waste units the opportunities for quantitative  $\gamma$ - spectroscopic determination are better because the geometry of the sample can be well defined. The uncertainty is then mainly associated with the representativity of the measured sample.

Direct Ge  $\gamma$  spectroscopic measurement on waste streams before conditioning gives also possibilities for measurement of some low-energy  $\gamma$ -emitters. Of special interest is the fissile isotopes  $^{235}\text{U}$  (186.7 keV, 57.5 % yield) and  $^{239}\text{Pu}$  (129 keV, 0.006 % yield). Such measurements must be carried out on relatively small samples of low density waste materials which do not contain significant amounts of other  $\gamma$ -emitters.

### 3.4 Neutron based systems

According to the classification system recommended by the IAEA, low and intermediate level radioactive waste (LILW) are subdivided into two classes, short lived waste (LILW-SL) and long lived waste (LILW-LL). In defining the boundary between these classes restrictions of the amount of long lived  $\alpha$ -emitting nuclides in short lived waste are suggested. For classification purposes it is therefore of interest to measure the amount of  $\alpha$ -emitting transuranic nuclides ( $Z > 92$ ) in waste packages.

Neutron based non-destructive measuring methods are based on the physical characteristics of even- and odd mass transuranic nuclides. Nuclides with even mass number generally have a comparable high spontaneous fission yield but low cross section for induced fission by thermal neutrons. Nuclides with odd mass number exhibit a low yield for spontaneous fission but have a high cross section for induced fission by thermal neutrons.

The two main neutron based measuring methods are based on these characteristics. In the so called "passive method" neutrons from spontaneous fission are measured. Generally this method is best suited to measure even mass transuranic nuclides. The other method is called the "active method". In this method neutrons from an external neutron source is moderated and used to induce fission in transuranic nuclides in the waste. Prompt neutrons from these fissions are then detected. This method is therefore best suited to measure odd mass nuclei in the waste. In the active systems the external neutron source can be an ( $\alpha$ ,n) reaction source (Am/Be, Pu/Be), a spontaneous fission source ( $^{252}\text{Cf}$ ) or a neutron generator.

Some systems are based on irradiation of the waste by an external neutron source for a short period and measurements of delayed fission neutrons after the source has been removed. The irradiation and measuring process can then be repeated until acceptable measuring statistics has been reached.

Instruments for measurements on 200 litre drums based on both methods are available on the market. Since neutrons are able to penetrate large objects with high density measurements can be performed on waste in lead or concrete shielded waste drums. Some knowledge of the isotopic composition of nuclides in the waste is preferable in order to evaluate the amount of various even and odd mass transuranic elements from one or the other method.

Since most transuranic elements are  $\alpha$ -emitters they will induce ( $\alpha$ ,n) reactions in light elements producing high energetic single neutrons, while spontaneous fission produces 2-3 coincident neutrons. The contribution of neutrons from ( $\alpha$ ,n) reactions can be suppressed or reduced by using appropriate coincidence detection techniques.

In order to reduce the amount of external neutrons from cosmic radiation instruments are equipped with a neutron shield (cadmium). Waste containing fission products often exhibit high  $\gamma$ -dose rates. Interference with the neutron detectors is prevented by the use of shields close to the waste (lead or tungsten).

Passive and active systems have been developed to detect both thermal neutrons and fast neutrons. In systems based on detection of thermal neutrons gas filled detectors containing  $^3\text{He}$  and  $\text{BF}_3$  are often used. Fast neutrons from spontaneous fission or prompt neutrons from neutron induced fission are then moderated in a moderator (polyethylene or boron loaded polyethylene layer). The detectors and the moderator are enclosing the waste during measurements. Fast neutrons are detected by  $^4\text{He}$  detectors or scintillation detectors. Scintillation detectors have a

rather high sensitivity of  $\gamma$ -radiation which reduces the applicability in case for  $\gamma$ -emitting waste.

### 3.5 Advanced radiometric methods

Various image construction methods based on radiometric measurements can be applied to the non-destructive examination of nuclear waste packages [Reimers 93]:

- Digital radiography (DR)
- Computerised tomography (CT)
- Microtomography (MCT)

Radiography using X-rays or  $\gamma$ -rays represents a well proven technique for the non-destructive examination of various objects in medicine and many fields of material testing. Used on waste packages radiography gives a picture of inhomogeneities within the waste matrix. Typical qualitative information are: waste form (cemented, supercompacted etc.), presence of inner structures (inner container, shielding, void, etc.), presence of free liquids. Typical quantitative information are: dimensions of inner structures, mean attenuation coefficients and matrix density.

In digital radiography a collimated beam from an external  $\gamma$ -radiation source passes through a given object and is measured by a detector. Moving the source and detector up and down and the drum stepwise horizontally from one side to another and visualising the measured intensity of the attenuated  $\gamma$ -beam at each of the resulting grid points gives the digital radiograph of the content in the drum. As the radiograph is 2-dimensional a mean attenuation and a mean density is calculated through the waste matrix at each point between the source and the detector.

Two types of computerised tomography are used: transmission computerised tomography (TCT) and emission computerised tomography (ECT). By transmission computerised tomography energy rays from an external source (X-rays,  $\gamma$ -rays from  $^{60}\text{Co}$  sources or very hard X-rays from accelerator irradiation of a suitable target) is passing through various pathways of a plane section of the drum at a selected height. Horizontal movement is combined with rotation of the drum. One or more detectors (typical scintillators) measure the signal after passage through the drum. The density distribution throughout the horizontal cross-section of the drum is then calculated from the obtained attenuation information.

By emission tomography the activity distribution in the waste is measured by one or several detectors (typical Ge-detectors). The specific and the total activity in the investigated section of the drum is then calculated from the obtained radiation and attenuation information.

When specimens of a suitable size are available (down to the order of few mm) such smaller objects can be investigated by microtomography. This method gives information about porosity, permeability or crack formation in the specimen.

To be used as a daily routine operation in waste characterisation digital radiography and computerised tomography have different advantages and disadvantages:

- Digital radiography gives an overall view of the contents of the total waste drum in relatively short time. By use of more than one detector (multi detector mode) the operation time can be reduced without effecting the resolution but increasing the expenses of the equipment.
- If good resolution is needed the operation time of computerised tomography is several hours for the examination of one waste drum. If only a few cross-sections are needed the operation time is reduced. Computerised tomography is able to give the most correct measurement of the activity of drums containing non-homogeneous waste, but at the same time the equipment is very expensive.

### **3.6 Estimation of chemical contents**

The chemical composition of the waste may influence the safety at final disposal. Metals will e.g. produce gases from corrosion and complexing agents may influence the migration of radionuclides from the repository.

It is comparatively easy to estimate the general chemical composition of the waste in some cases e.g. for precipitation sludges from treatment of liquid waste by co-precipitation with ferrocyanides or for ion exchange resins from water cleaning. The chemical composition of the waste can in both these cases be based on the raw materials used in the processes.

Estimations on gross quantities of metals in the waste can favourably be made during the treatment procedure. The amounts of stainless steel, zircaloy, carbon steel, zinc and aluminium can then be determined by weighing. These data combined with information regarding density and thickness for the metallic parts can be used for calculations of surface areas available for corrosion [Lindgren 94].

Information on minor quantities of chemically toxic materials must be based on destructive analyses or administrative information about the waste stream.

### **3.7 Other methods**

There are several methods based on mass spectrometry that may be used for radionuclide analysis. These are destructive methods based on e.g. aqueous solutions of samples of the material in question. Traditionally, mass spectrometry has been used for analysis of different organic substances, often combined with gas chromatography. These methods, however, may be suited for e.g. carbon-14 dating analysis, but their application to waste processing is not very feasible. On the other

side, some of the more recently developed methods are extremely powerful for small quantities of radionuclides, in particular may Inductively Coupled Plasma Mass Spectrometry (ICP-MS) be found well suited.

ICP-MS is based on the well known techniques of ICP analysis and mass spectrometry for research and industrial purposes. However, when the two techniques are combined, the ionisation power of the ICP gives an excellent feed of ions for the mass spectrometer leading to extremely low detection limits. With careful preparation and in clean rooms, these limits can be pushed down to the femtogram level even for the actinide elements. The size of the injected solution sample is quite small, typical 50 microlitres.

For strong  $\gamma$ -emitting nuclides, this method is not necessary. The method is much better suited for  $\alpha$ - and  $\beta$ -emitters of low energy which demand lengthy chemical purification procedures before counting. However, nearly all types of nuclides can be measured as long as the interference with the matrix solution does not preclude it. In addition information about contents of chemically stable toxic elements can be obtained.

Like other destructive methods, ICP-MS demands taking samples from the waste and dissolving them in some acid aqueous media. The removal of high concentrations of inorganic salts from the solution may also be necessary, e.g. if concrete or bone tissue has been dissolved, the calcium salts may need to be removed. This can be achieved by ion exchange.

ICP-MS is a technique well suited for environmental samples where the contents of radionuclides may be expected to be small, but it can also be used for determination of small amounts of uranium, neptunium and plutonium in waste before the waste is further processed. After preparation of the sample solution, the measurements consume only a few minutes in the ICP-MS instrument, which means much shorter measuring times than the counting of  $\alpha$ -disintegrations which may take days and even weeks to complete.

### 3.8 References

[Lindgren 94]

M. Lindgren, K. Brodén, J. Carlsson, M. Johansson and K. Pers, Low and intermediate level waste for SFL 3-5. Swedish Nuclear Fuel and Waste Management Co., Sweden 1994. Technical Note 94-32.

[Müller 97]

W. Müller, M. Noe, A. Morales, R. Gens, A. Yeates, M. Gilil. "Development of Methods to Provide an Inventory of Important Radionuclides: Analytical Methods and Correlation Data." T. McMenamin ed. Fourth European Conf. on Management and Disposal of Radioactive Waste. EUR 17543 EN, 1997, p. 272-285.



[Reimers 93]

P. Reimers. "Non-destructive examination of nuclear waste packages by advanced radiometric methods", Annual progress report 1993, Bundesanstalt für Materialforschung und -prüfung.

## **4 Administrative methods**

Administrative methods are used routinely to combine and store information about recently produced waste units. Administrative methods may in some cases also be used exclusively or together with measuring methods for estimations of contents in old waste packages. The routines for measuring and documentation used for old waste comply usually not with the requirement on the treatment procedures used today. It is however sometimes possible to get information about waste origin, dose rates etc. from e.g.

- production journals
- transport documents
- safeguard documents
- interviews with personnel involved in the treatment

These data may be supplemented with  $\gamma$  spectrometric measurements on selected packages.

A data base computer program may help a lot in the work if the amount of data is great and the data from different sources should be combined [Öberg 94].

### **4.1 References**

[Öberg 94]

L. Öberg, "Methods to determine data about drums with low-level waste." (In Swedish.) Studsvik RadWaste AB, Sweden 1994. Technical Note RW-94/51.

## 5 Use of correlation nuclides

Several studies have been undertaken to investigate the possibility to correlate difficult to measure radionuclides to other key radionuclides that are easy to measure [Brenk 95, SKB 97, Müller 97]. The conclusion from many of the investigations are that the task is difficult and great care is needed. The following aspects have e.g. to be considered:

- The estimated correlation factors are often waste and site specific
- The time aspect must be taken into account, especially for short-lived radionuclides
- The material composition including impurities (and the irradiation condition) determine the radionuclide contents in the case of induced activity
- Chemical separation of radionuclides may occur during production or treatment of the waste

The radionuclides  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{239}\text{Pu}+^{240}\text{Pu}$  have been selected as key nuclides for estimations of radionuclides in low and intermediate waste from CLAB (Central interim storage for spent fuel) and EP (Encapsulation plant) to SFL 3-5 [SKB 97]. Activation products are correlated to  $^{60}\text{Co}$ , fission products are correlated to  $^{137}\text{Cs}$  and transuranics to  $^{239}\text{Pu}+^{240}\text{Pu}$ .  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  from measurements are often reported together since they emit  $\alpha$ -particles with almost the same energies. The correlation factors used are presented in Table 5.1.

The  $^{239}\text{Pu}+^{240}\text{Pu}$  activity and the  $^{241}\text{Am}$  activity in ash drums from Studsvik are assumed to correspond to 1 % of the  $^{137}\text{Cs}$  activity and 4 % of the  $^{137}\text{Cs}$  activity, respectively [SKB 97].

The content of  $^{239}\text{Pu}+^{240}\text{Pu}$  in hot cell waste from Risø is assumed to correspond to 0.8 % of the  $^{137}\text{Cs}$  activity at the time of removal from the reactor. Recent measurements of bitumenized evaporator waste at Risø gave, however, a correlation factor 1.5 to 2 times smaller than this.

Table 5.2 shows correlation factors from the nuclear power plants in Finland. The factors include both Finnish nuclear power stations, so, the values may vary a lot. Also, correlation factors have been determined for various kinds of water and ionexchange resin samples. Occasional, very extreme values have been left out from this table.

**Table 5.1**

Correlation factors for radionuclides correlated to  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{239}\text{Pu}+^{240}\text{Pu}$ . The factors have been used for estimation of radionuclides difficult to measure in waste from CLAB and EP [SKB 97].

Radionuclide	Correlation factor correlated to $^{60}\text{Co}$	Correlation factor correlated to $^{137}\text{Cs}$	Correlation factor correlated to $^{239}\text{Pu}+^{240}\text{Pu}$
$^{59}\text{Ni}$	$10^{-3}$		
$^{63}\text{Ni}$	0.5		
$^{93}\text{Zr}$	$10^{-6}$		
$^{125}\text{Sb}$	1		
$^{90}\text{Sr}$		0.1	
$^{99}\text{Tc}$		0.005	
$^{129}\text{I}$		$4 \cdot 10^{-7}$	
$^{134}\text{Cs}$		1	
$^{135}\text{Cs}$		$5 \cdot 10^{-6}$	
$^{152}\text{Eu}$		0.1	
$^{154}\text{Eu}$		0.03	
$^{155}\text{Eu}$		0.01	
$^{238}\text{Pu}$			50
$^{241}\text{Pu}$			200
$^{242}\text{Pu}$			$3 \cdot 10^{-3}$
$^{241}\text{Am}$			5
$^{243}\text{Am}$			0.5
$^{243}\text{Cm}$			1
$^{244}\text{Cm}$			1

**Table 5.2**

Correlation factors from the nuclear power plants in Finland  
[Puukko 92, Puukko 88].

Radionuclide	Reference nuclide: <sup>60</sup> Co	Reference nuclide: <sup>137</sup> Cs	Reference nuclide: <sup>239+240</sup> Pu
<sup>63</sup> Ni <sup>90</sup> Sr <sup>99</sup> Tc <sup>129</sup> I <sup>238</sup> Pu <sup>241</sup> Am <sup>243</sup> Cm+ <sup>244</sup> Cm <sup>242</sup> Cm	0.03 - 1.4	0.01 - 0.65 0.001 - 0.002 2.3 - 4.6 × 10 <sup>-7</sup>	0.1 - 0.9 0.02 - 0.8 0.03 - 0.14 0.03 - 2.3

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## 6 Measuring equipment

### 6.1 Measuring equipment used in the Nordic countries

#### 6.1.1 Denmark

Table 6.1 outlines methods and equipment used at Risø for measuring the content of  $\gamma$ -activity in waste units or waste samples.

**Table 6.1**

Measuring equipment used for  $\gamma$  measurements on units of low level waste to be stored at the interim storage facility at Risø and equipment used for measuring activity in samples of waste.

Waste category	Non-destructive measurements on	Measuring equipment	Comments
Steel drums with bituminized evaporator waste or compacted solid waste	A 100-115 litre drum embedded in a 210 litre drum using concrete.	NaI-detector ( $\gamma$ measurements of $^{60}\text{Co}$ and $^{137}\text{Cs}$ )	The drum is measured in four different positions on a turntable - $90^\circ$ between each
Steel drums with bituminized evaporator waste or compacted solid waste	An overpacked drum. A standard unit embedded in a 280 litre drum	NaI-detector ( $\gamma$ measurements of $^{60}\text{Co}$ and $^{137}\text{Cs}$ )	The drum is measured in four different positions on a turntable - $90^\circ$ between each
Unknown activity of closed disused sources	Disused closed sources	Geiger-Müller detector (External $\gamma$ -radiation)	Knowing the type of isotope the activity can be calculated using exposure rate constants

**Table 6.1** cont.

Sample category	Measurements on sample of waste	Measuring equipment	Comments
Samples of distillate, concentrate, bitumen, ion exchange or earth collected during operation	1. 50 ml in a standard plastic container 2. 1.5 litre in a standard plastic container 3. About 1 litre of bituminized waste in a glass container	Ge-detector ( $\gamma$ measurements)	
Sample of bituminized evaporator waste or concentrate from the evaporator plant	A sample of few gram which is treated in a acid solution before electrolysis and $\alpha$ measurements	$\alpha$ spectrometric measurements of $^{239}\text{Pu}$ + $^{240}\text{Pu}$ and $^{238}\text{Pu}$	Correlation between content of $^{239}\text{Pu}$ + $^{240}\text{Pu}$ and $^{137}\text{Cs}$ is calculated

Standard units containing low level compacted solid waste or bituminized evaporator waste are measured by a NaI-detector before being stored in the facility for low level waste. The unit is measured in four different positions. The activity of  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  is calculated assuming a homogeneously distribution supplemented by three point sources near the wall in the midplane of the inner drum. For various reasons the measurements are likely to underestimate the activity and this is therefore increased about 15-20 % to obtain the most probable value inside a band of uncertainty [Catugati 96]

External  $\gamma$ -radiation from disused sources is used together with exposure rate constants to estimate the activity. The sources are stored in a interim facility (Centralvejslageret) placed in bore holes in a concrete block.

A lead shielded Ge-detector calibrated for different geometries is used on various samples from the waste management plant to determine releases as well as activity contents in waste streams.

Content of plutonium is calculated in samples of evaporated waste water by use of  $\alpha$  spectrometric measurements. Using  $^{242}\text{Pu}$  as internal standard calculation of  $^{239}\text{Pu}$  +  $^{240}\text{Pu}$  and  $^{238}\text{Pu}$  are possible. The measurement is very time-consuming because of the extensive preparation before the sample can be counted.

### 6.1.2 Finland

Dose-rate measurements are typically made with Geiger-Müller detectors. These are usually calibrated with a  $^{60}\text{Co}$  or  $^{137}\text{Cs}$  standard. Dose-rate measurements yields, however, only preliminary information for handling the waste. Spectroscopic measurements are used for obtaining more exact information for individual nuclides.

Gamma spectroscopic measurements are made with a semiconductor detector. In Loviisa NPP and at VTT, portable detectors with small containers for liquid nitrogen are used. Since these only contain nitrogen for one day's operation they are usually coupled to larger containers, see Figure 6.1. Most of the detectors are modern, pure Ge-detectors.

The activities in all drums, except those with very low or not measurable activity contents, are measured with a  $\gamma$  spectrometer that contains: a semiconductor detector with preamplifier, accessories (linear amplifier, A/D converter), multi-channel analyser, and a computer.

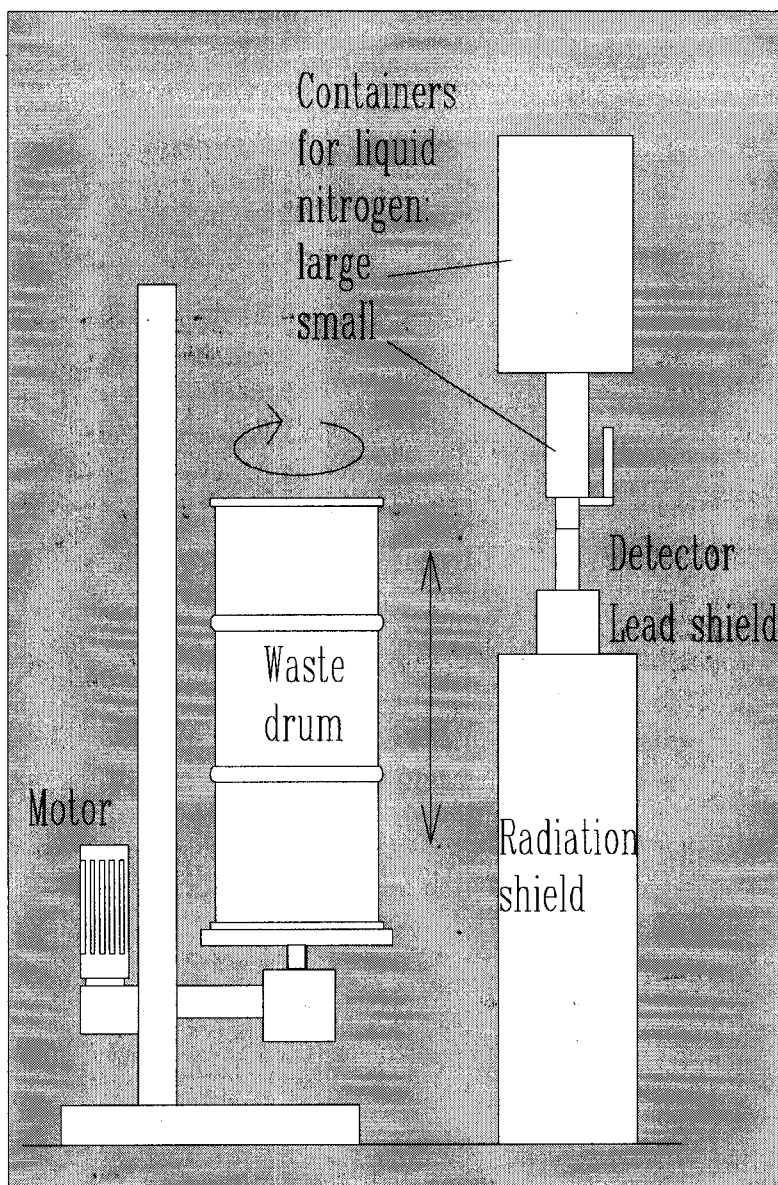
The scanning geometries used in the measurements of the activities in steel drums are in principle the same at the Loviisa and Olkiluoto NPP's and at VTT. Minor differences are found in the calibration procedures and in the use of standards.

At Loviisa a drum is used in which the calibration sources can be placed in various places. The drum contains representative waste, e.g. paper, clothing, plastic etc. Three sources are used:  $^{133}\text{Ba}$ ,  $^{137}\text{Cs}$  and  $^{60}\text{Co}$ . Intermediate waste has not yet been packed in Loviisa.

At Olkiluoto several calibration sources are used for various waste packages:

- The scanning measurements of plastic sacks are calibrated with a  $^{152}\text{Eu}$  point source.
- The  $^{152}\text{Eu}$  standard has been mixed with pure resin and is used for measuring low-active steel drums.
- The activity content in a drum with intermediate-level active ion-exchange resin has been measured in small portions in the laboratory. The measured resin has then been bituminized and can be used as standard.





**Figure 6.1**

Schematic description of the scanning equipment used at VTT. Similar equipment is used at the Loviisa and Olkiluoto NPP. The rotating platform is moved vertically upwards and downwards in front of the detector. The detector is lowered into the large radiation shielding during measurements of small samples.

At VTT point sources are used which are fixed at the wall of the drum. A measurement is made without the point source and if there is any activity, then a second measurement is made with the source attached. If the energy of the source

differs from the waste, then one measurement with the attached source is sufficient. This type of measurement is planned for the future.

The  $\gamma$  spectroscopic measurement makes it possible to observe nuclides with energies above approximately 100 keV.

The long-lived pure  $\beta$ -active nuclides that are present in the waste from nuclear reactors are  $^{63}\text{Ni}$  and  $^{90}\text{Sr}$ . The actinides U, Pu, Am and Cm are  $\alpha$ -active and emit also relatively few  $\gamma$ -quanta with low energy. The estimation of the activity of these nuclides, which are difficult to measure, is based on reference nuclides. The reference nuclides used are  $^{60}\text{Co}$  or  $^{137}\text{Cs}$ .

The power plants have an extensive book-keeping for the waste, which can be used for updating correlation factors for various nuclides and waste categories at different times. This makes it possible to adjust total activity estimates in a storage, if, for example, it is found that the previously used scaling factors were erroneous during a certain period.

A possibility to enhance the accuracy of  $\gamma$  spectrometry at low energies is provided by the use of a guard detector and the anticoincidence technique. The main idea of the arrangement is to reduce the background by ignoring the counts which are recorded simultaneously by the primary and the guard detector. These simultaneous incidents may happen due to Compton scattering. The primary photon impacts the semiconductor detector and is scattered with lower energy to be absorbed by the guard detector.

### 6.1.3 Norway

Radioactive waste delivered to the Waste Treatment Plant at IFE, Kjeller, shall be accompanied by information on the nuclide content, the activity levels and the chemical composition. Because this information often is incomplete or missing it is necessary to perform analysis and measurements. Analysis, measurements and calculations are performed by IFE which is operating the only radioactive waste treatment plant in Norway.

After evaporation of small samples on metal discs the total  $\alpha$ - and  $\beta$ -activity in liquid waste are measured by LEAB Universal Printing Spectrometer with a thin window GM-detector. The nuclide content and activity levels are calculated based on available information from waste producers or from radiochemical analysis or  $\gamma$  spectroscopy.

In radiochemical analysis of liquid or semi liquid waste separation and plating/filtering is followed by  $\alpha$  spectroscopy or  $\beta$  spectroscopy. Standard equipment based on Si surface barrier semiconductor detectors are used. Beta spectroscopy is performed by equipment based on anti coincidence technique. The quality of the radiochemical methods are tested in Nordic and international

calibrations tests. The measuring equipment are calibrated by use of standard calibration sources.

Samples of waste are often analysed by  $\gamma$  spectroscopy in order to obtain information of the nuclide composition and activity levels. The measuring equipment is based on Ge-detector systems and NaI-detector systems. Calibration of these systems are performed by use of standard calibration sources for different measuring geometries in use.

After waste processing and conditioning in waste containers dose-rate measurements are performed outside the containers. Based on available information on the nuclide composition supplied by the waste producer or gained by radiochemical analysis and/or  $\gamma$  spectroscopy the activity level for each nuclide inside the container is calculated. For ion exchange resins from the two research reactors in Norway the nuclide composition is well known. Measurements of the dose-rates outside shielded containers are made assuming that  $^{60}\text{Co}$  is the only contributor to these measurements. Calculation of the activity levels for other nuclides are based on the measurements of this reference nuclide.

In the near future the Waste Treatment Plant at IFE, Kjeller, will install a Ge-detector system for  $\gamma$  spectroscopy of processed waste inside waste containers.

#### 6.1.4 Sweden

The main part of the packages with low and intermediate level waste produced at Studsvik will in the future be disposed of in the deep repository SFL 3 [SKB 97]. Table 6.2 gives a summary of measuring equipment used for the waste.

Gamma spectroscopic measurements with Ge-detectors are performed on:

- 80 litre drums with intermediate level solid waste
- 100 litre drums with low level solid waste (ashes, refuse and scrap)
- Soft plutonium containing waste in plastic bags
- Sludge samples

The  $\gamma$  spectrometric measurements on waste drums gives generally less than  $\pm 40\%$  deviation or in the worst case, for very inhomogeneous waste,  $\pm 70\%$  deviation [Sundgren 94, Sundgren 89].

Total  $\alpha$  measurements with a proportional counter are performed on smear samples from hard plutonium contaminated components. The results from the measurements are used for calculation of the plutonium content on each

component [Brodén 91]. Total  $\alpha$  measurements are also performed on sludge samples. Then a zinksulfide scintillation detector is used.

**Table 6.2**

Measuring equipment used for long-lived low and intermediate level waste from Studsvik to the deep repository SFL 3.

Waste category	Measurement on	Measuring Equipment	Comments
Five holes concrete containers with intermediate level waste	80 litre drum	Ge-detector ( $\gamma$ measurements)	The drum is placed on a rotating table with a hydraulic lift Collimators are used
Five holes concrete containers with plutonium waste	1) Plastic bag with soft Pu-waste 2) Smear sample from hard Pu-contaminated components	1) Ge-detector ( $\gamma$ measurements) 2) Low-level planchet proportional counter ( $\alpha$ measurements)	
Steel drums with solidified sludge	Sludge sample	Ge-detector ( $\gamma$ measurements) Zinksufide scintillation detector ( $\alpha$ measurements)	
Steel drums with ashes in inner drums surrounded by concrete	100 litre drum	Ge-detector ( $\gamma$ measurements)	The drum is placed on a rotating table
Steel drums with refuse and scrap in inner drums surrounded by concrete	100 litre drum	Ge-detector ( $\gamma$ measurements)	The drum is placed on a rotating table

## **6.2 Survey of measuring systems and equipment on the market**

Measuring systems and equipment for measurements and analysis of nuclide content and activity levels in unprocessed and processed waste are commercially available from several producers and suppliers. In order to obtain information on available systems and equipment the AFA-1 work group has submitted a questionnaire given in Appendix A to several organisations. This has resulted in answers from:

- SGN
- BNFL instruments
- Canberra Nuclear Industries
- Gamma Data

Summaries of the received answers are given in Appendices B-E. The prices are however excluded.

Appendix F gives information on a European network of testing facilities for quality checking of radioactive waste packages.

## **6.3 Other measuring systems and equipment**

The commercial development of complete systems for waste unit characterisation is supplemented by more fundamental research efforts aimed at improving and extending the range of application of non-destructive measurement methods. Comparative studies to validate measurements using different - or a variety of the same measurement principle - on the same waste unit are also carried out. Projects within this field are typically found at the larger nuclear research centres.

Examples taken from the EU research programmes on Management and Disposal of Radioactive Waste are given in the following without any attempt to make an exhaustive listing [Odoj 97].

- Round robin comparison of  $\gamma$ -scanning characterisation was carried out under the previous and will be repeated under the ongoing EU-programme with participation of a large number of laboratories.
- Research systems for active or passive neutron investigation of contents of fissile material in waste units. The aim is to improve sensitivity and to be able to handle large units with difficult composition.
- Tomography of high level waste glass gives information about internal cracking, voids and other defects. It has been demonstrated for inactive full size glass containers but similar measurements on fully active units are difficult due to the intense  $\gamma$ -radiation. Extensive development of

accelerator driven tomography using Cerenkov detectors have been carried out and seems to solve the problem.

- Tomography of large, high density waste units (e.g. cementitious waste) may also require the high energy radiation available in accelerator driven systems.
- A general aim in development of the various imaging techniques are decreased counting time at reasonable investments in equipment, thereby making the methods more suitable for routine use.

Destructive methods are used to obtain detailed information about the contents of difficult to measure radionuclides and for verification of non-destructive measurements. Obviously, destructive methods can only be used on relatively few waste units. Sampling of conditioned radioactive waste is not simple and equipment for the purpose have had to be developed. The use of imaging technique on the waste unit before coring may improve the representativeness of the obtained samples. The necessary equipment can only be expected to be available at specialised laboratories and detailed characterisation of selected waste units must therefore be centralised.

#### 6.4 References

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O. Sundgren. "Waste management at Studsvik. Measurement of drums with low- and intermediate level waste." (in Swedish.) Studsvik Nuclear AB, Sweden 1989. Technical Note NP-89/101.

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O. Sundgren, "Gamma-ray counting of medium-activity waste." Radioactivity and Radiochemistry Vol. 5, No. 4, 1994.

## **7 Recommendations**

### **7.1 Advice for characterisation of waste under treatment**

The first precaution for waste of unknown radioactive content should be dose rate measurement to ensure safe handling. The sample can then be classified according to dose rate, its origin as well as physical and chemical composition. Depending on the size and geometry of the sample, different types of analyses could then be performed on whole samples or a sweep samples.

The following provides some general advice concerning the treatment of waste with regard to sample-taking, activity measurements, etc. The recommendations forms a basis on which more detailed instructions can be formulated.

Recently packed waste should be measured with a standard method (usually  $\gamma$  spectrometry). However, it is advisable to get information of waste under treatment which is not yet packed or is under packing. Collection of such information includes, e.g. taking samples which are used to determine the correlation coefficients for isotopes for which direct measurement is difficult.

#### **7.1.1 Sampling and sample analyses**

Sampling should generally be comprehensive enough in order to take into account statistical inaccuracy of the measurements. This implies that several samples or sweep samples should be taken when the results are used for activity estimations.

The position and frequency of sampling depends on the specific plant. Samples are desirable for e. g. evaporator concentrates and ion exchange resins. Samples of evaporator concentrates may be separated into clear solution and sludges which normally are present in such samples. Samples from ion-exchange resin should be taken regularly in order to take into account changes of isotopic ratios and correlation factors that are influenced by fuel leaks and water chemistry.

Alpha spectroscopy can be utilised when there is some uncertainty about the isotopic composition of the samples. Alpha spectroscopy is conveniently applied to small samples or smear samples, but precise measurements requires use of radiochemical methods.

#### **7.1.2 Direct measurements**

Dose-rate measurements can give a good estimation of activity when the principal radioactive isotope is known. This can be utilised especially to estimate the activity



of large samples. Usually the activity of metal parts like pump parts, tubes, valves etc. can be assumed to emanate from isotopes of cobalt.

Gamma scanning is the primary method for characterisation of waste packages in nuclear power stations and other laboratories handling considerable amounts of radioactive waste. The measurement arrangement depends on the activity and size of the package, so precise advice is unnecessary.

The use of a neutron dose meter is recommended for preliminary measurements if the presence of a considerable amount of neutron emitting isotopes is suspected.

Passive or active neutron counting in waste packages are relatively new techniques developed from the work originally carried out within safeguards analysis. The methods are presently not used within the Nordic countries and future use must depend on whether they are needed for the types of waste available and whether the sensitivities are sufficient.

### **7.1.3 Physical and chemical composition**

Classification of the waste according to physical and chemical composition is most simply achieved during the treatment. However, when radioactive waste is handled, the dose-rate measurement should be the first precaution prior to any other procedures.

### **7.1.4 Documentation and traceability**

Treatment of the waste is the natural and easiest stage from which to start creating documentation of the waste. Each package should be identified and documentation on activity as well as physical and chemical composition should be produced. Other factors to be included in the documentation should be, e.g. possible surface contamination of the package.

Reference nuclides are used for the analysis of isotopes difficult to measure. The analysis is done by combining results from  $\gamma$  spectrometry with documentation from correlation analysis. The temporal change of correlation factors should be taken into account.

If waste from other sources is handled these should usually have documentation indicating the desired knowledge of the sample. If documentation is missing, the reason for this may be that the sample is of low activity. However, if there is doubt about the contents of the sample, the sample should be treated as an unknown sample and follow the procedures indicated above.

## **7.2 Advice for characterisation of older waste packages**

New regulations for the inventory of a repository may demand a new assessment of radioactive waste packages. The existing documentation of a waste package is the primary source of information. Half-life correction for reported isotopes is trivial. If new knowledge about the correlation factors or isotopic composition becomes available, the documentation of previous measurements can be used and the activities can be corrected. However, if  $\gamma$  spectrometric measurement can be made with reasonable effort this is recommended.

Supervising of a waste storage might include taking sweep samples of the packages in order to detect leaks and contamination. Alpha,  $\beta$ - or  $\gamma$  spectrometry should be used according to the package documentation.

Dose-rate measurement may be used to confirm the expected isotopic composition if the decrease in external dose corresponds to decay of the radioisotopes. Interference from other waste packages in the store may have to be taken into account.

If a neutron-counting device becomes available it may also be used to check some older units.

Radiographic or thomographic methods can be used to assess the activity and density distribution on selected waste units, but the equipment is costly and time consuming in use.

For large and heavy concrete packages drill-core samples can be used to verify the results of existing documentation. Core analyses can then be used to re-estimate activity contents assuming package homogeneity. Due to the contamination risks when taking drill core samples, it is advisable to use a portable spectrometer to measure this kind of packages.

## **Appendix A**

### **Request regarding available measuring systems and equipment**

Dear Sirs,

The five Nordic countries (Denmark, Finland, Iceland, Norway and Sweden) are co-operating within a program called the Nordic Nuclear Safety Research (NKS). One part of this program is called AFA-1 and deals with long lived low- and intermediate level radioactive waste. The work group for this part of the program will in 1997 be responsible for preparing a report on measuring devices and equipment for assessing the nuclide content and activity levels in unprocessed radioactive waste and in old waste containers. In our report we plan to describe various measuring systems and equipment available from various producers and suppliers.

This request to you is made in order to obtain necessary relevant information on available measuring systems and equipment. We would therefore kindly ask you to answer the following questions in relation to each system available from your firm/organisation.

1. Please give a short description of the measuring system/equipment.
2. Is this measuring system/equipment based on destructive or non-destructive measurement principles?
3. Please give a short description of the physical principles for operation of the system/equipment.
4. What are the physical dimensions of the system/equipment?
5. Is the system stationary or mobile?
6. What are the requirements for installation and operation?
7. What are the detection limits for specified nuclides?
8. How are measurements stored and presented to the operator, is the data pre-processed or condensed in some way before presentation?
9. What are the demands for calibration and maintenance?
10. What are the personnel requirements for operating the system/equipment?
11. How much time will be required for one typical measurement?

12. Please give a short description of the radiation shielding/shielding principles incorporated in the system/equipment and doses to the operator in a specified measuring situation.
13. Is the system/equipment available for sale or is it still under development ? If still under development or if it is an research instrument what is the stage of development and where has the instrument been developed (name of the firm/organisation/institute and address)?
14. Can the producer/supplier give information on existing installations of the system/equipment?
15. If the system/equipment is available for sale please give us a price estimate for purchase.
16. Can the system/equipment be hired ? If so, please give us a price estimate for this.

We thank you for your co-operation in answering this questionnaire.

Yours sincerely

## **Appendix B**

### **Information about available measuring systems and equipment from SGN**

#### **General**

##### *Non-destructive measurements*

The systems presented are based on non-destructive measurement principles ( $\gamma$  spectroscopy, coincident passive and active neutron measurement, dose rate meter...)

##### *Mobility*

The systems are normally stationary but some of them are able to be mounted on long trays.

##### *Detection limits*

Betasys has around 300 Bq detection limit for  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  in 5 minutes of measurement.

EM 200 neutron passive assay has 0.8 mg of  $^{240}\text{Pu}$  equivalent detection limit in 10 minutes counting time.

EM 1800 has 50 mg plutonium as detection limit within 30 minutes measurement time.

##### *Calibration and maintenance*

Calibration is mostly required weekly or monthly depending on the operational conditions. Maintenance operations are currently processed one or two times per month (nitrogen refilling, etc.).

##### *Personnel requirements*

Assay softwares are designed for non specialist operators. Maintenance operation needs trained operators. Eurisys Mesures training is currently proposed to the customers in order to get their operational independence.

### *Time consumption*

Typical measurements need between a few minutes and half an hour. Detection limits are largely (but not only) depending on the measuring time.

### *Radiation protection*

Neutron passive assay includes polyethylen shielding. Gamma spectroscopy uses low background lead shield with copper X-ray reduction.

### *Stage of development*

The systems proposed are available for sale. New developments are also studied in a continuous improvement process.

### *Hiring*

The equipment is normally not for hire.

## **Betasys: Waste Measurement System For Beta Measurement Using Gamma Spectroscopy For 120 Litre Barrels**

### *Physical principles and dimensions*

A low background lead enclosure, 150 mm thick, with internal add-on copper 10 mm thick, including:

- 3 coaxial Ge detectors, N-type, with 20% relative efficiency, for low activity measurements (100 Bq to 20,000 Bq),
- 3 planar Ge detectors, 2000 mm<sup>2</sup> area, 13 mm thick, for high activity measurements (20,000 Bq to  $2 \times 10^{19}$  Bq).

### *Data presentation*

An XPRESS industrial PC 4486 type computer, with optical 600 Mb data disk recorder, digital parallel interface with the programmable automaton, and ETHERNET/TCP-IP network connection.

## **Em 200: Combined Gamma And Neutron Measurements On 100 And 200 Litre Barrels**

### *Physical principles*

3 different types of measurements are made:

- Gamma spectroscopy,
- Passive neutron counting,
- Dose rate calculation.

These operations may be either simultaneous or sequential, either manual or fully automated.

### *Data presentation*

The measurements using both  $\gamma$  spectroscopy and dose rate calculation are processed by a PC compatible microcomputer equipped with an Ethernet network interface. IEEE interface and RS232 input-output interfaces allow connection of the computer with the spectroscopy system, the dose rate system and the programmable logic computer which manages the conveying mechanism.

The  $\gamma$  measurement computer uses the following application programs:

- a complete  $\gamma$ -acquisition and processing software (automatic peak search, radionuclide identification using LARA isotope table, detector efficiency correction, etc.)
- a Quality Control software which ensures the surveillance of normal operation,
- a plutonium isotopic composition computing software, with U and Pu mass calculation,
- a specialised software for complementary data acquisition (weighing, barcode),
- control and command software for rotating and lifting mechanisms.

The neutron data are processed using the same type of computer; this computer manages the signals from the PMCCM module; the software is working with:

- passive neutron acquisition capability with neutron multiplicity process under Windows
- spontaneous fission neutron analysis with multiplicity process using BONDAR and HAGE/CIFFARELI methods.

This computer is connected with the supervisor by mean of the Ethernet network.

*Time consumption*

This system can perform up to 10 complete measuring cycles a day, i.e. 2 000 barrels during a complete year, with a total measuring duration of 25 minutes for each barrel.

**Em1800: Gamma Spectroscopy Systems For Measurement Of High Capacity Conditioned Barrels And Containers**

*Data presentation*

The software includes standard as well as dedicated programs:

- InterWinner, for acquisition and processing of  $\gamma$ -spectra (network version).
- Interfût, dedicated software for waste management, takes into account the measurement geometry and the choice of corresponding detector efficiency curves. depending on the status of the trunk positioning contacts.
- The on line Quality Control is performed using a complementary software; energy calibrations, resolutions and area calculations are controlled and compared with pre-defined thresholds; the resulting diagnosis are showed on the operator display unit in case of dysfunction.
- The plutonium, uranium and americium masses are calculated using a specialised program after determination of their isotopic composition, starting from the standard software.
- The evolution of plutonium and americium isotopic composition is evaluated using different processing methods; the masses are calculated by mean of the infinite energy method.
- The activities of fission and activation products are calculated using a dedicated software, starting from parameters which take into account measurement geometries, screens and matrices.



## **Em200fa: Complete Waste Measurement System Using Gamma Spectroscopy For Low Activity 200 Litre Barrels**

### *Physical principles*

Simultaneous dose rate and X-ray radiography before each measurement.  
Alpha and Beta activity of main isotopes with matrix effect correction.

### *Data presentation*

The whole  $\gamma$  spectroscopy system is connected to a PC type microcomputer; this computer is connected via an Ethernet network (under TCP/IP protocol) to a completely computerised system:

- PC server placed in a control room
- PC computer for displaying the barrel radiographs
- X-ray measurement system
- PC supervisor for the whole system
- programmable logic computer which controls the conveyor

Thus, the operations are under the operators control at any time: locally (near by the measurement system) or at a distance (from a control room which may be located within a few tens of meters away from the measurement system).

Push-button operations (start, stop, etc.) can be made locally by the operator.

The measurement computer is equipped with the following application programs: Complete qualitative and quantitative  $\gamma$  analysis software; GammaWinner under Windows (automatic peak search, isotope tables, efficiency curves, etc.)

## **Gammasys: Waste Management System Using Gamma Spectroscopy**

### *Physical principles and data presentation*

The detection system includes:

- an hyper pure Ge coaxial detector of 20 % efficiency or more (according to the detection limit), featuring a good compromise between efficiency and resolution in the energy range 40 keV to 2 MeV. the use of such a detector is essential for plutonium and uranium peak measurement.
- a cooling cryostat with LN<sub>2</sub> dewar for the detector (30 litre capacity) ensuring an autonomy greater than 2 weeks, with horizontal

streamlined configuration. The transistor reset preamplifier for high count rate capabilities is integrated to the detector end cap.

- a low activity lead shielding surrounds the detector, allowing ambient noise attenuation.
- as an option, Geiger-Müller dose rate meters (MN/DB).
- a manual or optional motorised collimator which allows, depending on the barrel activity, to measure either totality or slices of the barrel.
- a manual or motorised optional platform holding the detector and adjusting the distance between barrel and detector. The combination (large dynamic range - slited collimator and motorised detector platform) allows large dynamic measurement with reasonable measuring times
- a lead brick shielding located behind the barrel
- fast acquisition and management NIM electronics including:
  - a 7244 fast spectroscopy amplifier with pile-up rejection
  - a 7186 alarm module. Connected to the LN<sub>2</sub> level gauge of the Ge detector, shuts-off the HV power automatically as soon as the LN<sub>2</sub> lower level is reached
  - a 7161 HV power supply for detector polarisation (5 KV)
  - a 7601, 8K channel ultra fast fixed dead time ADC (3µs)
  - a NIM bin and power supply (12 units)
  - a high count rate multichannel analyser (PC board), INTERFAST type - 32K channels or NIM PCA MULTIPORT module
  - a PC compatible microcomputer with coprocessor, running under MS DOS
  - a laser printer
  - an interface rack for programmable controller boards, connected with a TTL board integrated into the PC
  - automatic weighing, barcode reading, electrically cooled Ge-detectors may be delivered as options.

### *Dimensions*

This mechanism ensures reliable loading and positioning of the barrels. The system is about 1.2 meter wide, 1.2 meter long and 2.80 meter high (with barrel at high position).

An automatic weighing system can be added at the end of the access conveyor.

## **Translab: Neutron Slab Waste Counter**

### *Physical principles*

It detects coincidence neutrons from the spontaneous fission of the even numbered isotopes of plutonium.

An AMN 10 neutron multiplicity coincidence analyser is used for the measurements.

### *Dimensions*

Overall size: 145 x 475 x 405 cm<sup>3</sup>.

### *Data presentation*

The detection device is connected to a PC portable computer in which the AMN 10 neutron multiplicity analyser is plugged. This computer is placed on a shelf fixed to the slab.

## **Em1000r: Waste Measurement System Using Gamma Spectroscopy And Neutron Counting**

### *Data presentation*

The  $\gamma$  spectroscopy measurements are made using a PC compatible microcomputer working under Windows. An IEEE interface, RS2342 input/outputs and a TTL parallel interface for connection to the spectroscopy system, the neutron counting system and the programmable logic computer (management of the waste positioning).

This measurement computer is implemented with the following programs:

- complete acquisition and processing software for  $\gamma$  spectroscopy (automatic peak search, radionuclide identification using the LMRI/LARA isotope table, detector efficiency correction, etc.)
- a Quality Control software for the survey of the acquisition system good operation
- a software for plutonium and uranium isotopic composition and mass calculation
- a software for control command of the rotating and lifting mechanism

- a software for neutron acquisition with multiplicity calculation.

*Time consumption*

The whole system is capable of making a complete measurement (neutron and gamma) in less than 20 minutes, thus allowing measurement of 2500 parcels per year; these parcels are divided into 4 classes with a 3 % accuracy on the total plutonium mass. This mass may reach a maximum of 1 kilogram.

## **Appendix C**

### **Information about available measuring systems and equipment from BNFL Instruments**

#### **General**

##### *Data Presentation*

The computer can store nearly whatever desired. Videopictures are transferred to tape.

##### *Personnel requirements*

Ordinary engineers and technicians can operate the equipment. Necessary training is provided as courses.

##### *Radiation protection*

Doses to the operator are generally very low. However, for the RadScan instrument, the doses are dependent on the time required for installation in a radiation environment.

ILW and repacked waste may give rise to large extra costs in order to reduce doses to personnel.

##### *Stage of development*

All equipment is fully developed. Development of new models, however, is a continuous activity.

##### *Hiring*

Both sale and hiring are possible. The more complex and expensive the equipment is, the more attractive the hiring option is. Quite often even hiring may cost several million crowns.

## **TRU Crate/Box Monitor**

### *Description*

Fast, accurate and efficient plutonium analysis of large redundant plant items allowing selection of appropriate waste disposal option.

Allows compliance with criticality safety cases and transport regulations.

Spatial location of Pu within a box by gamma/neutron imaging.

Automatic identification and correction for PuF<sub>4</sub> contaminated wastes.

### *Physical principles*

Passive neutron coincidence counting combined with high resolution  $\gamma$  spectrometry.

### *Operation*

Prior to loading into the main neutron counting chamber the crate passes between two HRGS detectors. This data is automatically combined with the neutron coincidence counting data to enable matrix and isotopic corrections and provide an accurate total Pu mass assay.

### *Detection limit*

Under typical operating conditions: <1g Pu

### *Data presentation*

A rugged key system and monitor are used to operate the software driven menus. This provides a simple and convenient interface which is easy to decontaminate. All measurement parameters are automatically stored to disk and may also be directly printed. Diagnostic and maintenance operations are password protected.

### *Time consumption*

4 hrs assay time.

## **RadScan 600: Gamma Scanner**

### *Description*

The BNFL Instruments' RadScan 600 Gamma Scanner reduces the requirement for health physics area monitoring particularly in unknown or high dose fields. The system is useful in saving cost and dose uptake for any application where an operator requires information on the spatial distribution of radioactive material. A typical application is in building area contamination surveying prior to the decommissioning or clean out.

### *Physical principles*

Detector: CsI (Tl) scintillator photodiode with built-in main amplifier.

### *Mobility*

Transportable.

### *Operation*

The RadScan 600 can be deployed in many ways including mobile crane, tripod and manipulator. The system uses one of two interchangeable detector heads. These are optimised for plutonium and fission product  $\gamma$ -energies. Set-up is achieved within a few seconds of the scanner being placed in the viewing location. System self-checking is automatic. The RadScan 600 is then capable of surveying all objects within its environment. The system is operated from the comfort and safety of a remote location through a dedicated PC-based power and control module. On- screen information is presented through Windows 3.1 compatible software and includes a high definition colour video picture of the area being surveyed overlaid by colour software graphics. The overlaid output is recorded on a VCR.

The system is controlled by use of a rollerball and cursor keys. Communications between control module and scanning head is via a single cable.

### *Detection limit*

<10 $\mu$ Ci for  $^{137}\text{Cs}$  in field of view at 1 m.

### *Data presentation*

*Count Mode* displays the raw count rate at the detector from objects within the current viewing zone with no correction for distance. This can be for up to 3 user-defined ROIs.

*Range Corrected Mode* allows the operator to correct the real-time count rate(s) to what it would be if the object were at a range specified by the user.

*Normalised Mode* allows the operator to normalise the real-time count rate(s) to the count rate from an object specified by the user and to correct for range. This enables the operator to estimate whether the zone contains more or less activity relative to the normalisation zone.

#### *Calibration and maintenance*

Periodic energy calibration is performed using a sealed source. This is menu-driven and allows energy calibration of the spectral analysis package.

#### *Measurement Precision*

The viewing zone circle displayed on-screen is colour coded to indicate the reliability of the displayed count rate information. When the count rate at the detector is relatively high, the circle defining the field of view will become green within a second or two of the scanning head coming to rest. This indicates that enough counts have been received to enable the count rate to be determined within operator pre-set uncertainty limits.

When the count rate at the detector is low, the circle will remain red until enough counts have been received. If the count rate exceeds the limit of the detector system, the operator is informed.

#### **Mobile Assay System**

##### *Destructive or non-destructive?*

Measurement needs are immediate and other non-destructive assay instrumentation is unavailable.

##### *Physical principles*

Highly sensitive active neutron measurement is performed using pulsed Zetatron neutron source and imaged differential decay analysis.



High resolution  $\gamma$ -ray spectroscopy identifies and quantifies radioactive constituents.

Neutron detectors are  $^3\text{He}$  proportional counter which surround the sample chamber.

Gamma-ray analysis is performed with a collimated high purity Ge-detector.

#### *Dimensions*

10' x 40' (3 m x 12 m) trailer houses all measurement systems, computers, and supporting electronic equipment.

#### *Mobility*

On site set up time is less than one day.

#### *Detection limit*

< 1 mg total plutonium.

#### *Data presentation*

Full Windows 3.1 and NT compatible.

#### *Personnel requirements*

Measurements are performed by our experienced operators or training for your operational staff will be provided.

#### *Time consumption*

15 min - 30 min assay times.

## **TRU Drum Monitor**

### *Physical principles*

Passive neutron coincidence counting combined with high resolution  $\gamma$  spectrometry.

### *Operation*

System boot-up, test and background determination is automatic. In normal operation the system prompts the operator to load and unload drums. Once loaded the drum is rotated during the measurement to allow a better all round view of the drum by the mechanically integrated high resolution  $\gamma$  spectrometry (HRGS) detector.

### *Detection limit*

20 mg total plutonium, in 1000 second count.

### *Data presentation*

This is via a rugged display unit, built into the monitor housing (remote operation is also possible as an option). Large and rugged keys are used to operate the software driven menus. The displays have been designed for clarity and ease of use. both screen and keys are designed for easy decontamination. Maintenance and diagnostics modes are password protected.

On-screen outputs include drum identification (confirmation), weight, Pu mass and overall error and Pu isotopic composition (either measured or default). Full assay information, including  $\gamma$ -spectrum and neutron count information is stored onto the system logger (hard drive) and can be copied to floppy disk or an optional optical disk drive.

### *Calibration and maintenance*

At preset intervals the monitor prompts the operator for a calibration check. An approved Pu calibration standard is then loaded into the measurement chamber and the calibration routine automatically carried out.

### *Time consumption*

An operator preset confidence interval for the assay result can be used to determine count times for individual drums, depending on drum activity levels. This optimises monitor utilisation across a broad range of drum activities.

## **Uranium Drum Enrichment Monitor**

### *Description*

The Uranium Drum Enrichment Monitor (UDEM) allows the fast and accurate verification of the contents of sealed uranium product drums without the need for expensive sampling routines.

### *Physical principles*

UDEM identifies product drums containing higher enrichment than normal product using  $\gamma$ -detection techniques.

High resolution  $\gamma$  spectrometry.

Measurement control, including automatic regular self-checking of  $\gamma$ -detector and counting electronics.

### *Detection limits*

Typical measurement range: 0 - 2%  $^{235}\text{U}$

### *Calibration and maintenance*

Calibration of UDEM is carried out using sealed uranium oxide standards of known %  $^{235}\text{U}$  enrichment contained in purpose made drums of similar construction to those used to contain the uranium product.

### *Personnel requirements*

All routine operator interactions with UDEM are via the plant process computers and their displays. Non-routine measurement control or fault diagnosis is provided via a dedicated maintenance terminal. This also allows the operator to enter an off-line maintenance mode where the instrument operating parameters, e.g. count time, may be viewed or altered.

*Time consumption*

Measurement time: 600 sec.

*Existing installations*

Considerable operational experience has been gained with these systems through their successful use on the THORP reprocessing plant on the UK Sellafield site. BNFL Instruments has worked closely with the plant designers to allow the instrument to be integrated seamlessly into the drum handling plant operations.

**DISPIM**

**Decommissioning In-Situ Plutonium Inventory Monitor**

*Description*

Total plutonium assay of redundant plant items prior to removal from plant process line. Flexibility to monitor wide range of object sizes and shapes up to 5 m<sup>3</sup>.

Easily relocates to any part of the plant via optional self-propelled transporter.

Protection against all forms of electrical interference which may occur in an industrial plant environment.

Easily decontaminated.

Individual neutron detector modules easily lifted by personnel.

Optional  $\gamma$  spectrometry system incorporates plutonium isotopic analysis and PuF<sub>4</sub> analysis software.

*Physical principles*

Comprise up to 30 polyethylene moderated <sup>3</sup>He detector modules combined with charge amplifiers and neutron coincidence counting electronics, interconnecting cables.

### *Operation*

Individual neutron detector modules are located adjacent to the available surfaces of the target plant item. A simple yet rugged cabling arrangement links all detector modules to the neutron coincidence counting electronics and an easy-to-follow system standardisation confirms the correct functioning of all system elements. Following calibration of DISPIM for the particular target plant item (see "Calibration") a measurement is carried out. The resulting calculation of  $^{240}\text{Pu}$  equivalent mass is automatically interpreted to give total plutonium content and error values using measured or default plutonium isotopic composition data. Enhanced precision and lower detection limits can be obtained by extending the particular measurement counting time.

### *Detection limits*

The performance of such a system is dependent on the conditions relating to a particular measurement (detector geometry, matrix characteristics, background radiation levels and plutonium isotopics).

However, for a typical plutonium contaminated glove box monitoring exercise, with a count time of the order of a few hours, a detection limit of a few grams Pu is usually achieved.

Detection limit:  $<1\text{g } ^{240}\text{Pu}$  equivalent (total Pu content if grade is known or determined by optional  $\gamma$  spectrometry system).

### *Data presentation*

The display has been designed for clarity and ease of use. The system is suitable for use on a routine basis by operators with minimal training. Maintenance and diagnostics are password protected.

### *Calibration and maintenance*

In-situ calibration of the system is performed via the "add-a-source" technique using a  $^{252}\text{Cf}$  source of known activity. This will determine the neutron detection efficiency of the particular arrangement for fissile material in a variety of locations within the plant item being monitored. For criticality safety measurements, worst case calibration is always used by the software, such that the plutonium mass calculated is based on the lowest efficiency measured during the calibration exercise. However, a knowledge of the likely hold-up areas in target plant items can also lead to a "best estimate" of plutonium content. A mathematical modelling service is available for calibration of plant items that are unsuitable for the "add-a-source" technique.

### *Existing installations*

Considerable operational experience has been gained with these systems through their successful use in a number of decommissioning projects on the UK Sellafield site. They are rugged, reliable and flexible. DISPIM systems have also been supplied to operational plant for special investigations or special measurements, such as in-situ drum counting of problem drummed wastes. In addition, a DISPIM system has been configured to monitor decommissioned plant items stored in boxes/crates.

### **IPAN/GEA Drum System**

#### *Description*

The PSC IPAN/GEA Drum System is the best option when you have the following requirements:

- TRU waste assay for safeguards, criticality, transport and disposal purposes.
- Reliable and efficient assay.
- Continuous operation in industrial environments.
- Wide range of different waste matrices and/or Pu isotopic compositions.
- Safe and confident use by operational staff with minimum training requirements.

#### *Physical principles*

- Active neutron imaging using pulsed Zetatron source and differential dieaway analysis.
- Passive neutron imaging.
- Active and passive neutron multiplicity counting using PSC PMCCM electronics.

#### *Operation*

System boot-up, test and background determination is automatic. In normal operation the system prompts the operator to load and unload drums. Once loaded the drum is rotated during the measurement to allow a better all around view of the drum by the mechanically integrated high resolution  $\gamma$  spectrometry (HRGS) detector.

#### *Data presentation*

- Full Windows 3.1 and NT compatible.
- Network compatible.
- Allen Bradley and April PLC compatible
- Full diagnostic capabilities.
- Routine and custom result reporting.
- Local, network and PLC operational interfaces.

On-screen outputs include drum identification (confirmation), weight, Pu mass and overall error and Pu isotopic composition (either measured or default).

Full assay information, including  $\gamma$ -spectrum and neutron count information is stored onto the system logger (hard drive) and can be copied to floppy disk or an optional optical disk drive.

#### *Calibration and maintenance*

At preset intervals the monitor prompts the operator for a calibration check. An approved Pu calibration standard is then loaded into the measurement chamber and the calibration routine is automatically carried out.

### **IPAN/GEA Crate System**

#### *Description*

Fast, accurate and efficient analysis of large TRU and low level waste items allowing selection of appropriate waste disposal option.

#### *Physical principles*

Active neutron imaging using pulsed Zetatron source and differential decay analysis. Passive neutron imaging.

Active and passive neutron multiplicity counting using PSC PMCCM electronics.

#### *Operation*

While inside the main neutron counting chamber, the crate passes between 16 NaI detectors and 1 HRGS. This data is automatically combined with the neutron imaging and multiplicity counting data to enable matrix and isotopic corrections and provide an accurate total Pu mass assay.

#### *Data presentation*

- Full Windows 3.1 and NT compatible.
- Network compatible.
- Allen Bradley and April PLC compatible.
- Full diagnostic capabilities.
- Routine and custom result reporting.
- Local, network and PLC operational interfaces.

A rugged key system and monitor are used to operate the software driven menus. This provides a simple and convenient interface which is easy to decontaminate. All measurement parameters are automatically stored to disk and may also be directly printed. Diagnostic and maintenance operations are password protected.

#### *Calibration and maintenance*

At preset intervals the monitor prompts the operator for a calibration check. An approved Pu calibration standard is then loaded into the measurement chamber and the calibration routine is automatically carried out.



## **Appendix D**

### **Information about available measuring systems and equipment from Canberra Nuclear Industries**

#### **General**

##### *Systems available from Canberra*

- Free release systems  $Q^2$  based on Gamma Spectroscopy
- Segmented Gamma Scanners (S.G.S.) based on Gamma Spectroscopy using vertical moving Ge-detector and transmission source
- Passive Neutron Systems based on detection of  $n_0^1$  from transuranics. Systems are available with or without "add a source" option.
- Active neutron systems based on activation with neutron source or neutron generator.

##### *Destructive or non destructive measurements?*

Non destructive measurements.

##### *Mobility*

Can be both stationary or mobile (in truck, trailer or container).

##### *Requirements for installation and operation*

No special requirements, LN is needed for Ge-detectors. A 220 V power source is needed.

##### *Storage and presentation of data*

This is flexible. A multi ways of reports are available or possible.

### *Calibration and maintenance*

Calibrations are depending on the system. The calibration is done at the factory (Canberra US) using Canberras own calibration standards (200 litre drums) for different waste densities. Calibrations can also be done at the customer site using their own or Canberra employees.

### *Personnel requirements*

Normally one operator is needed with normal skills.

### *Time required for one typical measurement*

Typical 10 minutes.

### *Radiation shielding*

All systems come with necessary shielding and/or collimators. Concerning shielding to the operator this has been constructed depending on the environment and the dose.

### *Stage of development*

Systems are available for sale, leasing or renting. Most of all the systems are standard systems already delivered in many places. Canberra is continuously developing and adapting the systems to new standards. The construction and design is completely done at Canberra Industries (USA) and/or Canberra Europe (Belgium).

### *Hiring*

The system can be hired.

## **WM 2110 series Q<sup>2</sup> Low Level Waste Assay System**

### *Area of use*

Quantitative and qualitative  $\gamma$ -assays for unprocessed waste in 200 litre drums.

### *Physical principles and dimensions*

Drums are measured in a low background 10 cm steel shielded chamber. The shielding can be upgraded to 15 cm steel. The drum is rotated at approximately 10 rpm during measurements. The system is equipped with 3 coaxial Ge-detectors mounted vertically along the side of the chamber. The system is also available with two large area NaI-detectors. Calibration is done at the factory for turnkey operation on includes a QA check source and holder or can be done by the customers with their own calibration standards.

Size: 152 x 173 x 122 cm

Weight: 10 cm shield: 7260 kg  
15 cm shield: 9980 kg

### *Sensitivity/detection limits*

111 Bq at 0.1 g/cm<sup>3</sup> material density and 446 Bq at 1.7 g/cm<sup>3</sup> material density for nuclides with  $\gamma$  energies from 300 to 1500 keV achieved in 10 minutes measurements for Ge-detectors and in 1 minute for NaI-detectors.

### *Data processing, storage and presentation*

Utilise a PC-based menu driven and multitasking system for control, analysis and presentation. All data, set up parameters and analysis results are stored for review and reanalysis. The system perform automatic weighting for matrix attenuation correction due to sample density. By using customer-provided correction the systems calculates the activity of alpha/beta emitters that cannot be measured directly. Calculation of plutonium and uranium isotopes can be made using optional Multi-Group Analysis (MGA) Code. Customer editable report formats are available.

## **Segmented Gamma Scanning System (S.G.S)**

### *Area of use*

Automatic and continuous waste analysis of up to 20 waste drums per hour. A "Super Compactor" can reduce a 200 litre drum to a height of 15 cm in order to achieve waste volume reduction.

### *Physical principles*

All drum handling and assay operations are automated including weighting, rotation and  $\gamma$ -scanning. The system is equipped with four lead collimated Ge-detectors for  $\gamma$ -assays and four dose rate meters for measurements of dose rates at the surface of the drums. The system is also equipped with four  $3,7 \cdot 10^5$  kBq  $^{152}\text{Eu}$  transmission sources in order to assay matrix densities.

### *Sensitivity/detection limits*

< 30 kBq for a homogenous content density of  $0,2 \text{ g/cm}^3$ .

### *Data processing, storage and presentation*

The system uses two 486 processors. The operator can choose fully automatic operation including report generation and data base update for minimum operator interaction or other options of intervention. Calculation of plutonium and uranium isotopes can be made using optional Multi-Group Analysis (MGA) Code. Customer editable report formats are available.

## **Model WM 3210 Passive/Active Neutron Cf-252 Shuffler system**

### *Area of use*

Assessment of  $^{235}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in radioactive waste.

### *Physical principles*

During measurements the drums are located inside a chamber shielded by tungsten, high-density polyethylene and boron-loaded polyethylene.

In the active mode the system utilise a  $8 \cdot 10^8 \text{ n/s } ^{252}\text{Cf}$  neutron source for irradiation of the sample. Measurements of delayed neutrons from fission and  $^{239}\text{Pu}$  and  $^{235}\text{U}$  are performed approximately 3,5 seconds after the source has been removed to a shielded position. The process of irradiation and measurements is repeated until a sufficient good counting statistics have been achieved. By removing the Cd-lining of the counting chamber thermal neutrons can be measured increasing the sensitivity of the system.

Measurements of spontaneous fission neutrons from  $^{240}\text{Pu}$  are made in passive mode with the  $^{252}\text{Cf}$  source in the shielded position.

Neutrons are measured by 54  $^3\text{He}$ -detectors by using a coincidence detection technique.

*Sensitivity/detection limits*

For a 1000 second count time:

- In active mode
  - Fast neutrons : 300 mg  $^{235}\text{U}$
  - Thermal neutrons : 3 mg  $^{235}\text{U}$
- In passive mode : 4 mg  $^{240}\text{Pu}$

**Installed waste systems - World wide summary**

System	Europe	North America	Pacific Rim/Asia
JCC-21 Passive Drum Counter	1	2	3
WM3110 Passive Counter	1	1	3 - Del. 8/95
High Efficiency Passive Counter		joint devel.	
WM3210 Cf-252 Shuffler		3	
Combined Thermal Epithermal Neutron System		joint devel	
Differential Die-Away Counter	1 - joint with CEA		
Passive Neutron Slab Counters	1	1	
WM2100 Q <sup>2</sup> Low Level Waste Assay Systems	7	14	1
WM 2200 Segmented Waste Assay System	19	44	2
Multi-detector Segmented Scanner	1	2 - Del. 3/96	
Isotopics System.	1		3 - Del. 8/95
WM4100 Mobile Neutron/Gamma Scanner		1	
WM 4200 M.A.D.A.M.		3	
Soil Conveyor Monitors		4	

## **Appendix E**

### **Information about available measuring systems and equipment from Gamma Data**

#### **General**

*EG&G ORTEC System for measurement and analysis of radioactive waste and Safeguards applications*

The system consists of:

- Special SGD (SafeGuardD) detectors
- Unmatched data acquisition units; portable or stationary
- A complete package of software dependent on application

#### *Non-destructive measurements*

Since the waste is expected to emit gamma-rays, the measuring system is based on non-destructive measurement principles.

#### *Principles for operation*

The system is designed as a standard Ge detector-based system with portable detector and electronics and can easily be used in field.

#### *Portable system*

The system is portable but can also be provided as a stationary laboratory system.

#### *Requirements for installation and operation*

No special requirements for installation and operation. General knowledge of high-resolution gamma spectroscopy is valuable. Liquid nitrogen supply dependent on application.

#### *Detection limits*

Overall detection limits dependent on physical conditions and measuring time. The choice of detector size is, of course, also of great importance. Using ISOTOPIC, a lot of data and experience are available from Richard Haugenauer at the Oak Ridge K-25 facility.

### *Storage and presentation of data*

The system is PC-based (Windows95/NT) and data stored and presented accordingly.

### *Calibration and management*

Calibration is done on site by the user using a point source. Stored calibrations can also be used. No maintenance other than filling cooling-medium is required.

### *Personnel requirements*

The personnel should be trained in general high-resolution gamma spectroscopy.

### *Time for one measurement*

Typically a few minutes. Typical time requirements dependent on physical conditions such as total activity and shielding of the sample.

### *Shielding*

The most applicable shielding principles chosen by the user. The ISOTOPIC software corrects for different collimators and shielding.

### *The system is available for sale*

The hardware is developed and manufactured by EG&G ORTEC.

### *Information on existing installation*

ISOTOPIC is extensively used by U. S. DOE facilities.

### *Hiring*

The system can be hired according to agreement.

## **Description of the system**

### *The Basics of the Measurement Problem*

The issue is the control of "Special Nuclear Material" (SNM), be it plutonium, uranium, waste, reactor fuel or scrap. The ultimate measurement problem to be solved is determining the mass in gram of the special nuclear material. To determine the absolute mass of material present requires that two different measurements be made: one of the so-called isotopic ratios and the other, either measurement of total heat output by calorimetry, or the neutron flux.

### *Peak Ratio Methods*

One can directly determine the isotopic ratios of all gamma-emitting isotopes of a given element by measuring the ratio of the corrected count rates in their respective gamma peaks. The corrections are necessary to account for the effects of geometry, absorption and detector efficiency. Since the peaks are close together in energy requiring the very best stability and resolution from the detector and electronics are essential, see below.

### *SGD-detectors*

High-performance Germanium Detectors optimized for Safeguards.  
Planar HpGe detector for low energy gamma-rays and Coaxial HpGe detector for high energy gamma-rays. Both detectors exceed the demanding resolution requirements of isotopic software codes. New, high-stability, low-power preamplifier. Stationary or portable dewar. Electrically cooled as option.

### *Electronics*

DSPEC	Digital Gamma-Ray Spectrometer
DART	Portable High-Resolution Gamma Spectrometer

DSPEC and DART are industry leaders in regards of peak-position stability over a wide range of count rates which is very important when measuring waste. See enclosed paper from Phoenix INMM Meeting for an overview.

### *Software*

#### **General**

Due to the overall measurement problem discussed, it is impossible to apply a general-purpose analysis program. Depending on the physical conditions and constraints, different programs have to be used instead. Packages such as



ISOTOPIC, MGA and PC/FRAM were developed specifically to determine isotopic ratios.

ISOTOPIC Spectroscopy software for HpGe Gamma-Ray Analysis of Poorly-Characterized Materials.

ISOTOPIC helps characterize material when a standard cannot be prepared to match the physical characteristics of the sample. Such sample occurs often in waste management and NDA applications. ISOTOPIC is used for determination the weight (gram) or activity of all gamma-emitting isotopes in such poorly characterized matrices. This program is in daily use in a number of U. S. DOE facilities and built into the program are geometries and attenuation features. Different geometries are stored in a database and the user may interactively change container material, thickness, matrix density etc. until all activity results are consistent. The program is extremely user friendly and can be used with any detector and collimator. The attenuation parameters are fine-tuned by the user and an external (and expensive) calibration is not necessary. A large number of matrices can be applied. The result can be normalized to other methods.

MGA++ Software for Non-Destructive Analysis

The MGA software was originally developed at Lawrence Livermore National Laboratory (LLNL). The software is used for analysis of the isotopic composition of plutonium mixed with other actinides, or for analysis of uranium mixtures. MGA++ is the code most commonly used by international Safeguards inspectors for accountancy measurements. All future enhancements to the software will come directly from LLNL.

PC/FRAM Software for Non-Destructive Analysis

Fixed-energy Response-function Analysis with Multiple efficiency was originally developed at the Los Alamos National Laboratory. The program is used for analysis of heterogeneous Am/Pu pyrochemical residues. Compared to MGA++ is more useful when attenuation problems are present and low-energy lines may be undetectable. PC/FRAM can handle sample inhomogeneity by the use of multiple-efficiency curves.

MGA-CZT

As part of a joint development with ORTEC, LLNL has produced a version of MGA to work with plutonium sample spectra from CZT detectors. Further information on request.

E-Meter

As option to the above Peak Ratio methods this method utilizes a stabilized NaI detector and portable electronics (DART or MicroNOMAD). This method is widely used for the measurement of uranium enrichment, being the measurement of the ratio of U-235 to U-238. The problems which emerge, however, are:

- Requires Calibration Standards
- Easily biased if interference's are present
- Difficult to use accurately when matrix or container attenuation effects are present.

While of limited applicability, the E-meter method is still used extensively for measuring uranium enrichment and EG&G ORTEC can offer a system based on this method.

## **Appendix F**

### **European network of testing facilities**

A European network of testing facilities for the quality checking of radioactive waste packages was formed in 1992 on the initiative of the Commission of the EC as a natural extension of the Community's R&D programmes on radioactive waste management, in particular the waste characterisation and quality assurance studies. Its objectives are to promote and facilitate collaboration in the development, application and standardisation of quality checking of radioactive waste packages so that conformity of conditioned radioactive waste with regulatory specifications and criteria can be verified.

A laboratory belonging to a country of the EU can be a member of the network if it is regularly performing quality checks on radioactive packages, is ready to share the relevant technical information, and willing to pay an annual fee of 1000 ecu covering workshops and publications.

Major laboratories from Austria, Belgium, Germany, France, the Netherlands, Italy, Spain, UK and the EC's own Joint Research Centre are presently contributing to the 'Network'.

Five working groups have been created covering the following topics:

- non-destructive testing by gamma assay
- measurement of volatile releases from waste packages
- general quality assurance/quality control procedures
- active and passive neutron assays
- chemical and radiochemical destructive analysis

External laboratories have participated in workshops arranged by the first working group, among others VTT from Finland. Otherwise the Nordic countries have so far not been involved with the activities of the Network. However, it obviously covers much of the same ground as discussed in this NKS report and further efforts should therefore be co-ordinated to avoid duplication.

Technical reports from the working groups are so far not available, but a general status for the system was presented at the Luxembourg conference in 1996 [1]. The annual report for 1995 has recently become available [2].

- [1] P. Van Iseghem, G. Brunel, C. Lierse, A. Morales, R. Odoj, F. Troinani, M. Hugon. "The European network of testing facilities for the quality checking of radioactive waste packages: Objectives and status." T. McMenamin ed. Fourth European Conf. on Management and Disposal of Radioactive Waste. EUR 17543 EN, 1997, p. 286-295.

- [2] European Network of Testing Facilities for the Quality Checking of Radioactive Waste Packages. Annual Report 1995. Nuclear Science and Technology. EUR 17559 EN, 1997.