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Classification, characterization and management of radioactive waste in Italy

N. Cherubini([1]), G. Gandolfo([2]), L. Lepore([2]) and G. A. Marzo([2])(*)

([1]) *ENEA, Technology, Facilities and Materials for Nuclear Fission Division, Casaccia Research Centre - Rome, Italy*
([2]) *ENEA, Nuclear Material Characterization Laboratory and Nuclear Waste Management, Casaccia Research Centre - Rome, Italy*

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Summary. — Radioactive waste produced in Italy belongs to two main categories according to production routes. These are radioactive waste generated from the nuclear fuel cycle and those radioactive waste deriving from non-electronuclear activities, *i.e.*, from nuclear medicine activities, industrial, and scientific research. The latter are responsibility of the Integrated Service for the management of radioactive waste: a group of authorized Operators supervised by ENEA (Italian National Agency for New Technologies, Energy and Sustainable Economic Development). In this paper, the Italian regulatory framework, the classification of radioactive waste and the most used characterization techniques, both destructive and non-destructive, are outlined.

1. – Introduction

Radioactive waste produced in Italy belongs to two main categories, depending on their origin. The radioactive waste that comes from the activities of the nuclear fuel cycle, *i.e.*, from plants involved in the production of electricity and related facilities, are part of the decommissioning activities under the responsibility of Sogin (a State company owned by the Italian Ministry of Finance). Radioactive waste deriving from non-electronuclear activities, *i.e.*, from nuclear medicine activities, industrial, and scientific research activities, are responsibility of the Integrated Service for the management of radioactive waste. This is a group of Operators, authorized by Law, supervised by ENEA (Italian National Agency for New Technologies, Energy and Sustainable Economic Development). ENEA is the public entity that becomes the final owner of the radioactive waste and the dis-

(*) Corresponding author. E-mail: Giuseppe.marzo@enea.it

used radioactive sources produced at national level and delivered through the Integrated Service Operators until their final disposal at the Italian National Repository. Sogin is in charge of designing and building the Italian National Repository, a Near Surface Disposal Facility structure that will allow the disposal of waste originated both from the decommissioning of both the Italian nuclear fuel cycle facilities and from medical, industrial and research activities.

The Integrated Service was established by ENEA with a resolution on June 4th 1986, following specific provisions of the Italian Government, which attributed to this public body the responsibility of intervening in the sector of low and medium activity waste, produced nationally, in order to ensure collection, custody and management and in particular for medium and low level waste from industrial and healthcare activities. This attribution was then reconfirmed by Legislative Decree 52/2007 and, more recently, by the Legislative Decree 101/2020, where the Integrated Service is recognized as a technical-operational tool capable of taking charge of all phases of the management cycle of the sources that are no longer used.

Before the Italian National Repository, the aforementioned Integrated Service is the only entity on the national territory since the 1980s, capable of playing an essential role in the management of radioactive waste of non-electronuclear origin. The proper management of radioactive waste is relying upon the IAEA classification of radioactive waste [1]. In the context of this classification, each Member State (and stakeholders involved in the technical management of such a waste, *i.e.*, collection, treatment, conditioning, storage, etc.) establishes regulations and technical guides for the proper management of radioactive waste. In this framework, the radiological characterization plays an essential role in discriminating and classifying waste streams within the interna- tional and national classifications, as long as other criteria, such as chemical composition of the waste matrices, that may produce sub-classification or modifications in disposal approaches.

In this paper, the Italian management of radioactive waste is summarized. In particular, the national regulatory framework, the classification of radioactive waste, the most used radiological characterization techniques, both destructive and non-destructive, and the waste disposal approach are outlined.

2. – Classification of radioactive waste

This section summarizes some historical evolution steps in radioactive waste management within the Italian regulatory framework, as long as some definitions and practical concepts playing a fundamental role in the field.

2˙1. *The international reference*. – Any use of radioactive materials generates radioactive waste, inevitably. Radioactive waste consists of material, solid or liquid, contaminated by radioactive substances distributed in the mass of the waste. Different radioactive waste are associated with different origins in terms of radionuclides contained, in concentration and toxicity, and even waste produced in the same installation can significantly differs from one another.

The variability of the characteristics of radioactive waste makes it necessary to adopt a classification system, above all for the purposes of defining criteria and requirements for the various phases of their management and, more generally, to make information exchange efficient.

Depending on the specific purpose, the classification systems can refer to various characteristics of the waste, for example the radionuclides contained, their concentration, decay time, the type of radiation emitted, the waste matrix, the potential production of heat. The classification system which the different countries refers to is proposed by the IAEA (International Atomic Energy Agency) and described in the General Safety Guide No. GSG-1 Classification of Radioactive Waste [1].

The basic principle is the subdivision of radioactive waste based on the type of confinement necessary to safely store them which, in turn, is based on the radiological content that characterizes each class of waste, both in terms of activity and half-life.

2˙2. *Technical Guide No. 26*. – The specific classification adopted in Italy was introduced in 1987 with the Technical Guide No. 26 [2] issued by the ENEA-DISP, the former Italian control authority (subsequently it became ISPRA, Higher Institute for Environmental Protection and Research, and now ISIN, Ispectorate for the Nuclear Security). [2] grouped radioactive waste into three categories. Such a guide coupled the requirements for management to indications for disposal within each category.

Shorter-lived radioactive waste is classified in the *first category*, *i.e.*, waste requiring up to a maximum of a few years to decay at radioactivity concentrations lower than those established by the legislation for exemption from authorization for disposal in the environment, or those which such permission has been granted for. With the concentration dropped below such levels, waste was allowed to be managed and disposed of as no longer radioactive, but in compliance with the regulations about hazardous waste. Most of the first category waste was produced in the medical use of radioactive substances.

The *second category* included radioactive waste requiring from a few tens to a few hundreds of years to decay at concentrations of radioactivity close to the environmental background or even waste with very long-lived radionuclides originally in concentrations comparable to those occurring in the environment. Second category waste was potentially produced by the use of radioisotopes for medical, industrial or research purposes, and in nuclear power plants and related fuel cycle.

Waste decaying to the environmental background level in thousands of years or more was classified in the third category. Third category waste was produced in nuclear power plants and related facilities, especially in those dealing with the reprocessing of the irradiated fuel, *i.e.*, the burned fuel at the end of its life cycle within a nuclear reactor core, to recover and reuse fissile Uranium and Plutonium isotopes to produce Mixed OXides (MOX) fuels. The same irradiated fuel was considered as third-category waste when not intended to be reprocessed.

2˙3. *Decree of August 7th 2015*. – In order to make the Italian classification of radioactive waste consistent with international references and compliant with [1], the Italian Ministry of the Environment and the Protection of the Land and Sea, jointly with the Italian Ministry of Economic Development, with the Decree of August 7th 2015 reformulated the classification of radioactive waste on the sole basis of their radiological content. Therefore, the Decree does not provide indications on the management of waste belonging to the specific categories which remained outlined in the Technical Guide No. 26 [2], pending a new technical guide or a regulatory integration.

2˙4. *Technical Guide No. 33*. – The gap between waste classification and its management has been filled by the Technical Guide No. 33 [3], the current Italian reference in

radioactive waste management. It establishes the criteria that must be respected for a correct management of radioactive waste, taking into account the nature and radiological risk associated with each type of waste, and defines the minimum requirements for radioactive waste packaging for their acceptance to the disposal or long-term temporary storage.

The guide [3] guarantees the compliance to current international standards, it incorporates the recommendations of the IAEA, and the safety requirements developed by WENRA, the European Association of Nuclear Safety Authorities, on the treatment and conditioning of radioactive waste and on disposal.

2˙5. *Dismissed radioactive sources.* – A radioactive source is a radioactive material specially packaged in a suitable form in order to exploit the specific goal for which it has been produced, as defined by the Italian Legislative Decree 101/2020. In Italy, a radioactive source that ceases to be of use to its owner or holder should be: i) reused by a new owner or holder for whom the source is still valuable and usable; ii) returned to the manufacturer to be refurbished (*e.g.*, re-activation of Cobalt pellets in nuclear reactors to produce ^{60}Co). When these two possibilities are not feasible, the source becomes a radioactive waste falling within the radioactive waste classification. In general, sources may have radiological characteristics requiring level of containment and insulation greater than those provided by Near Surface Disposal Facilities requiring additional shielding or conditioning without heat dissipation systems. This class of waste includes all sources with a high activity concentration (as defined in [4]), sealed and unsealed, with a special form certificate or without it.

2˙6. *Practical classification.* – Waste classification established by the international standards, taking into account the activity level of the radionuclides and their half-lives, allows to identify the management system more appropriate with respect to their radiological characteristics. However, it might not take into account certain degrees of complexity which lead to the retention of a management system different from that corresponding to the class which the waste is assimilated to. Other criteria, such as package stability over time or the presence of toxic chemicals, must also be taken into account, as long as other needs, *e.g.*, to reduce the volume and harmfulness of ultimate radioactive waste. Two important aspects concerning the classification of radioactive waste should be emphasized:

- there is no single classification criterion for determining the class of a waste. It is indeed necessary to study the radioactivity of the various radionuclides present in the waste to assess its most appropriate classification (*e.g.*, for certain radionuclides, the item could be part of a class, and for some others it could be part of another class: a decision criterion could be strongly related to the nature of the waste and its effective features);

- a waste may fall within a defined category but may not be accepted in the corresponding management chain due to other characteristics (*e.g.*, its chemical composition, potential production of gases, etc.).

Moreover, thanks to the improvement of the knowledge of the radioactive waste as well as the progress of the studies carried out on the optimization of the modes of treatment and conditioning, the options of waste management can evolve.

Concretely, current good practices distinguish a limited number of categories of radioactive waste on the basis of the different disposal routes, the latter depending also on the characteristics and operating requirements of the plants used for treatment, conditioning and, more generally, management operations. These different types of waste correspond to different costs associated with their disposal too and can include more than one category of waste as identified by the classifications described or, within the same category, two different management paths. Such practical classification is reported in table I, where it is also compared with the formal international and national classifications. This classification does not refer to fuel from the decommissioned Italian nuclear power plants.

3. – Characterization of radioactive waste

The objective of the radiological characterization is the identification and quantification of the radionuclides occurring in the radioactive waste. The characterization of radioactive waste is one of the fundamental elements of waste management, for classification purposes as long as for practical reasons such as safety assessments during transport or storage, determination of waste treatment and conditioning methods, as well as improvement of quality controls during these operations. An adequate design of the disposal site of conditioned radioactive waste also obliges the bodies in charge of it to be aware of the chemical-physical characteristics and the degree of radiological toxicity associated with the waste, which implies the use of data coming from global characterization operations (*i.e.*, radiological and physico-chemical). Finally, a correct management of radioactive waste that takes into account the necessary safeguards of health and environmental protection poses the need for the definition and classification of radioactive waste, as well as that of the legal determination of limits below which the waste can be unconditionally released without constraints on their destination or limitations on their use. The activities related to the waste characterization are carried out during different stages of the life cycle of the radioactive waste: during generation, during handling, treatment, conditioning, and during the disposal and storage phases. The type and importance of characterization actions depend on many factors, such as:

- the type and physical form of the waste;
- the regulatory regime;
- the amount of ancillary information available;
- the type of physical quantities to be measured and parameters to be determined.

From a radiological point of view, the characterization of the waste can be carried out through different methods, destructive and non-destructive. Non-destructive techniques consist in measuring the intensity of the emitted radiation, spontaneous or induced, for the purpose of qualitative and quantitative analysis of the radioactive material. They do not alter the physical state and chemical composition of the material, but have relatively low sensitivity and are often influenced by strong matrix effects. These types of techniques can be performed both in the field and in the laboratory, and they rely almost entirely upon gamma and neutron detection. Destructive techniques consist of the analysis of samples carried out in the laboratory, using physical (*e.g.*, thermal) and chemical treatment methods to set the sample in a form suitable for the subsequent measurements (usually the liquid form). They are more sensitive and accurate than non-destructive techniques even if far more time consuming and require that the samples analysed are uniform and sufficiently representative of the original material.

TABLE I. – *Practical classification of radioactive waste currently adopted in Italy in comparison with formal classifications.*

Description	IAEA	Decree 7-Aug-2015	Technical Guide No. 26
Radionuclides with half-life <75 days for which disposal as hazardous waste is foreseeable	Exempt Waste, Very Short Lived Waste	Exempt Waste, Very Short Lived Waste	First category
Beta and beta-gamma emitting radionuclides with half-life > 75 days, with activity concentrations such as to guarantee activity concentration values lower than the clearance levels within 2 years of delivery	Very Short Lived Waste, Very Low Level Waste Exempt Waste	Very Short Lived Waste, Very Low Level Waste	First category
Beta-emitting radionuclides, with activity concentrations such as to guarantee activity concentration values lower than the clearance levels within 300 years of delivery	Very Low Level Waste, Low Level Waste	Very Low Level Waste, Low Level Waste	Second category
Beta-gamma emitting radionuclides, with activity concentrations such as to guarantee activity concentration values lower than the clearance levels within 300 years of delivery	Very Low Level Waste, Low Level Waste	Very Low Level Waste, Low Level Waste	Second category
Beta-emitting radionuclides, with activity concentrations such as to guarantee activity concentration values lower than the clearance levels within 300 years of delivery	Very Low Level Waste, Low Level Waste	Very Low Level Waste, Low Level Waste	Second category
Beta-gamma emitting radionuclides, with activity concentrations such as to guarantee activity concentration values lower than the clearance levels within 300 years of delivery	Very Low Level Waste, Low Level Waste	Very Low Level Waste, Low Level Waste	Second category
Beta-gamma, alpha or neutron emitting radionuclides with high activity concentration and sources of high activity	Intermediate Level Waste	Intermediate Level Waste, High Level Waste	Third category

3˙1. *Non-destructive techniques.* – This type of analysis consists in leaving the sample to be characterized as it is, without any appreciable physical or chemical alteration. The characterization takes place through the detection of gamma or neutron radiation emitted spontaneously by the sample or induced by an external radiation source.

3˙1.1. In situ gamma spectrometry. The material to be identified is initially characterized in the field, using portable or transportable equipment.

The former are measurement systems consisting of a single detector, calibrated experimentally (by using certified sources) or mathematically (by using Monte Carlo-type calculation codes), suitable for the detection of gamma, X and neutron radiation. Their main objective is to estimate the activity of the radioactive material and possibly to identify the occurring radioisotopes. The evaluation of the radionuclide activities within the material of interest can be carried out more accurately by means of the latter. Transportable instrumentation can be gamma spectrometry systems calibrated with the aid of mathematical methods and specialized software suites. The most widely used of these systems is ISOCS (In Situ Object Counting System). It operates with a detector consisting of a Germanium semiconductor crystal, couple to a matrix of pre-calculated geometrical configurations and sources distribution within predefined matrices obtained by a Monte Carlo code. The efficiency calibration is determined using the MCNP (Monte Carlo N-Particle Transport Code [5]) calculation code during the execution of the actual measurement of each sample. MCNP simulates the interaction of the emitted photons with the surrounding environment and, in particular, with the detector. To do this, it requires a punctual description of the measurement geometry and the type of materials present, including the detector itself. The description of the measurement geometry and of the materials present is carried out by specifying the geometric parameters (*e.g.*, distances and thicknesses), the compositions and the density for each material under consideration.

The measured spectrum analysis algorithm allows the identification of the significant photopeaks and it provides the activity and the statistical measurement uncertainty. In case of absence of peaks, the system allows the evaluation of the decision threshold, *i.e.*, the limit that allows to verify if there is an activity contribution in the sample within the counted events and of the minimum detectable activity proportional to the limit of detection (Detection Limit [6]) both expressed in activity units.

3˙1.2. Gamma scanning spectrometry and tomography. Different types of measurements fall into this category which are typically applied to the case of radioactive waste packaged in drums of different volumes. The fundamental instrumental components are always one or more gamma detectors, often a gamma radiation source (*i.e.*, transmission source), a spectrometric acquisition and analysis chain, and systems for the continuous rotation of the sample to be analysed. The flexibility of the system allows the application of four different measurement techniques, each with its own particular field of application as described below.

Open Geometry (OG), for homogeneous waste.

The collimation and waste drum-to-detector distance are such that only a slice of the drum can be seen by the detector while the drum is rotating. The measurement procedure consists of transmission measurements to calculate the gamma attenuation factor for each segment and associated emission measurements to calculate the activity (corrected for attenuation) for each segment: the sum of all the contributions gives

the total activity inside the drum. The basic assumptions are: uniform distribution of activity and homogeneous distribution of density for each segment into which the drum is divided.

Gamma Scanner (SGS), for almost homogeneous waste.

The collimation and waste drum-to-detector distance are such that only a slice of the drum can be seen by the detector while the drum is rotating. The measurement proce- dure consists of transmission measurements to calculate the gamma attenuation factor for each segment and associated emission measurements to calculate the activity (corrected for attenuation) for each segment: the sum of all the contributions gives the total activity inside the drum. The basic assumptions are: uniform distribution of activity and homogeneous distribution of density for each segment into which the drum is divided. *Angular Scanning (AS)*, for waste with uneven activity distribution. The geometric configuration is the same as the SGS. The rotating drum is divided into m angular steps $\Delta\theta$ such that $m\Delta\theta = 2\pi$. A gamma spectrum is recorded for each of these steps. Analysis of these spectra allows peak count rates to be plotted as a function of angular displacement. The shape of this diagram depends on the spatial distribution of the nuclide activity dispersed in the volume of the segment involved. This gives the experimental profile, from which the possible hot-spot can be identified (number, position and activity). The basic assumption is the uniform density distribution for each cylindrical segment of the drum.

Transmission and Emission Computerized Tomography (TCT-ECT), for any kind of waste.

Transmission tomography (TCT), by means of combined axial and rotational movements of the drum relative to the detector-transmission source direction, is required to obtain the spatial distribution of the gamma absorption coefficient within the drum. The procedure consists of n steps along the axis perpendicular to the detector-source direction, each subdivided into m rotation spectra, in order to obtain the desired spatial resolution. This procedure is repeated for each of the slices along the vertical axis of the drum. The transmission tomography reconstruction can be carried out with different mathematical methods [7].

The three-dimensional activity distribution of the nuclide is reconstructed by means of emission tomography. In this case the same measurement procedure used for TCT is adopted, without the presence of the transmission source. The reconstruction of the emission image is carried out with the same methods adopted for TCT [7].

3˙1.3. Multi-group analysis. The Multi-Group Analysis (MGA) gamma spectrometry system allows to determine the isotopic compositions of Uranium and Plutonium samples by means of gamma spectrometry carried out with Germanium detectors. The MGA algorithm [8] determines the relative isotopic abundances of Plutonium by exploiting the complex region XK (94-104 keV) of the gamma spectrum by performing a spectral deconvolution. MGA automatically calculates the energy calibration and peak shape using the characteristic energies of the Plutonium-Americium blends at 59, 129 and 208 keV and takes into account several physical processes such as the detection efficiency as a function of energy and the photon self-absorption.

The measurement geometry and the intrinsic efficiency curve obtained by the MGA return also information on the presumed distribution of activity within the sample.

3˙1.4. Passive neutron techniques. Passive neutron techniques aim at the quantitative determination of the fertile material (^{238}Pu, ^{240}Pu, ^{242}Pu) occurring in the waste based from the detected neutrons. The main components of this system are:

- neutron detectors, generally ^{3}He;
- neutron moderator, typically high density polyethylene;
- Cadmium liner for removing thermalized neutrons from the matrix.

The detected neutrons can be due to spontaneous fission, (α,n) reactions on light materials caused by alpha particles produced by the decay of heavy nuclei, fission induced on the material under examination by neutrons deriving from spontaneous fission or from (α,n) reactions.

The rate of neutrons emitted due to spontaneous fission is proportional to the mass of fertile material and measured using temporal correlation techniques (Neutron Coincidence Counting [9], NCC, or Neutron Multiplicity Counting [10], NMC): these techniques are able to discriminate the origin of neutrons and therefore attribute the correct weight to the characteristic spontaneous fission component in fertile nuclei.

The main feature of the NCC technique is the use of a Shift Register connected to the detector head: for this reason, in the context of neutron measurement techniques, the terms Neutron Coincidence Counting and Shift Register are considered synonyms. This technique uses two registers whose content indicates the number of pulses counted in the relative inspection interval: one, R+A, relates to the interval opened immediately after the arrival of a pulse, the other, A (called coincidences accidental) refers to the time interval open after a certain delay time from the activation of the first interval. The data that the Shift Register supplies are, therefore: total counts, real and accidental coincidence counts (R+A) and accidental coincidence counts (A). The difference between the contents of the R+A and A registers depends exclusively on the correlated neutrons (*i.e.*, neutrons from one single fission event) and therefore is an index of the spontaneous fission activity present in the sample.

The NMC technique represents an extension of the NCC type analysis as it allows to consider, in addition to the correlated doublets, also the correlated neutron triplets. Each detected neutron signal opens up a set of time slots of ever-increasing duration; at the end of the measurement a multiplicity distribution table is constructed which gives the frequency of having $0, 1, 2, \ldots, n$ signals in each interval. The calculation of the first and second moments of this multiplicity distribution respectively provides the counts of the correlated doublets and triplets, from which it is possible to derive the spontaneous fission rate (or the amount of fissile present in the sample).

3˙1.5. Active neutron and gamma interrogation techniques. Active neutron techniques aim at the quantitative determination of the fissile material (^{241}Pu ^{235}U) occurring in the waste based on the detected neutrons. The sample to be measured is periodically interrogated with a pulsed beam of neutrons which, when suitably thermalized, generate induced fissions on the fissile material. The detection system is activated at the end of each interrogation pulse, to reduce the contribution of source neutrons to the measured signal. The discrimination of the source neutrons from those of induced fission is based on the different decay times of the interrogation and induced fission neutrons (Differential Die-Away time Analysis, DDAA).

The basic intrumental components are the same as those of the passive techniques with the addition of a neutron source which can be a neutron generator, or an electron

accelerator coupled to suitable targets to convert electrons to photons, and then photons to neutrons by photo-induced reaction.

A high-energy electron accelerator could be also used as interrogation source as is, by inducing fission events on the sample by means of photo-fission reactions directly. The waste is irradiated with high energy X-rays electromagnetic radiation in order to produce photofission reactions. In this case it is possible to determine the total mass of the actinides occurring in the sample (fission is induced on both fissile and fertile nuclei), by gamma and neutron detection techniques. Several photofission methods can be exploited, and they are based on detection of fission by-product gamma emissions or on detection of the emitted prompt-fission-neutrons and delayed neutrons. Neutron-based methods can vary, according to the maximum energy of the electron accelerator:

- low energy irradiation (< 7 MeV) followed by counting of photofission prompt neutrons;
- high energy irradiation (> 15 MeV) followed by counting of the delayed neutrons only.

The basic components of a gamma interrogation system are the same as those for active neutron interrogation except that the source must be an accelerating electron machine coupled to a suitable target (*e.g.*, Tungsten) in order to obtain Bremssttrahlung X-radiation with energy high enough to be induce fission events within the sample. Moreover if neutron-detection-based methods are used with a gamma-detection-based method a High Purity Germanium detector is usually adopted.

3˙2. *Destructive techniques*. – A comprehensive summary of these techniques is provided by [12]. The general outline of a destructive analysis includes the following steps:

- sampling;
- homogenization of the sample;
- solubilization of the sample (*e.g.*, acid digestion or mineralization);
- addition to the solubilized sample of a tracer of the radionuclide of interest;
- purification of the radionuclide of interest from possible interferents by chemiphysical treatments (*e.g.*, precipitation, selective extraction by means of suitable resins).

An adequate sampling of the original material has to be carried out to identify and analyse a portion of the original sample that is a representative quantity of the original waste.

If the waste is already in liquid form, the preparation procedure has still to be evaluated accordingly to the origin of the waste and its content in organic, inorganic, dispersed or insoluble solid materials. The solid and dispersed substances can be removed following filtration to undergo a specific physical-chemical process (*e.g.*, acid mineralization); any organic substances present are generally separated by extraction. The remaining liquid, mainly consisting in inorganic constituents, can be used for instrumental analysis after acidification of the sample, if required by the selected analytical technique.

3˙2.1. Liquid scintillation counting. The identification and quantification of α-emitting radionuclides plays a fundamental role in the characterization of nuclear materials and in radiation protection in general. The alpha spectrometry procedure is divided into two main parts: the first is sample preparation and the second is the spectrometry itself. Sample preparation is, in general, a long and complex procedure which constitutes a fundamental phase for obtaining correct, reliable and reproducible results. From an operational point of view, the final product of the preparation procedure is a metal plate which the analyte has been electrodeposited on by means of an electrolytic cell to obtain a layer as thin as possible (ideally a monoatomic layer). The electrodeposited metal plate and the detector are placed into a chamber in which suitable dynamic vacuum conditions have been imple- mented (*i.e.*, <100 mTorr). The reduced range that characterizes the interaction of alpha particles with matter leads, in fact, to a significant attenuation both within the sample itself and within each material interposed between the sample and the detector. From a spectrometric point of view this translates into a deformation of the peaks which assume an asymmetrical shape characterized by a low energy tail, which limits the achievable instrumental performances. The main steps for sample pretreatment in alpha spectroscopy are as follows:

- homogenization and sample preparation for subsequent chemical processes;
- chemical separation: used to isolate the elements of interest;
- production of the thin source, typically by electrodeposition.

The detection of the α particles emitted by the thin source is carried out by means of surface barrier semiconductor radiation detectors or Passivated Implanted Planar Silicon (PIPS) detectors. Each radioactive element that emits alpha particles has a unique energy pectrum through which a qualitative and quantitative determination of the radioisotopes present in the sample can be carried out.

3˙2.2. Gamma spectrometry. Gamma spectrometry is one of the main methods for quantifying the concentration of natural and artificial radioactivity occurring in different types of samples. The detectors used are able to discriminate the gamma emissions of the various radionuclides of interest at different energies. In the case of gamma spectrometry, the energies and the probabilities of emission of the photons asso- ciated with the decays of each radionuclide are recorded using the measurement system in special libraries; the use of these libraries therefore allows the individual radioisotopes to be unambiguously identified, even in the case of the simultaneous presence in the sample of a relatively high number of gamma-emitting radionuclides.

Typically, for this application, a hyperpure Germanium semiconductor detector is adopted, similar to that used for non-destructive analysis techniques, connected to a digital electronic chain for the acquisition and analysis of the gamma spectra.

The sample can be analyzed as it is or suitably treated to improve the measurement sensitivity. The treatment procedures depend on the type of sample and its activity. In general, the sample is subjected to homogenization to make it liquid or semi-liquid and possibly to subsequent processes of evaporation, drying and calcination in an oven to increase the concentration of radioisotopes in the sample which will be subjected to measurement.

The measurement geometries to be adopted are linked to the detection limits to be ob- tained, to the quantity of sample available, to the yield of any pre-concentration process. The most common containers are Marinelli beakers of various sizes, generally 0.5 liters or 1 litre, or glass vials of various capacities.

3˙2.3. Inductively coupled plasma mass spectrometry. The Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) technique is a type of mass spectrometry that is highly sensitive and capable of determining different metallic and non-metallic elements, and sometimes specific isotopes (*e.g.*, ^{238}U in biologic samples). It exploits the use of a plasma torch to produce the ionization and a mass spectrometer for the separation and detection of the ions produced. The ions are separated according to their mass-to-charge ratio by passing through a magnetic quadrupole and a signal proportional to their concentration is produced. The concentration can be determined by calibration with standards or by isotopic dilution. Other mass analyzers used in ICP-MS include magnetic-electrostatic dual focusing systems, both with single and multiple collectors, and time-of-flight sys- tems with axial or orthogonal accelerators.

The ICP-MS technique can be used both for elemental analysis, with a range of determinable chemical elements between the atomic mass of Lithium up to that of Uranium (in theory atomic masses between 7 and 250) and for isotopic analysis. Unlike atomic absorption, which can identify only one element at a time, ICP-MS instead allows the simultaneous determination of the elements with the consequent advantage of speeding up this type of analysis.

3˙3. *Limits of detection for characterization techniques.* – Non-destructive gamma spectrometry is characterized by detection limits ranging from a few tens of Bq to a few kBq, in relation to the containment matrix and the type of waste in question.

These limits differ for neutron techniques, where the detection limits are much higher than for gamma techniques: from 2 to 20 mg of ^{240} Pu_{eff} (^{240} Pu_{eff} = ^{240} Pu + 1.68 ^{242}Pu + 2.52 ^{238}Pu) for passive neutron techniques, and from 0.5 mg to 5 mg of $^{239}Pu_{eff}$ 460 (^{239} Pu_{eff} = $CU_{235\ 235}U$ + $CU_{238\ 238}U$ + $CPu_{238\ 238}Pu$ + $CPu_{239\ 239}Pu$ + $CPu_{240\ 240}Pu$ + $CPu_{241\ 241}Pu$+$CPu_{242\ 242}Pu$, where C are constants that depend on the waste matrix and the interrogation source adopted) for the techniques active neutrons, in relation to the containment matrix and the type of waste in question.

Destructive characterization techniques are much more sensitive than non-destructive techniques. Below are the orders of magnitude of the sensitivity of some characterization techniques typically achieved in normal laboratory activity and therefore corresponding to sustainable measurement times.

A sample specially prepared for gamma spectrometry has detection limits of 1 to 10 Bq. Typically considering 10 g of material present in the sample, this technique is capable of revealing specific activities <1 Bq/g.

The LSC technique has a detection limit of about 0.04 Bq per 4 ml of sample, corresponding to about 0.01 Bq/ml.

Alpha spectrometry is the most sensitive technique of those described up to now, having detection limits, for a spectrometer under normal operating conditions, in the order of 0.004 Bq. This corresponds to different specific activity limits depending on the activity of the source material. In fact, for materials with high activity (>10 kBq) it is necessary to subject small quantities to measurement (<0.1 g) and this translates into detection limits of about 0.04 Bq/g in terms of specific activity, while for environmental matrices or materials with low activity it goes down to 0.004 Bq/g.

Another extremely sensitive characterization technique is mass spectrometry, capable of revealing concentrations lower than 1 in 10^{12} (ppb, parts per billion) and therefore of the order of 1 ng/l of specific elements or isotopes.

4. – Management of radioactive waste

4˙1. *Packaging*. – Homogeneous radioactive waste must be packaged in polyethylene bags placed in approved containers (typically 60-litre or 220-litre steel drums). Solids and liquids must be completely separated and the single package must respect the radiological limits imposed by the receiving treatment plant or deposit.

The container that will receive the liquid waste must be lined with suitable absorbent material in order to avoid spills and contained in a polyethylene bag. Solid waste containing explosive, pyrophoric substances and putrescible organic substances cannot be delivered to any plant or deposit.

For the transfer of waste that presents possible health risks of an infectious nature, the production facilities must provide a certificate of sterilization with the description of the methodologies used. The sterilization methods must satisfy the provisions of the Law in force.

4˙2. *Delivery*. – In compliance with the regulations on the transport of radioactive material, the containers packaged in this way are delivered to the plants authorized for treatment, conditioning and temporary storage. Each container will be accompanied by adequate documentation compiled by the Radioprotection Expert of the waste producer or the Integrated Service Operator who carried out the waste collection which shows both the physical-chemical characteristics of the waste and the radiological content in terms of quality of the radionuclides present and in terms of activity.

4˙3. *Treatment*. – The radioactive waste treatment operations are carried out in authorized plants. These include the operations of opening the delivered packages, any sorting, transferring the waste into other containers and the actual treatment operations such as the compaction of the solid waste.

The treatment phase is common to all types of waste, regardless of their origin or radiological content.

4˙4. *Conditioning*. – The conditioning operations must be carried out in plants authorized by Law and include all the phases that lead to the inertization of the radionuclides present in the waste in order to prevent their dispersion in the environment. Conditioning is a typical phase for category II waste, as defined by Technical Guide No.26, and currently classified as Very Low Level Waste and above (table I). Such process might involve the solidification of liquid waste, including COD (Chemical Oxygen Demand) abatement processes and the incorporation in inerting matrices of both the latter and solid waste.

Conditioned waste must have a series of mechanical, physical and chemical characteristics that make it suitable for confinement in surface deposits and therefore comply with all the packaging requirements required by [3]. In the conditioning process, particularly in the case of envisaged transport in bulk, the requisites required by the regulations in force on the national and international transport of radioactive materials with which the manufactured articles will have to satisfy at the time of their production must be kept in mind, or directly, or by means of additional shielding components. The level of external radiation of the product without the aid of additional and removable shielding components, must not in any case exceed a contact dose equivalent of 10 mSv/h at the time of production.

4˙5. *Custody*. – The packages containing treated and conditioned radioactive waste must be suitably filed and labelled. The immediately available information must include: the manufacturer of the product or packaging, mass, size, density, chemical composition, type of matrix (typically cement, glassy matrix only for irradiated fuel), activity and activity concentration, radionuclides, level of radiation on the surface, transferable surface contamination, date of packaging and a unique identification code that allows the final container to be associated with the content originally delivered.

The site used for storage must be selected and built to minimize the possibility of waste leaching and the return of any contaminated water to the surface or, in general, to the biosphere. The climatic, geographical and geomorphological characteristics of the site must be such as to exclude significant erosion processes, especially by rainwater and surface water. The possibility of instability (landslides) and floods must also be excluded. In choosing the site, the uses of the territory, the presence of dangerous activities, the presence of works liable to modify, following accidents, the characteristics of the site itself must be taken into consideration.

For the purpose of complying with the objectives of health protection and environmental protection, engineering works must be provided on the disposal site and on the relative deposit, designed to prevent or delay direct contact between waste and the host environment, with the consequent possible release of radioactivity. The design of these works must tend to avoid maintenance interventions.

The characteristics of the deposits must guarantee the direct or indirect inspection of the packaged waste; their protection by meteoric agents; the protection against external events, including natural ones and human sabotages and attacks; drainage systems on the floor with the possibility of collecting and sampling the drained liquids; fire detection and prevention systems commensurate with the existing fire load; non-accessibility of the public during the surveillance period.

An environmental monitoring network and a radiometric surveillance service must be provided on the site. Such a regime must be maintained even after the site capacity to receive radioactive waste for disposal has been exhausted, and the facility is closed, till the end of the surveillance period.

5. – Conclusion

This paper summarizes the Italian experience in the classification, characterization and management of radioactive waste. Starting from the international references, the Italian regulatory framework has been described highlighting the most relevant Decrees and Technical Guides for the definition of criteria for classification of radioactive waste, and showing how these criteria are converted in good practices in realistic situations.

A well-performed characterization is crucial for a correct and safe management of radioactive waste. Beside a chemical-physical characterization underling the presence of hazardous and toxic materials, the different methods for performing the radiological characterization of these kind of waste were described distinguishing between destructive and non-destructive techniques and underling the field of application and limits of detection for each technique. Based on the characterization results, the different steps of management of radioactive waste were illustrated, starting from packaging until the custody in the temporary storages waiting for the Italian Near Surface Disposal Facility that will allow the disposal from the decommissioning of Italian nuclear fuel cycle facilities and radioactive waste originating from medical, industrial and research activities.

REFERENCES

[1] International Atomic Energy Agency, *Classification of Radioactive Waste*, in *General Safety Guides* (IAEA, Wien) 2009, IAEA Safety Standards Series No. GSG-1, STI/PUB/141.
[2] ENEA-DISP, *Guida Tecnica n. 26* (ISPRA, Roma, Italy) 1985.
[3] ISIN, *Guida Tecnica n. 33, Criteri di sicurezza nucleare e radioprotezione per la gestione dei rifiuti radioattivi* (ISIN, Roma, Italy) 2023.
[4] International Atomic Energy Agency, *Categorization of Radioactive Sources*, in *General Safety Guides* (IAEA, Wien) 2005, IAEA Safety Standards Series No. RS-G-1.9, STI/PUB/1227.
[5] Sood A., *The Monte Carlo Method and MCNP – A Brief Review of Our 40 Year History* (LANL, Los Alamos, USA) 2017, LA-UR-17-25633.
[6] Currie L. A., *Anal. Chem.*, **40** (1968) 586.
[7] Martin E. R., Jones D. F. and Parker J. L., *Gamma-Ray Measurements with the Segmented Gamma Scan* (LANL, Los Alamos, USA) 1977, LA-7059-M.
[8] Gunnink R., *MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances*, Vol. **1** (LLNL, Livermore, USA) 1990, UCRL-LR-103220.
[9] Hooton B. W., *A review of plutonium measurement by passive neutron coincidence counting*, in *Safeguards and Nuclear Material Management* (CEC) 1991, EUR-13686.
[10] Langner D. G., Stewart J. E. and Pickrell M. M., *Application Guide to Neutron Multiplicity Counting* (Los Alamos National Laboratory, USA) 1998, LA-13422-M.
[11] Marleau P., Nowack A., Clarke S., Monterial M., Paff M. and Pozzi S., *Gamma/Neutron Time-Correlation for Special Nuclear Material Detection Active Stimulation of Highly Enriched Uranium* (Sandia National Lab, USA) 2013, SAND2013-7442.
[12] L'Annunziata M. F. (Editor), *Handbook of Radioactivity Analysis* (Academic Press) 1998.