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ASPECTS OF RADIOACTIVE WASTE MANAGEMENT

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ABSTRACT: The origin and types of radioactive waste, the objective and the fundamental principles of radioactive waste management and the classification of radioactive waste are presented. Problems of the radioactive waste management are analyzed.

Key Word: radioactive waste

1. Introduction

In the fifties and sixties there was a strong development of nuclear energy and nuclear techniques. Three main risks are associated with the nuclear field: major accidents with transboundary effects, handling of nuclear waste and proliferation. Minimization of these risks involves high amounts of money. In the last two decades of the 20th century the interest in nuclear field gradually declines mainly because of financial aspects: investments in other fields became more attractive.

The global warming put a new challenge to political decision-makers: finding solutions to rapidly decrease emission of greenhouse gases. The Kyoto Protocol [1], ratified by Romania, put clear limits in this respect. France, which produce its electricity mainly by nuclear power plants, generates half as much carbon dioxide per unit of energy consumed as Denmark, who produce electricity mainly in coal or lignite plants [2]. The development of new clean energy sources is not sufficient to reduce significantly global gas emission. The comeback of a renewed nuclear field is therefore obvious.

2. Origin and Types of Radioactive Waste (Radwaste)

Radwaste is produced in a great variety of activities like nuclear fuel cycle, production and use of radionuclides, decommissioning of nuclear facilities and also in non-nuclear activities [3].

2.1 Nuclear Fuel Cycle

The major steps generating radwaste are mining and milling, fuel supply and power reactor operations.

In the case of mining and milling, the waste results from the production of uranium. It contains low concentrations of uranium and is contaminated principally by its daughter products like thorium, radium and radon.

In the case of fuel supply, the waste results from purification, conversion and enrichment of uranium and the fabrication of fuel elements. It includes contaminated trapping materials from off-gas systems, lightly contaminated trash and residues from recycle or recovery operations. This radioactive waste generally contains uranium and, in the case of mixed oxide fuel, also plutonium.

In the case of power reactor operations, the waste results from spent fuel, treatment of cooling water and storage ponds, equipment decontamination and routine facility maintenance. The spent fuel contains uranium, fission products and actinides. It generates significant heat after being removed from the reactor. Usually, spent fuel, after a certain period of cooling in storage ponds, is reprocessed. Reprocessing operations generate mainly solid and liquid radwaste. Solid radwaste contains activation products, as well as some undissolved fission products, uranium and plutonium. Liquid radwaste contains in principal nitric acid solutions with high activity fission products and actinides in high concentrations.

2.2 Production and Use of Radionuclides

Research activities, radioisotopes production and applications produce radwaste, but in a smaller amount as the fuel cycle.

In the case of research activities deployed in facilities like research reactors, accelerators and laboratory activities, the type and volume of waste depend on the research conducted.

The radioisotopes production generates usually a small amount of waste but specific activities might be significant.

Applications of radioisotopes generate a small volume of radwaste.

2.3 Decommissioning of nuclear facilities

At the end of the useful life of a nuclear facility, adequate actions have to be taken to retire it from service, leading finally to the unrestricted use of the site. After decontamination, the results are solid and liquid radwaste, which contain the radionuclide spectrum, which has been used or generated in the respective nuclear facility.

2.4 Non Nuclear Activities

Industrial activities where raw materials (phosphate ore, oil, gas, etc) are involved generate small quantities of natural radionuclides through concentrations procedures during processing. This radwaste is present in different gaseous, liquid or solid streams.

3. Objective and Fundamental Principles of Radwaste Management

The goal of the radwaste management is to store the nuclear waste until it is no longer harmful. The fundamental principles of radwaste management are the following [4]:

Principle 1: Protection of human health

Radwaste shall be managed in such a way as to secure an acceptable level of protection for human health.

Principle 2: Protection of the environment

Radwaste shall be managed in such a way as to provide an acceptable level of protection of the environment.

Principle 3: Protection beyond national borders

Radwaste shall be managed in such a way as to assure that possible effects on human health and the environment beyond national borders will be taken into account.

Principle 4: Protection of future generations

Radwaste shall be managed in such a way that predicted impacts on the health of future generations will not be greater than relevant levels of impact that are acceptable today.

Principle 5: Burdens of future generations

Radwaste shall be managed in such a way that will not impose undue burdens on future generations.

Principle 6: National legal framework

Radwaste shall be managed within an appropriate national legal framework including clear allocation of responsibilities and provision for independent regulatory functions.

Principle 7: Control of radwaste generation

Generation of radwaste shall be kept to the minimum practicable.

Principle 8: Radwaste generation and management interdependencies

Interdependencies among all steps in radwaste generation and management shall be appropriately taken into account.

Principle 9: Safety of facilities

The safety of facilities for radwaste management shall be appropriately assured during their lifetime.

4. Classification of Radwaste

The following properties of radwaste are used as criteria for classification [3]: origin (see chapter 2), criticality, radiological properties (half-life, heat generation, intensity of penetrating radiation, activity and concentration of radionuclides, surface contamination, dose factors of relevant radionuclides), other physical properties (physical state, size and weight, compactibility, dispersability, volatility, solubility, miscibility), chemical properties (potential chemical hazard, corrosion resistance, organic content, combustibility, reactivity, gas generation, sorption of radionuclides), biological properties (potential biological hazards).

The International Atomic Energy Agency (IAEA) [3] recommends the following classification of radwaste: exempt waste, low and intermediate level waste and high level waste.

Exempt waste (EW) includes that waste which generates an annual dose to members of the public less than 0.01 mSv. The corresponding activity levels are dependent on the individual radionuclide and range from about 0.1 Bq/g to about 10^4 Bq/g.

Low and intermediate level waste (LILW) includes that waste whose activity levels are above those for EW and the thermal power is below about 2kW/m^3. Low waste requires no shielding during normal handling and transportation (a contact dose less than 2 mS/h).

Intermediate waste requires shielding (a contact dose higher than 2 mSv/h) but needs little or no provision for heat dissipation. LILW is classified in short lived waste and long lived waste. For the short lived waste (LILW-SL) long lived alpha emitting radionuclides are limited to 4000 Bq/g in individual waste packages and to an overall average of 400 Bq/g per waste package. In the case of long lived waste (LILW-LL), long lived radionuclide concentrations are exceeding limitations for short lived waste.

High level waste (HLW) includes that waste which generates thermal power above about 2kW/m^3 and contains long lived concentrations exceeding limitations for short lived waste. Typical activity levels are in the range 5x10^4 to 5x10^5 TBq/m^3 corresponding to a heat generation rate of about 2 to 20 kW/m^3. Significant heat can be generated by radioactive decay for several centuries.

The European Commission recommends to its member states the following classification [5]: transition radioactive waste, low and intermediate waste and high level waste.

Transition radioactive waste is a type of radwaste (mainly from medical origin), which will decay within the period of temporary storage and may then be suitable for management outside of the regulatory control system, in compliance with clearance levels.

Low and intermediate waste is a type of radwaste whose concentration of radionuclides generates sufficiently low values of thermal power during its disposal. These acceptable thermal power values are site-specific following safety assessments. Low and intermediate waste is classified in short-lived waste and long-lived waste. In the case of short-lived waste the nuclides half-life is less than or equal to those of ^{137}Cs and ^{90}Sr (around 30 years) with restricted alpha long-lived radionuclide concentration (limitation of long-lived alpha emitting radionuclides to 4000 Bq/g in individual waste packages and to an overall average of 400 Bq/g in the total waste volume). For long-lived waste the long-lived radionuclides and alpha emitters concentration exceeds the limits for short-lived waste.

High level waste is a type of radwaste with such a concentration of radionuclides that generation of thermal power shall be considered during its storage and disposal.

In Romania are still valid the obsolete Instructions for radioactive waste issued by the former State Committee for Nuclear Energy in 1980 [6]. According to these instructions, radwaste is placed in three categories: high, intermediate and low radioactive waste, based on the specific activity and surface dose rate. The current classification should be reviewed in the near future, based on the IAEA system and EC recommendations [5].

5. Radwaste Management

The basic steps in radwaste management are: pretreatment, treatment, conditioning and disposal [4].

Pretreatment of radwaste is the initial step in waste management that occurs after waste generation. It consists of collection, chemical adjustment and decontamination and may include a period of interim storage. This initial step is extremely important because it provides in many cases the best opportunity to separate waste streams, for recycling within the process or for disposal as ordinary non-radioactive waste when the quantities of radioactive materials contain are exempt from regulatory controls. It also provides the opportunity to separate radwaste, for near surface or geological disposal.

Treatment of radwaste includes those operations intended to improve safety or economy by changing the characteristics of the radwaste. The basic treatment concepts are volume reduction (incineration of combustible waste or compaction of dry solid waste), radionuclide removal (evaporation, filtration or ion exchange of liquid waste streams) and change of composition (precipitation or flocculation of chemical species). Often, several of these processes are used in combination to provide effective decontamination of a liquid waste stream. This may lead to several types of secondary radwaste to be managed (contaminated filters, spent resins, sludges).

Conditioning of radwaste involves those operations that transform radwaste into a form suitable for handling, transportation, storage and disposal. The operations include immobilization of radwaste, placing the waste into containers and providing additional packaging. Common immobilization methods include solidification in cement or bitumen of low and intermediate level liquid radwaste and vitrification of high level radwaste in a glass matrix. Immobilized waste is packaged in containers, depending on the nature and concentrations of the radionuclides. In many instances, treatment and conditioning take place in close conjunction with one another.

Disposal is the final step in the radwaste management system. It consists mainly of the emplacement of radwaste in a disposal facility with reasonable assurance for safety, without the intention of retrieval and without reliance on long term surveillance and maintenance (recently, the possibility of retrieval of the radwaste by future generations start to be discussed). The safety is mainly achieved by the isolation of suitably conditioned radwaste in a disposal facility. Isolation is attained by placing barriers around the radwaste in order to restrict the release of radionuclides into the environment. The barriers can be either natural or engineered and an isolation system can consists of one or more barriers. A system of multiple barriers gives greater assurance of isolation and helps ensure that any release of radionuclides to the environment will occur at an acceptable low rate. Barriers can either provide absolute containment for a period of time, such as the metal wall of a container, or may retard the release of radioactive materials to the environment, such as a backfill or host rock with high sorption capability. During the period when the radwaste is contained by the system of barriers, the radionuclides in the waste will decay. The barrier system is designed according to the disposal option chosen and the radwaste forms involved. Although it is planned to dispose of most types of radwaste by concentration and containment, disposal may also comprise the discharge of effluents (for example, liquid and gaseous waste) into the environment within authorizes limits, with subsequent dispersion. For all practical purposes this is an irreversible action and is considered suitable only for limited amounts of specific radwaste.

5.1 High Level Waste Management

After being discharged from the reactor, spent fuel is cooled on site for several years in deep-water pools. Unfortunately, these pools are rapidly (up to 10 years) reaching capacity.

On site interim dry storage is a solution for another about 50 years. The spent fuel is put in special casks, which could be used also for transport. The casks weight range between 25-100 metric tons [7]. These casks must withstand serious accidents, no loss of shielding and no loss of containment upon a 9 m free-fall drop onto an unyielding surface, a puncture test of a least a 1 m free-fall drop onto a 15 cm diameter steel pin, exposure to a thermal environment of 80^0C and immersion in water for 8 h [7]. The casks are invariably made from steel composite with lead and uranium thick enough to meet structural and gamma shielding requirements. Additional elastomer or resin material may also be included for neutron shielding [7]. Attention should be paid to the transfer of heat.

Several countries don’t use interim storage. They reprocess the spent fuel. There are several separation stages. The resulting waste is highly radioactive. But more than 95% of the radioactive content of the waste appears in the first separation stage. Gaseous effluent comprises fission product gases, Kr and Xe and some volatile species like iodine and ruthenium. The liquid discharges are radiologically more important than the gaseous discharges. The main management problem is solidification of the liquid waste. Vitrification (borosilicate glass) or ceramic materials are used. These materials should have a reasonably low melting point to ease production problems, high thermal conductivity, good mechanical strength and good resistance to radiation damage and to leaching with water [8]. A slow leach rate is the most important characteristic because of the need of safety against groundwater in a repository. α-decay generates the most damage and produce release of helium. But the leach rate change only by a factor of two after an α irradiation time equivalent to one million years [8]. The leach rates of ceramic materials are from 10 to 100 times lower than in glasses. Management of HLW implies also reduction in volume. France (in La Hague) and UK (in Sellafield) reprocess spent fuel from their nuclear power plants (NPP) and from Germany, Belgium, Netherlands, Sweden, Switzerland and Japan. Russia reprocesses spent fuel from its NPP and from those countries which use soviet design NPP. USA use reprocessing only for military purposes.

Some other countries like USA, Canada, Spain use direct disposal of spent fuel.

The geological repository is the solution for spent fuel and reprocessed HLW disposal. The safe disposal of spent fuel means safe storage until the radioactive isotopes decay to non-radioactive elements or to no hazardous levels of radioactivity. The typical storage time is tens of thousands to millions of years, much greater than the design lifetimes of the casks. Therefore the solution of a deep (around 500 m) geological repository is obvious. The typical candidates are salt, clay, crystalline granites and tuff (fused volcanic deposit). Each option has advantages and disadvantages. Isolating the waste from ground water, long-term stability of the geologic formation, difficulties in mining, possibility of waste retrieval and last but not least the public acceptance are several criteria which should be investigated before the final choice is made.

Several countries, which major nuclear programs, are advanced in choosing sites and designing geological repositories. United States of America is the first country, which has already built a repository for spent fuel and high-level waste in a tuff formation at the Yucca

Mountain Site in Nevada. The average depth is 300 m. Recently in USA has become operational the Waste Isolation Pilot Plant, near Carlsbad, New Mexico, designed for the permanent isolation of the defense-generated transuranic wastes. This repository, 650 m deep, is in a salt formation which is about 900 m thick. Sweden is considering a 500 m deep underground repository into crystalline rock. In Romania geological studies for a deep repository has already been started. Meantime an on-site interim storage for spent fuel will become operational till 2003. The storage capacity will be 300,000 fuel bundles [9].

5.2 Intermediate and Low Level Waste Management

Sorting is the first step in the pretreatment of LILW. Known contaminated material can be sorted into combustible material, waste suitable for decontamination, compactable waste and noncompactable waste. In case of decontamination, the surface contamination of the solid waste is either removed or reduced. The goal is to reduce the contamination to such a level where the reuse of the waste is feasible or sending the waste to a non-radioactive disposal is possible. Decontamination includes mechanical (steam, water, or sand blasting), chemical (acids, alkalis) and ultrasonic (vibrating liquid bath) methods [7].

The most common way of treatment of LILW is volume reduction. Liquid wastes can be concentrated and their volumes reduced by evaporation, incineration (especially combustible organic materials), ion exchange, filtration, reverse osmosis and precipitation. In case of incineration the reduction ratio could be 100 to 1. Volume reduction of solid wastes can be done either by incineration or by mechanical volume-reduction techniques (baling, shredding and compaction-supercompaction). A usual reduction ratio is 10:1[7].

Solidification is generally used for conditioning of LILW. The waste to be treated can be liquid, slurry, sludge, wet solid, or dry solid. In most cases, the material to be stabilized is simply mixed with an appropriate solidification agent and allowed to solidify in the final disposal container. Some of the most widely used solidification agents are cements, asphalt, polymers and molten glass depending of the form of the waste and of the chemical and thermal stability of the waste [7].

The standard 200 liters drums with LILW are sent to the final disposal. There are above-ground, near-surface underground, intermediate depth and deep underground repositories. The above- ground repository consists of reinforced concrete vaults or buildings constructed directly on the surface. The Drigg repository in UK is such an example. The near-surface repository consists of trenches or bunkers about 10 m deep. After a disposal unit is filled, a cap composed of single or multiple layers of soil or impermeable synthetic membranes is used to seal the unit from precipitation and ground-water runoff. The Centre de L'Aube in France is such an example. The intermediate depth repository is placed either in a mine or in augered holes in the earth's surface. In Romania the Baita-Bihor site in a former uranium mine is the repository for LILW. In USA there are several sites for LILW involving the construction of concrete, or metal lined holes, up to 3 m in diameter and up to 20 m or more in depth, in suitable soil [7]. The deep underground repositories are perfect for disposal of HLW, but some European countries (like UK, Sweden, Germany) consider using such repositories for the disposal of LILW. The advantage of such a repository is the excellent barriers against the migration of radionuclides into the environment and against human intrusion into the disposal unit. The disadvantage is the huge cost of such a repository.

Sea/ocean disposal of radwaste above a certain radioactivity level was proposed to be prohibited in 1972. The so-called "London dumping convention" (Convention on preventing see pollution by the discharge of waste and other materials, open for signature in London on December 29th 1972) has not entered into force up to now. Romania didn't sign it yet.

6. Safety Aspects and Legal Framework of Radwaste Management

All the radwaste management stages are under the rigorous "on-line" supervision of the national regulatory body. In the case of Romania, no supplementary exposure of the population due to radwaste management operations has been detected [10,11,12].

In the case of repositories, the "on-line" supervision is no longer sufficiently. A reasonably quantitative estimate of the maximum radiological burden to the public is mandatory. Usually a 10,000 years period is considered. The reason is that this period represents a time over which basic geology and climate is expected to be reasonably stable. Recently, calculations have been extended to 100,000 years and even to 1,000,000 years, taking into account models of climate change. The proposed population dose limit varies between 0.05 mSv/y in Canada and 0.25 mSv/y in USA. In many countries it is 0.1 mSv/y [8] and this value adds to the natural exposure. Despite the existence of fluctuations in the natural exposure, it is generally agreed that the reference natural exposure is 2.4 mSv/y [10,11,12].

Another safety aspect of the radwaste management is related to the radiological risks of transporting radioactive materials. The risks can be determined using probabilistic safety assessment [13]. Two types of risks were estimated: those resulting from normal (accident-free) transport and those resulting from transportation accidents involving radioactive shipments.

A very interesting approach is either to destroy the radwaste completely by converting it to non-radioactive waste, or to convert it to short-lived nuclei that, although still radioactive, could decay relatively quickly (30 years or less). Studies to use modern particle accelerators for transmutation are on the run [14, 15, 16]. The idea is to use the neutron bombardment in the reactor to convert the radwaste to benign or short-lived nuclei. The key idea is to introduce more neutrons from another source: a proton beam accelerator. The accelerator could be a 0.8-1.0 GeV linac [14,15], or a 1 GeV superconducting cyclotron [16].

In order to enhance the safety of radwaste management, the Vienna Convention [17], ratified by Romania, was adopted. Romania, as Contracting Party, must impose an authority to implement the Convention. This authority, according to Law no.111/1996, republished [18], is the National Commission for Nuclear Activities Control, which works as a regulatory body. Each Contracting Party shall establish and maintain a legislative and regulatory framework to govern the safety of spent fuel and radiation waste management. According to [18] a law on the safe management of radwaste is under preparation.

Compared to other human activities, the nuclear field and especially radwaste disposal is an extremely sensitive issue for the public. A direct consequences are the reserved political decisions compared to the scientific and technical expertise.

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