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NUCLEAR WASTE MANAGEMENT - AN INTERNATIONAL APPROACH

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Rezumat. *În acest studiu, tipurile de soluții pentru fiecare tip de deșeuri nucleare au fost analizate, precum și procedurile de implementare a acestora. Superioritatea anumitor matrice de imobilizare a deșeurilor (forma de imobilizare a deșeurilor) este evidențiată cu precădere în această lucrare, care precizează argumentele pro și contra pentru diferitele forme de imobilizare a deșeurilor, precum și o analiză asupra combustibilului uzat și a metodelor de producție.*

Majoritatea tipurilor de deșeuri nucleare este imobilizată cu sticlă borosilicată, pentru care am făcut o analiză mai amplă și am oferit de asemenea soluții pentru deșeurile greu de incorporat în borosilicat. Pentru acest tip de deșeuri este preferată o soluție ceramică sau vitrificarea, din considerente economice. De asemenea, au fost prezentate tipuri de configurații de depozit final și o analiză comparativă între depozitatrea de suprafață și depozitarea geologică profundă a deșeurilor nucleare. În final, au fost furnizate mai multe sugestii pentru cercetări viitoare în acest domeniu.

Abstract. *In the study we presented the types of solutions for each type of nuclear wastes which have been analyzed, as well as the procedures for their implementation. The superiority of the waste immobilizing matrices (waste forms) are outlined mostly in this paper, stating the pros and cons of different types of waste forms as making an analysis of the spent fuel and the production methods.*

The waste majority of the nuclear waste is immobilized with borosilicate glass, on which we made a broader analysis, as well as providing solutions for the wastes difficult to incorporate in borosilicate. Fr this type of waste a ceramic or glass solution is preferred, because of its economical benefits. It was also presented the final deposit disposal configuration, and a comparative analysis between surface disposal and deep geological disposal of the nuclear waste. Finally, several suggestions for future research in this area have been provided.

Keywords: nuclear waste management, borosilicate glass, surface disposal, deep burial, decontamination.

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1. Introduction

The situation of radioactive waste worldwide is very delicate and must be seen from the point of view of the impact on the environment, the population and the operating personnel, their final storage practices and their optimization in order to ensure radiological safety, in accordance with the regulations and recommendations at national and international levels.

A key factor in the management of nuclear waste is its treatment and storage in accordance with 3 principles to be followed.

Environmental protection, economic efficiency and process flexibility are the factors whose compliance depends on the success or failure of the final treatment and disposal.

At present there is an international consensus that the management of radioactive waste cannot be considered as totally resolved, making technical and socio-political conditional development of the application of nuclear energy and the maximum use of its benefits.

2. Focusing on the Radioactive Waste Risks

One of the main concerns about nuclear energy is the unjustified fear of radioactive waste. Radioactive wastes have particular characteristics, they have low volumes, for example, compared to those produced by the burning of coal, and the health hazard is due to the emission of ionizing radiation.

A comparison is made of the risks associated with waste generated by the operation of a coal-fired power plant and those produced by nuclear power. Assuming that coal burning takes place at 100 MW without any pollution control measures, each year it will generate 75 deaths.

If the radioactive waste is vitrified (current technology), then it is arbitrarily thrown into the sea (an unacceptable solution), it will produce 0.6 deaths for each nuclear power plant, spread over millions of years [1].

To quantify the risk associated with deep geological storage, we will resort to the analogy between buried nuclear waste and host rock, as the groundwater movement in geological environments is well studied [2].

From measuring the speed with which rivers carry dissolved material into the oceans can be estimated that on the average $1.4x10^{-5}$ meters are eroded each year. Hydrogeologists estimate that 26% of the transported material is recovered from groundwater, the rest being due to surface water. The thickness of the bark dissolved annually in the groundwater is therefore $0{,}26x10^{-4} = 3{,}64x10^{-6}\,m$.

Knowing that the underground waters at a depth of 500-600 m have an incidence probability of $2,6x1x10^{-4}$, it results that the annual dissolved thickness at this depth is of $3.6x10-6x10^{-4} = 1x10^{-9}m.$ If $1x10^{-9}$ are dissolved each year, it results that an atom has a chance of 1 in a trillion of reaching the surface, that is, “the probability that an atom from the water surface to reach the human body by dividing the amount of water intake of the population to the total amount of water in rivers”. The value obtained is of $1/10000$. Combining the two probabilities we get the chance that an atom located in the underground rock to reach the human stomach is of $10x10^{-9}x10^{-4} = 10^{-12}$” [1].

3. Biological Risks of Radiation

With regard to the risks associated with the biological hazard of nuclear waste, we can say that fears are most often unfounded. Although ionizing radiation as the primary damaging effect of nuclear waste is hazardous, a dose that is quite intense to cause effects is required. Ultimately these effects are divided into deterministic “effects” and stochastic “effects”. The impact on the human body determined by deterministic effects is easy like, balding and erythema. This type of impact is measurable, and has a maximum on a scale, with the rising dose, is raising the effects. Stochastic impacts are the ones referring to the probability of the event and a measured dose radiation which infers that these sorts of impact can never be completely eliminated, just the event can be limited. Here is standing as examples are fatal cancer and severe hereditary diseases in offspring [3].

4. An International Approach on Nuclear Power Plants and Nuclear Power Plants Generated Waste.

The definition of radioactive waste is given by the International Atomic Energy Authority [5] as “any material that contains or is contaminated by radionuclides or radioactivity levels higher than exempted quantities established by the competent authorities and for which no use is foreseen”. This definition has a certain degree of ambiguity, given that in some countries it is considered waste, materials that other states refold. It is the case of irradiated used fuel and scraps and uneradicated residues for the reuse of fissile material.

In considered nuclear waste, disposal is according to IAEA, [5] definition of “the disposal of waste and an approved facility without the intention of retrieval”. It should be carefully thought when throwing nuclear material, because with the development of new technology fissionable material can be reused more and more so that the decision to integrate it into a certain category remains in government interpretation, with some countries requiring retrievability to be a post disposal option.

4.1. A European approach on nuclear power plants and nuclear power plants generated waste

Currently, over 150 reactors are operational in the European countries [4] which produce about 35% of the energy needed in Europe, yet the biggest challenge is the maintenance of nuclear power plants to produce electricity. Construction, maintenance and decommissioning costs greatly reduce the revenue generated by the production, distribution and sale of electricity.

The necessarily strict regulation of reactor design and operations, coupled with the relatively high end-of-life decommissioning costs, is beginning to be an additional burden. The current trend in nuclear power generation is therefore very much associated with plant lifetime extension and also the approval of specially designed spaces to decontaminate the resulting debris. As well, to be noted that France, Belgium, Bulgaria, and Lithuania have a high dependence on nuclear power (all over 50%), so they rely more than other countries on nuclear generated energy [4].

4.2 International Regulation and competent Authorities

The International Atomic Energy Agency (IAEA) *found* by the United Nations in 1957 to ensure world cooperation for the peaceful use of nuclear [6]. It develops the safety guidelines associated with all stages of the nuclear lifecycle. The member States develop their own regulation with respect to IAEA regulations. Currently there are 113 member countries.

The International Commission on Radiological Protection (ICRP). ICRP publishes universal recommendations on the effects of radiation exposure on health. Its activity begun in 1928, and adequate radiological protection data are regularly updated. ***The OECD Nuclear Energy Agency (OECD NEA)*** has the purpose to encourage the cooperation between member States to develop sustainable nuclear power in a non-polluted environment.

In collaboration with other international nuclear organizations the aim of the organization is to help the development of national regulatory policies on the safety of nuclear installations, protection ionizing radiation protection, preservation of the environment and rehabilitation, radioactive waste management, and nuclear third party liability and insurance.

Exchanges of scientific and technical information, particularly through participation in common services, consist of developing international research and development programmers [7]. It includes all European Union Member States, as well as Australia, Canada, the Czech Republic, Hungary, Iceland, Japan, the Republic of Korea, Mexico, New Zealand, Norway, Poland, Switzerland, Turkey, and the USA.

The European Commission. All Member States of the European Union have developed the legislation, regulation according to the principles, standards, and requirements related to nuclear and associated environmental matters stated upon the 1957 Treaty of the European Atomic Energy Community (Euratom), the 1957 Treaty of the European Economic Community (EEC), and the single European Act of 1987. Recommendations made by the ICRP, IAEA, and OECDNEA form the basis of specific Community Directives [6].

5. International Situation of Radioactive Sources

All units using radioactive sources can be considered as generating radioactive waste. Areas where such radioactive materials are used are vast and include: energy production, medicine, research, industry, production, and so on.

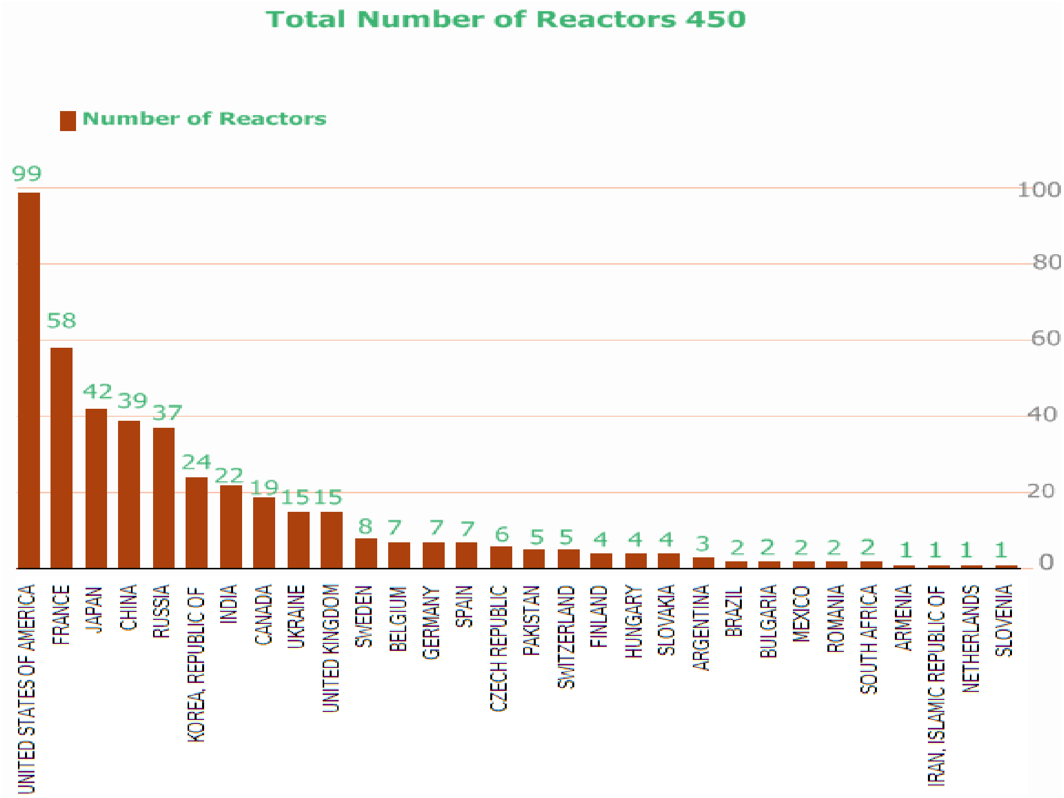

Fig.1. World Number of operational reactors.

Source: Author based on, International Atomic Energy Agency, PRIS (Power Reactor Information Systems), Regional Distribution of Nuclear Power Plants [8].

At international level IAEA counts almost all the world's states that have activities in the field to report the status of radioactive waste, keep it under control and the development of new technologies with regard to the situation of radioactive waste. In 1957, 56 States, including Romania, set up the agency, and then 106 other Sates joined the IAEA, resulting in a total of 169 Member States by December 2017 [8].

According to the data centralized by the International Atomic Energy Agency, on nuclear research reactors (IAEA, 2009, pp.10) [2], there are States with a single nuclear research reactor, as is the case with 26 States.

Economically developed States are generally the ones that use the most nuclear research reactors, reaching the most advanced States in this respect, to talk about 65 nuclear reactors in Russia, 42 nuclear reactors in the US or 16 nuclear reactors in the case of China.

Production of radioactive waste may result either from the normal activities of a nuclear power reactor (spent nuclear fuel) or from activities specific to radiological stations or nuclear research reactors, where an estimate of the quantities of radioactive material is almost impossible, the scenario of the experiments carried out being difficult to predict.

By centralizing the data on nuclear research reactors we can find out that there are a total of 773 reactors in the world. Out of these 450 are decommissioned and 166 are stopped, awaiting the start of decommissioning. 19 nuclear research reactors are temporarily stopped and work has been interrupted in 8. Also 55 reactors are under construction and 12 are planned to be built in the near future. It follows that there are still 247 nuclear research reactors in use that are currently in operation.

On the other hand, the situation of nuclear power reactors is slightly different. Nuclear power reactors in operation are much more global than research ones [9].

Due to the fact that they are used with only one purpose, well defined, the production of electricity, their situation is not so nuanced like that of the research ones. Nuclear energy reactors are usually used for a longer period of time. Worldwide, there are only two NPP-type reactors declared but not yet decommissioned.

Analyzing the results of power reactors in terms of electricity production, an increase in the quantities produced by NPP has been observed in recent years. There are States that rely heavily on nuclear power generation.

In the case of Romania, of the total electricity produced, 19.82% is produced with the help of nuclear energy. States with significant activity in the field revolve around 15-25%. For example, in the US 19.44% of total electricity is nuclear. In Russia, the percentage is 17.52%, in Spain it is 19.73% and in Germany the percentage is 15.45%. At the opposite end, there are almost all northern countries, such as Finland with 33.31%, Sweden with 42.72%, Belgium with 52.08% and Switzerland with 36.41% [9].

6. Types of Radioactive Waste

Different natural and man-made radioactive substances produce significant quantities of radioactive waste (s) with half-lives and different toxicity, which often also contain significant amounts of inactive substances.

There are various classifications made in order to classify the radioactive waste: short- and long-lived, low-level waste, intermediate-level waste and high-level nuclear waste (HLW) are the most used but not universal, definitions. The most dangerous waste of large-scale nuclear origin is HLW, resulting from the reprocessing of the spent fuel or the spent fuel itself.

The low type of waste usually resulted from reactor operations, decontamination of radioactive samples, hospital wastes, spent sources, mining operations, and forming the vast majority and having the greatest volume of radioactive waste. Currently HLW is considered as the most dangerous waste of large-scale nuclear origin is HLW, resulting from reprocessing of the spent fuel or the spent fuel itself. The current definition of HLW is given by its method of production and so HLW cannot be converted to low-level waste by dilution.

That makes HLW hard to manage and their disposal in the oceans is forbidden for instance. The Radioactive activities of this kind of waste are around 1000 Ci/l Ci=curie, the activity of 1 gm of 226Ra, 3.761010 disintegrations/second (Becquerel's, or Bq) [10]. The most quantity of HLW arises from spent fuel or reprocessing waste from power plant fuel, existing, though in other forms like military applications.

7. Treatment of Radioactive Waste

The liquid waste obtained from the treatment of spent nuclear fuel is processed in several ways: evaporation or distillation, precipitation, co-precipitation or flocculation, absorption and ion exchange.

Evaporation or distillation applies to large volumes of radioactive solutions when highly concentrated and highly radioactive solutions are obtained. The method is very expensive, but quite efficient.

Sometimes evaporation takes place up to seconds, when high-level radioactive waste is produced.

Precipitation, co-precipitation or flocculation are classical methods that generally apply to water depollution. They consist of the treatment of radioactive solutions with various chemical reagents that lead to the formation of floccous precipitates, capable of tracing, especially by coprecipitation, the radionuclides present in the solution.

8. Design Drivers for High-level Nuclear Waste Forms

When designing a high level waste form three key drivers we should take into account: environmental economic and process flexibility.

The environmental aspect is one of the most important in terms of waste forms of disposal, it must have several characteristics like resistance to aqueous leaching, resistance of the material that seals the waste, a relatively accessible position, but still a safe one, in the event of future use of the buried material. Geological repository, to minimize environmental risk.

8.1. Borosilicate glass vitrification

Borosilicate glasses have been the most popular material adopted for the immobilization of both HLW and LLW. This waste form poses both the chemical durability, mechanical and thermal stability [10].

Borosilicate glasses are flexible with waste loadings and possess the capability of incorporating most of the waste elements. The material has been studied for many years, before it becomes baseline for HLW immobilization [13].

Its chemical conformation containing seven or eight cations, they ensure allow melting temperature around as well as maximizing durability.

Usually produced by Joule melters they use a technology where glass is poured into the disposal cans, Joule melters have been considered obsolete due to finite lifetimes electrode span, corrosion and the limitation of temperature to maximum ~1150^{o}C. Among other inconveniences associated with the off gas systems, Joule melters produce large amounts of secondary radioactive waste.

There is as well the problem of waste ions with very limited solubility in borosilicate glass like Ti, Zr, Cr, Mo, sulphate, etc.

Decreasing the waste loading could solve the problem, with price ineffectiveness and increased costs. As well with a Joule melter a relative constant temperature must be maintained in order not to alter the glass viscosity and process to function properly.

Glass viscosity is as well crucial when using a cold-crucible melting (CCM) instead of a Joule melter. The ecology is a little different in CCM's using oxide charge heated by radiofrequency (rf) field. This is based on the conductivity of the charge, and when needed carbon or metal is added to induce the heating.

In the process, the carbon is oxidized to gas or the metal forms the oxide and becomes part of the melt. With the CCM technology a variety of glass forms can be attained and as well as the evasion of corrosion by using water-cooled coils that allow the containment of the melt.

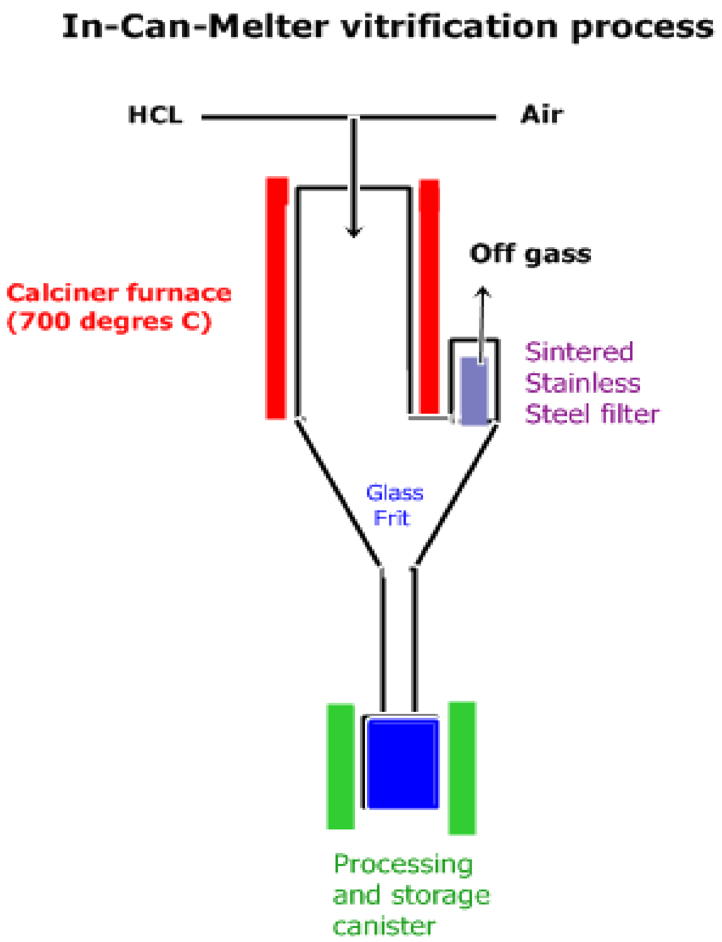

Fig.3. Example of the vitrification process is a technique called "In-Can-Melter"
Source: Author, adapted after M. I. Ojovan and W. E Lee, W. E., *An Introduction to Nuclear Waste Immobilization* [15].

The canister is as a glass manufacturing vessel, and prevents the contact between waste and groundwater.

Thermal decomposition occurs in the calciner, oxysalts (as nitrites and hydroxides) being converted to oxides which are incorporated into the waste form.

In order to form glass, the calcine frit mixture is heated to around 1000°C by the zoned furnace.

The canister will be removed and the process will continue by replacing the canister with an empty one.

To be noted that the canisters are corrosion-resistant and proven one of the best long-term disposal solution [14].

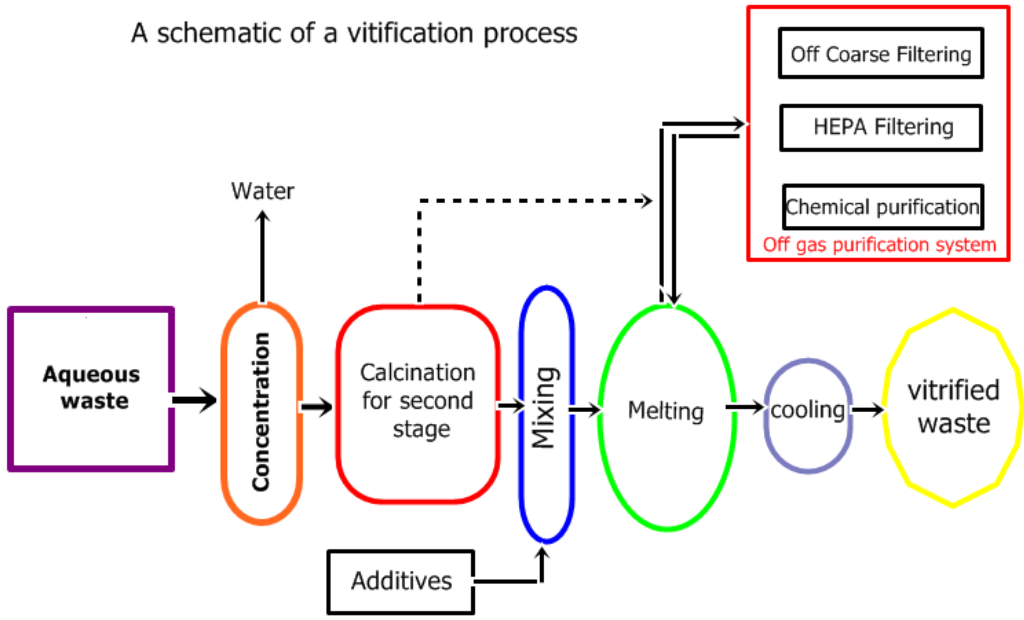

Fig.4. A schematic of vitification, using borosilicate glass.

Source: Author, adapted after (M. I. Ojovan and W. E Lee, W. E., "*An Introduction to Nuclear Waste Immobilization"* [12].

The first stage of the vitification process describes the batch formation from forming additives mixed with the liquid waste, and form a paste. After this process the batch enters a melter where water is evaporated followed by the calcination process and glass melter.

In the second stage of the process, the waste is put into the calciner, and after the calcination, additive like (glass frit) are added in order to form the glass. At the end of the process it results the glass melt, carrying the most of the radioactivity and the gas flow. The glass is poured into carbon steel canister, and then put into a cooling annealing furnace, in order to prevent the stress in the glass and avoid further cracking [12]. The gas flow is passed through a complex filtering system that removes the restant radionuclides and the restant chemical compounds [10].

8.2 Phosphate glasses

They were first developed in America, in the mid '80s and were unsuccessful due to unattractive from the environmental point of view, even though the waste form properties were satisfactory and the melting temperatures were considered low (~800^{o}C). In Russia, with the aid of the cold-crucible melter (CCM) technique sodium aluminophosphate glass were used for some HLW.

Table 2. Typical borosilicate glass compositions designed for HLW immobilization

No.	*Oxide (wt%)*	*Type of glass*		
		R7T7 (France)	*DWPF (US)*	*Pamela (UK)*
1	SiO_2	54.9	68	58.6
2	$B2O_3$	16.9	10	14.7
3	Li_2O	2.4	7	4.7
4	Na_2O	11.9	13	6.5
5	CaO	4.9		5.1
6	TiO_2			5.1
7	MgO		1	2.3
8	Al_2O_3	5.9		3
9	ZnO	3		

Source: Author based Ahn J., Apted M. Geological Repository Systems for Safe Disposal of Spent Nuclear Fuels and Radioactive Waste [10]

8.3 Ceramics: silicate, aluminate and phosphate

Single-phase ceramics have been widely used as a waste form driver Sodium zirconium phosphate (NZP) are most used the full range of FPs and actinides. Monazite, apatite and zircon have been in use for actinides, immobilization.

For Cs immobilization the common practice is to use pollucite and $CsAlSi_5O_{12}$.

Although broadly used, diluting with alumina, silicate or phosphate would increase heat, pro heat production and volatility losses, overcoming these difficulties Rockwell Science Center (RSC), which put forward hot isostatic pressing (HIP) as an innovative method [15].

The waste is immobilized in a pressure vessel surrounding an insulated resistance heated furnace. The can is filled with the calcined feed, then is evacuated and sealed.

Heating and pressure is applied after the can is placed into the HIP, immobilizing and consolidating the waste into a dense monolithic block can sealed.

The HIP has are zero gas emissions during the consolidation process, minimizing the secondary waste and the costs of the high temperature off-gas treatment.

Making it a reliable method, the ip process is used for more than 30 years.

8.4 Titanate ceramics

Multi-phase titanate ceramics based dense materials Synroc-C ceramic [16] are made by first mixing inactive precursors of Al, Ba, Ca, Ti and Zr oxides with liquid HLW. The technology further implies drying and calcining in an H_2/N_2 atmosphere at 750°C, mixing with 2 wt% of powdered Ti metal considering redox control and then isostatic pressured at 1100°C. The waste ions are dilutely incorporated in the titanate mineral making them more insoluble in water than the silicates and phosphates used in supercalcine.

8.5 Glass–ceramics

Glass–ceramics are combining the technologies and glasses and ceramics, being suitable for the waste streams contain glass formers, together with components difficult for glass immobilization. Sphener glass–ceramics was initially developed in Canada and consisted of sphene, $CaTiSiO_5$, in a durable aluminosilicate matrix [17]. Other silicate-based materials were celsian ($BaAl_2Si_2O_8$), fresnoite ($Ba_2TiSi_2O_8$), diopside ($CaMgSi_2O_6$) and calcium aluminosilicates [17].

9. Final Storage of Radioactive Waste

Final disposal is the last stage of the radioactive waste management system. This mainly consists in the placement of radioactive waste in storage facilities, ensuring reasonable security, without the intention of being moved and without ensuring long-term surveillance and maintenance. Security is mainly achieved by concentration and containment involving the containment of conditioned radioactive waste in the final repository.

Gaseous waste containing 41Ar, 85Kr, 131Xe, 3H 131I resulting from the 235U fission in the current practice is evacuated directly into the atmosphere at high heights where it is diluted with atmospheric air. In cases where these gaseous emissions have a very high radioactivity, exceeded or exceeded by radioactive aerosols, before sending them into the atmosphere, they pass over a special filter system capable of retaining them [11].

Radioactive aerosols with excess activity may not be discharged directly into the atmosphere. For their retention, the polluted air passes through electrostatic filters, porous filter materials (filter paper, cellulose, asbestos), or through filters made of glass fibres, synthetic materials, etc. Low-grade liquid wastes, after dilution and storage in special basins, are discharged into surface waters such as rivers, lakes, and oceans.

The most attractive storage options are storage in tanks buried or submerged in oceans and seas is the most widely used method in countries with developed nuclear industry.

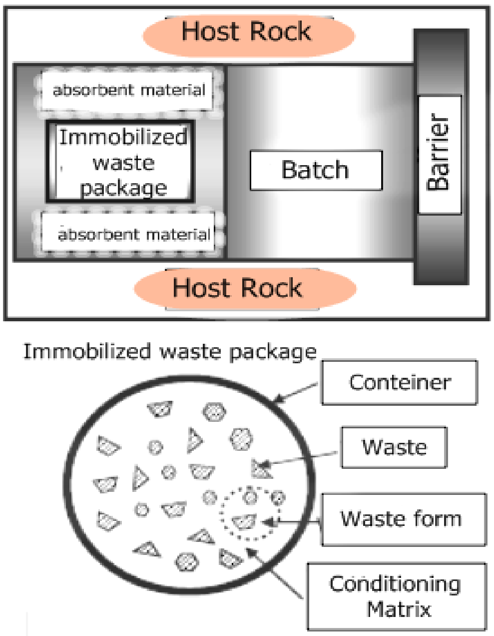

Fig. 5. Components of the storage system.

Source: Adapted after Alan Carolissen, Overview Radioactive Waste Management in RS [18]

Requirements for the location of radioactive waste dumps [19]. In establishing the optimal site for the construction and operation of a radioactive waste repository, the fundamental principles regarding the safe management and storage of radioactive waste are taken into account. The objective of designing a storage facility is to ensure that the facility can be built, the waste accepted, handled and stored without risk to the health of the population and the environment, both during and after the facility is closed.

9.1 Surface storage options

Activities such as the decommissioning of nuclear power plants and other nuclear facilities as well as non-restrictive decontamination, decommissioning or releasing operations of associated installations and sites can lead to significant amounts of low-activity waste with low trans uranium content and/or long-term activation and fission products [20].

For this type of waste, landfill storage, with engineering barriers limited to / or near the site where the wastes originate is an attractive safety and economic option. Isolation of radionuclides from waste is guaranteed by their location, properly confined, above the groundwater and by limiting or avoiding rainwater percolation with a sufficiently tight covering/closure system.

Waste with a high content of long life radionuclides is usually stored in geological formations of depth. The depth of location of these types of geological deposits varies between several hundred meters. The forms of conditioned waste accepted at a specific landfill depend on the characteristics of the waste and the site. Waste treatment and conditioning provide both physical barriers and chemical barriers in terms of radionuclide migration and their disposal into the environment [21].

The deposit may take the form of a tunnel, a room or a silo. It can be specially built for this purpose or built / placed in an existing mine. The walls can be covered, for example, with cement-based material (hollow), and the free spaces can be filled with a low permeability material, e.g. bentonite, to control underground water movement. In principle, deposits located in caverns / mining tunnels provide a higher level of containment compared to surface deposits.

10. Conclusions

Conclusion (1). This paper aims to draw attention to the potential risk posed by radioactive waste. Some clarifications have been made as to the properties of radioactive waste and its particularities. Real data on the situation of nuclear centres in the world were presented.

Conclusion (2). This study explores new ideas on radioactive waste management, such as the reduction of radioactive waste by compaction, or the use of a wet air polluting radioactive air filtration facility, in combination with a liquid radioactive waste treatment facility makes treatment of radioactive mud instantly or by accumulation in special tanks.

Conclusion (3). We have tried to present the methods of radioactive waste disposal, presenting the advantages and disadvantages of each of them, the focus was on the vitrification techniques and especially the borosilicate vitrification technique. Also, other convenient alternatives as to the immobilization of nuclear waste were presented when vitrification was not feasible. All aspects presented can be widely debated and can be the subject of further interesting studies that will certainly lead to the development of the field of radioactive waste management.

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