

ECOLOGICAL AND TECHNICAL REQUIREMENTS OF RADIOACTIVE WASTE UTILISATION

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Received: 2012.11.15
Accepted: 2012.12.17
Published: 2013.01.15

ABSTRACT

The paper presents a survey of radioactive waste disposal technologies used worldwide in terms of their influence upon natural environment. Typical sources of radioactive waste from medicine and industry were presented. In addition, various types of radioactive waste, both liquid and solid, were described. Requirements and conditions of the waste's storage were characterised. Selected liquid and solid waste processing technologies were shown. It was stipulated that contemporary methods of radioactive waste utilisation enable their successful neutralisation. The implementation of these methods ought to be mandated by ecological factors first and only then economical ones.

Key words: radioactive waste, utilisation, storage, solidification, vitrification.

INTRODUCTION

The implementation of safe technologies of radioactive waste utilisation is currently a vital task for environmental engineering. Until the beginning of 1980s, huge quantities of low-level-radiation waste were dumped into seas and oceans. At present this type of waste is stored in waste dumps or is recycled. Treatment of radioactive waste strives for the maximum reduction of the volume of the waste, ensuring resistance to external conditions and minimising their influence upon natural environment [Gołofit 2003]

Radioactive waste may be harmful to people due to the emission of radiation. Currently, radioactive elements penetrate some plants which animals feed on. When consumed, meat of such animals introduces these elements into our organisms. Laplanders, for example, as a consequence of ingesting reindeer meat, have got elevated levels of polonium in their organisms [Hryniewicz 2001]. By inhaling cigarette smoke, smokers also introduce polonium into their organisms as tobacco leaves contain this element. Inhabitants of areas rich in uranium ore are also exposed to ra-

diation as such ores, when stored in piles, are the source of radioactive isotopes of lead, bismuth and thallium.

The preparation of waste for utilization requires a different approach depending on their state (liquid, solid), activity (low or high level of radiation) and mass. The majority of high-radiation-level waste originating from the processing of spent fuel is stored in liquid state in tanks. Processing of such waste poses considerable problems. Solid waste is utilized by the reduction of their volume and solidification or vitrification [Colombo et al. 2003, Jezierski 2006]. After that, these are frequently stored in special containers.

The paper's aim is to present the useful technologies of radioactive waste utilization and to define their influence upon environment.

SOURCES OF RADIOACTIVE WASTE

The main sources of radioactive waste are the following:

- uranium ore mines and ore's processing plants,

- remains from reactor fuel production and processing of spent fuel,
- energy and research reactors,
- nuclear reactors under liquidation,
- radioactive isotopes in medicine, agriculture, industry and scientific research,
- military sites producing nuclear weapons.

A considerable amount of radioactive waste in countries with well-developed power industry originates from nuclear power plants. Nuclear reactors utilise fissile isotopes for fuel i.e. heavy isotopes whose nuclei are easy to split as a result of low-energy neutron bombardment - most frequently uranium (^{235}U , ^{233}U) and plutonium (^{239}Pu , ^{241}Pu).

In Poland, radioactive waste originates from the use of radioisotopes in medicine, industry and scientific research, during the production of open and closed sources of radioactivity and in the operation of research reactors used for the production of radioisotopes among others [Skalmowski 2009]. Considerable sources of radioactive waste in Poland are “Maria” research-production nuclear reactor and radioactive isotopes production plant Research and Development Institute in Świerk near Warsaw. The plant prepares radioactive substances for recipients from various branches of industry, medicine and science.

Filters from purification and ventilation systems, post-decontamination waste, spent elements of reactor devices and appliances constitute reactor-originating waste. A specific type of radioactive waste is spent fuel. Initial high activity of spent fuel and heat generation bring about the necessity of its periodic storage in water storage tanks before being ultimately stored or processed [Meshik 2005]. In Poland, there are no suitable sites for spent fuel storage. It is stored in the place of origin, initially in wet storage containers and then temporarily in dry storage containers. Due to the possibility of its military use, spent fuel is protected from any unauthorized access throughout the process.

In medical therapy, radioactive sources of two types of constructions are used in order to utilize the properties of emitted radiation. The first construction constitutes radioactive sources encapsulated in a hermetic container. The second is the open type construction where the radioisotope, in a suitable chemical form, is introduced into tissues or organs. Radioac-

tive sources used in medical therapy are characterized by a relatively high level of activity and, frequently, relatively long half-life. Due to constant spontaneous isotope's breakdown, the encapsulated sources lose their therapeutic properties in time and are handed over as radioactive waste [Zdrojewicz & Bielawska-Bień 2004]. Medicine most frequently utilizes the following:

- radium sources (Ra-226),
- cobalt (Co-60) and caesium (Cs-137) sources.

Spent therapeutic radioactive waste is handed over for utilisation in protective containers. In medical visual diagnostics radioactive sources are used in the form of radiopharmaceuticals introduced intravenously or orally into the organism. Currently, more than 50 types of radiopharmaceuticals are used [Hryniewicz 2001]. For diagnostic purposes the following are applied:

- iodine (I-131) and technetium (Tc-99) sources – used in visual techniques, imaging conducted by means of gamma-cameras or scintigraphy in order to localise lesions;
- iodine (I-125) sources – used in in-vitro in order to carry out tests of assay for biologically active substances (e.g. hormones) which are present in body fluids in a billionth part of a gram.

Various types of materials and instruments necessary during medical procedures and contaminated elements of devices and apparatus are also considered radioactive waste.

Radioisotopes of cobalt (Co-60) and iridium (Ir-192) have found their application in several branches of industry. These isotopes are used in gamma defectoscopy which examines material defects in particularly important metal elements of machines and devices.

Measuring devices containing encapsulated sources of radioactivity are used in automation of industrial processes. The devices include isotope scales, level gauges and hygrometers used in chemical, food, metal and other types of industry. The greatest group of industrial waste consists of smoke detectors used to signalise fires. The detectors are the source of radioactive americium (Am-241).

However, the most dangerous radioactive waste originates from spent radioactive materials from nuclear energy and military applications.

CLASSIFICATION OF RADIOACTIVE WASTE

Radioactive waste is classified according to the following categories:

- state of matter – gas, liquid and solid waste;
- activity concentration – low, medium and high;
- types of radiation – α -, β -, γ -, neutron radioactive and containing fissiles;
- half-life – short ($T_{0.5} < 30$ years), long ($T_{1/2} > 30$ years), intermediate (in 3 year's time radioactivity reaches low level);
- level of radioisotopes – low (III or IV class of radiotoxicity), high (I or II class of radiotoxicity).

Liquid waste mainly consists of liquid solutions and suspensions of radioactive substances. Nuclear power plants create liquid waste (radioactive sewage) chiefly due to leakages in the cooling system of the reactor's core. The acceptable leakage level amounts to 0,1%. Radioactive sewage is usually the solution of fission products obtained from fuel where radioactive isotopes constitute approx. 15% [Włodarski 2002a].

Encapsulated radioactive sources rank among the solid radioactive waste. The following constitute examples of solid waste: personal protective equipment contaminated with radioactive substances (rubber gloves, protective uniforms, footwear), laboratory materials and equipment (glass-

ware, elements of equipment, lignin, cotton wool, foil, radium needles), used tools and elements of technological devices (gauges, fragments of piping, pumps' elements), sorption and filtration materials used in the process of purification of radioactive solutions or air released from isotope reactors (used ion-exchange resin, post-precipitation sludge, filtration inserts, etc.). Elements of fuel systems and fuel jackets also belong to the highly active waste [Włodarski 2002b].

An exemplary classification of solid radioactive waste put forward by the International Atomic Energy Agency (IAEA) was shown in Figure 1.

Waste of the lowest radioactive level is usually stored in metal barrels, which, having been filled, undergo pressing in order to reduce their volume. In addition, melting of metal waste and burning their combustible part while retaining the radioactive ash is utilized.

Waste of slightly higher level of radioactivity is stabilized by concreting or immersing in resins providing resistance to possible wash out. After that, such waste is encapsulated in moisture-proof protective containers.

Long-lived medium-activity waste usually originates from disassembled internal structures of the reactor's core. These also encompass control rods which control the flow of nuclear reactions and other rods limiting the reactivity of fresh fuel. Such waste is flooded with cement and

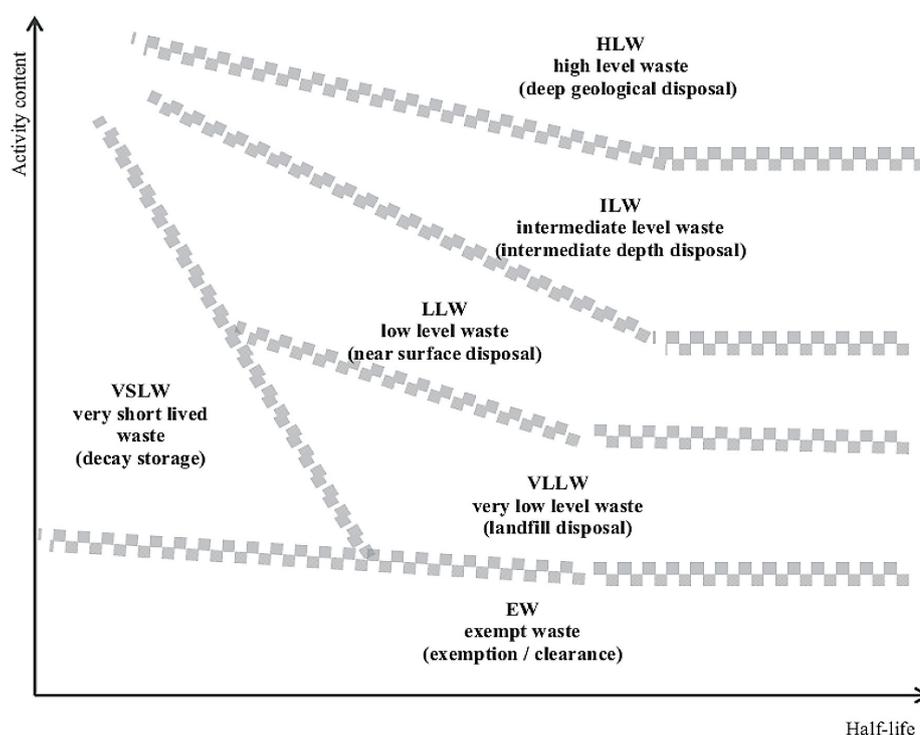


Fig. 1. Classification of solid radioactive waste in terms of their activity [IAEA 2009]

then placed in concrete containers. In some cases and additional barrier is required in the form of metal shields [Włodarski 2002b].

Gas-state waste constitutes a small percentage of all radioactive waste. Its radioactivity is defined by the presence of xenon and krypton. In practice, such waste is not classified, but defined as regards their activity and contents of isotopes.

STORAGE OF RADIOACTIVE WASTE

The storage of radioactive waste is relatively simple and boils down to conforming with the following requirements:

- the volume of the waste ought to be reduced to maximum,
- waterproofness and prevention of dispersion ought to be provided,
- storage ought not to endanger natural environment.

In order to meet all these requirements, numerous protective barriers are utilized. Natural and man-made barriers can be distinguished. The man-made barriers include:

- chemical barriers – difficult to dissolve chemical compounds binding radioactive waste;
- physical barriers – a binder (concrete, asphalt, organic polymers, ceramic mass) for solidification of radioactive waste in order to prevent spills, dispersion, spraying and wash out of radioactive substances;
- engineer barriers I – containers isolating radioactive waste, preventing mechanical damage,

protecting against elements and water; usually metal drum-shaped containers are used;

- engineer barriers II – concrete structures providing additional protection, in particular, against elements and corrosion of metal containers;
- bituminous or other impregnating layers – outer layers in order to prevent the penetration of precipitation waters to the storage area, container corrosion and wash out of radioactive substances.

Natural barriers include geological structures, favourable topography and also its non-seismicity. Suitable geological and hydrogeological conditions prevent the spread of radionuclides into the soil and their penetration into ground and surface waters [Frankowski & Mitreġa 1998].

Numerous countries possess waste dumps dedicated for storing radioactive waste. The choice of localization of such a dump must take into account geological and hydrogeological features of the area. The level of ground water ought to be lower than the depth of the dump and ground structure ought to prevent the migration of radionuclides (Figure 2). Local population must receive protection against the radioactive waste being released. The protection ought to be provided both during the operation of the dump as well as after it has been closed down.

In Poland, there exists only one waste dump conforming with the above requirements – National Radioactive Waste Dump (NRWD) in Rózanie near Pułtusk [Tomczak 1998].

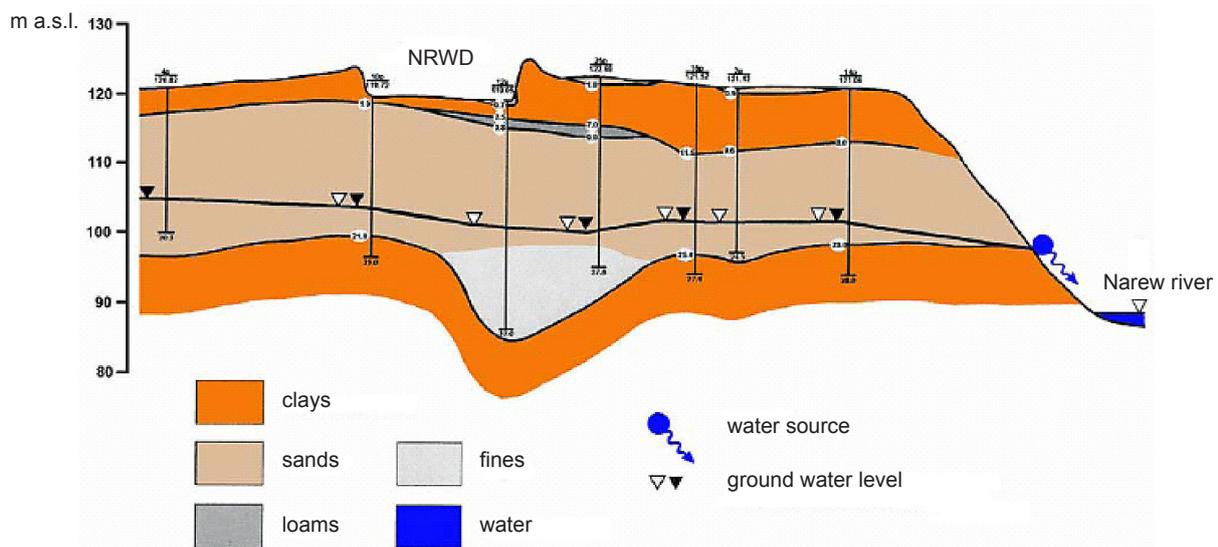


Fig. 2. Hydrogeological system profile of National Radioactive Waste Dump in Rózanie [Madaj 2010]

TECHNOLOGIES OF RADIOACTIVE WASTE PROCESSING

Liquid waste

The main purpose of liquid waste processing is the purification of output streams, reduction of waste volume and condensation of radioactive substances. During the process of utilization, components which, owing to their properties, can be recycled, ought to be separated. In order to process liquid waste the following methods are implemented:

- purification with inorganic sorbents,
- rotary evaporation,
- purification by means of ion-exchange resin filters,
- solidification by means of cement, asphalt and plastics.

A large volume reduction is obtained in the process of rotary evaporation and then filtration to the solid state. Radioactive nuclides contained in the sludge are separated by means of the following processes: chemical precipitation, ion exchange and distillation. Precipitation is a relatively simple method with quite low efficiency. It is suitable for liquids containing large amounts of salt and contaminated with low-activity substances. Ion exchange is more efficient when applied to the purification of relatively clean sewage containing little salt. The most universal and effective but expensive method of sewage treatment is evaporation. In case of high-activity sewage, an additional water treatment may be required e.g. by means of ion exchange or redistillation. The treated water is released or reused in technological processes

Sludge and concentrates which remain after evaporation must be processed to obtain solid state e.g. by means of cement solidification. The method involves mixing of these concentrates with cement mass and filling metal drums with it. The method is relatively simple and inexpensive, however it has its limitations:

- the volume is considerably increased as compared to the deposited concentrates;
- it requires limiting the contents of salt and boric acid which damage the structure of concrete;
- wash out of radioactive nuclides from concrete is possible while storing;
- it requires limiting of sludge's radioactivity (due to risk to personnel);

Solidification is widely applied to waste of low and intermediate activity. The utilization of liquid highly active waste requires the application of a different technology. Currently, the majority of highly active solutions are stored in tanks in liquid state. While stored, the solutions' activity considerably decreases- by approx. 90% in the first five years. During the first 150 days of spent fuel's storage, before undergoing processing, fuel's activity decreases by 30% as regards its initial value.

Storage containers are usually vertical cylinders made of stainless steel and their capacity amounts to 1200 m³. Containers are placed in heavy concrete underground chambers or ones at least partly immersed in the ground. Chambers are inlaid with stainless steel sheets. The container itself is placed on special supports located at the bottom of the chamber. Containers are fitted with cooling systems (pipes with water induced circulation immersed in a concentrate), systems of the concentrate mixing (in order to prevent the collection and solidification of residues at the bottom of the container), systems for pumping the concentrate from one container into another, measurement systems (measurement of pressure, temperature, level, concentration etc.), monitoring and alarm systems (excessive temperature, leakages), ventilation systems. Time of operation of such containers equals 20-30 years.

Rotary evaporation installations are used to process waste of intermediate activity. In the evaporation process chemical contaminants (e.g. heavy metals) are also removed. As a consequence, such method is particularly environment friendly because the purity of the distillate can be compared with rivers and lakes classified as clean. Releasing the treated sewage into the sewage system is always preceded by radiometric measurements whose result is the basis for further action. Post-evaporation concentrate can undergo solidification by means of cement.

In Institute of Chemistry and Nuclear Technology in Świerk a technology of sewage treatment by means of three-fold installation of reverse osmosis was introduced. The technology is used for treatment and solidification of liquid radioactive waste with low salt contents. Water obtained in the process is virtually free of radio-nuclides. Efficiency of sewage treatment amounts to 99,9% with the output of 2 m³/h. The volume of treated radioactive waste totals 45 m³ per annum. Solid waste fractions are concrete solidified and placed in drums [Gołofit 2003].

Solid waste

Processing of solid radioactive waste is carried out by means of the following:

- fragmentation,
- pressing,
- combustion,
- cement and plastics solidification,
- vitrification.

Waste which is impossible to store directly undergoes fragmentation. Fragmentation, basically separation into smaller parts, may lead to the creation of dusts and must usually be conducted in controlled environment.

Pressing by means of hydraulic presses leads to a considerable reduction of waste volume. The efficiency of the process increases along with the increase of press pressure. In 1990s, the technology of solid waste pressing which decreased waste volume by squashing the mass in a 12-ton-pressure hydraulic press, was implemented. Depending on the type of waste, reduction coefficients amounted to 1,5-3,0 [IAEA 2006].

For combustible waste its combustion is the most effective method of utilisation. Post-combustion ash is mixed with binders (cement and water) and the mixture is loaded into barrels. Combustion is carried out in special furnaces whose exhaust gasses pass through mechanical and electric filters in order to draw any dusts and chemical compounds which may contain radioactive nuclides.

Solidification

The following methods of solidification of solid waste are implemented:

- solidification in urea-formaldehyde resins which is used for the consolidation of biological waste,
- solidification in epoxy resins used for solidification of used ion-exchange resins from nuclear reactor's water cooling system,
- cement solidification.

Solidification involves mixing the radioactive waste with the proper proportion of a special cement or resin mixtures containing supplements improving solidifying properties. Mixing is carried out in drums, which at the same time constitute the container. Drums are layered in properly prepared chambers and flooded with thin cement-based mortar.

Since 1980s various methods of radioactive waste solidification have been implemented:

1. Spent radium materials are placed in a glass ampule and then in a brass casing. After that, the casing is hermetically encapsulated in a stainless steel container placed in a protective housing of a lead container.
2. Utilisation of smoke detectors involves their disassembly, control and separation of radioactive material (Am-241, Pu-239 and Pu-238) as waste. The waste is solidified in containers and stored. Savings as regards the area of storage amount to at least 95%.
3. Utilisation of spent reactor fuel involves two steps:
 - recycling of fuel,
 - deep geological storage.

The utilization of spent fuel proceeds according to the following plan. Spent fuel is stored in a water pool at the power plant for the period of 10 years. After this time, it is transported to a treatment plant where individual products are separated. Remains of plutonium and uranium are recycled, as these can be reused as nuclear fuel. Spent fuel contains 95% U-235 uranium and 1% PU-239 plutonium. 28 kg of plutonium containing approximately 65% of fissile plutonium isotopes can be obtained from recycling 2,5 ton of spent fuel [Skalmowski 2009].

The remaining portion of spent fuel (approx. 4% of the initial mass) is classified as waste which, having been solidified, is stored geologically 500-1000 m below ground.

Vitrification

The technology of vitrification of highly active waste in industrial installations for the treatment of spent reactor fuel originating from water reactors (pressurized and boiling) was implemented in France (La Hague), Great Britain (Sellafield), Japan (Tokai, Rokkashomura) and Russia (RT-1 and RT-2). The technology is based on providing a considerable amount of heat required for the development of amorphous structure and then rapid cooling. Single and two-stage installations are used in the process. In the single-stage installations, special heating crucibles are used to heat the substance to 1300-1450 °C [Ojovan & Lee 2011]. A partial liquefaction of waste occurs in the temperature. Heating is continued until the migration of components is complete.

The installation requires the use of glass as a flux, usually as paste, which facilitates liquefaction. In the two-stage process, individual heating stages are carried out in separate devices. During the initial heating stage, apart from chemical components, also organic components are neutralized. The process requires extensive amount of heat and special heating installations. Single and two-stage installations are utilized for the process of vitrification. Single and two-stage installations are used in the process [IAEA 2006]. In the single-stage process, special heating crucibles are used to heat the substance to 1300-1450 °C. A partial liquefaction of waste occurs in the temperature. Heating is continued until the migration of components is complete. The installation requires the use of glass as a flux, usually as paste, which facilitates liquefaction. In the two-stage process, individual heating stages are carried out in separate devices. Ceramic chamber furnaces or crucible furnaces are used in the heating process where the material is melted by means of high-intensity inductive electric current. Waste which is difficult to treat with other methods undergoes vitrification.

Ultimately, the waste takes up a vitrified-block shape of the container it was cooled in. Attempts at modifying the process by granulation of fine-grained waste fractions were undertaken [Colombo et al. 2003]. The granulate is heated in rotary or grate furnaces. Having undergone melting and phase transitions, the granulate is cooled down which leads to the development of tight vitreous coating. As a consequence, the initial shape of the lump is retained, as opposed to block melted waste. Vitrified radioactive waste is characterised by considerable resistance to mechanical damage, low chemical reactivity and insignificant release of dangerous compounds to environment [Connelly et al. 2011].

CONCLUSIONS

Contemporary technologies enable successful utilisation of radioactive waste. Poland has not developed its own nuclear power plant, thus the problem of utilizing spent reactor fuel is non-existent, with the exception of Maria nuclear reactor in Świerk used for research purposes. The greatest amount of radioactive waste originates from medical and industrial sectors, in particular from spent smoke detectors.

The most popular method of radioactive waste utilization is storing these in designated dumps in a non-threatening way. Liquid waste in the form of contaminated radioactive sewage most frequently undergoes evaporation in order to decrease its volume. Sludge obtained in the process is mixed with cement mass and stored in barrels.

Solid waste is usually cement or resin solidified and then mixed in a drum container. Such containers are stored at designated dumps or placed deep underground in closed down mines. Solidification, in terms of ecology, presents extensive limitations due to a possible damage to the concrete's structure and wash out of radioactive nuclides during storage.

Vitrification of radioactive waste is becoming more popular. Waste components are neutralized both in the crystalline structure and by hermetisation, which provides extensive protection against the release of dangerous compounds. Unfortunately, further development of the method requires extensive funds. However, in this case, positive ecological influence ought to surpass any other factors as regards the implementation of vitrification of radioactive waste.

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