

# The Management of High-Level Radioactive Wastes

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## WHAT ARE HIGH-LEVEL WASTES

The terms, low-level, medium- or intermediate-level and high-level radioactive wastes are being universally used, implying different concentrations of radionuclides or radioactivity in the waste. These terms originated in the 1950's for operational purposes and generally indicated how the waste was being handled or treated and what was being done with it under the local operating conditions at a particular site rather than precisely defining radioactivity concentrations or contamination. This still holds true today with the defined and/or quantitative divisions between the three categories differing from country to country and even amongst institutions in the same country Refs [1, 2].

There has been considerable agitation, during the past ten years, for quantitatively defining low-, medium- and high-level waste from a radioactivity standpoint in order to avoid confusion. On the other hand, there is considerable resistance against quantitatively defining these terms because the handling and effects of a curie of radiostrontium is not the same as a curie of plutonium which is not the same as a curie of tritium, and so on for the different radionuclides. Furthermore, definitions for low-level, medium-level and high-level wastes would not be the same from a health physics standpoint as from an operating standpoint as from a transportation standpoint as from a sea dumping standpoint, etc. However, the terms low-level, medium- or intermediate-level and high-level, within their broad and overlapping ranges, are generally understood by those involved with radioactive waste management and its technology.

High-level waste is characterized, of course, by high radiation levels but probably its most distinctive feature is that it requires special handling and considerations, such as thick biological shielding and engineered cooling systems because of the radiodecay heat load. The term high-level waste, generally implies the raffinate (liquid effluent) from the first cycle of fuel reprocessing operations that recover the plutonium and unburned uranium. The term also is extended to any matrix that contains a high enough concentration of fission products to require cooling, which, unless they are separated from the waste, includes the actinides (the alpha-emitting transuranium elements). Examples are solidified high-level

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waste or a "throw-away" spent fuel and possibly cladding hulls. Other examples of what sometimes is considered a high-level waste are removed highly irradiated reactor components, such as control rods, piping or flow orifices, and a container with several millions of curies of the gaseous fission product, krypton-85.

My remarks here concern mostly the management of the highly radioactive nuclear waste generated within the nuclear fuel during its burning (irradiation) in a reactor. Also, unless otherwise identified, my generalities will apply to LWR fuels although in principle they will hold true for the nuclear waste generated in other types of reactor fuels.

The major consideration in the management of high-level waste is to ensure its isolation from the biosphere and avoid any significant release of radionuclides, at least in concentrations that may be hazardous to man, over the extended time scale during which this possibility exists. It is widely agreed by nuclear waste management experts that a satisfactory high-level waste management scheme can be based on the following sequence of operations:

- interim storage in liquid form (if liquid);
- solidification (if liquid) and packaging the waste;
- engineered storage of the solid waste;
- disposal of the solid waste into geological formations (either on land or beneath the ocean floor).

Figure 1 illustrates this sequence. My remarks will follow the operations indicated by Figure 1. Alternative strategies also are possible and will be mentioned. With the exception of the French vitrification process that recently started full-scale operations at Marcoule, present industrial practice for power reactor fuels has not progressed beyond the first stage of the sequence but considerable research and development work is being directed to solidification processes and engineered interim storage facilities. There also is considerable activity in evaluating potential disposal sites or waste repositories in geological formations. These aspects will be discussed later.

## FUEL REPROCESSING AND HIGH-LEVEL WASTE

The current reprocessing flowscheme for power reactor fuels and probably the one that will be used for some time to come, consists of chopping or shearing the fuel element into short pieces, or otherwise removing the cladding, and dissolving the fuel in an aqueous nitric acid solution. (The remaining or removed fuel cladding is commonly termed "hulls".) The plutonium and unburned uranium (fissionable materials) are recovered by chemical separations. The remaining nitric acid solution which contains chemicals added during the process, greater than 99% of the nonvolatile fission products, together with impurities from cladding materials, corrosion products, several tenths of a per cent of originally dissolved plutonium and uranium and most of the actinide elements, constitutes the so-called high-level waste. Around 5–10 m<sup>3</sup> of this high-level liquid waste is produced per tonne of fuel reprocessed, depending on the fuel and the reprocessing flowscheme. The waste is treated to remove any remaining organic solvents and eventually is concentrated by evaporation to around 5–15 per cent of its initial volume for storage in specially designed waste tanks, the degree of concentration depending on the nature of the waste, the formation of precipitates, and the cooling capability of the waste tanks. The eventual result for

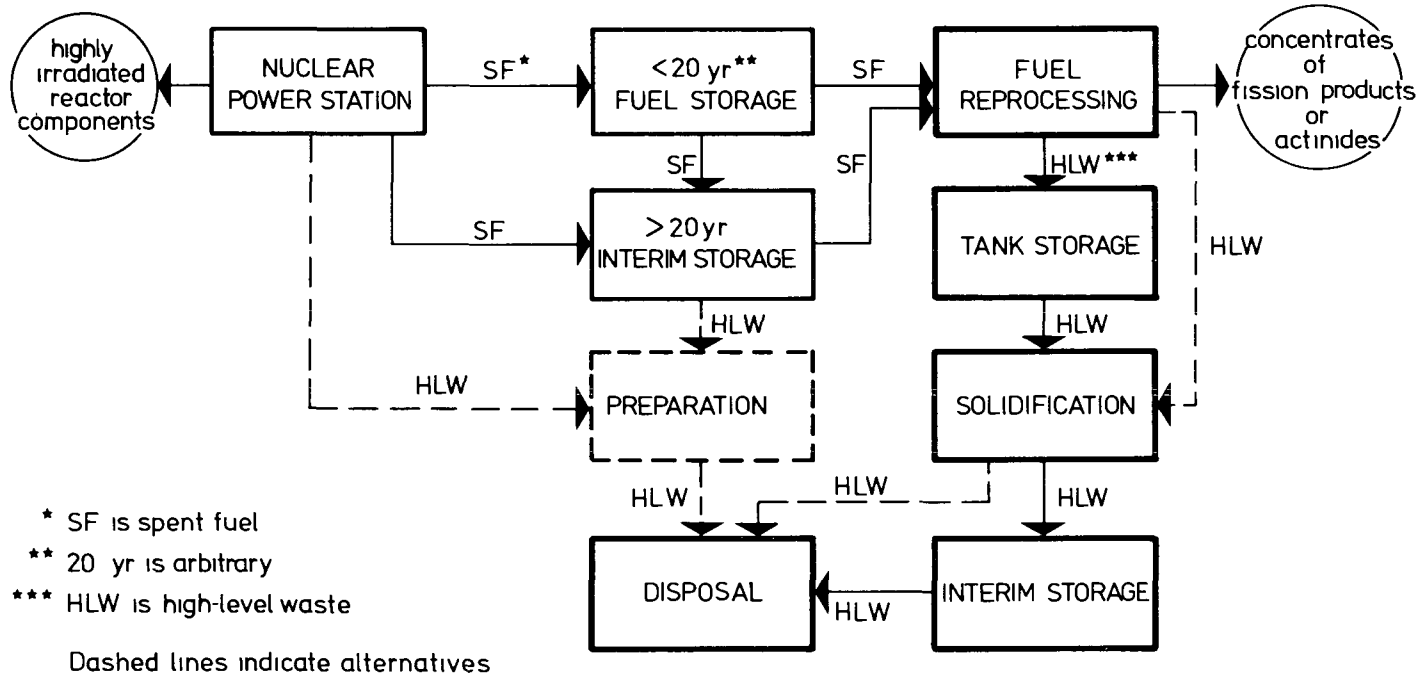


Figure 1. Handling High-level Radioactive Waste

reprocessing light-water reactor (LWR) fuels is around 15 m<sup>3</sup> of stored high-level liquid waste for each 1000 MW(e) generated annually.

Table 1 gives an indication of the high-level wastes from different reactor types and fuel reprocessing techniques. The management of these wastes must be integrated with the operation of the reprocessing facility and cannot be placed on a short-term basis. It must be anticipated, analysed and planned many years in advance. It is generally agreed that the high-level liquid waste should be solidified to a less mobile form at some time in the future.

## STORAGE OF HIGH-LEVEL LIQUID WASTE

At present, it is the general opinion that the high-level liquid waste concentrates resulting from the reprocessing of power reactor fuels should be kept in their acidic condition in high-integrity stainless steel tanks. (Neutralizing the acidic waste with caustic and storing it in carbon steel tanks is not considered the best practice since it increases the volume considerably, forms precipitates and sludges and limits the flexibility for subsequent treatment of the waste )

The waste containers (tanks) are in thick-walled concrete cells or underground vaults for appropriate shielding and are either double-walled or have a steel-lined vault to retain any leakage from the primary holding container(s). The capacity of this outer container should be capable of retaining the entire contents of a holding tank. Stainless steel tanks in use range from 70 to as much as 1000 m<sup>3</sup> capacity. Cooling systems (water) with standby emergency cooling facilities are provided to remove decay heat, the practice being to keep the temperature of the solution below 65°C to reduce corrosion of the stainless steel. Storage tank systems (tank farms) should also provide for in-tank agitation, ventilation, monitoring, solution transfer from both inner and outer containers, vapour condensation, removal of gases produced by radiolysis, and off-gas filtration.

In designing and constructing a tank farm system for storing high-level liquid waste, one should keep in mind that there should be a capability to fill and empty the tanks repeatedly, there should be a minimum of obstructions which would interfere with emptying or decontaminating both the inner and outer containers and the vault, and enough space should be allowed between the inner and the outer containers and the vault for adequate inspection and any maintenance. There should be an abundance of tank monitoring devices and instrumentation to detect tank levels, tank and liquid temperatures, and possible leakage. Gravity flow for liquid transfer is better than pumping. There should be sufficient tank space, preferably an empty tank, maintained at all times for an emergency. Waste tanks should be planned and constructed far enough in advance so that they will be available when needed. Management of a tank farm for liquid radioactive wastes requires careful planning several years ahead.

The technology for safe storage of high-level liquid waste in tanks over periods of many years have been demonstrated in several countries. Some leakage has occurred from tanks in the USA. This, however, was from mild steel tanks of early design. No leakage or other important problems have been encountered from storing the acidic waste in stainless steel tanks which has so far proven safe and convenient. It offers operating flexibility and leaves open the widest range of options for future treatment of the waste. It also can be

satisfactory for a considerable length of time. However, waste tanks do have a finite life and require the operation of heat removal and other auxiliary systems. Consequently, adequate surveillance must be maintained, with spare tanks available to deal with unexpected failures. Eventually the tanks will have to be replaced if their use continues.

However, it is generally thought that storage of high-level waste in the liquid form should be only an interim measure and that solidification of the waste should be undertaken when practicable. Solidification will reduce the mobility of the waste and its potential for dispersion. Furthermore, the solid form will be more suitable for transportation, storage and/or disposal.

## SOLIDIFICATION OF HIGH-LEVEL WASTE

As noted above, the primary aim of any solidification process is to convert the high-level waste solution to a solid form that is less mobile, more stable, requires less surveillance and is more suitable for transport, storage and disposal. Ideally, the final solid form should satisfy certain criteria, e.g. it should have high thermal and radiation stability together with good thermal conductivity and a high melting point. It should maintain its mechanical integrity, be resistant to shock and have a very low rate of leaching by water. However, depending on what is done with it, the solid form may not necessarily have to meet all of the above criteria nor would all forms need to meet the same requirements.

Several approaches have been developed for the solidification of high-level liquid waste, including the use of fluidized beds, stirred beds, rotary kilns, pots and spray chambers Ref [3]. They all essentially involve de-watering and denitrating the waste and heating the residue to between 400 and 1200°C, driving off most of the volatile constituents, and leaving a calcined solid. However, most calcined wastes are moderately soluble in water and are usually not considered to be a preferred form for storage or disposal. Consequently, in most cases, borosilicate or phosphate glass-forming constituents are added either in the calcining stage or as a second stage of the solidification process, incorporating the fission product and actinic oxides into a glass melt which cools to a vitrified product, with leaching rates similar to pyrex glass. The basic vitrification processes generally produce monolithic glass blocks in stainless steel containers. The volume equivalent to the annual generation of 1000 MW(e) in LWRs ranges from 2 to 4 m<sup>3</sup> with waste oxides content ranging from 20 to 30% depending on the radiodecay heat load that can be tolerated. This, in turn, depends on the desired centerline and surface temperatures for the filled containers.

While the solidification of high-level liquid waste has been investigated for about 20 years in several countries which are engaged in the reprocessing of irradiated fuels, only two of the processes have reached the state where they are being applied on a routine basis. The Idaho Chemical Processing Plant (ICPP) has been operating a fluidized bed calcination process on a production basis for about 15 years, and a vitrification plant, using a rotating drum calciner followed by a melting furnace to produce a borosilicate glass, recently has started production-scale operations at Marcoule, France (AVM process).

Besides basic calcination and vitrification, other, more novel, processes involve the fabrication of calcine or glass beads and coating them with pyrolytic carbon, silicon carbide and thin metal layers. There are processes that involve incorporating the granular calcine

or vitrified product into a wide range of matrices, such as metal and metal alloys and graphite. Other techniques, which have been or are being studied, are exothermic reactions which lead to virtually insoluble radionuclide products in metallic or ceramic phases, the production of crystalline and synthetic minerals which incorporate the radionuclides within low solubility crystalline lattices, pressed and sintered calcines, and sorption on zeolite or clay-like sponges followed by a high temperature treatment to produce a ceramic product. While many of these processes may provide final solid forms superior to the simpler vitrification process, they could be much more complicated from an operating standpoint, and many of them produce significantly larger volumes of the final solid form per unit of electricity generated.

Regardless, most high temperature solidification processes, especially calcination and denitration, involve evaporation and denitration of the liquid waste to a solid residue. This brings up treatment of the off-gases which should be mentioned. In addition to steam and oxides of nitrogen, the off-gases usually contain some radioactive and other fine particulate carry-over and volatile radionuclides, such as ruthenium. The off-gas treatment must trap and remove radioactive constituents in the off-gas and ensure that any discharge to the atmosphere is within acceptable limits.

The major amount of process equipment in a high-level waste solidification facility involves the off-gas treatment system which can be the most complex and sensitive section of a solidification facility. The treatment system generally consists of a complex arrangement, in series, of filters, condensers, scrubbers, fractionators and even dry bed absorbers. Most of the condensate and wash streams can be recycled, others might be discharged after treatment for removal of radioactivity to acceptable levels.

## INTERIM STORAGE OF SOLIDIFIED HIGH-LEVEL WASTE

Disposal concepts for the solidified high-level waste are currently being evaluated. Implementation of the most suitable concept or concepts for even demonstration purposes may require up to ten years or more. During the meantime, any solidified high-level waste is going to have to be stored in some readily retrievable and safe manner. Furthermore, even longer storage may be considered desirable in order to cool (radiodecay) the waste further and reduce the heat generation rate in the waste prior to its disposal, especially for disposal in a geological repository. While some may advocate disposal of the high-level waste immediately after its solidification, nevertheless some storage capability probably will be required between solidification and transfer to a disposal facility.

The difference between storage and disposal should be made quite clear. Storage is the emplacement of waste with the intent of retrieval. Consequently, a storage facility is located, designed and operated to facilitate eventually moving the waste somewhere else. Disposal is the emplacement of waste with no intent of retrieval, at least not after an initial demonstration of the disposal concept when the waste would be isolated from any easy access.

Storage facilities being considered for solidified high-level waste so far fall into three concepts: (a) water basins, (b) air-cooled vaults, and (c) shielded, air-cooled casks or canisters. The water basins and air-cooled vaults can be either located on the ground surface, below grade, or placed in underground excavations. While there are obvious benefits and

objections to the use of each location, each can be engineered to a satisfactory degree of safety. The choice depends on local environmental conditions and personal preferences. The use of shielded, air-cooled casks or canisters is a surface storage arrangement, requiring much larger land areas than the other two concepts.

The experience gained from storing irradiated fuel elements in water basins or air-cooled vaults is applicable to the storage of high-level wastes, with the greatest experience existing for water basins. While several variations of each concept have been studied, the only storage facility in actual operation is an air-cooled vault at Marcoule for storing the AVM product. A similar air-cooled vault for vitrified high-level waste is reported to be under construction in India. Also, the granular product from the fluidized bed waste calciner at the ICPP is being stored in large (3.7 metre diameter by 14 metre high) bins in an air-cooled vault. This is a special case because of the relatively low fission product content (heat generation rate) of the calcine. Recognizing that there are many design variations for the three basic storage concepts, a general description of each one follows.

(a) **Water basins:** The waste containers are generally, but not necessarily, sealed in stainless steel overpacks which are then stored in stainless steel-lined concrete basins filled with demineralized water. The water provides a cooling medium and transparent radiation shielding. Decay heat, removed by circulation of the water, is transferred to a secondary cooling loop where the heat is rejected either by cooling towers or to a pond or some other heat sink. The cooling system requires redundancy, an adequate cooling water supply, continual surveillance and operation, and close control over the water chemistry to minimize corrosion, particularly for long-term storage. However, the water does provide an additional contamination barrier for any leakage from a waste container and its overpack, and also a heat sink in case of a short-time failure of the cooling system.

(b) **Air-cooled vaults:** The waste containers are sealed in carbon steel overpacks which are suspended in concrete vaults and surrounded either by steel sleeves or by baffles for directional control of the cooling air. The filtered cooling air from a lower distribution plenum either passes upwards through annuli between the overpacks and the steel sleeves, or is appropriately distributed around the overpacks by baffles, and is collected by an upper plenum. After passing through a high-efficiency filter, the warm air is discharged through a stack. Pressure drops and good cooling capability (capacity) requires forced air circulation. Nevertheless, if the cooling requirements are low enough, there is no reason that an air-cooled vault cannot be operated using natural draught cooling. Radiation shielding is provided by either the thick concrete construction or the location of the vault. The concept involves high-integrity overpacks, continual surveillance of the air circulation and, where forced air circulation is required, an assured standby system. One drawback is the possibility of airborne contamination should both the waste container and overpack fail.

(c) **Shielded, air-cooled casks:** One or more waste containers are sealed within a steel cask. The cask is surrounded by an annular concrete gamma-neutron shield with an air space between and transported to an outdoor storage area where it is mounted vertically on a concrete base in such a way that the heat is dissipated by natural convective air flow through the annulus between the steel cask and the concrete shield. The flexibility of this concept is attractive as the facilities can be modified readily to adjust to the nature of the wastes, the dimensions of the waste containers and their number. The system is practically immune

to damage because of the massive concrete shielding and requires a minimum amount of operation and operational surveillance but like other storage concepts requires continual monitoring for the detection of any escaping radionuclides. Defective casks or concrete shields can be quickly identified and either repaired or replaced. As indicated, a disadvantage of this concept is its large land requirements, estimated to be an order of magnitude greater than that of the other two concepts.

Choice of a suitable concept for a high-level solid waste storage facility depends on the site, environmental considerations, costs and continuing reliability of the materials. Another obvious consideration is the estimated interim storage time or how long the storage facility will be used. It has been estimated that the total costs of constructing and operating an engineered storage facility, including surveillance for 100 years after the last waste delivery, are comparable for the two air-cooled concepts but about 50% greater for the water-basin concept because of the additional costs for power and water and more staff. Currently available engineering materials and technology should be capable of providing a facility and storage system that will perform adequately for at least 100 years, if necessary. However, it is anticipated that suitable disposal methods for the high-level waste should be available within the next 30 years, hopefully before the end of the century. On the other hand, as mentioned previously, there may be technical and economic advantages to a long interim storage period for radiodecay thus reducing both the waste temperature and the heat dissipation problems and effects in a waste repository.

## DISPOSAL OF HIGH-LEVEL WASTE

Disposal of the high-level nuclear wastes, including the very long-lived transuranium radionuclides, is one of today's most popular topics and a public issue. It is a difficult topic to summarize or debate because, in my opinion, it seems to have been emotionalized out of perspective. The primary reason there is not a disposal method for high-level waste in operation today is that there has not been a quantitative need for one, and probably won't be for another decade or so. Nevertheless, many are insisting that an assured and demonstrated disposal system for nuclear waste should be available as one condition for accepting nuclear power.

Owing to its accessibility, disposal of radioactive waste in engineered facilities on or slightly below ground-surface grade can be considered essentially more of a storage mode. For more remote isolation from man and his environment, several disposal concepts for high-level and transuranic-containing wastes have been proposed Ref.[4]. They essentially fall into the following classification.

- (a) Terrestrial (on earth)
  - (i) in geological formations deep under land areas Refs.[5,6] or beneath the ocean floor,
  - (ii) on the ocean floor;
  - (iii) in glaciated areas.
- (b) Into space.
- (c) By nuclear transmutation.

All concepts for disposal of radioactive waste on earth are aimed at isolating the waste or its radionuclides from the biosphere to the extent that there will occur no unacceptable levels



of exposure to man or other biological species. This objective can be obtained by providing containment with an adequate degree of reliability for the requisite time period or by ensuring an adequate retention or delay mechanism and the availability of a dispersal medium to dilute to acceptable levels any radionuclides that do reach the biosphere.

It is generally agreed that for disposal in a terrestrial environment there would be less risk with the high-level waste being in a solid leach-resistant form. However, the USSR has demonstrated the possibilities of injecting high-level liquid waste directly into a porous water-bearing strata, having appropriate isolation, although it currently has a very active programme that is developing the technology for the disposal of vitrified waste in a geological repository. My remarks concern the disposal of only solidified high-level waste.

Fortunately, the hazard of a radioactive source, unlike many hazardous non-radioactive elements, such as mercury, cadmium and lead, declines with time. Required isolation or confinement periods for radionuclides depends upon their respective initial concentrations and their respective half-lives for concentrations to reach acceptable levels. A "rule of thumb" for estimating the time that a high concentration of a radionuclide in a fixed radioactive source may reach an acceptably non-hazardous level is twenty times its half-life.

Suggested periods of integrity for the isolation of high-level waste have been based upon the relative concentrations of the radioactive fission products and the remaining actinides in the high-level waste. There has been the *tacit* assumption that cesium-137 and strontium-90, each with approximately a 30-year half-life, are the determining fission products while plutonium-239 with a half-life of about 25 000 years is the determining actinide. Consequently, when one considers the desired isolation period, it extends to about 1000 years for the fission products and on the order of a 100 000 years for the transuranium actinides in the high-level waste. While projections of isolation over this latter time period may seem somewhat presumptuous, it is within the periods of time during which geological events should be predictable with reasonable certainty for many locations.

The basic requirement for the suitability of any geological formation for a repository for the disposal of radioactive waste is its capability to contain and isolate the radionuclides from the biosphere until they have decayed to non-hazardous levels. The predominant natural mechanism by which the radionuclides in a waste could be moved from a carefully selected deep repository into the biosphere is by action of groundwater. Hence, a very important consideration is related to the protection of the waste from circulating groundwater. Consequently, a dry geological formation, while so maintained, should provide ample protection. The integrity of a geological formation against circulating groundwater can be further enhanced by surrounding geological barriers. Other barriers that can contribute to the confinement of the radionuclides need not limit a repository to a completely dry formation as long as the formation's groundwater migration rates are stagnant or extremely slow over long distances. These other barriers are:

- The form of the waste, e.g. a relatively insoluble product.
- A high-integrity container.
- Groundwater composition and chemistry.
- The integrity of surrounding geological barriers against groundwater migration.
- Retardation of radionuclides migrating from the disposal zone by various mechanisms in the surrounding geological strata, such as ion exchange, filtration, surface adsorption and precipitation. The magnitude of the retardation effect can vary considerably with the

- nature of the geological formation or strata and the radionuclide. It should be studied closely prior to giving it a credible safety factor.
- Artificial mechanisms and/or engineered devices to retard groundwater migration and waste dissolution rates.

The independent nature of each of the foregoing barriers illustrates the large measure of redundancy that can be incorporated into schemes aimed at isolating wastes in a geological formation.

Thick beds or diapiric intrusions of **rock salt** are currently considered one of the more promising geological media for the disposal of high-level waste and waste that is mostly contaminated with the very long-lived transuranics. This results largely from the considerable studies that have been carried out in the United States and the Federal Republic of Germany. The major advantages of rock salt as a medium for waste disposal are its ease of mining, its very low water content, its relatively good thermal properties and its plastic characteristics enabling it to flow and seal the man-made penetrations or any faulting that may occur. On the other hand, rock salt is water soluble, decrepitates at relatively moderate temperatures and is rather corrosive to most metals.

Some **argillaceous formations** (clays), which have a high plasticity, are being investigated in some countries. The plasticity of clays is approximately proportional to their water content. In order to be sufficiently plastic, suitable clays probably would have to contain 15–20% water which, however, moves with extremely low velocities or not at all. While clays have a much lower thermal conductivity than rock salt and may present excavating (mining) difficulties, they offer, on the other hand, certain advantages such as high sorption capacity, insolubility and very little corrosion of the waste containers.

Investigations also are being carried out regarding the possibilities of locating repositories for radioactive wastes in **crystalline or so-called "hard" rocks**, including granite, basalt, limestone, and metamorphic rocks. Such rocks are impermeable when massive but frequently are intersected by a network of joints or fractures that are able to transmit large amounts of groundwater. However, hard rock formations containing very little or no circulating groundwater do exist, usually as a result of particular geological situations, such as secondary sealing (mineral deposition) of the fractures or isolation from aquifers by impermeable formations. Even if faulting were to lead eventually to a limited contact between waste and groundwater, this would not necessarily imply a serious loss of isolation because further barriers can be afforded by the container, the waste form and by retarded migration of the radionuclides through the surrounding strata.

A discussion of the comparative merits of different types of geologic formations, including their surrounding strata, for the location of a radioactive waste repository can go on and on. The point is that there should be various acceptable locations for waste disposal in deep rock media.

Disposal in geological formations under the ocean floor is not conceptually different from disposal in geological formations under land. The essential difference lies in the waste emplacement technology and the ease of waste retrieval, if that should ever be necessary. While technology exists for under-land operations, the details for emplacement under the ocean floor still are conceptual although similar activities are carried out in sub-marine explorations. However, disposal of the waste in formations under the ocean floor has the

inherent advantages of remoteness from man's anticipated activities, high ion exchange capacity of any overlying sediments, and the enormous dilution of any radionuclides that may escape.

The emplacement of carefully conditioned high-level waste, including the transuranics, on the ocean floor has been proposed by some. The technical feasibility of the concept depends on the ability to produce solid matrices and/or containers capable of providing the required long-term containment with the necessary reliability. An attractive feature is the cooling capability of the surrounding water environment.

Various emplacement concepts have been proposed for the disposal of wastes in continental ice sheets, especially the Antarctic ice cap that apparently has been in existence for several millions of years. It is theorized that the warm waste containers would melt through the ice to the ice-rock interface and be sealed off by the refreezing of the melt behind them. The reliability of the containment could be further enhanced by emplacement of the waste in the bedrock underlying the ice sheet, which, in a sense, would be disposal in a geological formation. There currently is little interest in the possible use of glaciated areas due to the mobility of the ice caps, a limited understanding of the mechanisms controlling the long-term climate of the earth, and the more favorable aspects of accessible geological formations.

Disposal of high-level waste into space would provide its most complete isolation from man's environment. However, the cost of launch energy currently limits the practicality of the concept to small special volumes. Launch reliability and the consequences of the waste-containing capsules re-entering the atmosphere are other issues of concern.

Nuclear transmutation is regarded by some as the ideal solution, in principle, to the problem of disposing of the long-lived radionuclides in the high-level waste. It involves their conversion into much shorter-lived or even stable nuclides. The transmutation of the long-lived fission products is not considered to be feasible within the limits of foreseeable technology, although transmutation of the transuranium actinides could be feasible. An attractive idea is to remove the transuranium alpha-emitters (actinides) with the very long half-lives from the bulk of the wastes, especially the high-level waste. The remaining fission product contaminated material then supposedly would need to be isolated only up to around 1 000 years. Without the high-heat generating fission products, the low-heat generating transuranics could be disposed of in some more suitable manner or recycled back to nuclear reactors for burning (transmutation) to the shorter-lived fission products or stable elements.

While it appears to be technically feasible to remove the transuranics to negligible concentrations in the high-level waste, there also are some very long-lived fission products remaining that need to be evaluated from a biological standpoint. Furthermore, the high-level raffinates from fuel reprocessing represents less than one-half of the transuranics in nuclear fuel cycle waste, and probably would represent much less if the transuranics are to be recycled for burning in reactors. There now is considerable doubt regarding the benefits of handling a large circulating load of transuranics in the nuclear fuel cycle.

The disposal of high-level waste in geological formations currently appears to be the only feasible concept under today's technology while maintaining the option during an initial period for potentially retrieving the waste, should this be necessary Ref [7]. Probably the principle area of uncertainty concerns the effect of the temperature of the high-level waste on the repository environment. This, of course, can be modified by the concentration of the

heat-generating fission products in the waste, a suitable aging period prior to its disposal and the density of the waste emplacement.

## IRRADIATED FUEL ELEMENTS

There has been considerable thought lately about the possibility of not reprocessing spent nuclear fuels in order to recover and recycle their fissile and fertile material (plutonium and unburned uranium) but rather to dispose of the spent fuel elements as a waste. The extent that such disposal of a potential national resource will ever actually be carried out is rather speculative, at least until there is assurance of an acceptable source of energy to replace nuclear fission. Nevertheless, in today's climate, the disposal of spent fuel elements as a waste product should be considered. In addition to the fission products and transplutonium elements normally constituting the high-level waste, the spent fuel elements contain around 99 times more plutonium and uranium than the equivalent high-level waste that would result from reprocessing them. There also is 7–10 times the volume of material in the spent fuel element for disposal.

What has been discussed regarding the storage and disposal of solidified high-level waste also holds for the storage and disposal of spent fuel elements. As mentioned, proposed storage modes for solidified high-level waste are extrapolated from the technology, experience, and concepts of storing irradiated reactor fuels, i.e. water basins, air-cooled vaults and air-cooled individual canisters. There is little doubt that suitably overpacked irradiated reactor fuels can be stored for long periods of surveillance, with air-cooled concepts probably being preferable when the fuels are cool enough.

**Table 1. A Comparison of the Thermal Power and Radioactivity of Spent Fuel (SF) and High-Level Waste (HLW)**

Time since discharge of spent fuel (Years)	Thermal power (watts/MTHM)*		Radioactivity (Ci/MTHM)*	
	SF	HLW	SF	HLW
10	1200	1000	410 000	320 000
100	290	110	42 000	35 000
1000	55	3.3	1 800	130
10 000	14	0.47	480	42
100 000	1.1	0.11	58	21
1 000 000	0.39	0.15	21	10

\* MTHM is metric tons of heavy metal originally charged to the reactor.

Table 2. Nature of Radioactive Liquid Wastes from Different Reactor Types

Reactor Type	Fuel		Characteristics of Waste Solution Entering Evaporator						Initial Characteristics of Concentrate to be Stored			
	Type of Fuel	Type of Cladding	Typical Burn-up (MWD/t)	Minimum Time of Cooling before Reprocessing (T <sup>1</sup> )	Volume l/t Heavy Metal	Activity at Time (T <sup>1</sup> Ci/l)	Heat Content at Time (T <sup>1</sup> W/l)	Possible Concentration Factor	Volume (after evaporation)(l/t)	Activity at Time (T <sup>1</sup> Ci/l)	Heat Content at Time (T <sup>1</sup> W/l)	Acidity (N)
LWR (USA)	UO <sub>2</sub>	Zircaloy 2 or 4	29 000*	3a*	5 200	150	0.6	14	380		8.3	4-7
LWR (France)	UO <sub>2</sub>	Zr	33 000	1a	9 800	230	0.93	20	540	4 100	18.5	2.5
LWR (UK)	UO <sub>2</sub>	Zircaloy	33 000	150d	6 250	860	3.2	16.5	400		5.3	0.5-1.0
LWR (India)	UO <sub>2</sub>	Zircaloy	15 000	150d	7 200	300	9.0	9	800	3 000	0.9	2-3
LWR (Japan)	UO <sub>2</sub>	Zircaloy	28 000	180d	5 500	210	0.59	16	350	3 100	9.3	2.5
VVER (USSR)	UO <sub>2</sub>	Zr	28 000	3a	5 500	60	0.27	13	420	730	3.3	4-6
Gas-cooled (Fr)	U Mo	Mg	3 000	1a	7 600	70	0.24	70	110	4 800	17	0.8
Gas-cooled (Fr)	U/Si/Al	Mg	5 000	1a	5 400	100	0.34	50	100	5 400	18	2.5
Magnox	U (Nat)	Magnox	1 300 3 500	125d	4 500	460	0.25	50 100	90 45		12	3
AGR	UO <sub>2</sub>	SS	37 000 18 000	1a	5 000	600	0.33	20	100		16	
Candu	No plans for reprocessing											
HTGR	UC <sub>2</sub> /ThC <sub>2</sub>	Graphite, SiC	100 000	180d	5 700	1 700	7.5		3 600			
PFR	(U,Pu)O <sub>2</sub>	SS	60 000	180d	9 100	900 (A) 1 200 (B)	4 (A) 5 (B)	2.5	3 600		10 (C) 12 (C)	3
MTR	U-Al	Al	200 000	~ 1a	400 000	200	0.8	1.3	300 000		1	-1

\*Probable equilibrium values for USA (Currently there is no reprocessing of commercial nuclear reactor fuel)

- Notes (A) 1st cycle, mean of outer and inner  
 (B) 6th cycle, mean of outer and inner  
 (C) Assuming evaporation immediately (in practice, evaporation will be delayed)

While it may be technically reasonable to dispose of spent fuel, or one that has been overpacked, directly into a geological formation, relying on geological containment, it is debatable whether, from a waste management standpoint, this will ever be acceptable as minimizing risk to the extent practicable. First, one should recognize that nuclear fuel currently is designed for optimum performance within a reactor and not to meet any disposal criteria. Furthermore, gaseous fission products are within the fuel element under considerable pressure and one has to consider the radiological impact of their escaping during the handling of the fuel element or in the disposal medium, at least until radiodecay has reduced their concentrations to biologically acceptable levels. On the other hand, there is the possibility that the fuel elements can be confined and disposed of in suitably designed cases or coffins that would be expected to maintain their integrity for on the order of a hundred thousand years or so that their contents remain hazardous. As a matter of fact, the plutonium content of the spent fuels probably extends this time period considerably. In the long run, it may be desirable to de-gas and to process the irradiated fuel into some form for disposal, such as chopping or shearing the fuel elements into smaller pieces and placing them in a relatively insoluble matrix. Techniques suitable for the disposal of spent fuel elements have been neither studied extensively nor agreed upon.

## OTHER HIGH-LEVEL WASTE

Figure 1 identifies highly irradiated reactor components and fission product concentrates as possible high-level waste. These can be covered rather quickly.

Highly irradiated reactor components can be considered high-level waste because of their induced radioactivity. They initially require heavy shielding for radiation protection but ambient air-cooling generally is sufficient. The principal heat-generating radionuclides do not have relatively long half-lives and, in time, decay to levels which may require precautions but cause no problems for the disposal of these components either underground or elsewhere. The current technique of handling such radiologically hot components is to store them in a heavily shielded, well ventilated area for a suitable cooling period, at least until the heat generation rate no longer is of concern during their preparation for disposal and their disposal.

Fission product or actinide concentrates result from removal of selected fission product fractions or certain actinide fractions, for some reason, from fuel reprocessing streams or the high-level waste stream. They also can be evaporator or concentrator bottoms resulting from the evaporation and concentration of medium-level liquid waste streams. Unless required for some useful purpose these liquid concentrates can go to the high-level liquid waste storage tanks. Here, one must ensure that these waste streams, particularly the evaporator bottoms, do not add some component incompatible with the planned solidification process. It may be desirable or necessary to solidify these highly radioactive liquid wastes by a separate process.

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