

10/7-18-95 JS (1)

LBL-37333  
UC-~~600~~ 2010



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### Geological Aspects of the Nuclear Waste Disposal Problem

N.P. Laverov, B.I. Omelianenko, and V.I. Velichkin

June 1994



Prepared for the U.S. Department of Energy under Contract Number DE-AC03-76SF00098

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**GEOLOGICAL ASPECTS  
OF THE  
NUCLEAR WASTE DISPOSAL PROBLEM**

**N.P. Laverov**

Russian Academy of Sciences

**B.I. Omelianenko, V.I. Velichkin**

Institute of Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry  
of the Russian Academy of Sciences, Russia

June 1994

Lawrence Berkeley Laboratory  
University of California  
1 Cyclotron Road  
Berkeley, California 94720, USA

Under the auspices of the Department of Energy, Office of Environmental Management, Office of Technology Development (DOE/EM-OTD) and the Department of Energy, Office of Energy Research, Office of Basic Energy Sciences (DOE/ER-BES) under Contract Number DE-AC03-76F00098.

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# **GEOLOGICAL ASPECTS OF THE NUCLEAR WASTE DISPOSAL PROBLEM**

**N.P. Laverov, B.I. Omelianenko, V.I. Velichkin**

## **Abstract**

For the successful solution of the high-level waste (HLW) problem in Russia one must take into account such factors as the existence of the great volume of accumulated HLW, the large size and variety of geological conditions in the country, and the difficult economic conditions. The most efficient method of HLW disposal consists in the maximum use of protective capacities of the geological environment and in using inexpensive natural minerals for engineered barrier construction. In this paper, the principal trends of geological investigation directed toward the solution of HLW disposal are considered. One urgent practical aim is the selection of sites in deep wells in regions where the HLW is now held in temporary storage. The aim of long-term investigations into HLW disposal is to evaluate geological prerequisites for regional HLW repositories.

## **Introduction**

The principal requirement for nuclear waste (NW) disposal is to isolate NW from the biosphere as long as it is potentially dangerous to people. Thus the design and building of a NW repository that can fix radioactive elements in place is a desired goal. It is obvious, therefore, that the NW disposal problem is above all a geologic one. At the same time, one must recognize that the disposal topic has practically been ignored in Russian scientific literature. The authors of this article hope to make up for this deficiency, even if only in part.

## **Nuclear waste as a cause of radioecological danger**

The first years of atomic industry development in the USSR were characterized by a serious underestimate of the danger associated with NW. In the 1949–1951 nuclear weapons development period, radiochemical liquid waste products in the “Mayak” region (between Cheliabinsk and Ekatarinenburg in the South Ural) were directly discharged into the river Techa. The radioactivity of NW discharged during this period amounted to 2.8

million Ci [Drozhko et al., 1993]. About 70% of the radionuclides were sorbed by bottom sediments. The tragic consequences of this policy ultimately brought about an end to the uncontrollable discharge of liquid NW into the river. Subsequently, middle-level (MLW) and low-level (LLW) radioactive wastes were discharged into closed reservoirs and HLW was accumulated in special containers. The design of storage facilities and the quality of process instrumentation did not correspond to the level of danger, however, and in 1957 an 80-ton container filled with liquid HLW exploded. As a result, 20 million Ci of HLW were released. About 2 million Ci were lifted to an altitude of 1 km and scattered by the wind. An area 120 km long and about 5 km wide was contaminated. This area was called "East-Uralian Track" [Nikipelov and Drozhko, 1990].

After the accident, the technology of HLW storage was substantially modified. The problems of processing, consolidating, and disposing of HLW remained unsettled, however, and as a consequence nuclear industry development became dependent on nuclear waste disposal decisions. It is obvious that because of the cruel aspiration of the USSR leadership to achieve parity with the U.S. in nuclear weapons, slowing down or ending nuclear weapons production was impossible. At the same time, economic conditions did not allow the use of expensive foreign technologies for processing, storing, and disposing of NW. The simple and economical strategy of disposing of liquid NW in deep water-bearing sites was the soviet scientist's answer to this dilemma [Spitsin et al., 1972, 1978; Kedrovski et al., 1991]. This decision made it possible to use the integrated chemical plant in Siberia (Tomsk), the integrated mining-chemical works in Krasnoyarsk, and the Research Institute of Nuclear Reactors in Dimitrovgrad. About 50 million cubic meters of liquid waste, including HLW, was disposed of in the water-bearing repositories over this entire period. The total radioactivity of buried waste amounted to 2 billion Ci. This method of disposal does not meet IAEA standards, which apply especially to waste containing transuranic elements. The IAEA standards were set because of the unreliability of forecasts over long time periods. At the same time it must be admitted that the hazards associated with buried waste are much less than hazards associated with surface disposal.

Unfortunately, the technology of disposing of liquid NW in water-bearing underground locations was unfit for the "Mayak" region. In that region, waste was discharged into open reservoirs. As a result 4 million cubic meters of waste with a total radioactivity of 120 million Ci was allowed to accumulate in "Karachai" lake, and about 10 million cubic meters (20 million Ci) were deposited in Reservoir 17. A good deal of LLW was deposited in other reservoirs.

Like the disposal of NW in underground water-bearing sites, storage of NW in open reservoirs is a serious violation of IAEA standards. Scattering of aerosols during strong winds, the almost-certain underground water contamination, risk of waterspouts, and other factors contribute to the high probability of radioecological catastrophes. Before much passage of time the theoretical hazards developed into actual events. After saturation by radionuclides, the bottom muds and underlying loams of the "Karachai" lakes actually prevented the penetration of radionuclides into underground waters. But particularly severe contamination was caused by direct penetration of radioactive water from "Karachai" to underground water through fractures of crystalline rocks in 1961, when considerable expansion of the reservoir occurred during the rainy summer. By the time of this writing, 5 million cubic meters of polluted water had infiltrated water-bearing strata from "Karachai." As a result, a lens of polluted water of 14 km<sup>2</sup> was formed. Contaminated groundwater is spreading to the south and to the north at a speed of 80 m per year, threatening to enter water intakes and rivers.

In the dry summer of 1967 the water level in "Karachai" fell and the bottom mud, oversaturated with radionuclides, was exposed to sunlight. When the mud dried, winds scattered these radionuclides and an area of 2700 km<sup>2</sup> was contaminated with radioactivity totaling 6 million Ci. Waterspouts, fairly common in the region of the Urals, posed a serious danger. Fortunately, the reservoirs of "Mayak" region were not affected by waterspouts until recently. In recent years the solidification of liquid HLW and MLW into an alumino-phosphate glass matrix was introduced at "Mayak." Since that technology was introduced, about 100 million Ci of the waste has been solidified.

Another hazardous situation has been created in areas where atomic-powered ships are based. Much spent fuel has accumulated near these ports. Unfortunately, concentration and solidification of liquid waste from these sources has not yet been introduced. Eighteen reactors from ships have been sunk in the oceans, six containing spent fuel. A good deal of LLW has also been discharged into the ocean.

It is obvious that a strong necessity exists for developing strategies, and a policy based on those strategies, that will address the overall NW disposal problem. This policy will have to take into account the presence of huge amounts of waste and the real economic state that Russia finds itself in. A number of suggestions and recommendations are considered below.

## **Geological prerequisites for nuclear waste disposal decisions**

Various methods for removing NW from the biosphere are under consideration: transmutation of radionuclides, dispatching space rockets with NW into outer space, dipping refractory containers with HLW into the Earth's mantle through the melt formed by HLW heat release, and underground disposal of NW. The last method is recognized as the most realistic and practical.

The conclusion that underground disposal is the best choice clearly follows from the experience of mineral deposits research. It has been determined that mineral deposits containing uranium ores, under specific geological conditions, can be stable in interior parts of the Earth for many millions of years without any migration to the surface. One of the prerequisites for high safety in dealing with concentrations of uranium minerals is limiting the geological conditions where they are placed. Uranium deposits are destroyed by the action of oxygenous water. This process is particularly intense in the presence of sulfides that form sulfate solutions. If NW were composed solely of uranium it would be far more manageable in a repository. Unfortunately, NW consists of many radionuclides with various properties, which means that conditions that might create immobility for one radionuclide might not immobilize the others. For some radionuclides immobilizing conditions can be created by specific values of pH or Eh, for others by isomorphic entry into internal crystal framework, or for others by the presence of minerals with high sorption properties.

The Oklo deposit is a unique natural analog of a NW repository. A natural reactor was active there 2000 million years ago for 500 thousand years at a depth of approximately 3.5 km. Enclosing rocks were heated to 600 °C. Most of the radionuclides were kept within the site of their origin by isomorphic entry into the internal crystal framework of uraninite. The stability of uraninite contributed to limiting the geological variables in the area.

A number of articles by foreign scholars have been devoted to examining conditions favoring the isolation of radioisotopes within the limited volume of uranium deposits [Brookins, 1978; Chapman, McKinley, 1988; Cramer et al., 1987]. It is necessary to analyze the data for uranium deposits of the former USSR from this point of view. Natural uranium deposits are not the only analogs of a NW repository. Sulfide ores with a high content of such a toxic element as arsenic are widespread in nature and in most cases are safely isolated from the biosphere by geological media. Other natural analogs of liquid NW repositories are deposits of oil and mineral water. The main prerequisite of repository safety is the absence of water exchange between water-bearing zones and the surface.



On the whole, the observations of natural repositories are evidence of the presence in the Earth's crust of regions characterized by high insulating properties of geological media. The task of the geologist is to provide a selection of such sites for building NW repositories.

### **Types of nuclear waste and the methods of their disposal**

Of principal importance in repository site selection is the period of isolation required and the NW action on the insulating properties of the geological media. Obviously, NW composition will determine the requisite period of its isolation. When transuranic elements are present in NW one must plan for a storage duration that is practically unlimited. A reliable prognosis of geological events that can cause radionuclide penetration into the biosphere is impossible for the entire period of isolation. In the U.S., the design of an underground repository for HLW was mandated by a 1982 act of Congress to provide safe underground storage for a period of 10,000 years. The intention is that available geological and engineering barriers have to provide an even longer period of isolation [Krauskopf, 1988].

Three categories of HLW containing transuranic elements are recognized:

- Liquid HLW obtained by different methods, including the waste remaining after extracting plutonium and uranium in the reprocessing of spent fuel.
- Solidified HLW formed as a result of concentrating and vitrifying liquid HLW.
- Spent fuel that is not subject to reprocessing.

The influence of HLW upon geological media is the second factor that should be taken into account in the selection of sites for repositories. Reliable information on such influence can be obtained from the underground laboratories. Such laboratories function in many countries but have not yet been created in Russia.

The main potential factors that can affect the geological media are temperature and radiation. Investigations show that radiation in most cases does not influence the insulating properties of the geological environment [Krauskopf, 1988]. Heat release exerts a more substantial influence. Under heat exposure, thermal expansion, porosity, and permeability increase; minerals dehydrate; underground waters move as a result of heat convection; the interaction between HLW and groundwater is stimulated; the preservative matrix decomposes—though it is enough, this enumeration of the possible negative effects of temperature is incomplete. Heat release is the cardinal obstacle that both prevents repository designers from concentrating the volume of HLW and that prevents them from

decreasing the distances between canisters in repositories; thus the prospect of heat release stands in the way of lowering expenses. When calculations are made of the maximum concentration of radionuclides in units of volume of HLW and of the minimum distances between canisters, the calculations are derived from the value of the ultimate permissible temperature. The maximum temperature in a repository will be reached in some dozen years; after that it begins to decline gradually. Temperature conditions over any period of time can be calculated as a function of the variety and age of waste, the concentration of heat-generating elements, the distance between canisters, and the thermal conductivity of rocks. In designing a repository, one can use different combinations of these variables to predict temperatures. Reducing temperatures is possible only by increasing repository dimensions, which of course increases the repository cost. Researchers do not agree on what the optimum temperature conditions are for repositories. In Europe and Canada, standard repository design limits the allowable temperature to 100 °C. In the U.S. this limit is 250 °C. These divergencies in the values of allowable temperatures underline the uncertainty and disagreement that exists over the influence of temperature on geological media. This uncertainty cannot be eliminated, even though we have results from underground laboratory investigations, because the effects of temperatures over a very long period of time can be quite different from short-term effects.

Classifying NW by the level of radioactivity into LLW, MLW, and HLW is widely accepted. The degree of danger in NW for the environment depends, however, both on the amount and concentration of radioactivity and on such characteristics as toxicity, solubility in underground water, the half-life of radionuclides, and the probability of radioisotopes changing to a soluble state. For problems associated with underground disposal of NW it would be useful to create a classification of NW that takes into account all these factors. Unfortunately, such a classification was not established until recently. The classification inadequacy has not been very serious, however. In spite of the shortcomings of classifying NW solely by level of radioactivity, on the whole such classification does reflect the level of hazard of NW for the environment. The greatest concern is for HLW, which contains a complete set of long-lived, highly toxic, and highly soluble radionuclides.

Methods of safe disposal of LLW and MLW are now well developed. A great volume of LLW and MLW is associated with radiochemical manufacture, most of which is in liquid state. As an environment for LLW and MLW disposal the clay zones occurring near the surface are usually used. Trenches or quarries for waste are constructed and, before loading, the wastes are concentrated, then cemented or put into canisters. After loading,

the reservoir is covered by a concrete slab and filled with clay. Rarely, the waste canisters are placed into underground proving holes in salts or other rocks.

It should be noted that up to 1958 in the former USSR, the safe disposal of LLW and MLW was not given proper attention. These wastes were dumped and buried near the surface without warning signs. As a result, radioactive materials in soils and rock were sometimes dug up and used on building sites, public gardens, columbaria, and so on. Collection and disposal of LLW and MLW are strictly regulated now, and repositories are monitored.

In contrast to other countries, where liquid wastes are solidified, the processing of liquid waste in Russia is badly organized. At the enterprises where it is possible, liquid wastes are injected into water-bearing zones. A huge amount of liquid waste at "Mayak" is in open reservoirs. LLW from atomic-powered ships is dumped directly into the ocean; MLW is kept in overfilled tankers. It is evident that the most urgent problem in LLW and MLW management is organizing production to concentrate and solidify the wastes. As to the burial of solidified LLW and MLW, providing their secure isolation for 500–1000 years does not present a difficult challenge. The most complicated and, until now, unsettled problem is the safe disposal of HLW.

#### **Peculiarities of HLW that determine the conditions of its disposal**

In considering safe disposal conditions for HLW one must take into account that such wastes contain radioisotopes of both cesium-strontium and the transuranic group. The requirements for isolating the radionuclides of each group are substantially different; that makes separation of HLW highly desirable. As work in this direction proceeds, it is expedient to briefly define the differences in disposal requirements for radionuclides in each group.

Radioisotopes  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are exceptionally toxic; the maximum permissible concentrations of these elements in drinking water are  $2 \times 10^{-12}$  and  $2 \times 10^{-10}$  g/l, respectively. These values for  $^{234}\text{U}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ , and  $^{237}\text{Np}$  are, respectively,  $5 \times 10^{-6}$ ,  $1 \times 10^{-9}$ ,  $8 \times 10^{-8}$ , and  $4 \times 10^{-6}$  g/l. If one notes that  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are characterized by high solubility, it is increasingly obvious how hazardous the penetration is of these radionuclides into groundwater. In the HLW vitrified into aluminum-phosphate glass, cesium and strontium are the basic heat-producer elements. But compared to the transuranic elements, the half-lives of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are short (about 30 years). Therefore, the disposal of radionuclides in the cesium-strontium groups for a period of 1000 years guarantees complete decay. In the technologies accepted in many countries,

isolation of cesium-strontium for this period of time is ensured by corrosion-resistant containers. If HLW contained only cesium and strontium it would be enough for safe disposal to place them near the surface in clay zones in such containers. Even if the containers leak before 1000 years, penetration of radionuclides into the biosphere would be completely prevented by clay mineral sorption.

It is necessary to use a completely different approach in solving the problem of disposal for radionuclides in the transuranic group. The required period of their isolation is hundreds of thousands and even millions of years. Such time spans make even expensive engineering barriers insufficient. The principal means of isolating these radionuclides lies in the geological media.

One of the most important factors in appraising the fitness of geological media for repository siting is the prognosis of geological events over a long period of time. Such events should not permit outcropping of the HLW, destruction of the repository, unfavorable changing of hydrogeological features of the site, etc. It is evident that the nearer HLW is placed to the surface the higher the probability it will rise to the surface as a result of different events. Accordingly, it is desirable to dispose of HLW containing transuranic elements at a maximum depth within the most stable geological blocks.

The combined presence in HLW of radioisotopes of both cesium-strontium and the transuranic groups complicates of the problem HLW disposal. High heat generation can destroy the preservative matrix, fracture rocks and augment their permeability, reduce the sorptive properties of the buffer, and accelerate chemical reactions, including corrosion. It is obvious that the task of HLW disposal would be much easier if the radionuclides of different groups were separated. The reality, however, is that for at least a few dozen years we shall have to deal with HLW of varied composition.

### **Safe geological disposal of HLW**

Universally recognized for the safe disposal of HLW is the multibarrier concept of isolating waste [Bieniawski, 1990; "Underground Nuclear Waste Disposal," 1981; Chapman, McKinley, 1988; Krauskopf, 1988]. For consolidated HLW such barriers are a corrosion-resistant container, a preservative matrix, and a buffer consisting of subpermeable sorptive material. In spent-fuel disposal the waterproof barriers consist of corrosion-resistant material and concrete.

Authorities from many countries consider the container one of the most important barriers. Containers will prevent contact of radionuclides with underground water for 300–

1000 years. That amount of time is sufficient for lowering  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  concentrations to safe levels. The total radioactivity of HLW at that time will reach values comparable to uranium ore. Many studies have been published on containers and their corrosion by underground water. Materials recommended for containers are stainless steel, titanium, zirconium alloy, copper, corrosion-resistant cast iron, and others.

The containers with aluminum-phosphate glass at "Mayak" are made of common steel, with these dimensions: diameter—60 cm; height—80 cm; thickness of walls—3 cm. Three such containers are installed in canisters made of stainless steel with a diameter of 63 cm, a height of 340 cm, and thin walls. It is obvious that such containers and canisters are unable to secure the long-term separation of HLW from underground water. The second barrier is the preservative matrix, which because of its low solubility in underground water controls the spread of radionuclides. In most countries boron-silicate glass is the preservative matrix used. Investigations into creating mineral matrix materials of high isomorphic capacity are being conducted.

In Russia aluminum-phosphate glass is the preservative matrix used for HLW. Its shortcoming is its relatively high solubility in water ( $10^{-6}$  g glass/cm<sup>2</sup> in 24 hours at 25 °C and  $10^{-4}$  g/cm<sup>2</sup> at 45 °C) as well as its rapid crystallization above 200 °C. If the strontium transition out of glass is limited by the presence of a newly formed phase—goyazite ( $\text{SrAl}_3[\text{PO}_4]_2[\text{OH}]\text{H}_2\text{O}$ ), for example—then practically all cesium will pass into solution. These shortcomings of glass are stimulating active investigations into more reliable matrix materials. Nevertheless, a considerable portion of liquid HLW is still being consolidated into aluminum-phosphate glass, and thus investigations continue into the physical-chemical processes of HLW–rock–underground water systems with respect to aluminum-phosphate glasses.

The third barrier is the sorption material filling the space between the container and the walls of the cell (or concrete revetment) where HLW is placed. Bentonites and zeolites are usually considered candidates for such material. The results of radiogeochemical investigations of authors testifies that material from the weathering of the crusts of basic rocks has important advantages.

### **Selection criteria for geological environments of HLW disposal**

The requirements for geological disposal environments become obvious if one consistently considers the reasons for radionuclides entering the biosphere. Such reasons are:

- Denudation of the repository as a result of erosive processes.
- Migration of radionuclides from the repository to the biosphere by underground water.
- Capture of HLW by intrusion of magmatic melt and its removal to the surface.
- Penetration of the repository by boreholes and exploratory openings as a result of prospective geological drilling.

The probability of unfavorable events depends on numerous factors, such as climate, rate of denudation, frequency and intensity of neotectonical movement, petrophysical characteristics of rocks, presence of useful minerals, and other variables. Obviously, geological investigation for a repository is directed toward selecting a site capable of securing the minimum probability of unfavorable events.

The scale of the site investigation and many other elements will be determined by the ultimate aim. For example, if the proposed site is for building a regional repository for disposal of HLW from several enterprises, the investigation will include an evaluation of zones within the prospective territory. The foundation of this investigation is the analysis of available geological-geophysical data. The compiling of a specialized map with subdividing of territory by degree of fitness of the geological environment for repository building is the final product.

On the basis of this map and socio-economic characteristics (density of population, presence of mineral deposits, transportation routes for the HLW, etc.) sites are selected for more detailed geological evaluation. Unfortunately, the probability of building regional HLW repositories now is very low because of the unavoidable opposition of local inhabitants and authorities to waste disposals in their areas, the high cost of repository construction, and the hazards of HLW transportation.

The disposal of HLW closer to the location of enterprises that generate the waste seems much more realizable. The aim of geological investigation in such cases is to identify the presence of suitable sites within the bounds of the enterprise's region and to make an objective appraisal of them. It is obvious that the characteristics of any single recommended site will not always be optimal. Nevertheless, any site that is chosen must meet the basic prerequisites of repository safety. Whether the proposed site meets the prerequisites is determined by a model that characterizes radionuclide migration. The model must show that even in the case of unfavorable events the penetration of radionuclides into the biosphere at rates exceeding permissible limits will not occur.

The elements of the geological investigation for repository sites are given in the following paragraphs:

A **geomorphologic study** of the site, focused on a calculation of the long-term relief development prognosis, is the first step in the investigation. On the basis of geomorphologic criteria, a site can be accepted as suitable if the repository located there will not be exposed through denudation within at least 10,000 years and the probability is very high that such exposure will not happen within millions of years. During the geomorphologic study the possibility of global climate change that can intensify denudation is taken into account. The importance of this factor becomes particularly obvious if one remembers that only 10,000 years have elapsed since the last glacial period. A failure to meet geomorphologic criteria is the reason most mountainous regions are unfit for HLW disposal into adits, which can be denuded. The same can be said about nondeep levels of rising geological blocks. The combination of relief-formation and climate can create the basis for a prognosis of hydrochemical radionuclide migration from the repository to the biosphere.

The **seismic regime**—the frequency and intensity of neotectonic movement—is an important criterion in the siting of atomic plants and HLW repositories. The obvious shortcoming of regions with high seismic activity is the high probability of repository destruction during the building stage and operation, the probability of hydrodynamic conditions and water composition changing, and the low reliability of long-term prognoses of geological events. All of this is cause to consider regions of high seismic activity as undesirable for HLW repository building. An exception to these seismic considerations might be made in such countries as Japan, where stable blocks are absent. The seismic shortcomings of a site can be compensated for by engineering barriers, in particular durable and corrosion-resistance containers. But such methods increase the cost of a repository. For Russia, regions of high seismic activity have to be recognized as unfit for HLW disposal.

**Structural-geological investigations** attempt to identify geological blocks that are favorable in size, the tectonic structure of the blocks, and the composition of rocks in the blocks. At the early stage of the investigation analyses are made of available geological materials of different scales, geological-geophysical explorations are conducted, and a specialized map is compiled showing preliminary zonation—zones are defined by the degree of fitness of the geological environment within each zone for repository building. On the basis of this map, selection of prospective local plots for more detailed study can be made. The purpose of the structural-geological study of the plots is to gather representative

geological data for selecting the most suitable sites. Dimensions of favorable rock massifs, the intensity of tectonic disturbance, the degree of permeability of structural elements, the probability of the presence of mineral deposits in the zone, etc., are taken into account. The information about geological structure at a depth of 400–1200 m where the repository will be situated has special significance. Such information can be obtained from complex geophysical investigations and structural drilling. Basic graphical materials used in the investigation are specialized geological maps, scale 1:25000-1:10000, and series of sections.

**Geodynamical and structural-petrophysical appraisals** evaluate crystal rock massifs based upon old and recent analyses of the strained state of massif media, the nature and grade of rocks in deformational transformations, and the changing of their petrophysical parameters as a result of tectonic burden. The investigations select the blocks in which future tectonic phenomena will have a minimum effect.

The rocks are subdivided into three groups by petrophysic properties: elasticofragile (quartzite, granitoid, siliceous volcanite, crystalline schist); elasticoplastic (terrigenous carbonate rock mass, sericitic-chlorite slate); and viscostrong (gabbro, diabase, basalt) [Starostin, 1979, 1984]. Configurations where bodies of viscostrong rocks occur in surroundings of elasticoplastic rocks are favorable for repository building. Under different catastrophic deformations in the Earth's crust such viscostrong bodies will be protected from strong tectonic strains by the elasticoplastic rocks, which reduce breaking stress. On the whole, the basic rocks, by their petrophysical characteristics, are more favorable for HLW repositories than granites, gneisses, and others rocks of siliceous composition.

**Petrographical investigations** search for rocks that by their composition and textural-structural properties are favorable for HLW disposal. Since radionuclides are carried away from a repository to the biosphere mainly by underground water, rocks with a very low water content and in which water does not move attract the attention of investigators.

**Salt** falls into such a category. As an environment for NW disposal, salt was recommended in the U.S. as long ago as the mid-fifties ["The Disposal of Radioactive Waste on Land," 1957]. Salt in Germany is in essence the only medium in which LLW and MLW are disposed, and disposal of HLW is being considered in salt. The main advantage of salt is the absence of movement of underground water in the salt medium. Salt in mining and a depth of several hundred meters shows that the holes hold their shape for some years. Another advantage of salt is its high thermal conductivity. All conditions



being equal, the temperature in salt repositories will be lower than in repositories located in other rocks.

The shortcoming of salt is its high flowage rate, which increases with the heat generation of HLW. As a result, underground holes are eventually filled by salt. After such filling in a repository, HLW would become less accessible and its extraction for reprocessing or redispersing would be difficult to achieve. Reprocessing of HLW and its use in the future can prove to be economically attractive, especially for spent fuel containing substantial amounts of uranium and plutonium. Though as a whole salt undoubtedly makes a favorable environment for HLW repositories, blocks suitable in size and internal structure are not found very often. The danger of salt dissolution in groundwater and of brine penetration to the canisters requires a comprehensive consideration of action on rocks by mining and heating. The presence in salt of argillaceous beds and insufficient dimensions of the block limit the possibilities of using salt for HLW disposal. The problem of the influence of salt pile upon the environment has also been noted. All of these considerations force one to appraise the advantages of salt more conservatively than when it was first studied.

**Clays** are widely used for LLW and MLW disposal and in some countries disposal of HLW is planned in clay. The advantages of clays are low permeability and high sorptive properties as to the radionuclides. The shortcomings are low stability of excavations, high cost of mining works, and low thermal conductivity. At a temperature of more than 100 °C clay minerals undergo dehydration accompanied by loss of plasticity and sorptive properties, fracturing, and other negative consequences. Objective appraisals of the fitness of clays for HLW disposal can be made from investigations in underground laboratories and by comparing expenses of HLW repositories built in rocks of different composition.

**Shales** by their characteristics can be classed as the least suitable rocks for HLW repositories. Low durability and heat resistance, high permeability, and many other characteristics give shales a low rating in the opinion of all experts. It is evident that studies of blocks composed of shales can be justified only in countries that do not have more suitable geological environments.

**Tuffs** are classed as favorable rocks owing to relatively low permeability (accompanied by high porosity) and high sorption properties. Zeolitization peculiar to tuffs raises sorptive properties even more. The most favorable tuffs for mining excavations are welded tuffs, which are characterized by durability.

Extensive information on the insulating properties of tuffs is being gathered by American investigators at the Yucca Mountain site in Nevada where the possibility of a potential HLW repository is being investigated. The Yucca Mountain area is characterized by a very low groundwater level (a depth of 600 m). The repository will be located above the groundwater level in a zone free of water rocks. Such conditions practically exclude the interaction of HLW and groundwater and the migration of radionuclides to the biosphere. It is obvious that with respect to hydrogeologic conditions and native rocks the Yucca mountain region is unusually well-suited for HLW repositories. At the same time, the region's relatively high probability of earthquakes, magmatic activity, climate changes, and possible changes in groundwater levels partially offset its desirable qualities.

The U.S. is the only country where siting a repository in tuffs is permitted. There are many reasons for this, such as the small size of tuffs massifs, high heterogeneity, high water saturation, etc. The multiform mineral composition of tuffs, the size of fragments, porosity, permeability, sorption and other characteristics require very detailed investigations for selecting optimal sites, which increase the cost of prospecting work.

**Crystalline rocks.** In the foreign literature this term encompasses a wide number of rocks that are wholly composed of crystals. All holocrystalline magmatic rocks and all crystalline schists and gneisses fall into the category of crystalline rocks. Though salt, anhydride, and marble are holocrystalline rocks they are not included in this group. In Russian literature, the term "rocky" is usually used. All volcanic, plutonic, and metamorphic rocks are included in this term. We take the word "crystalline" to mean all magmatic and high-grade metamorphic rocks with the exception of volcanic glasses.

The merits of crystalline rocks are their high durability, tolerance to moderate temperatures, and good heat conduction. Proving holes in crystalline rocks keep their stability for almost an unlimited time. Underground waters in crystalline rocks usually have low concentrations of salts and a weakly alkaline reducing character that as a whole corresponds to conditions of minimum radionuclide solubility. In selecting a site for HLW disposal in crystalline massifs, the blocks with the highest durable properties and low jointing are recommended. The search for an exact place for a repository comes down to a selecting a block acceptable in size and low in permeability. Geological data demonstrate that blocks of crystalline rocks characterized by very low permeability and devoid of large fractures and sheeted zones do exist, but finding and delineating such blocks is rather difficult. The procedure of efforts that search for such blocks is reduced to mapping of faults, zones of jointing, crush zones, etc. The exploration of prospective sections in deep horizons is conducted by dense network well boring. Particularly careful work of this kind

was done in Sweden, where granites are under consideration as an environment for repository siting [Swedish Nuclear Fuel Supply Co., 1983].

It is strange that the influence of chemical composition on the insulating properties of geological media is relatively little discussed in the literature. At the same time, physical-chemical processes in the system of HLW, rock, and underground water can promote either an increase or a lowering of repository safety. HLW gives up enough heat to upset physical-chemical balances. For example, the temperature field in the vicinity of HLW canisters can affect water circulation and mineral formation. If the interaction is accompanied by a lowering of permeability and a rise in sorptive properties, the rocks can be considered favorable.

On the basis of petrologic investigations we can confidently contend that physical-chemical interactions will mostly influence hydration and carbonization reactions. It is important to underscore that mineral formation will take place mainly in the fractures and pores. Petrographical and experimental data also show that minerals are characterized by different relative stabilities with respect to the action of hot solutions. It is obvious that the rocks most favorable for repositories are those in which mineral-forming reactions are accompanied by the most intensive clogging of pores and fractures. Thermodynamic calculations and natural observations show that the more basic the rock the more it will meet these properties [Laverov et al., 1990, 1991; Omelianenko et al., 1993]. Thus, the hydration of dunite is accompanied by a 47% increase in volume due to newly formed, whereas the corresponding percentages for gabbro, diorite, and granodiorite are 16, 8, and 1% respectively. Therefore, the hydration of granite results in no significant clogging of fractures. Within the limits of temperature conditions in repositories the hydration reactions will be accompanied by the formation of such minerals as chlorite, serpentine, talc, hydromicas, montmorillonite, and various mixed-layered minerals. The high sorptive characteristics of these minerals will prevent the spread of radionuclides outside the repository.

Thus the insulating properties of basic rocks under the effect of HLW will increase, which allows one to consider these rocks as preferable for repository building [Laverov et al., 1991]. Peridotite, gabbro, basalts, basic crystalline schists, amphibolites, and basic alkaline rocks fall into this category.

**Hydrogeological and hydrogeochemical explorations** have a special significance because underground waters transport radionuclides from the repository into the biosphere. The factors determining the fitness of a geological environment in this regard are the intensity of water exchange, the direction and speed in which the

groundwater moves, the distance from the repository to the zone of discharge, and the composition and physical-chemical parameters of the water. Low alkaline reduction qualities correspond to the least solubility of radionuclides. For a repository site, favorable conditions are the absence of water exchange with the surface, or a very low speed of natural flow, and a long distance from the repository to the zone of discharge. Heat release can cause a change in the hydrodynamic regime. An impartial assessment of heat effects is possible in underground laboratories by creating temperature conditions that correspond to those in a repository.

**Metallogenic investigations** directed at appraising known mineral deposits and predicting potential deposits are an important part of repository site evaluation. Blocks are suitable for repository siting where mineral deposits are absent or their presence is unlikely. Such conditions not only avoid economic waste but also diminish the probability of repository violation by wells and exploratory openings in the future.

**An ecological prognosis** for the long term must establish the probability of radionuclides penetrating the biosphere. Modeling radionuclide migration relies on assembling a number of parameters. Some of these parameters have constant values (composition of rocks, radionuclide half-lives, their geochemical properties, etc.); others can change over time. In constructing a radionuclide migration model, it is important to account for the retardation coefficient, which depends on the ratio of water flow and the radionuclide's velocity. Knowing the distance to the groundwater discharge zone, the solubility of radionuclides, the water velocity, the retardation coefficient, and half-lives we can theoretically calculate the concentration of radionuclides in the water at the zone of discharge at any period of time. Data on the influence of temperature fields, the dispersion of water flow, and other characteristics are also taken into account. On the basis of such calculations, different alternatives of repository design, methods of disposal, and sites for repository construction can be evaluated and compared. If the radionuclide content in the water at the discharge zone within 10,000 years exceeds the ultimate permissible level, the conditions of the repository site are considered unacceptable.

The foregoing approach is based on an optimistic scenario that unfavorable events will not occur during the period of time under consideration. The probability of such events is derived from the sum total of geological data, such as seismic regime, geodynamical conditions, the history of geological development, etc. A consecutive accounting of the influence of each of these events and their overall interrelated effects allow one finally to appraise the level of repository safety. The results of computer simulations of radionuclide migration form the basis for an acceptance decision on repository construction.

### **The use of natural mineral mixtures as sorptive geochemical barriers**

In most of the designs for sorptive barriers it is anticipated that the bentonites with an admixture of quartz sand will be used. Sand is used for increasing thermal conductivity. An investigation of the literature showed, however, that material weathered from the crust of basic rocks is preferable. This material contains montmorillonite as well as chlorite, serpentine, various mixed-layered minerals, hydroxide of iron, titanium, manganese, etc. The diversity of minerals with their varied properties enhances the mixture to sorb a wide range of radionuclides. The cost of these materials is very low since they are not used for other purposes. Besides the sorption of radionuclides in the mix, the buffers and backfills, thanks to low permeability, prevent contact between the HLW and underground waters. The water-saturated material of buffer serves as a medium through which the migration of radionuclides takes place by means of diffusion. This mechanism promotes maximum utilization of buffer sorptive properties. With the buffers and backfills we can create conditions that prevent radionuclide migration. In the general case, a reductive weakly alkaline medium corresponds to such conditions. The lowering of reduction potential can be done inexpensively by adding coaly shale to the buffer and backfill. In the early stages of a repository's life the most hazardous radionuclides for the environment will be  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , which provide the cardinal contribution to the level of radioactivity and are characterized by high solubility in groundwater. The task of the expensive corrosion-resistant containers used for HLW disposal in foreign technologies is to isolate the radionuclides from underground water until the concentration of these elements reaches a safe level. As our investigation shows, this task can be completely achieved at a much lower cost by using sorptive materials. Such is the effectiveness of the crust of weathering products. The equilibrium concentrations of Sr and Cs in these products are a thousand times greater than in water solutions.

In some cases the enrichment of natural materials by one or another mineral phase can be useful.

### **The conceptual foundation of investigations into geological strategies for providing safe NW disposal**

The investigations of geological approaches to NW disposal bring practical results when they take into account the real socio-economic situation in Russia, the quantity and form of NW, the prospect of further accumulation of NW, and the NW storage conditions that now prevail. One difficulty in any discussion of the conceptual foundation of NW disposal is a lack of NW management law. The problem of liquid NW disposal is the most complicated. In the first draft of NW law a prohibition of liquid NW disposal was

envisaged. It is the authors' opinion that the prohibition is a reality only for NW containing transuranic elements. As to the MLW and LLW, their safe disposal into deep water-bearing horizons can be completely secured. The experience of liquid waste repositories is in full accord with this point of view.

The authors offer their conceptual foundation based on the following general propositions:

- The top priority for securing radioecological safety in Russia is moving NW out of the biosphere and safely isolating it for the required period of time.
- The most realistic method of NW disposal is localizing it deep in the earth.
- Fundamental factors in solving the NW disposal problem are a guarantee of safety and economic effectiveness.
- HLW must be disposed of only in a solidified state.
- Securing radioecological safety and high economic effectiveness for liquid MLW and LLW disposal can be achieved by pumping such wastes into water-bearing horizons containing stagnant waters unfit for any other use.

The factors that determine the investigation of tasks and their order of priority are:

- The presence in Russia of an enormous amount of NW.
- The difficult economic status of the country.
- The lack of conditions for safe HLW transportation and the lack of remote operating equipment for performing work in underground openings.
- The high probability of local inhabitants reacting against NW repository siting outside the nuclear enterprise areas.

Therefore, the most important task of geological exploration in the near future is the selection of optimum geological conditions for safe HLW disposal in the areas of nuclear enterprises. The most realistic way of implementing this decision is using well repositories. The construction of such repositories does not need considerable capital investment and would allow HLW disposal to begin relatively soon. Comparatively small blocks of favorable rocks can be used for building these repositories.

In arranging geological investigations it is necessary to take into account as well the tasks that are oriented toward a remote perspective. These tasks are determined by the following considerations:

- The inevitability of subsequent development of the nuclear power industry.
- The reality of introducing in Russia safe HLW transportation and remote operating technology, already in use by Western countries, for regional repositories.
- The wide variety of geological conditions in Russia that allow one to select optimum sites for repositories.
- The necessity for creating a highly productive, ecologically safe, and economically effective HLW disposal industry.

In connection with the foregoing, it is necessary to produce a scientific manual that establishes methods for selecting suitable geological environments for HLW disposal. It is also necessary to identify the prospective sites on Russian territory for regional HLW repositories.

The authors' basic concept of HLW disposal is to abandon expensive corrosion-resistant containers and use as basic barriers the geological environment and sorptive mineral mixtures. The geological investigations we have presented support decisions in line with this concept.

The complex geological investigations considered above allow one to determine the insulating properties of geological media at the present time as well as factors that can change those properties in the future.

### **Conclusion**

1. In solving the problem of safe NW disposal in Russia one must take into account the very difficult economic situation and the existence of a great volume of accumulated NW. The most efficient method of NW disposal consists in the maximum use of the protective capacities of the geological environment as well as in natural inexpensive minerals for engineered barrier construction.
2. The selection of geological environments with high insulating characteristics is achieved through the following:
  - Geological-structural, petrographical, petrophysical, geodynamical, and geophysical research that is directed to a selection of geological blocks characterized by high tectonic stability, low hydraulic permeability, and high mechanical tolerance.
  - Petrologic investigations for selecting rocks that on reaction with groundwater at high temperature increase their isolating properties.

- Hydrogeological and hydrochemical research that is aimed at selecting sites where the composition of water, intensity of water exchange, speed of flow, and distance to zone of discharge rule out the penetration of radionuclides to the biosphere.
  - Simulation of the processes of radionuclide migration in geological media for developing a long-term prognosis of ecological systems.
  - Experimental investigations of interactions in the HLW–rock–groundwater system and obtaining characteristics for simulation.
3. The immediate practical task of geological investigation is selecting sites for HLW disposal in deep wells in the regions where they are now temporarily stored. The long-term aim of these investigations is the development of geological prerequisites for creating regional HLW repositories. The topical task of Russian geologists is to work out a methodical scientific guide defining the geological aspects of the NW disposal problem.

## ACKNOWLEDGMENT

This paper was prepared under the auspices of Russian-American Center for Contaminant Transport Studies at the Lawrence Berkeley Laboratory. We appreciate the funding support from Department of Energy, Office of Environmental Management, Office of Technology Development (DOE/EM-OTD) and the Department of Energy, Office of Energy Research, Office of Basic Energy Sciences (DOE/ER-BES) under Contract Number DC-AC03-76SF00098. Discussions and encouragement from Chin-Fu Tsang, Sally Benson, Irag Javandel, Joe Wang and John Apps are gratefully acknowledged. We also thank John A. Apps and Harold Wollenberg for their review, comments, and editorial improvements.



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