Conf-820154--1

UCRL- 86582
PREPRINT

UCRL--86582

DE82 020176

Nuclear-Waste Disposal in Geologic Repositories

MASTER

D. Isherwood

Society of Civil Engineers
"The Geotechnics of
Waste Management"
Philadelphia, PA
January 19, 1982

August 2, 1982

Lawrence Livermore National Laboratory

NUCLEAR-WASTE DISPOSAL IN GEOLOGIC REPOSITORIES*

Dana Isherwood
Lawrence Livermore National Laboratory
Livermore, CA 94550

SUMMARY

Deep geologic repositories are being widely studied as the most favored method of disposal of nuclear waste. Scientists search for repository sites in salt, basalt, tuff and granite that are geologically and hydrologically suitable. The systematic evaluation of the safety and reliability of deep geologic disposal centers around the concept of interacting multiple barriers. The simplest element to describe of the geologic barrier is the physical isolation of the waste in a remote region at some depth within the rock unit. Of greater complexity is the hydrologic barrier which is determined by the waste dilution factors and groundwater flow rates. The least understood is the geochemical barrier, identified as a series of waste/water/rock interactions involving sorption, membrane filtration, precipitation and complexing. In addition to the natural barriers are the engineered barriers, which include the waste form and waste package. The relative effectiveness of these barriers to provide long-term isolation of nuclear waste from the human environment is being assessed through the use of analytical and numerical models. The data used in the models is generally adequate for parameter sensitivity studies which bound the uncertainties in the release and transport predictions; however, much of the data comes from laboratory testing, and the problem of correlating laboratory and field measurements has not been resolved. Although safety assessments based on generic sites have been useful in the past for developing site selection criteria, site-specific studies are needed to judge the suitability of a particular host rock and its environment.

*This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-Eng-48.

INTRODUCTION

The question "what I'll do with the nuclear waste?" is one the government and industry has been asking since the atomic age began in the 1940's. We are concerned with both the high level waste generated by commercial nuclear power plants and the nuclear waste generated by the military and referred to as "defense waste". Both types of waste require the long-term isolation provided by a deep geologic repository, but in this report we will consider only commercial high level waste.

Figure 1 outlines the fuel cycle. The front end of the cycle--that is the part of the cycle up to the reactor--begins with the mining of the uranium ore. This is followed by the milling, processing and fabrication of the fuel rods that go to the reactor. After the rods are used in the reactor they are stored at the reactor site in cooling pools. This temporary storage of fuel begins what is called the back end of the cycle. At the present time the cycle remains open. We are "stuck" at the reactor. The holding pools for the spent fuel are rapidly filling. For the present the storage capacity of the pools is adequate. In the long term we must solve the problem of disposal or further development of nuclear power must end. In California, for example, state law says that until nuclear waste disposal techniques are proven, no new nuclear power plants can be built. For those against nuclear power, delays in solving the waste problem are useful to their cause.

For several reasons nuclear waste is not just a scientific or engineering problem, it is also a highly political one. One of the problems has been deciding just what to dispose of in the geologic repository. Should we dispose of spent fuel directly, thus essentially throwing away an important uranium resource--spent fuel is 95% uranium oxide. Or should we process the spent fuel, store the plutonium and other radioactive waste products and use the uranium to make new fuel. Reprocessing spent fuel frees plutonium which can be used for nuclear weapons, thus unleashing political concern. Back in the days of the Carter Administration, President Carter, in an effort to control the proliferation of nuclear weapons,

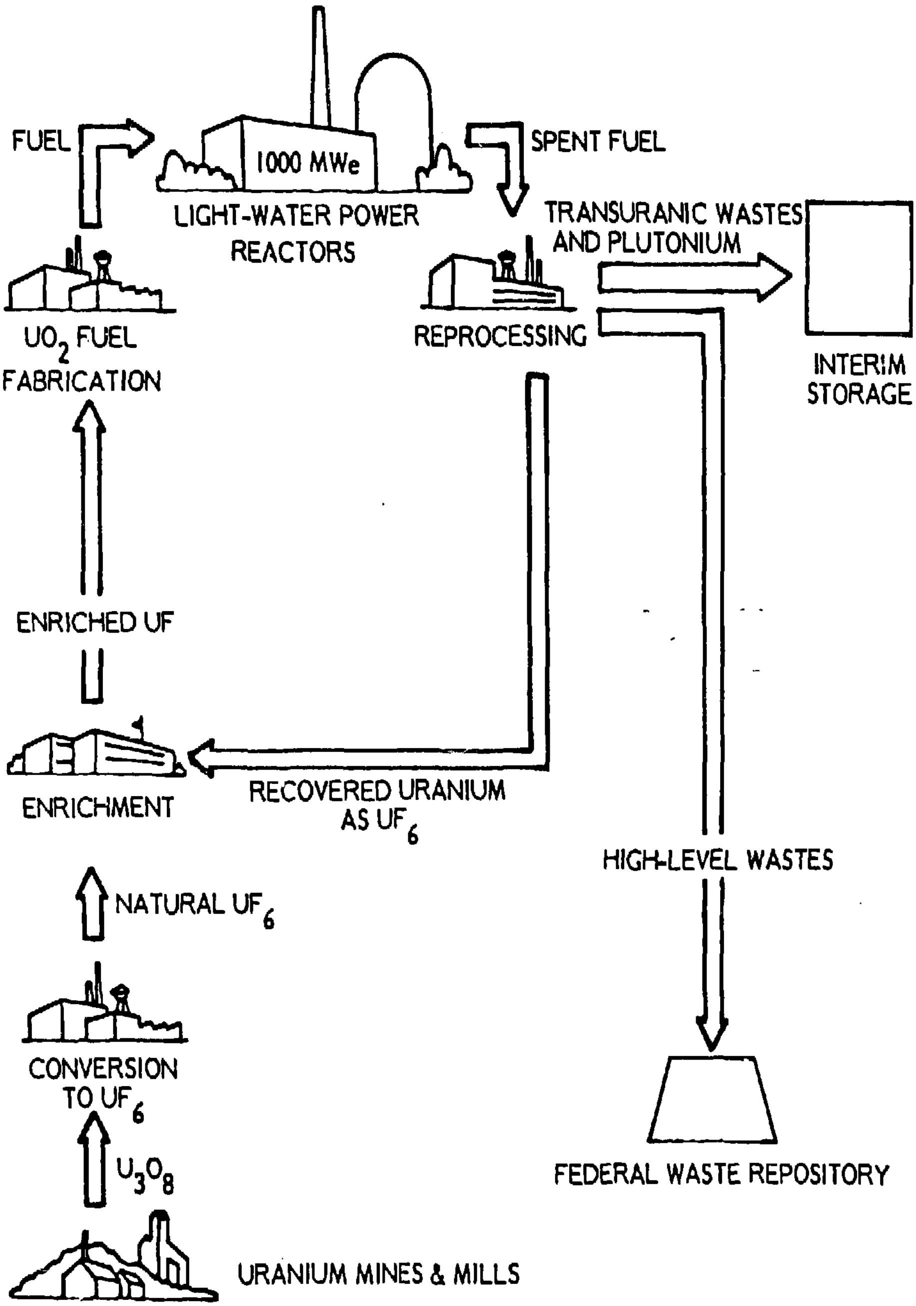

Figure 1. Nuclear fuel cycle.

decided that spent fuel would be disposed of directly. The rest of the world ignored Carter's concerns. Uranium resources are limited and the other countries simply couldn't afford the luxury of throwing away their precious supply. Today, President Reagan has reversed Carter's decision and the search for a suitable processed waste form is in progress. So, in answer to "what is nuclear waste?", the politicians have decided and probably will again.

We do know that no matter what the waste form is, its radioactivity will be around for a long time. Figure 2 illustrates the potential hazard of various radioactive components of spent fuel versus time. The units on the vertical scale are less important than the trend in the amount of radioactivity (i.e., hazard). Note that for the first 1000 years the waste hazard drops dramatically due to the rapid decay of the fission products, primarily cesium and strontium. For the next 500,000 years the hazard remains relatively constant, eventually dropping off. At about one million years the hazard approximates that of uranium ore, a level of hazard the public has learned to live with and might accept. Obviously nothing man can engineer is likely to last that long, thus the emphasis on deep geologic disposal.

ALTERNATIVE METHODS

Deep geologic disposal is not the only method considered at one time or another. Alternative methods include disposal in the seabed, polar ice caps and outerspace. It was suggested that we change the radioactive waste into a less hazardous form by transmutation. It was also suggested that we inject the waste into deep holes drilled into the ground where it will melt the rock by the heat given off by the waste and form an insoluble residue. Of all of these, only disposal in the seabed is actively being considered today.

For countries like Japan that are land poor, using the sea to solve the problem of nuclear waste holds promise. The results of research in the U.S. suggest that disposal in the seabed is plausible. The biggest problem will not be the development of the disposal method, but rather the public's

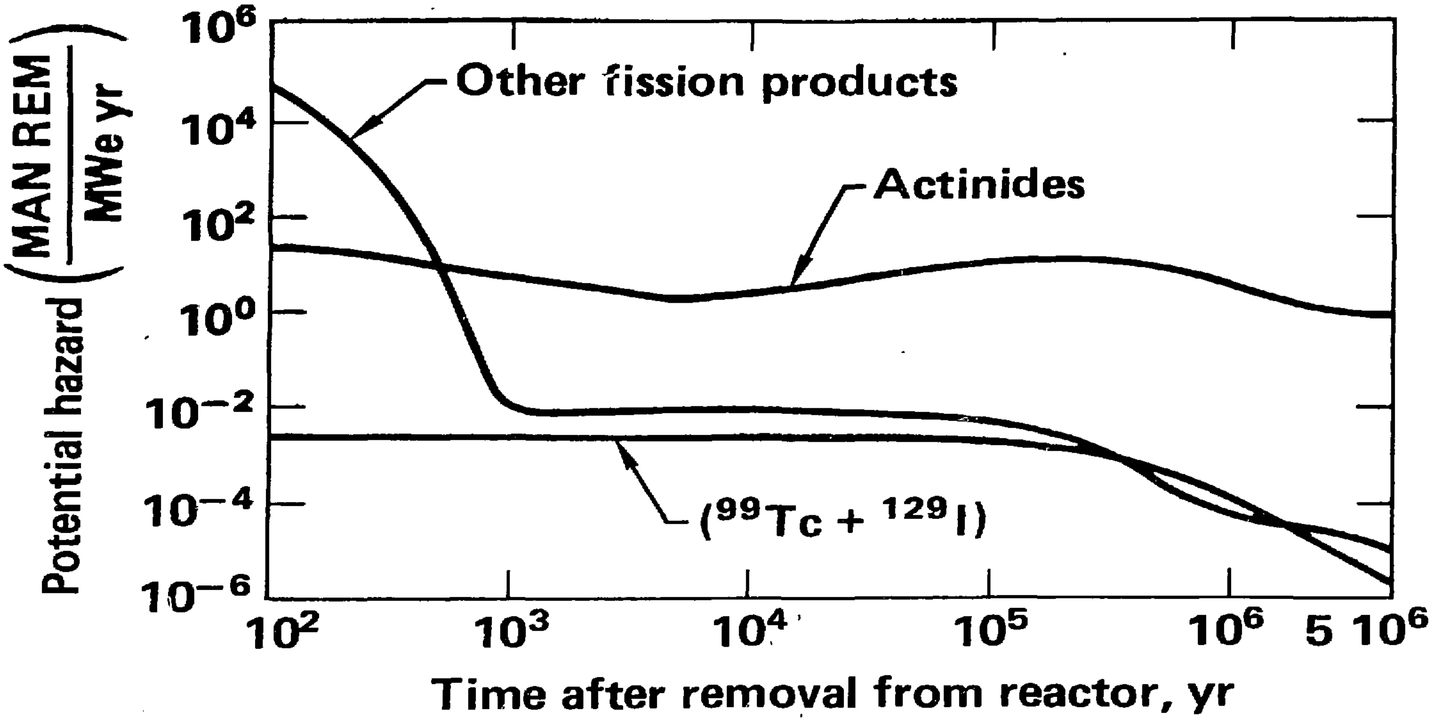

Figure 2. Potential hazard of the various radioactive components of spent fuel as a function of time.

perception of damage to the environment. The law of the sea, as used today, has difficulty dealing with fishing rights, let alone who can put nuclear waste where. In comparison, geologic disposal looks less threatening.

REPOSITORY SCHEDULE

When will we have a site? Our government has worked sporadically on the concept of deep geologic disposal since the 1950's. One recent timetable came from the Interagency Review Group (IRG). Their report, written for President Carter by a group of representatives from various government agencies, reflected a cautious approach. They recommended that 1) the choice of a site will not be made before 1984, 2) construction could be completed by 1992, and 3) initial operation of the site would be from 1992 to 1995. Since the IRG report was released, the National Waste Terminal Storage Program has developed a revised schedule. They are planning to 1) evaluate three specific sites, in basalt, tuff and salt by 1984, 2) select one of the sites in 1986, 3) begin construction in 1992, and 4) start up the repository sometime between 1998 and 2001.

DEEP GEOLOGIC DISPOSAL

What is meant by deep geologic disposal? As currently conceived, shafts will be constructed to depths of 1500 feet or more in a stable rock formation. Rooms will be mined out and the nuclear waste will be stored in canisters in holes in the room floor. Figure 3 is an artists's conception of Lawrence Livermore National Laboratory's Spent Fuel Test at the Nevada Test Site. The actual repository will be more complicated than what is shown in Figure 3, but the basic concept is the same. This test evaluates the feasibility of safe and reliable short-term storage of spent fuel assemblies at a plausible repository depth in granitic rock and retrievability of the fuel. This is one of several projects designed to study the basic concept of deep geologic disposal of nuclear waste.

Deep geologic disposal is based on the multiple-barrier concept. Each component of the system will form a barrier to radionuclide release to the environment. If one of the barriers is breached for any reason, the other

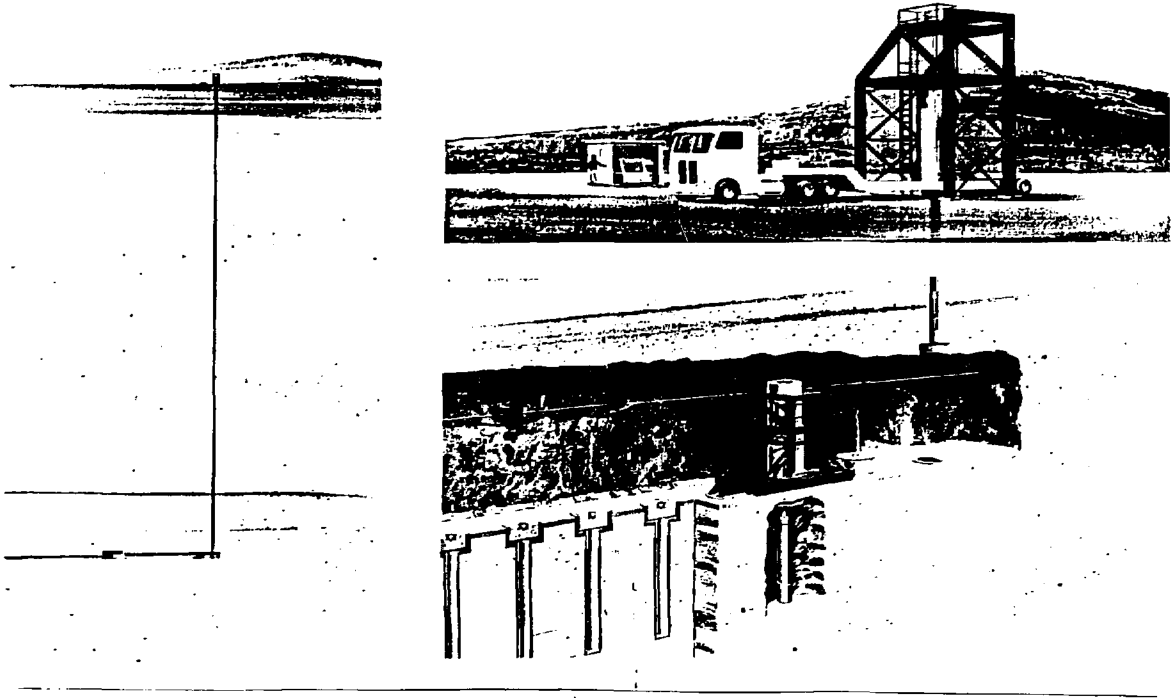

Figure 3. An artist's conception of Lawrence Livermore National Laboratory's Spent Fuel Test at the Climax granite, Nevada Test Site is similiar to the basic concept of a nuclear waste repository.

barriers will still be effective. The barriers are the waste form, the container around the waste form (i.e., the canister), the fill material around the container, and finally, the ultimate barrier, the geologic formation.

WASTE FORM BARRIER

First, let us consider the waste form. It will serve as a barrier if the waste form is stable relative to the environment, non-reactive and insoluble. In addition, federal regulations say it must be a solid. The waste forms investigated up to now include the spent fuel mentioned earlier, plus a number of reprocessed forms that incorporate the liquid waste from the reprocessed spent fuel: 1) Calcine, a powder is formed by evaporating the high level liquid waste. There are serious problems with its relatively high solubility, as well as the risk of handling and transporting a powder. 2) Borosilicate glass is formed by adding a glass powder to the liquid waste. The resultant liquid melt is allowed to solidify. Its main disadvantages are thermal instability and a tendency to devitrify. Borosilicate glass has already been chosen for reprocessed defense waste and is a strong candidate for commercial waste. 3) Super-calcine, a ceramic, is made by adding silicon, calcium, aluminium and strontium oxides to the liquid waste before calcining and hot pressing into pellets. An assemblage of crystalline phases is formed with greater stability than glass. 4) Super-calcine is superseded by Synrock, the newest waste form concept. To make Synrock, the liquid reprocessed waste is mixed with a carefully tailored mixture of components that, when cooled, forms a number of stable natural minerals, such as rutile, zirconolite, hollandite and others. Approximately 50% of the liquid waste can be incorporated into the mineral structures which is comparable to the borosilicate glass.

CANISTER BARRIER

The waste form will be placed in a canister. Suggestions regarding the material out of which the canister will be made include pure copper (a plan, now rejected, that would have taken 5% of the world's supply) and titanium (an expensive idea from the Swedish waste management program). The most likely material will be a stainless steel alloy or cast iron. The problem that must be addressed is the proposed requirement of the U.S. Nuclear Regulatory Commission that the canister must last for 1000 years. Corrosion testing is an important part of the evaluation of the various materials. The length of time a canister will remain intact will be highly dependent on its environment (i.e., rock chemistry, ground-water composition, heat of the waste, etc.)

BACKFILL BARRIER

The next barrier is the backfill material around the canister. The most often discussed material is a sand-bentonite mixture chosen for its high ion exchange capacity and physical characteristics. Various other clays and crushed rock from the mining operations have also been suggested. If ground water does reach the waste and both the canister and the waste dissolves, the radioactive species in solution could be adsorbed by the backfill material.

GEOLOGIC BARRIER

The final barrier is the geologic barrier which can be divided into three main interacting sub-barriers, geometric, hydrologic and geochemical. The geometric barrier defines the physical isolation of the waste - for example, the depth of the repository, the strength of the rock, and the total distance to man's environment defined as a river, a town, a farm, etc. The hydrologic barrier includes all those parameters which effect the ground-water flow rate: permeability of the rock, pressure gradients, dispersion, porosity, etc. The geochemical barrier includes all those chemical interactions that will determine if and in what chemical form the waste will enter and be transported by the ground water. These include reactions involving precipitation, solution, ion exchange and ion filtration. For example, if ground water reaches the waste and the waste dissolves, the radionuclides will be released into solution. One or more reactions might take place. Either the chemical species, say a strontium ion, might precipitate as a solid strontium carbonate, or the strontium ion might stay in solution and be adsorbed onto a mineral surface (i.e., ion exchange) thus retarding its movement away from the repository. The chemistry of the ground water and the surrounding rock will determine what chemical reactions will take place.

Several different rock types are being considered for a repository host rock.

Crystalline Rocks. Both basalt and tuff are now being evaluated. In Yucca Mountain at the Nevada Test Site, deep exploratory holes are being drilled into the tuff beds to determine the best location for a candidate tuff repository. In the basalt near Hanford, Washington, a similar search is also underway. A granite site will be evaluated only after the site of the first repository is selected.

Salt. Historically salt was the first choice for a nuclear waste repository. Back in the late 1950's the National Academy of Sciences recommended salt. It was chosen for its dryness and its plastic quality under stress. Once the repository was sealed it was believed that the salt would flow around the waste, trapping it forever. A site was chosen at Lyons, Kansas initially for research purposes, but it was later declared a prime candidate for a commercial waste repository. The site was eventually abandoned, the victim of politics and geology. Not only was there strong public opposition, but the area had been extensively mined for salt, there were many petroleum exploration holes through the salt, and the salt was not as dry as had been originally thought. Today, salt is sti a prime contender. Studies are ongoing in the Paradox Basin and the salt domes of the southern U.S.

The site most often in the news is in the salt beds of the Permian Basin near Carlsbad, NM. This site is referred to as the WIPP site (Waste Isolation Pilot Plant) and is designed to handle defense waste only. Evaluation of this site is well along by the Department of Energy and Sandia National Laboratory.

LICENSING

Licensing of a nuclear waste repository will require the cooperation of both the Department of Energy (DOE) and the Nuclear Regulatory Commission (NRC). The NRC takes the role of the regulator responsible for licensing and monitoring repositories in a position similar to the one it has for nuclear power plants. NRC regulates commercial waste, but not defense waste, although there have been periodic moves in Congress to place defense waste under NRC as well. The Department of Energy is the regulatee and must have NRC's permission to build a repository. Figure 4 outlines the various steps that DOE and NRC must take prior to the operation of a waste repository. NRC has written the regulations. They will be formally released this year. DOE, in the meantime, is evaluating sites. Once a site has been selected, a license application will be filed with NRC.

To write the regulations NRC needed to know what was important--what criteria to emphasize in the regulations. For example, should the depth of the repository be a prime consideration? How far from a city does a repository need to be to protect the public from an accidental release of radioactivity? The best way to decide these parameters would be for NRC to study the history of an existing repository. Since there are none, the next best approach is to model one. Figure 5 illustrates a generic repository in salt. This simple model involves a six-layered system and is one of many that have been used by NRC. In the model the repository is in a salt layer, with shale beds above and below and sandstone beds above and below the shale. Computer models that incorporate the hydrology, geology and geochemistry of the repository site are used to simulate different scenarios for release of the waste. In one simulation, the bottom sandstone bed is an artesian aquifer, meaning that flow is upwards through the repository into the upper sandstone bed and then along the sandstone to the river.

We can study the importance of the various parameters by changing the values assigned to them in the computer simulations. For example, we can change the permeability of the rock layers to study the effect of permeability on radionuclide release. We can also hold permeability

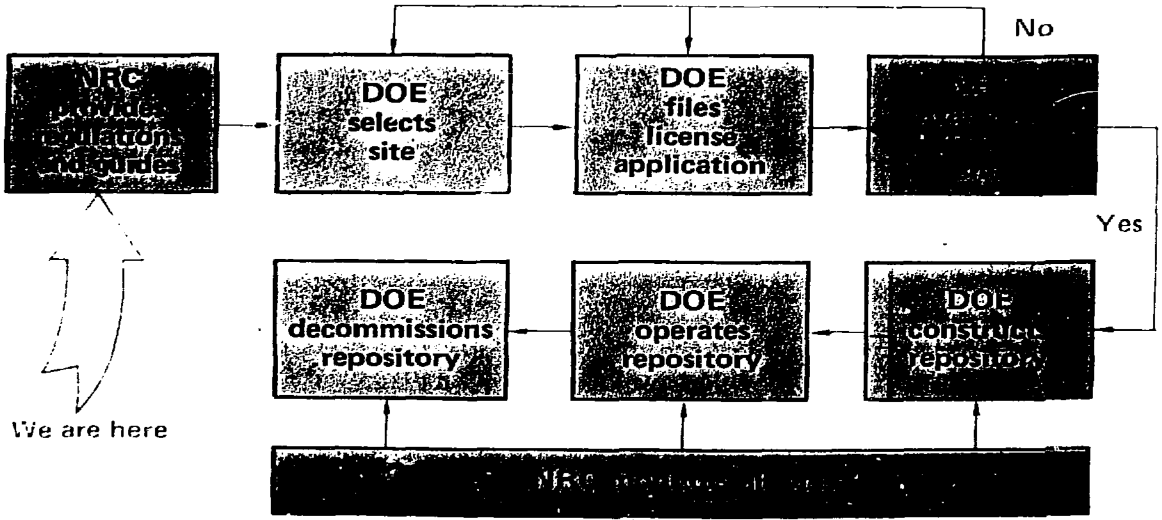

Figure 4. Licensing procedure and relationships between DOE and NRC for a nuclear waste repository.

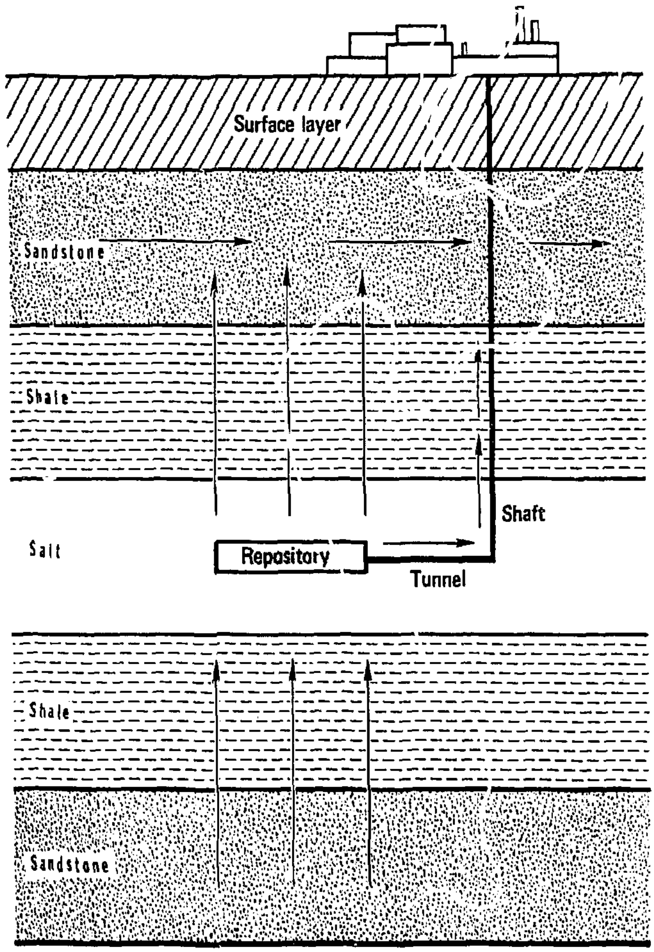

Figure 5. Simple physical model illustrating a generic repository in salt is used to evaluate the relative importance of the various parameters controlling radionuclide migration.

constant and vary pressure gradients. The results tell us whether permeability is important relative to pressure gradients. In another simulation we might change the chemical characteristics of the sandstone and see whether the resultant chemical reactions between the rock and the dissolved waste will significantly change the rate of movement of the waste. This will tell us the importance of choosing a repository site with slow ground-water flow rates over one which is chemically very reactive. To make these simulations we must, of course, assume that processes will occur that in a real repository we would not want to occur. Once a repository site is chosen, NRC and DOE will design computer models to simulate various scenarios for that specific site. In the meantime, the modeling of hypothetical or generic sites has allowed NRC to predict what aspects of a repository site are critical to its isolation and to write regulations addressing those aspects.

The predictive capabilities of the computer models are useful to the regulatory process, but remember that the models are only as good as the values we use in them. There are uncertainties in the parameter values used in the model simulations that need to be addressed. For example, how well can we measure dispersion in ground water? Do the laboratory measurements of chemical reactions really measure the type and extent of those that take place in the field? There are uncertainties created by the use of models. The physical model I used to illustrate a point was for a porous flow, but in some rocks fracture flow will dominate. Can a porous flow model adequately describe fracture flow? The fracture flow models are being developed. We are at the state-of-the-art in modeling. If it is necessary to predict the behavior of a repository for 100,000 years we must improve our modeling capability.

INTERNATIONAL ASPECTS

Nuclear waste is an international problem. Other countries with advanced programs in the disposal of nuclear waste are: Canada, Sweden, France, United Kingdom, Switzerland and Germany. These nations are conducting nuclear waste programs similar to the U.S. program--researching geologic disposal methods, rock types, potential sites, and package and repository designs. While the U.S. has put granite as the last rock type to be evaluated, all of these countries, with the exception of Germany, have chosen granite as their first choice. Although granite locations are widespread in the U.S., many are either in seismically active regions (e.g., Sierra Nevada, California) or in remote regions far from the centers of nuclear power (e.g., Maine, North Dakota, etc.). Politics also plays a part as seen in the local public opposition to studying granite in Wisconsin.

In Canada researchers are studying sites on the Canadian shield and have designed an underground research laboratory to study the characteristics of granite at depth in Manitoba. The English are developing an underground facility in Cornwall as well as other sites. The French have used boreholes to study radionuclide migration in two locations in France. The locations have not been identified due to political concerns and access problems that might occur if the nature of their research was known. In Sweden there is a major underground research facility at Stripa, an old iron mine in granite. Supporting laboratory work is ongoing at all their major universities. In Switzerland an underground laboratory is being developed in an existing pumped storage facility in the Alps. A repository site in northern Switzerland is being evaluated. In Germany the primary emphasis is on salt, with granite as their second choice. An underground repository for low and intermediate waste was constructed in an old salt mine outside the city of Brausweig. No high level waste is stored there, however, research related to the disposal of commercial waste is being conducted.

The U.S. has a strong commitment to cooperative international programs. Since most foreign waste management activities complement or parallel work in the U.S., cooperation between countries provides for a more efficient use of technical expertise, resources and funds.

DISCLAIMER