DISPOSAL OF RADIOACTIVE WASTES IN GEOLOGIC FORMATIONS*

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Abstract—The ultimate disposal of radioactive wastes into geologic formations has received increasing attention during the past 10 years. Due to the varying heat-generation rates of the different categories of wastes and the allowable costs, no single formation is capable of handling the entire spectrum of wastes produced. Therefore, three different methods of disposal have been investigated to handle the high-, intermediate-, and low-level wastes generated. High-level, small-volume wastes would be converted to solids and stored in a dry impermeable underground formation such as salt; intermediate-level, intermediate-volume wastes would be slurried with cement and additives and injected into slightly permeable formations using the hydrofracturing technique; and low-level, large-volume wastes would be injected into deep porous formations. The engineering-scale demonstration of the hydrofracturing concept has been completed, and a demonstration of the disposal of radioactive solids in a salt mine is underway.

A total of seven injections at depths from 800 to 1000 ft have been made at the Oak Ridge National Laboratory using the hydrofracturing technique. These injections have shown that horizontal conformable fractures are possible and have contributed valuable operating experience. In addition, a number of mixes have been developed and tested and are capable of satisfactorily retaining fission product waste.

Project SALT VAULT is a demonstration of the disposal of solidified high-level wastes in an out-of-service mine in Lyons, Kansas. In November 1965, fourteen Engineering Test Reactor fuel assemblies, containing 1,000,000 Ci of fission products, were placed in the mine floor. During the course of the 2-year test, four sets of fuel assemblies will be used to achieve a peak dose to the salt of about 8×10^8 rad and to obtain experience with the waste-handling equipment. The operation has proceeded smoothly, and most of the experimental objectives have been already achieved.

Laboratory studies on the flow path of radionuclides through a sandstone block have been carried out. However, it appears that a field scale demonstration of the deep well technique is some years away.

INTRODUCTION

In September 1955, the Atomic Energy Commission requested the Earth Sciences Division of the National Academy of Sciences, National Research Council to organize a meeting of geologists and engineers to discuss the possibilities of permanent disposal of radioactive wastes in geologic formations. At this meeting,[1] the disposal of high-level wastes in salt was recommended as having the greatest present potential. Disposal of diluted high-level wastes into deep porous formations was suggested as a possible method for the future. In subsequent discussions with representatives of the petroleum industry, it was agreed that it might be possible to dispose of radioactive wastes by pumping them into formations using hydraulic fracturing techniques. Research and development, leading up to demonstration experiments using radioactive materials, have been carried out on the disposal of high-level radioactive solids in salt formations and the disposal of intermediate-level liquid wastes by hydraulic fracturing. To date, no field demonstration has been performed

* Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

on injection of low-level liquid wastes into deep permeable formations.

DISPOSAL IN SALT FORMATIONS

Salt was suggested as a possible medium for high-level waste disposal because of its availability, geographic distribution, thermal conductivity, plasticity, impermeability, and low cost of mining.[2-5] Initially, studies on the direct disposal of liquid radioactive wastes in salt were performed, but in 1961 the liquid waste studies were terminated and solid waste studies were initiated. The solid waste to be stored in salt are those produced by the solidification or calcination of first-cycle wastes from reactor fuel reprocessing. In the United States a Waste Solidification Pilot Plant is currently under construction to demonstrate the potential methods for achieving the desired solidification. The solids produced are highly radioactive and generate significant amounts of heat.

There are two ways in which the waste containers can be stored in the mine; above the mine floor in racks, or below the floor in drilled holes. For storage above the mine floor, cooling can be obtained by convection but all handling operations would have to be performed remotely. Storage in the mine floor allows access to the rooms for transfer operations, but requires drilling of individual holes for each waste container. After detailed study, storage in the mine floor with heat dissipation by conduction through the salt adjacent to the containers was deemed most suitable. A computer program was written to determine the optimum spacing of waste containers to dissipate the heat in the salt and prevent overheating of the waste cans or the salt.[6] The computer results were checked by field experiments using electrical heaters and the calculated results agreed favorably with the experimental observations.

Allowable salt temperature rises are limited by the shattering of the salt due to increased vapor pressure in small bubbles containing brine ("negative crystals") located within the salt, and by an increased rate of plastic flow of the salt. Laboratory and field studies have shown that the problems of salt shattering and plastic flow can be minimized if the salt temperature is not allowed to exceed 200°C.

As a result of the theoretical calculations and experimental studies it appeared that it would be possible to dispose of high-level solid wastes in a salt mine. In order to determine the equipment and handling operations necessary in an actual disposal operation and the most economical design of the disposal facility, a full-scale field demonstration was needed. This demonstration, called Project Salt Vault, is currently in operation in a salt mine in Lyons, Kansas.[7]

The engineering and scientific objectives of Project Salt Vault are: (1) demonstration of waste-handling equipment and operating techniques; (2) determination of the possible production and release of radiolytically produced chlorine; (3) determination of the possible gross effects of radiation (up to 10^9 rad) on the uplift and salt-shattering temperatures in an area where salt temperatures are in the range of 100–200°C; and (4) collection of information on creep and plastic flow of salt at elevated temperatures which can be used later in the design of actual disposal facilities.

The Lyons mine was operated for a number of years before it was placed in an inactive status in 1948. In the existing mine space the marketable salt has been mined leaving the more impure salt in the floor and ceiling. Since the impure salt remaining in the mined out areas contains significant moisture bearing shale impurities, it was necessary to mine a new experimental area above the existing mine floor such that the fuel assemply canisters would be located in pure salt. Excavation of the new experimental area involved the mining of 19,000 tons of salt. The new mine level, which is about 14 ft above the old mine floor, is connected by a ramp having a 10% grade.

A schematic cross section of Project Salt Vault is shown in Fig. 1. The waste solids, after canning in Idaho, are loaded into a carrier and shipped on a specially designed truck trailer to Lyons, Kansas. At Lyons the carrier is removed from the trailer and placed vertically over a steel-cased charging shaft which extends from the surface to the mine level, approximately 1000 ft below. The waste canisters are lowered, one at a time down the shaft into a shielded container mounted on a mobile vehicle. This vehicle (Fig. 2) moves from the fuel assembly charging shaft to the experimental area where

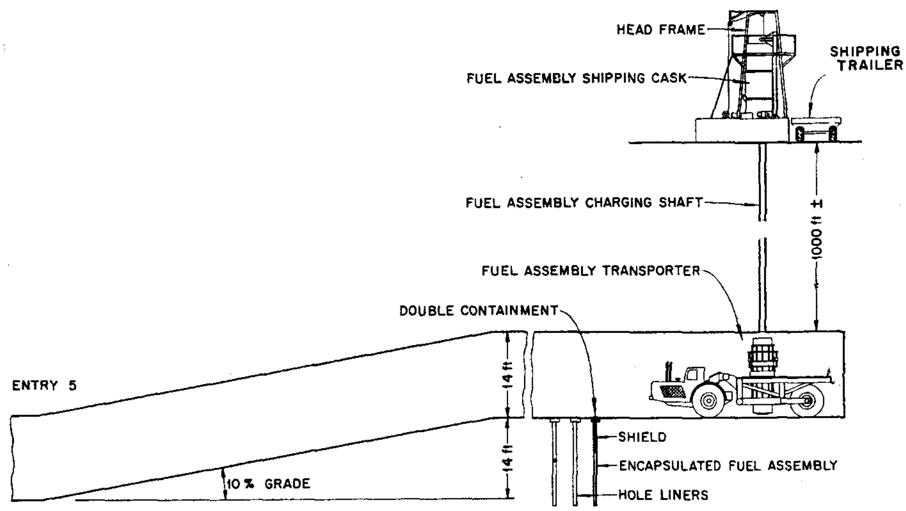

FIG. 1. Schematic cross-section of Project Salt Vault.

the canisters are to be lowered into the storage holes.[8]

Since high-level packaged solids were not in production at the time the demonstration was proposed, irradiated fuel assemblies from the Engineering Test Reactor (ETR) were used as the heat and radiation sources. In order to achieve a peak dose to the salt of about 10^9 rad, it is necessary to replace the fuel assemblies every six months with freshly irradiated assemblies. Each shipment from the Idaho Chemical Processing Plant (ICPP) consists of 14 ETR fuel assemblies contained in seven stainless steel canisters. The canisters supply the secondary containment system and are about 5 in. in diameter by $7\frac{1}{2}$ ft long. A depleted uranium shield plug is located at the upper end of the canister and thermocouples are installed to monitor fuel assembly temperatures during the experiment. When Project Salt Vault has been completed all canisters will be returned to Idaho for recovery of the fuel assemblies.

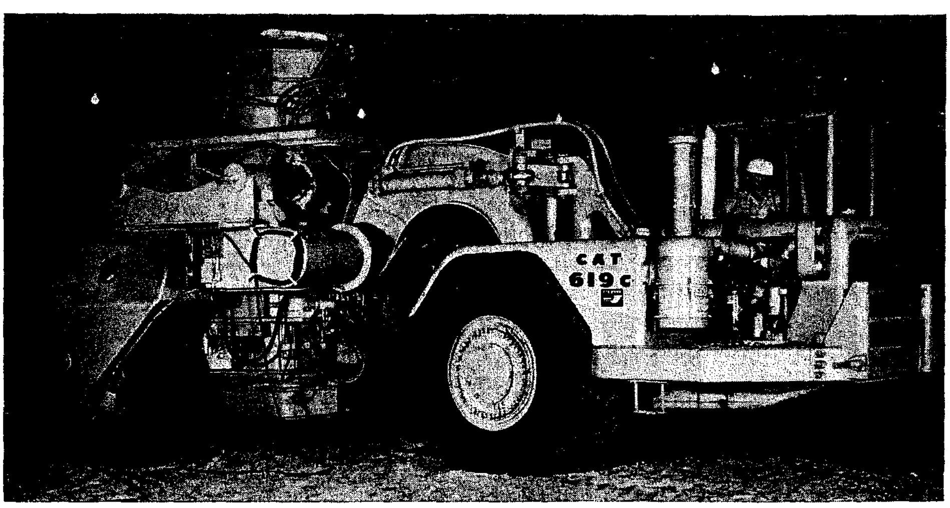

FIG. 2. Underground transporter used to transfer canisters at mine level.

Project Salt Vault is composed of four experiments: (1) an array of seven fuel assembly canisters located in pure salt in the newly mined area; (2) a non-radioactive control array using electrical heaters to study the effects of radiation on plastic flow and chlorine release; (3) a heated pillar experiment for plastic flow and mine stability studies; and (4) an array of seven canisters in the old mine floor to determine operational problems related to the use of abandoned mines for radioactive waste disposal. Their location in the mine is shown in Fig. 3.

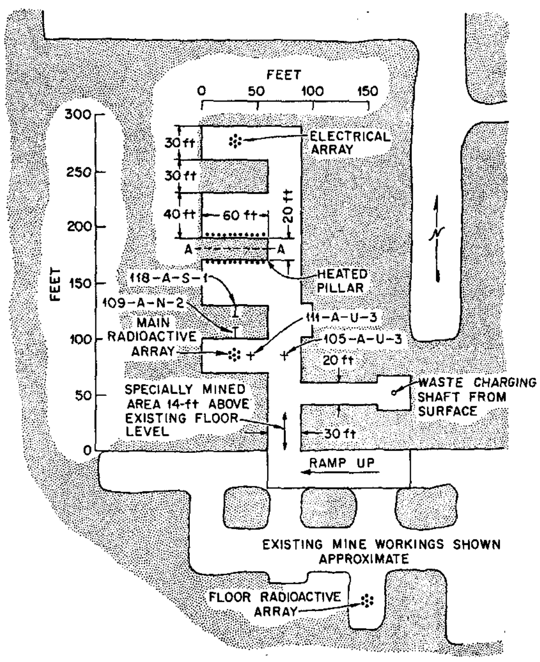

FIG. 3. Plan of experimental area.

The first transfer of fuel assemblies to Project Salt Vault occurred on November 17–19, 1965. Seven canisters were lowered into the mine and placed in the main radioactive array. The fuel assemblies were about 105 days out of reactor and contained approximately one million curies. The maximum radiation dosage received by the operating personnel during the transfer operation was 200 mr.[9]

Transfer of the seven canisters from the main radioactive array to the array in the existing mine floor, followed by the insertion of seven new canisters in the main array, took place on June 13-15, 1966. The seven new canisters contained assemblies that were 105 days out of the reactor. The second set of assemblies received higher irradiation in the ETR and their heat generation rate was 800 W initially and they contained about $1\frac{1}{2}$ million curies. As in the case of the initial loading operation, no handling problems were encountered, and radiation exposures of operating personnel were minimal.

Temperatures in both the electrical and radioactive arrays are reasonably close to those predicted by the theoretical calculations. Figure 4 shows the temperature rises in the salt $1\frac{1}{2}$ ft from the center line of the center holes in the arrays along with the calculated temperature rises. It can be seen that most of the temperatures are somewhat lower than the calculated values. This is due in part to heat loss from the salt to the mine room. The peak fuel element temperatures reached about 300°C soon after placement in the mine.

In Fig. 5(a) is shown the uplift profiles for room 1 (radioactive array), along the north–south and east–west axes of the room. It is apparent that thermal expansion of the material in the floor extends to 40 or 50 ft from the center of the array. The north–south uplift profile shows the restraining effect produced by the presence of the adjacent pillars.

Vertical thermal expansion of the floor in the center of the arrays had reached nearly an inch by the end of December 1965. The floor uplift (measured in feet) as a function of time for each array at the center and 10 ft from the center is shown in Fig. 5(b). It may be observed that the rate of rise in and near the array is slowing down. This is due to the fact that the rate of rise of the salt temperature is also slowing down. The total vertical expansion had reached about $1\frac{1}{4}$ in. in May 1966.

Project Salt Vault will continue, with fuel assembly changeouts every six months, until about November 1967. Following completion of the demonstration essentially all basic data necessary for the design of an actual disposal facility will have been obtained.

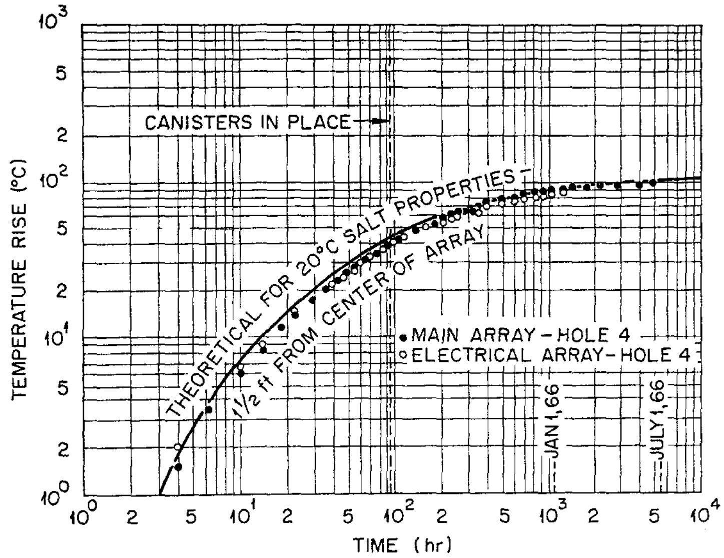

FIG. 4. Comparison of actual and theoretical temperature rises in radioactive and electrical arrays.

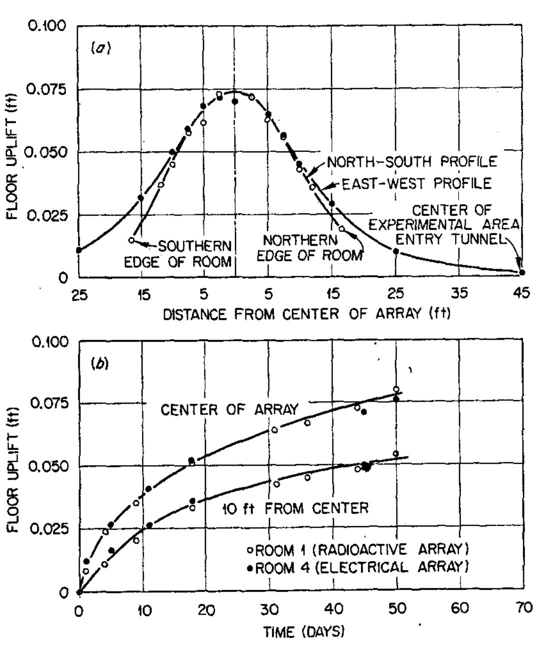

FIG. 5. Floor uplifts—Project Salt Vault. (a) Floor profiles in Room 1 as of December 30, 1965. (b) Floor uplift in array rooms.

DISPOSAL BY HYDRAULIC FRACTURING

Hydraulic fracturing has been used for many years in the petroleum industry to increase production from oil wells. Normally, a mixture of sand and water or sand and oil is forced out into a hydraulically produced crack in the formation where the sand is deposited, allowing the oil to drain into the sand layer and then out into the well. In the case of radioactive waste disposal, however, the problem is not one of increased oil production but rather one of pumping the material requiring disposal into the fracture.

The disposal well is possibly the most critical part of the disposal facility. The well must be drilled into the formation selected for disposal operation, cased, and cemented to prevent ground water from entering the well. When an injection is to be performed, this casing is slotted and water is pumped into the well until the pressure builds up producing a fracture or crack in the formation (see Fig. 6). The waste-cement mixture is then injected into this fracture. In the case of radioactive waste disposal, solid ingredients (such as cement, clay, and a retarder are mixed with the waste to produce a

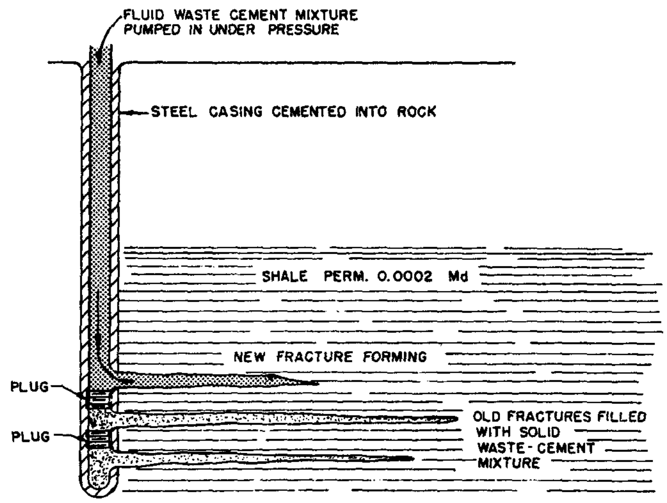

FIG. 6. Schematic cross-section of hydraulic fracturing operations for radioactive waste disposal.

slurry which will harden in the fracture and retain the fission products.

An essential prerequisite in the use of hydraulic fracturing for radioactive waste disposal is the need for producing horizontal fractures in the formation. Considerable controversy exists in the petroleum industry over the conditions necessary to produce horizontal rather than vertical fractures. Since experience in the petroleum industry is mainly with permeable formations and not the relatively impermeable formations proposed for radioactive waste injections it was decided to perform a series of test injections in the shale at Oak Ridge. The first experimental injection was made at a depth of 290 ft and consisted of 27,000 gal of a mixture of water, cement, and diatomaceous earth tagged with 35 Ci of ^{137}Cs. Subsequent coring and gamma-ray logging verified that the grout sheet had followed the bedding planes and that the fracture was essentially horizontal (see Fig. 7). The second experiment included two injections at greater depths. The final injection was made at a depth of 934 ft and consisted of

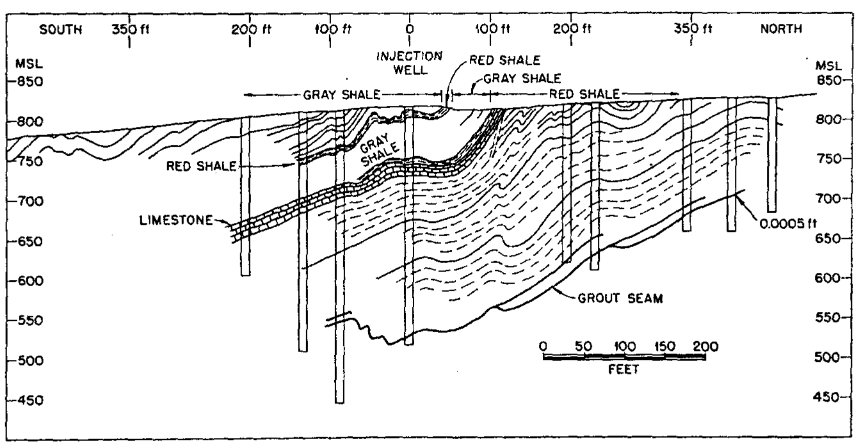

FIG. 7. Location of grout sheet—First experiment.

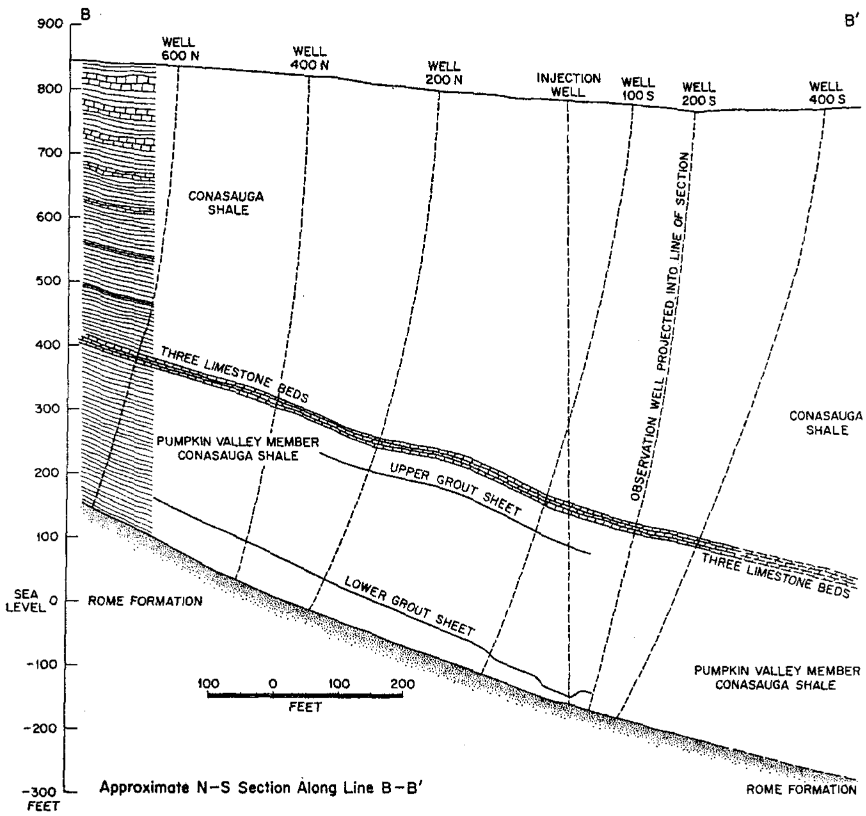

FIG. 8. Location of grout sheet—Second experiment.

91,500 gal of water, cement, and bentonite, tagged with 25 Ci of ^{137}Cs. Several days later a second injection was made in the same well at a depth of 700 ft. Core drilling and logging again verified that the grout sheets followed the bedding planes in the formation (see Fig. 8). During the course of these experimental injections, measurements were made on surface uplift and wellhead and observation well pressures in order to develop an understanding of the mechanics of fracture formation. [10, 11]

Desirable characteristics of a waste-cement slurry for the disposal of radioactive waste by hydraulic fracturing are: (1) low viscosity for the period of time the waste is injected; (2) sorption and retention of the radioactive liquid after the slurry sets; and (3) a mixture that is relatively cheap. Studies of the waste-cement slurries for use with ORNL intermediate-level waste have shown that it is possible to develop mixtures with these properties. For long pumping times a "retarder" such as calcium lignosulfonate (CLS) must be added; if the cement content is reduced a "suspender" such as attapulgite is required. For improved retention of radiocesium a clay material such as illite must be added. From the studies completed to date it is apparent that mixes can be designed having any range of physical properties and cost for any composition of radioactive waste. [12]

Based on the results obtained in the experimental injections, an experimental plant has been built at ORNL to dispose of intermediate-level wastes and evaporator concentrates. A view of the Fracturing Plant is shown in Fig. 9. The four large bins contain the mixture of solids used in the slurry. The solids are transferred pneumatically to the mixing cell in the

FIG. 9. View of facilities at fracturing plant.

concrete block structure where they are mixed with the liquid waste and the resulting slurry is pumped into the well by the high pressure pump shown in the foreground. In order to provide radiation protection, it is necessary to enclose the waste pump, mixer, wellhead, and injection pump in concrete cells.[13]

Geologic conditions at the site are shown in Fig. 10. Shale formations exist at several depths that are believed to be suitable for waste disposal injections. Economic considerations suggested the use of the shale located at a depth of 720 to 950 ft. All the formations are well below the deepest known water bearing formation (about 200 ft).

Experimental operation of the plant during the past two years has resulted in the safe disposal of approximately 430,000 gal of waste containing 11,500 Ci. A total of seven experimental injections ranging in size from 40,000 to 148,000 gal have been made. During these injections it has been shown that it is possible to halt the injection, clear the well and equipment, make repairs, and resume operation without undue hazard to the operating personnel. Core drilling has shown that the grout sheets from the first five experimental injections conformed to the bedding planes of the shale.

Cost estimates have shown that the cost of injecting 400,000 gal per year of radioactive waste would be $0.13 per gallon including solid ingredients, depreciation, and operating costs.[14] The estimated costs are based on one injection of 100,000 gal per slot and a well life of 10 years.

The major unknown in hydraulic fracturing is that of well life. As successive injections are performed and layers of the waste are built up in the formation, the earth's surface is slowly pushed up and stresses build up in the overlying formations. Exactly how many injections can be made until the stresses in the rock will produce failure in the system (vertical fractures) is currently under study.

At the present time the plant used at ORNL for experimental hydraulic fracturing is being upgraded to an operating facility for injecting intermediate-level wastes and evaporator concentrates on a routine basis. Routine operation of this facility is scheduled to begin this fall.

INJECTION INTO POROUS FORMATIONS

For a number of years the petroleum industry has used deep well injection as a means of disposal of waste brines. In the East Texas field alone, 7×10^9 gal of brine were injected in 1958, and the total volume of brine injected

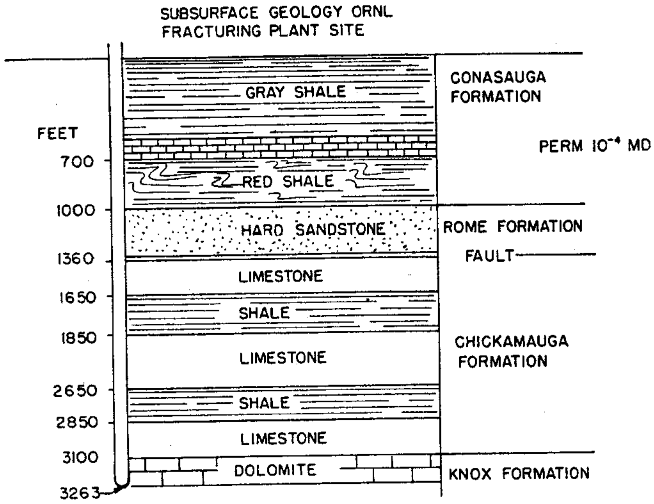

FIG. 10. Geology of shale fracturing site.

since operations were initiated in 1935 was 1×10^{11} gal.[15] At the present time, a number of companies are also using deep well disposal methods for their industrial wastes.[16, 17]

Before using deep well injection for radioactive wastes there must be an adequate disposal site and the waste must be compatible with the formation and its fluid.[18] An ideal site would be one in which a brine saturated permeable formation is bounded above and below by impermeable strata. The original concept was to store the wastes in the interstices of the formation, but the current philosophy allows injection into flowing formations since the normal flow is usually very slow; sufficient decay time should be available to reduce the radionuclide concentrations to safe levels. Radionuclides have been found to move much slower in the formation than the liquid due to ion-exchange and adsorption. However, nonhomogeneities in the formation could produce higher velocities in given areas and this may prove to be limiting.

In 1959 the American Association of Petroleum Geologists (AAPG) was requested to make a survey of potential sites where deep well injection could be performed. Their report[19] describes six basins which apparently would supply the necessary permeable formations. Three of the basins selected also contain shale reservoirs and salt deposits which might be used for intermediate- and high-level waste disposal.

The movement of the liquid and radionuclides

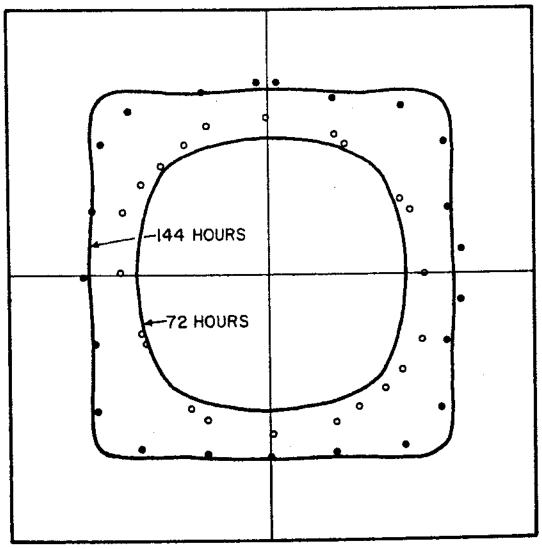

FIG. 11. Contours of mean ^{85}Sr movement in sandstone block as a function of time.

in homogeneous isotropic formations can be calculated.[20] Laboratory studies using a Berea sandstone block have verified the calculations.[21] Figure 11 shows the rate of movement of ^{85}Sr from a single injection well. It can be seen that the measured distribution agrees quite well with the predicted behavior.

Experience in the deep well injection of brines and other wastes has shown that unless the waste is compatible with the formation and its fluid the well will eventually plug. Plugging can also be caused by suspended solids or by bacterial growth. If the well were to plug by these mechanisms it might be possible to remove the plugged area by acid treatment, but the resulting waste containing radionuclides washed from the formation would present an additional disposal problem. It would appear simpler to pretreat the waste and filter prior to injection. The type and degree of pretreatment would be dependent on the characteristics of the waste and the injection formation and would have to be determined for each specific site.

At the present time there are no firm plans for a field scale demonstration of deep well injection. This is due, in part, to the large volumes of waste required to carry out a meaningful field study. It would also require a considerable period of time to complete a field study of any significant size. The results of a single field study could not necessarily be extrapolated to other sites due to local geologic peculiarities because the imperfections or non-homogeneities of the formation would probably control the degree of leakage.

CONCLUSION

Field scale demonstrations have shown that it is possible to safely and economically store high-level solidified wastes in salt formations and intermediate-level wastes in the slightly permeable shales at Oak Ridge. Disposal of low-level liquid wastes in permeable formation has not yet been demonstrated in field scale plants but the concept has been proven on laboratory scale models.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the contributions by the following individuals to the work described in this paper: R. L. Bradshaw, W. de Laguna, F. M. Empson, D. G. Jacobs, H. Kubota, W. C. McClain, W. F. Schaffer, Jr., R. C. Sexton, T. Tamura, and H. O. Weeren.

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