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H. C. Claiborne

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THE EFFICACY OF BACKFILLING AND OTHER ENGINEERED BARRIERS IN A RADIOACTIVE WASTE REPOSITORY IN SALT

H. C. Claiborne

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ABSTRACT

The early concept for the disposal of radioactive waste was simple and seemingly adequate — load the calcined waste (and later borosilicate glass) in a canister and bury it deep in a bedded salt formation. Corrosion and waste/rock interactions seemed unimportant since there had been no circulating groundwater for over 2×10^8 years in potential sites.

In the United States, investigation of potential host geologic formations was expanded in 1975 to include hard rocks. It is presumed that these crystalline rock repositories could fill with groundwater relatively soon after sealing. Presumably, potential groundwater intrusion was a factor in the development of the multibarrier concept and the proposed Nuclear Regulatory Commission (NRC) requirement that the waste container provide containment for a minimum of 1000 years. Apparently, this is leading to very conservative and expensive waste package designs to provide assurance of compliance. Recent studies have concluded that incentives for engineered barriers and 1000-year canisters probably do not exist for reasonable breach scenarios. The assumption that multibarriers will significantly increase the safety margin can also be questioned since any system failure will probably be of the common-cause type.

Use of a bentonite backfill for surrounding a canister of exotic materials was developed in Sweden to meet the requirements of their law. Apparently this same technique is being considered in the U.S. waste program for all geologic media even though the conditions are different. For example, the expectation that bentonite will remain essentially unchanged for hundreds of years for U.S. repository designs may be unrealistic. In addition, thick bentonite backfills will increase the canister surface temperature and add much more water around the canister than would be expected from brine migration in salt repositories.

The use of desiccant materials, such as CaO or MgO, for backfilling seems to be a better method of protecting the canister since the desiccants would react with any water and keep the canister dry, thereby greatly inhibiting corrosion. An argument can also be made for not using backfill material in salt repositories since the 30-cm-thick space will provide for hole closure for many years and will promote heat transfer via natural convection. In such a system, the canister surface could remain dry since the cooler salt surface will probably control the dew point.

It is generally concluded that expensive safety systems are being considered for repository designs that do not necessarily increase the safety margin. It is recommended that the safety systems for waste repositories in different geologic media be addressed individually and that cost-benefit analyses be performed for the addition of each redundant safety system.

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1. INTRODUCTION

The early concept for the disposal of nuclear waste was simple and seemingly adequate — load the calcined waste in a canister and bury it deep in a bedded salt formation. Corrosion and waste/rock interactions seemed unimportant since there had been no circulating groundwater for over 2×10^8 years in potential sites. With care in site selection and repository design, it seemed likely that additional millions of years would pass before circulating water reached the buried waste. The concept was modified to include a more leach resistant waste form; namely, the calcined waste was to be immobilized in a borosilicate glass. Replacing the slightly soluble calcine by a material with a solubility similar to Pyrex glass seemed to provide an additional large margin of safety for handling and ultimate disposal.

In the United States, investigation of potential host geologic formations for commercial high-level waste (CHLW) was expanded in 1975 to include granite, basalt, tuff, argillaceous rocks, and salt domes. With the exception of salt and perhaps argillaceous rocks, it was presumed that repositories in these host rocks would fill with groundwater relatively soon after the repository was backfilled and sealed. Even though these groundwaters would be naturally almost motionless, such repositories would not be leak-tight and might possess natural pathways to the biosphere as a normal condition. Presumably, this groundwater intrusion with its concomitant potential for transport of radionuclides to the biosphere was a factor in the development of the multibarrier concept and the U.S. Nuclear Regulatory Commission's (NRC) proposed rule[1] that the waste package provide containment for 1000 years.

In the following sections, the possible impacts of the proposed NRC regulations and the risks from radioactive waste repositories are discussed, and the need for engineered barriers and some consequences of the type of backfilling around a waste canister are examined.

2. IMPACT OF PROPOSED NRC REGULATIONS

The most stringent requirement of the proposed rules[1] for geologic repositories is the minimum 1000-year containment for all radionuclides by the waste package after permanent closure. This seems to be leading to unnecessarily conservative and expensive waste package design with a thick backfill of various materials in a desire to provide assurance of compliance.

It is interesting to note that backfilling around a waste package is not addressed and backfilling is mentioned in only two sections. In the definition section, it is stated that the "waste package means the airtight, watertight, sealed container which includes the waste form and any ancillary enclosures, including shielding, discrete backfill and overpacks." The other mention of backfilling is in the section on "Engineered Barriers," where backfill is defined as a barrier. The description of the backfill functions seems only to apply to the shafts, corridors, and disposal rooms. It is also stated that the backfill shall retard radionuclide migration.

In spite of the vagueness in regard to backfilling, the proposed regulations are generally being interpreted as requiring a thick backfill around the waste package that is capable of sorbing radionuclides in addition to protecting the waste package from corrosion by intruding water. A sorptive requirement for a backfill around the waste package does not seem to be a logical requirement since holding up the long-lived radioisotopes for a 1000 years or so will not significantly reduce their radioactivity, and the ^{90}Sr and ^{137}Cs would have decayed to relatively innocuous levels in 400 years.

In the section on "Waste Package Requirements," it is stated that synergistic interactions between the various factors that could compromise the waste package function must also be considered. It seems that protecting the waste package with a thick backfill could be interpreted as creating a synergistic effect in that the waste package temperatures could be increased substantially and that use of such materials as bentonite would add more water around a waste package than would migrate there in salt repositories.

3. REPOSITORY RISK IN PERSPECTIVE

The perception of the risk by the general public of the nuclear industry is apparently orders of magnitude greater than actual and estimated risks. In efforts to diminish this perceived risk, more and more engineered safety features and redundant safety systems are added to nuclear systems with little regard to cost-benefit analyses. Admittedly, placing a monetary value on a human life or on human misery is a repugnant and difficult thing to do, and it is considered immoral to do so by some people; however, such valuations are made indirectly by individuals with their choice of life-style and directly by the courts in lawsuits involving accidental injury or death.

In the addition of systems to power reactors, cost-benefit analyses are generally ignored which can lead to abnormally high cost per fatality averted.[2] For the current concepts of nuclear waste disposal, Cohen[3] has estimated that the cost of averting a fatality is \$200 million for defense high-level waste and \$18 million for commercial high-level waste; the cost increases to \$1 billion after 400 years when discounted at 1%. These results seem to be an indication of the greatly overestimated risks as perceived by society since such sums are not, and cannot be, expended to reduce the risks from the more mundane hazards to society (such as accidents, fires, explosions, drownings, etc.) that are much more prevalent.

In regard to the back end of the fuel cycle, the perceived risks by society also appear to be orders of magnitude greater than actual, even though a serious nuclear incident seems impossible without invoking accident scenarios of ridiculously low probability. Nevertheless, the nuclear industry has been pilloried because of the lack of a permanent waste disposal system, and criticism was heaped upon the early managers and scientists for ignoring the problem. The problem was not really ignored; geologic waste disposal did not seem to present any unusual technical difficulties or significant risks for a properly chosen site in bedded salt. This judgment has been vindicated by many studies;[4,5] the problems are not technical, but primarily social and political. For example, Erdmann et al.,[6] compared the risks from a waste repository with those from other parts of the fuel cycle. The results showed that the risk from latent-cancer fatalities from preclosure operation of a repository was 7 orders of magnitude less than that from the operation of nuclear reactors and that the long-term risk after closure was over 11 orders of magnitude smaller.

In one of the more recent reviews of geologic disposal of radioactive waste, Pigford[7] concluded, on the basis of the quantity of water required to dilute dissolved radioactive materials to drinking water standards, that the potential hazard of CHLW after decaying 600 years would be less than the hazard from the source ore and its mill tailings residue. Using reasonable assumptions, he also concluded that the radioactive hazard from the average coal-ash pile exposed to weathering would be greater than the maximum hazard from buried CHLW; he further pointed out that the hazard from the ash pile remains essentially constant because the ^{226}Ra is continuously generated in the uranium decay chain.

4. FUNCTION OF BACKFILL SURROUNDING THE WASTE PACKAGE

The early concept for geologic disposal specified that the diameter of the waste emplacement hole should be drilled about 10 cm larger than the canister to allow for deviations in the waste canister and hole alignment. The void space was to be packed with crushed salt, which would eventually consolidate and improve the heat transfer to the salt formation.

With the advent of the multibarrier concept, the backfill assumed a greater importance and many functions were assigned to it. Briefly these include:

1. act as a barrier to hydrologic intrusion to the waste package,
2. act as a sorptive barrier for radionuclides,
3. chemically buffer or modify the pH, Eh, or ionic composition of intruding groundwater or brine,
4. provide a mechanical stress buffer from hole closure, and
5. serve as a heat transfer medium (or even enhance it).

The Swedish KBS-project[8] under pressure of time to comply with a new law that required a demonstration of a safe method of waste disposal selected their only viable geologic medium, granite. The fractured and jointed characteristics of their granite formations with available groundwater prompted them to design an exotic waste package that included a thick protective covering of a bentonite-quartz mixture (and later highly compacted bentonite) to perform functions 1 and 2. The Swedish workers realized early that the most important function of the backfill was to keep water away from the canister. It was generally conceded that the waste package design would contain the waste for perhaps thousands of years in the Swedish granite environment. Functions 3 and 4 were generally American additions. Tailoring the backfill to perform function 3 seems to be unnecessary and of doubtful practicality. The Swedes were not particularly concerned with the heat transfer from the package because the much lower volumetric heat generation rate of the waste resulted in canister surface temperatures of less than 100°C. This quick technological fix, which seems adequate for Swedish conditions, has basically been incorporated into the U.S. waste program for all geologic media even though the heat fluxes from the U.S. waste canister are greater and the stability of bentonite for hundreds of years can be questioned because of the higher temperatures and radiation fields predicted for the U.S. program;[9] moreover, in the case of waste disposal in salt, no circulating groundwater will be present.

Prescribing the same regulations and engineered barrier requirements for waste packages for all geologic media does not seem to be sound practice. The point is strongly made in the following quotation from Verkerk:[10]

> For the case of granite where water is present in a repository in principle from the first day, all kinds of barriers preventing this water to reach the waste or the spent fuel have a much larger significance. It should therefore be pointed out with some emphasis that the present tendency in the discussion on long term risks to treat the same case in the same way as the granite or other hard rock cases is certainly not warranted. To prevent unnecessary burdens on the design criteria for repositories in salt each geologic formation type should be considered on its own merits.

5. EFFICACY OF MULTIBARRIERS

The need for the ultimate waste package, which includes a very low-leachability waste form, long-lasting containers with overpacks, and backfilling around the waste package in all situations, has been questioned either directly or indirectly in the past. The indirect questioning can be inferred from several studies[11-14] that concluded, after examining the consequences of breaching a hypothetical repository (and consequently all engineered barriers) in a generic site with subsequent groundwater intrusion, that no serious radiological consequences occur. Examples for specific sites are the evaluations of the long-term safety for the Waste Isolation Pilot Plant (WIPP)[15] and for a commercial high-level waste repository in the same general area.[16]

More direct questioning of the efficacy of the multibarrier approach results from studies by Hill,[17] Sutcliffe et al.,[18] Cloninger et al.[19] and Burkholder.[20]

Based on sensitivity analyses, Hill[17] concluded that the migration of radionuclides through the geosphere is of primary importance and the leach rate was only of secondary concern in a wide range of circumstances. She also concluded that the potential doses to the biosphere were insensitive to the lapse of time before leaching began if this time was greater than a few hundred years after emplacement and less than 10^5 to 10^6 years.

Sutcliffe et al.,[18] showed by a sensitivity analysis of the transport of radionuclides by groundwater from a generic spent-fuel repository that the maximum discharge rates to the biosphere were insensitive to the container lifetime.

In a systems study on engineered barriers for use in the disposal of spent fuel, Cloninger et al.,[19] came to the cautious conclusion that engineered barriers may not be beneficial. This view is best expressed in the following quotation from their report:

> There are several considerations that may limit the usefulness analyses of this type in providing an accurate basis for what is necessary or desirable for nuclear waste isolation. . . .
>
> [These] considerations should be kept in mind when reviewing the results and conclusions of this work, not as a detraction but as a realization that the analysis tends to maximize both the incentive for, and the resultant effectiveness of, the engineered barriers in the context of the overall repository system. In this way, the conditions under which an incentive is indicated for providing an engineered barrier and the degree of barrier effectiveness indicated are subject to some doubt. Conversely, a large degree of confidence can be attached to conclusions regarding conditions for which there is no apparent incentive or even a disincentive for providing engineered barriers.

The study by Burkholder[20] was aimed at the evaluation of the technical justification for the development and use of sophisticated engineered components in radioactive waste isolation systems located in domal and bedded salt, granite, and basalt. The most interesting result of this study was a demonstration that the site portion of the isolation system dominated the overall system performance to the point that even relatively poor sites were more than adequate to isolate the waste without the use of any engineered components. His results are summarized in the following quotation from the report:

> Based on the results of this study and other studies in the nuclear waste isolation literature, the use of long-lived waste containers and very low transport rate man-made subsystems seems unjustified from a technical standpoint and a geologic isolation point of view. The design objective for the waste container lifetime should not be greater than that required for retrievability (10 to 100 years), and the design objective for the man-made subsystem [nuclide fractional] transport rate should not be lower than about 1×10^{-4} per year in basalt systems, 3×10^{-4} per year in granite systems, and 1×10^{-2} per year in salt systems.

In spite of the results of the various studies, multibarriers are still generally recommended for use as redundant safety systems. This seems to be unjustified on the basis of reliability engineering theory. The multibarrier concept for waste repositories amounts to using a number of redundant safety systems in series, each with a very low probability of failure to perform as specified. Four different barriers to the release of radionuclides to the biosphere from the repository can be identified, namely, waste form, waste container, backfill, and the geologic medium. In normal practice, one redundant or backup component is usually considered sufficient when the component failure probabilities are very low. The resulting probability of failure for the operational as well as the backup component is already so low that additional backup components would not provide a significant additional safety margin because common-mode or common-cause failure can be expected to be operative if such a system would actually fail. Consequently, it seems unjustified to attempt to decrease the insignificant risk from a repository by using more than one redundant barrier.

6. THE EFFECT OF BACKFILL THICKNESS ON CANISTER TEMPERATURE

Figure 1 was prepared to demonstrate the effects of backfill thickness and thermal conductivity on the canister wall temperature. The calculations are for an infinite cylinder, an equivalent heat load of 2.16 kW per canister,[9] and a temperature of 150°C at the salt-backfill interface. This model, though simple, demonstrates a possible synergistic effect previously mentioned. It suggests that the backfill becomes an insulator for thicknesses greater than 10 to 20 cm (4 to 8 in.) in realistic ranges of an effective thermal conductivity (k) of 0.5 to 0.8 W/(m•K). The 30.5-cm (1-ft) backfill of bentonite and sand (k = 0.75 W/(m•K) specified in the preliminary conceptual waste package designs for spent fuel[21] and high-level defense wastes[22] would increase the temperature of a canister containing high-level waste by 70 or 90°C over that for a seemingly more reasonable backfill thickness of 5 to 10 cm. Higher or lower values of the thermal conductivity are possible, depending on the materials used and the conditions involved. In the case of bentonite, it is possible to produce high-density blocks of pure bentonite with a thermal conductivity >1 W/(m•K) by forming under high isostatic pressure.[8] The effective conductivity after emplacement will average somewhat lower than the blocks alone.

7. BENTONITE AS A BACKFILL MATERIAL

As previously mentioned, the Swedish waste program adopted bentonite as a backfill material under the urgency of a tight schedule to demonstrate the safe disposal of radioactive waste. The maximum temperature of 100°C for the canister surface and the lead or copper walls that reduced the absorbed radiation dose made bentonite more acceptable for that program than for the U.S. program because higher temperatures and less shielding of the backfill material are generally assumed in the latter program. Dehydration of bentonite can begin at 100°C, or even lower temperatures, under certain conditions. The potassium/sodium ratios in the waters that will eventually fill the void spaces in a repository in tuff are sufficiently high to cause replacement of sodium in bentonite by potassium; that, along with the higher temperatures, will promote the conversion of the bentonite to illite and possibly cause other changes.[23] Under these conditions, the behavior of the bentonite over hundreds of years may not be predictable with a reasonable degree of certainty.

The behavior of bentonite on exposure to the brines of a salt repository will be different, and the high sodium/potassium ratio will inhibit the replacement reaction. The long-term behavior in a radiation field is not known, but it seems logical that the hydrated-layered structure will be subject to disruption and changes over long time periods. In addition, Nowak[24] has found that neither bentonite nor hectorite is an effective sorber for cesium or strontium in WIPP brines. He suggests that activated carbon be added to the bentonite to enhance the sorbability for these and other radionuclides.

In the case of disposal in salt, the use of thick bentonite backfills will add much more water around the canister than that to be expected from brine migration.[25] A minimum of about 10% water is required to produce a sufficiently plastic bentonite that can be compressed and shaped. For example, a 30.5-cm-diam canister surrounded by a 30.5-cm-thick backfill of the 20% bentonite - 80% sand mixture assumed in the preliminary reference conceptual design[20] for spent fuel, with no overpack and 10% water in the bentonite, will contain 66 L of water. For pure bentonite, this increases to 330 L. Using the larger inner diameter of the backfill of the preliminary design to allow for the overpack increases the water content to 90 and 440 L respectively. For the preliminary reference conceptual design[22] for high-level defense waste (DHLW), which uses a 61-cm-diam canister, the corresponding amounts of water are 95 and 480 L. These values for the water content of the bentonite are more than an order of magnitude greater than the expected water influx by brine migration to an emplacement hole in a salt repository and less than the upper-bound estimates of Jenks and Claiborne[25] of 120 and 30 L for CHLW and spent fuel, respectively, for rock salt with 0.5 vol % brine inclusions. Extrapolating their results for DHLW would indicate an upper bound of <25L.

As a general comment, bentonite cannot keep the canister dry; it can only greatly inhibit the flow of water and/or vapor to the canister surface.

A large testing program involving the use of bentonite as a backfill is under way at the Sandia Laboratories.[26] The results of the tests were favorable with respect to the compatibility of bentonite with the proposed usage. The effects of a radiation field and the subsequent radiolysis of the water content are to be ascertained in the second phase of the program.

8. USE OF DESICCANT MATERIALS AS BACKFILL MATERIAL

The use of CaO mixed with some diluent as a backfill material was first suggested in an internal memorandum by W. R. Grimes, of Oak Ridge National Laboratory, in 1971. The CaO will swell by a factor of 1.89 on hydration to $Ca(OH)_2$ and form an impervious barrier that is stable at high temperatures; the vapor pressure of H_2O over $Ca(OH)_2$ is extremely low, only 0.35 kPa at 300°C. Jenks[27] also recognized the useful properties of CaO as a backfill material and suggested a mixture of about 32% CaO and 68% sand, and possibly mixtures of CaO and crushed salt.

Later Simpson,[28] after a literature search and evaluation of the properties of possible backfill materials, selected MgO, CaO, and calcined dolomite (CaO•MgO) as having the best potentials; he favored MgO because of the smaller probability of reaction with CO_2. Although the vapor pressure of H_2O over $Mg(OH)_2$ is higher than for $Ca(OH)_2$, it is still low, 84 kPa at 170°C. On complete hydration, MgO swells by a factor of 2.18.

Jenks and Claiborne[25] estimated that a sand-CaO or sand-MgO backfill could react with and absorb 43 and 63 kg of water, respectively, for the reference conditions (a 5-cm-thick backfill) in a commercial high-level waste repository in salt.[9] This greatly exceeds the amount of water expected to enter the emplacement hole by the brine migration process, particularly for a domal salt repository. Therefore, it seems that CaO or MgO, in contrast to bentonite, could keep the waste canister dry.

A possible advantage of oxide backfills is the pH buffering effect. Equilibrium contact of brine with CaO or MgO will produce solutions of about pH 14 or pH 12 respectively. In this high pH range, the corrosivity for water and iron or titanium is low,[29] and any HCl formed by hydrolysis of the $MgCl_2$ present in brine will be neutralized. It must be pointed out, however, that once the canister is breached, the solution will be very reactive with glass waste forms because of its high pH.

Under the actual conditions, hydroxy chlorides rather than hydroxides might be formed when a brine solution comes into contact with CaO or MgO. Formation of these compounds would neither decrease the water-reactive capacity of these oxides nor increase the vapor pressure of water over the hydrated material as long as some oxide is present. The amount of swelling when the hydroxy chlorides are formed would be different but could be determined experimentally. If the regulations that are finally adopted require that the backfill around a waste package be capable of sorbing the various radionuclides, additives such as activated charcoal can be used as has been suggested[24] for bentonite in contact with WIPP brines.

Because of the promising potential of MgO as a backfill material, an investigation of the kinetics and nature of the hydration reaction of MgO and also variants of Ca_2SiO_4 as a function of temperature and grain size using water and a synthetic brine is under way at Lehigh University by D. R. Simpson.[30]

9. USE OF CRUSHED SALT AS BACKFILL MATERIAL

In the case of a salt repository, crushed salt may be quite adequate as a backfill material as has been proposed in the past. The thermal conductivity of the crushed salt backfill, initially 0.4 to 0.5 W/(m•K), will gradually increase as consolidation and recrystallization occur. For the consolidation to be effective in causing the effective thermal conductivity to increase, the crushed salt lying above the heated zone must be able to move downward to the void spaces being formed, which may not be possible. Such a mechanism would contribute significantly to lowering the surface temperature of the canister and might possibly enhance its long-term integrity.

10. USE OF NO BACKFILL MATERIAL

A case can be made for using no backfill material in the space between the waste package and the host rock for salt repositories. Heat transfer will not be significantly affected by the thickness of the void space (see Fig. 1). Leaving a 30-cm-thick space will provide for hole closure for many years and promote natural convection heat transfer. A reservoir that is either sand-filled or open could be located at the bottom of the emplacement hole to catch any incoming liquid. Regardless of the complex solution or solid phases that form in the reservoir, a water vapor pressure will be exerted. Venting the emplacement hole during the retrieval phase will then allow a continuous water removal. The longer the room remains open, the less chance there is for liquid water to come in contact with the waste package in such a system. It is possible that refluxing can cause contact of the water with the waste package; however, it is more likely that the cooler salt surface and/or reservoir solution will control the dew point so that the surface of the waste package remains dry. Refluxing on the salt surface may cause enlargement of the emplacement hole by dissolution, but this will increase the heat transfer efficiency because of the decreasing salt temperature and increasing surface area.

After the room has been backfilled, water can no longer be effectively vented; however, the surface of the waste package should still remain dry because of the favorable dew point situation that should continue to exist until the emplacement hole effectively closes. For such conditions, a steel container could last hundreds of years since it is well known that steel will not corrode significantly for indefinite periods of time in a salt-mine atmosphere when free from contact with water.

11. CONCLUSIONS AND RECOMMENDATIONS

It appears that the waste disposal program in the United States has been unduly affected by the techniques developed in Sweden for a different set of conditions and is unnecessarily complicated by the uniform application of the multibarrier concept to the various geologic media under consideration. In the effort to ensure the safety of waste repositories, cost-benefit analyses are generally ignored, and redundant safety systems are added that seem to exceed the dictates of the logic of engineering reliability analysis and, in fact, may be counterproductive. Actually, the risks presented by the waste repositories are negligible compared with those in other parts of the fuel cycle, which in turn are small compared with the more mundane societal risks. The systems approach to the safety of repositories that has been adopted in principle should be implemented in fact; this should lead to significant reductions in complications and costs. In this vein, the following conclusions and recommendations are made:

1. Recognize that each geologic medium represents a specific situation that may require different waste package designs and engineered barriers, particularly in the case of salt.

2 In the case of salt, the use of a simple carbon steel container with a crushed salt backfill should be seriously reconsidered since a carbon steel container should remain essentially intact for a few hundred years in the anoxic conditions that will develop after sealing and the supply of water is insufficient to furnish the oxygen required for destructive corrosion. Without credit for the canister, the salt formation and a borosilicate glass waste form represents two safety systems, with the latter the redundant one.

3. If backfilling around a waste canister is required, serious consideration should be given to the use of carbon steel canister since the more exotic metals would represent a third redundant barrier.

4. The experimental program of a test facility in salt should include evaluation of both desiccant material backfills and no backfills.

5. A cost-benefit analysis should be performed for the addition of each redundant safety system.

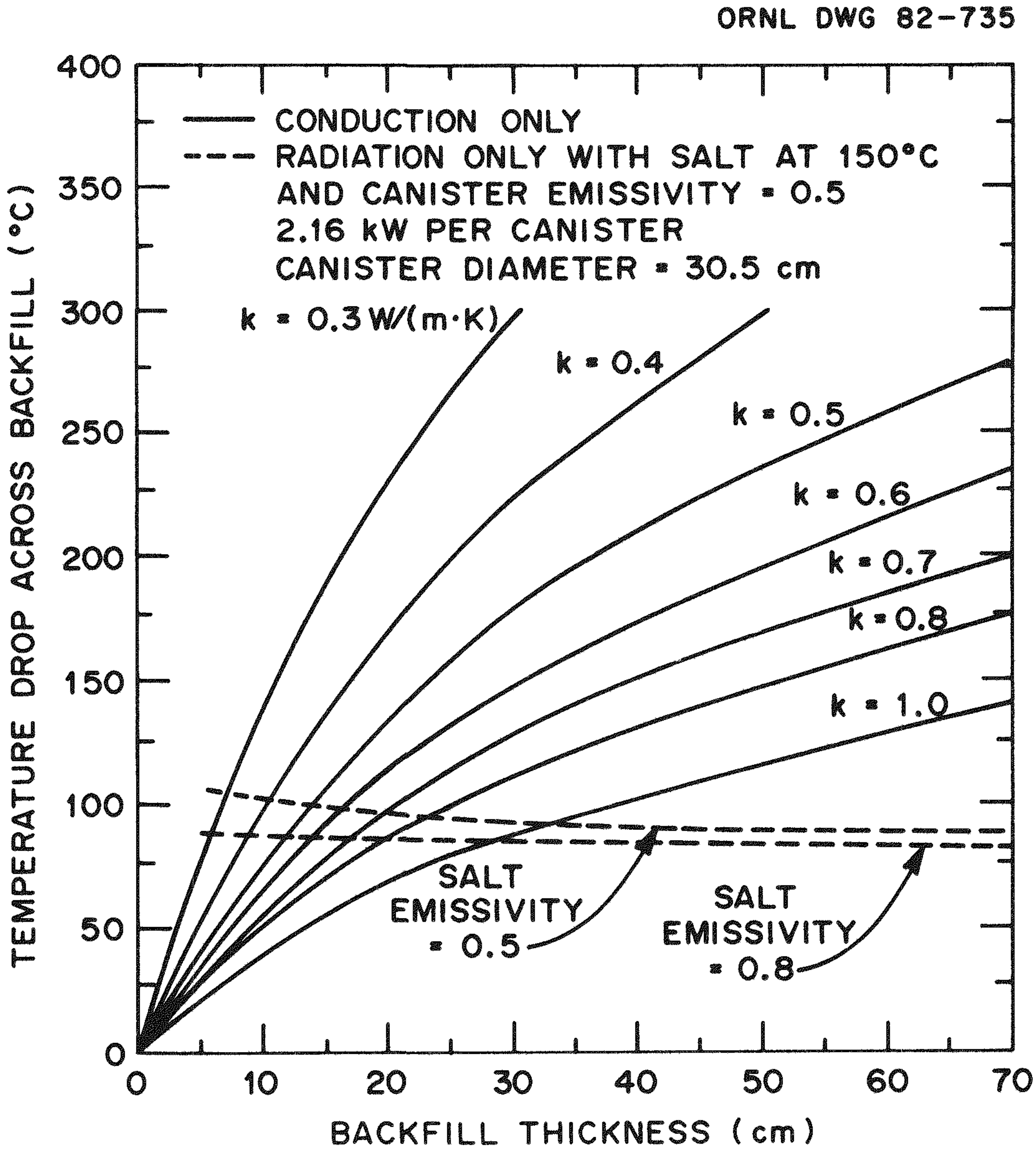

Fig. 1. Temperature drop across backfill as a function of thickness and thermal conductivity.

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608. Sally Rodgers, Friends of the Earth, New Mexico Ranch, Box 131, Santa Fe, NM 87501
609. D. M. Rohrer, Division of Waste Management, Nuclear Regulatory Commission, MS-687-SS, Washington, DC 02555
610. Eugene H. Rosenboom, Jr., U.S. Geological Survey, Mail Stop 959, Reston, VA 22092
611. Herbert N. Rosenberg, Energy Systems Group of TRW, Inc., 800 Oak Ridge Turnpike, Suite B-202, Oak Ridge, TN 37830
612. Eva Rosinger, Atomic Energy of Canada, Ltd., Pinawa, Manitoba ROE ILO, Canada

613. Jacob Rubin, U.S. Geological Survey/WRD, 345 Middlefield Road, Menlo Park, CA 94025
614. David E. Ruiter, NUS Corporation, 720 South Colorado Boulevard, Suite 900, Denver, CO 80222
615. James E. Russell, Department of Petroleum Engineering, Texas A&M University, College Station, TX 77843
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617. John L. Russell, U.S. Environmental Protection Agency, AW-45, Washington, DC 20460
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620. Dr. B. Sagar, Dames & Moore, 1100 Glendon Avenue, Suite 1000, Los Angeles, CA 90024
621. San Juan County Commission, Box 338, Monticello, UT 84535
622. Sandia Laboratories, Department 4510, Albuquerque, NM 87185
623. F. P. Sargent, Storage & Disposal Branch, Atomic Energy of Canada, Ltd., Pinawa, Manitoba, Canada ROE 1LO
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626. Dornest Schueler, Albuquerque Operations Office, U.S. Department of Energy, P.O. Box 5400, Albuquerque, NM 87185
627. Sheldon Schwartz, Assistant Director for Program Development, Office of State Programs, Nuclear Regulatory Commission, Washington, DC 20555
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629. W. E. Selph, Manager Program Development, IRT Corporation, P.O. Box 80817, San Diego, CA 92138
630. R. A. Sexton, Rockwell Hanford Operations, P.O. Box 800, 2704-E/200-E, Richland, WA 99352
631. Dr. Heyward Shealy, Conference of Radiation Control Program Directors, 1600 Bull Street, Columbia, SC 29201
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634. James N. Siltanen, Manager, Project Development, Advanced Projects Division, General Atomic Company, P.O. Box 81608, San Diego, CA 92138
635. Authur E. Slaughter, State Geologist, Michigan Department of Conservation, Geological Survey Division, Stevens T. Mason Building, Lansing, MI 48926
636. Alvin E. Smith, Manager of Nuclear Processing, The Ralph M. Parsons Company, 100 West Walnut Street, Pasadena, CA 91124
637. Lawrence J. Smith, TRU Waste Systems Office, Building T-796, Rockwell International, P.O. Box 464, Golden, CO 80401
638. Dr. Michael J. Smith, Manager Engineered Barriers, Basalt Waste Isolation Program, Rockwell Hanford Operations, Richland, WA 99352
639. T. H. Smith, EG&G Idaho, Inc., P.O. Box 1625, Idaho Falls, ID 83401
640. A. J. Soinski, California Energy Commission, 1111 Howe Avenue, MS-18, Sacramento, CA 95825
641. David L. South, Mining Department, University of Arizona, Tucson, AZ 85721
642. J. D. Spaulding, Center for Applied Isotope Studies, 110 Riverbend Road, University of Georgia, Athens, GA 30602
643. Ed Spitzer, Executive Director, Tennessee Energy Authority, Suite 707, Capitol Boulevard Building, Nashville, TN 37219
644. Lewis L. Staley, Supervisor of Energy Information, New York State Electric and Gas Corporation, 4500 Vestal Parkway East, Binghamton, NY 13902
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649. M. Steindler, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439
650. J. Carl Stepp, Director of Geosciences, Furgo, Inc., 3777 Long Beach Boulevard, Long Beach, CA 90807
651. Peter R. Stevens, Resident Geologist, U.S. Geological Survey, National Center (410), 12201 Sunrise Valley Drive, Reston, VA 22092
652. David B. Stewart, U.S. Geological Survey, 959 National Center, Reston, VA 22092
653. L. A. Stinnett, NUS Corporation, 720 South Colorado Boulevard, Suite 900, Denver, CO 80222
654. Ken Stefflet, 1706 Cameron Drive, Madison, WI 53711
655. Sheldon D. Strauss, Senior Editor, Power, 1221 Avenue of the Americas, New York, NY 10020
656. Arthur Sutherland, Ford, Bacon & Davis, Utah, P.O. Box 8009, Salt Lake City, UT 84108
657. R. T. Sutton, Commissioner, Louisiana Department of Conservation, P.O. Box 44275, Baton Rouge, LA 70808
658. M. J. Szulinski, Science Applications, Inc., 292 Torbett, Richland, WA 99352
659. Daniel M. Talbert, Sandia Laboratories, SEABED Programs Division 4536, P.O. Box 5800, Albuquerque, NM 87185
660. Charles L. Taylor, Woodward-Clyde Consultants, 3 Embarcadero Center, Suite 700, San Francisco, CA 94111
661. Dr. Irving H. Tesmer, Department of Geosciences NS270, State University College, 1300 Elmwood Avenue, Buffalo, NY
662. Burton J. Thamer, Ford, Bacon, and Davis Engineers Constructors, 375 Chipeta Way, P.O. Box 8009, Salt Lake City, UT 84108
663. John H. Thoman, Wisconsin State Energy Office, 1 West Wilson Street, Room 201, Madison, WI 53702
664. Dr. Jack Thomas, Tennessee Energy Authority, Suite 707, Capitol Boulevard Building, Nashville, TN 37219
665. L. S. Tobias, 662 Lincoln Avenue, Manville, NJ 08835
666. James R. Tomonto, Nuclear Analysis Department, Florida Power and Light Company, P.O. Box 529100, Miami, FL 33152
667. Charles Trautmann, 235 Forrest Home Drive, Ithaca, NY 14850
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674. Charles J. Vitaliano, Department of Geology, University of Indiana, Bloomington, IN 47401
675. Richard Waddell, U.S. Geological Survey WRD MS-416, Box 25046 DFC, Lakewood, CO 80225
676. J. L. Wallace, Atomic Energy Control Board, P.O. Box 1046, Ottawa, Canada KIP 559
677. Peter J. Walley, Mississippi Fuel and Energy Management Commission, Suite 228 Barefield Complex, 455 North Lamar Street, Jackson, MS 39201
678. Dr. Jack L. Walper, Professor of Geology, Texas Christian University, Department of Geology, Fort Worth, TX 76129

679. Gary Waymire, Manager, Safety Analysis, Reprocessing and Field Services Department, Exxon Nuclear Company, Inc., 777 106th Avenue NE C-00777, Bellevue, WA 98009
680. David A. Webster, DAWCON, Management Consulting Service, 310 Tara Trail NW, Atlanta, GA 30327
681. George Wehman, 3955 Georgia Lane, Ammon, ID 83401
682. R. K. White, 205 Program Manager Cress, SRI International, 333 Ravenswood Avenue, Menlo Park, CA 94025
683. Dr. Robert E. Wilems, Science Applications, Inc., 800 Oak Ridge Turnpike, Oak Ridge, TN 37830
684. W. A. Williams, Environmental Protection Agency (ANP-459), Washington, DC 20460
685. Richard M. Winar, Dames & Moore, 1550 Northwest Highway, Park Ridge, IL 60068
686. Rick Windholz, 1803 Laramie, Manhattan, KS 66502
687. Robert H. Wolle, Deputy Commissioner for the Environment, Tennessee Department of Public Health, 349 Cordell Hull Building, Nashville, TN 37219
688. D. E. Wood, Rockwell Hanford Operations, Energy Systems Group, P.O. Box 800, Richland, WA 99352
689. Warren W. Wood, U.S. Geological Survey, 12201 Sunrise Valley Drive, Reston, VA 22092
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691. Kenneth S. Wurm, Battelle Columbus Laboratories, Room 11-3082, X-4322, Columbus, OH 43201
692. P. A. Ybarra, Rockwell Hanford Operations, Energy Systems Group, P.O. Box 800, Richland, WA 99352
693. W. J. Ziefenbach, Battelle Columbus Laboratory, Room 11-2089, Columbus, OH 43201
694. Michael Zolensky, Pennsylvania State University, 236 Deike Building, University Park, PA 16802