Containment systems for storage of solidified high-level radioactive wastes in bedded salt deposits

Terence Joseph Sullivan
Iowa State University
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wastes in bedded salt deposits

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Terence Joseph Sullivan

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Approved:

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For the Major Department

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For the Graduate College

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Ames, Iowa

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TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>I.</td>
<td>NOTATION</td>
<td>iv</td>
</tr>
<tr>
<td>I.</td>
<td>INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>A.</td>
<td>Literature Review</td>
<td>1</td>
</tr>
<tr>
<td>B.</td>
<td>Safety and Recoverability</td>
<td>6</td>
</tr>
<tr>
<td>C.</td>
<td>Objective</td>
<td>8</td>
</tr>
<tr>
<td>II.</td>
<td>HIGH-LEVEL RADIOACTIVE WASTES</td>
<td>10</td>
</tr>
<tr>
<td>A.</td>
<td>Nature and Source</td>
<td>10</td>
</tr>
<tr>
<td>B.</td>
<td>Solidification Methods</td>
<td>11</td>
</tr>
<tr>
<td>C.</td>
<td>Selection of Model Waste</td>
<td>16</td>
</tr>
<tr>
<td>III.</td>
<td>BEDDED SALT DEPOSITS</td>
<td>22</td>
</tr>
<tr>
<td>A.</td>
<td>The Hutchinson Salt Section at Lyons, Kansas</td>
<td>22</td>
</tr>
<tr>
<td>B.</td>
<td>Hydrology</td>
<td>24</td>
</tr>
<tr>
<td>C.</td>
<td>The Local Environment</td>
<td>25</td>
</tr>
<tr>
<td>IV.</td>
<td>CONTAINMENT DESIGN CRITERIA</td>
<td>33</td>
</tr>
<tr>
<td>A.</td>
<td>Temperature Limits</td>
<td>33</td>
</tr>
<tr>
<td>B.</td>
<td>Diffusion Processes</td>
<td>34</td>
</tr>
<tr>
<td>C.</td>
<td>Migration Assisted by Moisture</td>
<td>38</td>
</tr>
<tr>
<td>D.</td>
<td>Time Required for Closure of the Salt Mine</td>
<td>40</td>
</tr>
<tr>
<td>V.</td>
<td>MATERIALS FOR CONTAINMENT</td>
<td>51</td>
</tr>
<tr>
<td>A.</td>
<td>Corrosion</td>
<td>51</td>
</tr>
<tr>
<td>B.</td>
<td>Methods and Materials to Overcome the Corrosion Problem</td>
<td>57</td>
</tr>
<tr>
<td>C.</td>
<td>Mechanical and Physical Properties</td>
<td>66</td>
</tr>
<tr>
<td>D.</td>
<td>Radiation Stability</td>
<td>68</td>
</tr>
</tbody>
</table>
E. Summary of Containment Systems 69

VI. HEAT TRANSFER ANALYSIS 72
   A. Previously Published Results 72
   B. Selection of Heat Transfer Model 73
   C. Temperature Profiles 86

VII. COST ANALYSIS 96
   A. Previous Cost Estimates for High-Level Waste Storage 96
   B. Costs of Mining and Waste Handling 97
   C. Cost of Materials 99
   D. Cost Comparison of Containment Systems 102

VIII. CONCLUSIONS 109

IX. RECOMMENDATIONS FOR FUTURE STUDY 115

X. BIBLIOGRAPHY 117

XI. ACKNOWLEDGMENTS 125

XII. APPENDIX A: MINE CLOSURE CALCULATIONS - PROGRAM CLOS 126

XIII. APPENDIX B: HEAT TRANSFER CALCULATIONS - PROGRAM HEAT 134
NOTATION

A Atomic weight
A_{ij} Matrix element in finite difference approximation
a Inner radius of annular region
B_{ij} Matrix element in finite difference approximation
b Outer radius of annular region
b_5 = \mu (R_o + d)
= \mu \sqrt{R_o^2 - d^2}
b_6 = \mu \sqrt{R_o^2 - d^2}
BFILL Fractional crushed salt backfill
C_{ij} Matrix element in finite difference approximation
c_p Specific heat
D Diffusion coefficient
D_{ij} Matrix element in finite difference approximation
d Distance from waste cylinder centerline
d\vec{a}_i Differential unit vector normal to surface \partial_i
E_{ij} Matrix element in finite difference approximation
EXT Fractional salt extraction
F_A Area or view factor for thermal radiation
F_\varepsilon Emissivity factor for thermal radiation
f_{o}(\Delta W, \Delta H) Function used in Newton's method
G Tabulated function in shielding calculations
G_{i} Vertical mesh spacing
g Acceleration due to gravity
g_{o}(\Delta W, \Delta H) Function used in Newton's method
H Original pillar height
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$H_j$</td>
<td>Radial mesh spacing</td>
</tr>
<tr>
<td>$h$</td>
<td>Height of waste cylinder</td>
</tr>
<tr>
<td>$h_1$</td>
<td>Distance from one end of waste cylinder</td>
</tr>
<tr>
<td>$h_2$</td>
<td>Distance from other end of waste cylinder</td>
</tr>
<tr>
<td>IR</td>
<td>Number of vertical mesh lines</td>
</tr>
<tr>
<td>IZ</td>
<td>Number of radial mesh lines</td>
</tr>
<tr>
<td>$K$</td>
<td>Thermal conductivity</td>
</tr>
<tr>
<td>$K_c$</td>
<td>Apparent thermal conductivity due to conduction and convection</td>
</tr>
<tr>
<td>$K_r$</td>
<td>Effective conductivity due to thermal radiation</td>
</tr>
<tr>
<td>$K_{ij}$</td>
<td>Thermal conductivity of mesh region</td>
</tr>
<tr>
<td>$k$</td>
<td>Boltzmann constant</td>
</tr>
<tr>
<td>$L$</td>
<td>Original pillar length</td>
</tr>
<tr>
<td>$L_o$</td>
<td>Thickness of horizontal layer</td>
</tr>
<tr>
<td>$L_1$</td>
<td>Length of annular region</td>
</tr>
<tr>
<td>$N_o$</td>
<td>Avogadro number</td>
</tr>
<tr>
<td>$N_c$</td>
<td>Number of waste containers required per year</td>
</tr>
<tr>
<td>$(N_{Gr})_L_o$</td>
<td>Modified Grashof number</td>
</tr>
<tr>
<td>$Q$</td>
<td>Waste thermal power density</td>
</tr>
<tr>
<td>$Q_o$</td>
<td>Activation energy for diffusion process</td>
</tr>
<tr>
<td>$Q_{ij}$</td>
<td>Thermal power density of mesh region</td>
</tr>
<tr>
<td>$Q_M$</td>
<td>Waste thermal power per unit area of mine</td>
</tr>
<tr>
<td>$R$</td>
<td>Outer radius of cylindrical unit cell in heat transfer calculation</td>
</tr>
<tr>
<td>$R_o$</td>
<td>Waste cylinder radius</td>
</tr>
<tr>
<td>$R_j$</td>
<td>Radial position of vertical mesh line</td>
</tr>
<tr>
<td>$R_W$</td>
<td>Resistance to heat flow of annular region</td>
</tr>
</tbody>
</table>
\( r \)  
Radial distance from waste centerline

\( S_{ij} \)  
Element of source vector

\( S'_{ij} \)  
Dummy source vector element

\( S_v \)  
Volumetric radiation source

\( T \)  
Temperature

\( T_{ij} \)  
Temperature at mesh point

\( t \)  
Time

\( t_M \)  
Mine lifetime

\( V \)  
Active volume of waste cylinder

\( V_i \)  
Volume of mesh region

\( W \)  
Weight fraction

\( W_P \)  
Original pillar width

\( W_R \)  
Original room width

\( Z \)  
Distance below surface

\( Z_i \)  
Vertical position of radial mesh line

\( \alpha_i \)  
Surface area of mesh region

\( \beta \)  
Volume coefficient of thermal expansion

\( \Delta H \)  
Change in pillar height

\( \Delta r \)  
Radial gap thickness

\( \Delta T \)  
Temperature difference

\( \Delta t \)  
Time increment in finite difference technique

\( \Delta x \)  
Spatial increment in finite difference technique

\( \sqrt{\Delta x} \)  
Root mean square displacement

\( \Delta W \)  
Change in pillar width

\( \dot{\varepsilon} \)  
Strain rate

\( \varepsilon_1, \varepsilon_2 \)  
Total emissivity of inner and outer surface, respectively
\( \mu \)  Linear attenuation coefficient  
\( \bar{\mu} \)  Mass attenuation coefficient  
\( \nu \)  Kinematic viscosity  
\( \rho \)  Density  
\( \Sigma_R \)  Macroscopic removal cross section  
\( \sigma \)  Average pillar stress  
\( \sigma_R \)  Microscopic removal cross section  
\( \sigma_S \)  Stefan-Boltzmann constant  
\( \phi \)  Radiation flux at waste surface
I. INTRODUCTION

Nuclear power is expected to assume an increasingly important role in meeting the nation's electrical energy requirements over the remainder of this century. Thus the quantity of radioactive wastes will increase by several orders of magnitude. The waste of primary concern is designated "high-level" and its source is the reprocessing of spent reactor fuels. High-level wastes consist principally of fission products with small amounts of actinide elements which are present as an inescapable loss to the waste stream during reprocessing. The long-lived fission products (primarily $^{90}$Sr and $^{137}$Cs) require periods of 500-1000 years to decay to nontoxic levels while the plutonium requires periods of up to 500,000 years. Since these wastes must be isolated from man's biosphere for such extended periods of time, it is desirable to implement storage concepts which have a minimal reliance on surveillance and periodic replacement of the containment vessel. Deep geologic formations are the primary candidates for such a storage system.

A. Literature Review

In 1955 a committee of geologists and geophysicists was established by the National Academy of Sciences — National Research Council (NAS-NRC) to consider the possibility of disposal of high-level wastes in geologic formations within the United States. In 1957 this committee concluded that bedded salt deposits offer the best potential for high-level waste disposal [28]. This recommendation was based on the many
characteristics of salt which are desirable for such an application. The presence of good quality bedded salt is evidence of the general tectonic stability of an area and of the absence of circulating ground water. Bedded salt has a high compressive strength but flows plastically at relatively low temperatures and pressures so that openings in the salt are eventually sealed. The thermal properties of rock salt are better than most other types of rock and this permits the dissipation of larger quantities of heat. Also salt has radiation shielding characteristics similar to concrete. Finally, salt formations are widespread and abundant and salt is relatively easy to mine.

From 1957 to 1961 Oak Ridge National Laboratory (ORNL) conducted laboratory and field studies on the feasibility of disposing of high-level liquid wastes in bedded salt [76, 77, 37]. This program indicated that liquid wastes probably could be deposited in salt but that there were a number of difficult problems that would require detailed examination. The results were reviewed by another NAS-NRC committee in 1961 and it was recommended that more extensive studies be directed toward solidified wastes [26].

During 1963-1967 ORNL performed extensive experiments with spent fuel assemblies from the Engineering Test Reactor in the mine of the Carey Salt Company at Lyons, Kansas, under the name Project Salt Vault [16]. In 1969 ORNL published a favorable estimate of the feasibility of storing solidified high-level wastes in bedded salt [17]. A preliminary conceptual design was undertaken in 1969 and is illustrated in Fig. 1. In this concept, cylindrical containers of high-level waste are inserted into holes drilled in the floor of a salt mine excavated
Fig. 1. Artist's conception of the demonstration repository [33, p. 23].
by the room and pillar method. The holes and rooms are subsequently backfilled with crushed salt. After a row of rooms is filled, the corridors are sealed with plastic and backfilled with crushed salt. When the total area of the mine has been utilized, the access shaft is sealed with layers of stone, concrete, and clay. The conceptual design was extended in 1970 to include the possibility of storing alpha wastes in a separate section of the mine.

Pierce and Rich assembled an extensive amount of information on rock salt deposits in the United States as potential repository sites [81]. The primary site selection criteria have been detailed in the environmental statement for the proposed Lyons repository [94] and summarized by Culler in his 1971 review of the technical status of the proposed repository [33]. The salt formation must be of the bedded type, approximately horizontal in attitude, and relatively undisturbed structurally. It should have a horizontal extent of at least several tens of miles, lie at a depth greater than 500 feet but less than 2000 feet, and be at least 200 feet thick. The formation should be situated in a zone of tectonic stability and not be associated with valuable reserves of oil, gas, or other mineral resources. Salt formations in central Kansas, west-central New York, and southeastern and northern Michigan satisfy these criteria. Based on a comparison of these areas with regard to the primary criteria and other secondary considerations such as the accessibility of a nonoperating mine, land values, population distribution, waste transportation costs, and public acceptance, the Lyons, Kansas, mine of the Carey Salt Company
was selected in 1970 as the tentative location for a demonstration waste repository.

In 1970 the NAS-NRC Committee on Radioactive Waste Management reviewed the ORNL results and issued a report approving of the concept of storage as developed by ORNL and of the selection of the Lyons site [27]. They recommended that further investigations be undertaken to study site geology, location of old oil and gas wells, subsidence, the possibility of a stored energy effect, movement of water in the neighborhood of the salt, thermal and mechanical properties of key geologic structures, and long term plans for monitoring and possible waste retrieval.

In 1971 the Kansas Geological Survey published its final report on the geology and hydrology of the Lyons site in which serious reservations were expressed concerning the large number of old oil and gas holes, the existence of water above and below the salt, possible deep seated structural problems, and the high potential for oil and gas development in the area [50]. Other problems concerning the solution mining operations of the American Salt Company within 1500-1800 feet of the proposed repository and the inability to locate and plug old holes were brought out in late 1971 [73]. As a result, the Kansas Geological Survey undertook a study of alternative sites in the state of Kansas. The results of this survey were published in early 1972 and several promising regions in the state of Kansas were discussed [1].

In May of 1972 the Atomic Energy Commission (AEC) announced that it would design and build surface facilities for the storage of high-level wastes [95]. However the advantages of storage in salt were reemphasized
and it was announced that, while investigation of questions concerning the Lyons site continue, ORNL with the aid of the U.S. Geological Survey is to expand its efforts in search of a suitable site for a pilot plant repository. The site for this demonstration facility is to be announced within a year.

B. Safety and Recoverability

The primary objections to the concept of storage in bedded salt center around the geology and hydrology of the Lyons site. It is expected that the difficulties at Lyons will be resolved or that an alternate site which is more suitable from a geologic and hydrologic point of view will be selected. There are, however, three safety related aspects of the ORNL concept which remain undefined and tend to amplify these objections.

The first concerns the form of the solidified waste. The current Code of Federal Regulations specifies that within five years after reprocessing spent fuel, the resultant high-level liquid wastes must be converted to a dry solid and placed in a sealed container [24]. Further, the dry solid must be chemically, thermally, and radiolytically stable to the extent that the equilibrium pressure in the sealed container will not exceed the safe operating pressure for that container during the period from canning through a minimum of 90 days after receipt at the Federal repository. The waste must be transferred to a Federal repository within 10 years after reprocessing. These criteria are open to a wide range of interpretation. Several methods of waste
solidification are currently available whose products apparently satisfy the above criteria. But the properties of these products vary over a wide range, especially with respect to leachability of radioactive nuclides by contacting liquids.

Secondly, Culler has stated that the required lifetime of the waste containers is still a moot question [33, p. 55]; but that the current acceptance criterion is that they be able to maintain their integrity for at least 90 days after they have been buried in salt [33, p. 3]. This is based on the premise that the salt serves as the primary containment. In fact, the salt is a poor container for an extended period of time after insertion of the waste due to the porosity of the crushed salt backfill and because of cracks and fractures introduced during mining and drilling operations. Depending on mining patterns and temperatures, it is generally acknowledged that it will take at least several decades for pillar deformation resulting from the overburden load to effect a complete closure and reconsolidation of the backfilled salt [33, p. 47]. Given a serious container failure, radioactive nuclides would be free to migrate by means of a number of processes including crystalline, grain boundary, and surface diffusion and liquid or vapor phase transport. It is probable that these nuclides would not escape the salt formation. However, positive control and localization of the waste would be lost.

The stability of the waste and containers ties in directly with the third area of concern which is the necessity of a plan for retrieving the waste. Walter Meyer (Department of Nuclear Engineering, Kansas State University) has argued from the point of view of safety that the
wastes should be recoverable in a reasonable time and at a reasonable cost; and, based on his report, the Kansas Governor's Nuclear Energy Council has recommended adoption of the principle of recoverability [71]. Schneider has argued that it may be desirable to retrieve the waste because of some unforeseen problem, because a truly ultimate disposal technique had been discovered, or because man might develop a different and vital need for the storage area [86, p. 394]. The Committee on Radioactive Waste Management also recommended that a waste retrieval plan be developed [27, p. 14]. Accordingly, in the environmental statement for the proposed Lyons repository, the AEC stated that the facility would be designed so as not to preclude retrieval and that concepts for retrieving the wastes would be examined in the course of the design of the facility [94, pp. 80-81]. The lack of a definite retrieval plan is a source of considerable criticism at this time. The development and implementation of such a plan would be greatly facilitated by the choice of a highly stable waste form and of containers with lifetimes extending to the point where plastic flow and reconsolidation of the salt effectively seals the waste in place.

C. Objective

Motivated by the above considerations, this study was undertaken to develop concepts for storage of high-level waste in bedded salt which provide for continuous containment of wastes in the immediate vicinity of the original container. This development involves the
choice of a suitable waste form, determination of the time required for the salt to reconsolidate and effectively seal the waste in place, and selection of materials for containers or secondary barriers which can maintain their integrity until the salt can act as the primary containment. A heat transfer analysis is carried out to determine how much waste heat can be stored in a given area of mine for each set of materials. Finally, an economic comparison of the various materials systems based on costs of mining and materials is presented.
II. HIGH-LEVEL RADIOACTIVE WASTES

A. Nature and Source

The extraction of fissionable material from spent fuel results in high-level waste streams which contain essentially all (> 99.9%) of the nonvolatile fission products and smaller amounts of uranium, plutonium, and other transuranium elements. These aqueous wastes are usually nitric acid solutions. They are characterized by very intense, penetrating radiation and high heat generation rates.

By the year 1974 there will be three commercial reprocessing plants in operation in the United States with a combined capacity of 2700 metric tons of uranium (U) per year. The Nuclear Fuel Services, Inc., plant at West Valley, New York, began operation in 1966; and, currently, over 600,000 gallons of high-level liquid wastes are stored at this site [86, p. 392]. The General Electric (GE) plant at Morris, Illinois, received its first fuel shipment in early 1972 and plans call for immediate solidification of high-level wastes by means of the fluidized bed calcination process [42]. The Allied-Gulf Nuclear Services Plant at Barnwell, South Carolina, should be operational by 1974 and will initially store its high level wastes in liquid form in stainless steel tanks. In addition, AEC reprocessing plants at Richland, Washington, Savannah River, South Carolina, and the National Reactor Testing Station, Idaho, store over 80 million gallons of radioactive solutions and sludges in tanks [86, p. 392].
B. Solidification Methods

The solidification methods available in the United States at this time are pot calcination, fluidized bed calcination, spray melter and in-pot melting variations of spray solidification, and phosphate glass solidification. Schneider [86] and McElroy et al. [67] have discussed the current status of these processes. Key process considerations and product properties are summarized in Table I.

Pot calcination was developed at ORNL [22] and engineering scale tests with high-level wastes were carried out by Batelle-Northwest (BNWL) [68]. Liquid waste is added to a pot which is heated in a multiple zone furnace. A scale of calcine cake forms on the walls of the pot and grows inward until the pot is full. Pot temperatures of the order of 850-900 °C for up to 40 hours are required to boil all of the liquid to dryness. The processing vessel is also the final container for the solidified waste.

The fluidized bed calcination process initiated at Argonne National Laboratory (ANL) [62] and extensive development through two full scale processing campaigns has been carried out by Idaho Nuclear Corporation (INC) at the National Reactor Testing Station [25, 64]. Liquid waste is injected through pneumatic atomizing nozzles into the side of a heated (400-600 °C) bed of granular solids (activated alumina) which is continuously agitated by a suitable fluidizing gas sparging upward through the bed from inlets at the bottom of the reactor. When the waste solution contacts the hot bed, nitrate salts decompose with water vapor and volatile gases flashing from the spray droplets.
<table>
<thead>
<tr>
<th>Development</th>
<th>Pot calcine</th>
<th>Fluidized bed calcine</th>
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<tbody>
<tr>
<td>Process system</td>
<td>ORNL-BNW</td>
<td>ANL-INC-GE</td>
</tr>
<tr>
<td>Type</td>
<td>Simple</td>
<td>Simple</td>
</tr>
<tr>
<td>Throughput capacity, liter/hr</td>
<td>Batch</td>
<td>Continuous</td>
</tr>
<tr>
<td>Scaleup Potential</td>
<td>Limited</td>
<td>Very good</td>
</tr>
<tr>
<td>Flexibility for waste variations</td>
<td>Good</td>
<td>Very good</td>
</tr>
<tr>
<td>Product form</td>
<td>Scale</td>
<td>Granular</td>
</tr>
<tr>
<td>Description</td>
<td>Calcine cake</td>
<td>Amorphous</td>
</tr>
</tbody>
</table>

**Volume of solid waste, liters per metric ton of U processed**

<table>
<thead>
<tr>
<th>Process system</th>
<th>Pot calcine</th>
<th>Fluidized bed calcine</th>
</tr>
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<tbody>
<tr>
<td>Throughput capacity, liter/hr</td>
<td>12-14</td>
<td>10-400</td>
</tr>
<tr>
<td>Scaleup Potential</td>
<td>Limited</td>
<td>Very good</td>
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<tr>
<td>Flexibility for waste variations</td>
<td>Good</td>
<td>Very good</td>
</tr>
<tr>
<td>Product form</td>
<td>Scale</td>
<td>Granular</td>
</tr>
<tr>
<td>Description</td>
<td>Calcine cake</td>
<td>Amorphous</td>
</tr>
</tbody>
</table>

**Pot properties**

- **Calcium**
- **Granular**
- **Amorphous**

**Fluidized bed calcine properties**

- **Calcium**
- **Granular**
- **Amorphous**

**Max thermal power W/liter, 8 in. pot**

- **Pot calcine**
  - 85
- **Fluidized bed calcine**
  - 70

**Max stable T, °C**

- **Pot calcine**
  - 900
- **Fluidized bed calcine**
  - 600

**Leachability in cold H2O, g/(cm²-day)**

- **Pot calcine**
  - $10^{-1} - 10^{-2}$
- **Fluidized bed calcine**
  - $10^{-1} - 10^{-2}$

**Hardness**

- **Pot calcine**
  - Soft
- **Fluidized bed calcine**
  - Moderate

**Friability**

- **Pot calcine**
  - Crumbly
- **Fluidized bed calcine**
  - Moderate

**Container**

- **Pot calcine**
  - Stainless steel
- **Fluidized bed calcine**
  - Mild or stainless steel
<table>
<thead>
<tr>
<th>Spray melter</th>
<th>Spray in-pot melting</th>
<th>Phosphate glass</th>
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<tbody>
<tr>
<td>BNWL Complex</td>
<td>BNWL Moderately complex</td>
<td>BNL-BNWL Complex</td>
</tr>
<tr>
<td>Continuous</td>
<td>Semi-continuous</td>
<td>Continuous</td>
</tr>
<tr>
<td>11-18</td>
<td>11-18</td>
<td>13-26</td>
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<td>Good</td>
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<td>Good</td>
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<tr>
<td>Limited</td>
<td>Limited</td>
<td>Limited</td>
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<td>Monolithic</td>
<td>Monolithic</td>
<td>Monolithic</td>
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<td>Glass (Borosilicate)</td>
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<td>30-60</td>
<td>50-100</td>
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<td>200</td>
<td>190</td>
</tr>
<tr>
<td>Phase separation</td>
<td>900</td>
<td>Devitrifies 500</td>
</tr>
<tr>
<td>$10^{-3}$-$10^{-5}$</td>
<td>$10^{-4}$-$10^{-5}$</td>
<td>$10^{-4}$-$10^{-6}$</td>
</tr>
<tr>
<td>Hard</td>
<td>Very hard</td>
<td>Very hard</td>
</tr>
<tr>
<td>Tough</td>
<td>Brittle</td>
<td>Brittle</td>
</tr>
<tr>
<td>Mild or stainless steel</td>
<td>Stainless steel</td>
<td>Mild or stainless steel</td>
</tr>
</tbody>
</table>
leaving the metallic oxides deposited uniformly on the surface of the bed particles. Solid product is transported pneumatically from the calciner to the storage containers.

Spray solidification was developed by Battelle-Northwest and has undergone two engineering scale tests [10, 11]. In the spray melter variation, liquid waste is fed through a pneumatic atomizing nozzle into the top of a heated cylindrical tower where it is sequentially evaporated, dried, and calcined to a powder as it falls into a continuous melter. The melter is heated to 800-1200 °C and the molten calcine flows through an overflow weir into the receiver-storage container below. Container temperatures range from 450-500 °C for up to 40 hours. In the in-pot melting variation, the melter is eliminated and a borosilicate flux is added to the calcine with subsequent melting carried out in the final container. Container temperatures of the order of 900-950 °C for up to 40 hours are required.

The phosphate glass process was developed at Brookhaven National Laboratory [35] and two series of engineering scale tests were carried out by Battelle-Northwest [69, 70]. Liquid waste containing all melt making additives is fed to an evaporator where it is continuously concentrated and denitrated, forming a thick slurry. The slurry is fed to a continuous melter and heated to 1000-1200 °C. The molten glass flows through an overflow weir to the receiver-storage container below. The receiver is heated to 400-600 °C to match the slump point of the melt and insure uniform filling.

From a processing point of view, the fluidized bed technique appears to be the best. It is simple and continuous and the product
can be transported pneumatically, eliminating considerable remote manipulation of containers. The throughput capacity of a solidification process is limited by the rate at which heat can be supplied. Idaho Nuclear has demonstrated the use of internal heating in a fluidized bed by means of heater tube bundles with NaK as a heat transfer fluid. This, coupled with the excellent heat transfer properties of the fluidized bed, leads to high potential throughput capacities for a single unit. This process also has the most understood scaleup technology. By the use of additives, a number of various waste types can be successfully calcined leading to the best overall flexibility.

However, calcine products are inferior to glass or microcrystalline products in several key respects. Due to void fractions of the order of 50%, the thermal conductivity of calcines ranges about 75% lower than glass products. For a given container size, the thermal conductivity determines how much waste heat can be put in the container consistent with maximum allowable waste and container temperatures. As indicated in Table I, the maximum allowable thermal power in a calcine waste is about 35-45% of that allowed in a glass. Thus a greater number of containers would be required for the same total stored waste heat. Even in the current concept the final container is one of the most expensive single items [79, p. 28]. If a concept of longer lived and hence more expensive final containers were implemented, the economic effects would be even greater.

More significantly, the calcines exhibit leach rates several orders of magnitude higher than glasses when contacted by a liquid. For comparison with the data in Table I, the leach rate of Pyrex glass
is about $5 \times 10^{-7}$ g/(cm$^2$-day) [86, p. 411]. Paige has demonstrated that the fluidized bed alumina calcine can be easily converted to a good quality glass by adding appropriate fluxing agents and applying heat (900 °C) [75]. Leach rates of this alumina-phosphate glass were of the order of $10^{-4}$-$10^{-6}$ g/(cm$^2$-day). A comparison of leach rates of $^{137}$Cs and $^{90}$Sr in percent fission product leached per day is presented in Fig. 2 for alumina calcine and alumina-phosphate glass. Results indicated that about 95% of the Cs and 35% of the Sr were leached from the calcine within seven weeks while only 0.005-0.01% of the Cs and Sr were leached from the alumina-phosphate glass. Note also that even after 95% of the Cs and 35% of the Sr had been leached from the calcine, the leaching rate of the calcine was still three orders of magnitude higher than for the glass.

Therefore the inadvertent contact of liquid with a calcined waste during transportation or storage leads almost immediately to a high-level liquid waste. While long term contact of a liquid with the glass product would certainly not be desirable, the extremely low leaching rate offers a considerable time cushion during which the glass could be safely recovered from the deleterious environment.

C. Selection of Model Waste

The considerations in the previous section indicate that from the point of view of safety and economics it would be highly desirable to solidify high-level wastes in the form of a glass. The phosphate glass system is the most highly developed system in the United States
Fig. 2. Comparison of leachability of cesium and strontium from calcine and glass at 25 °C in distilled water [75, p. 11].
and McElroy et al. [67, p. 80] have concluded that this process is feasible and essentially ready for commercial application. Sulphate and sulphate-free wastes from light water reactor (LWR) fuels irradiated up to 45,000 MWD/metric ton and a liquid metal fast breeder reactor (LMBR) core fuel have been successfully solidified. Mendel has summarized the advantages of the phosphate system over other glass forming systems for solidifying high-level wastes [87, Ch. 4].

It is unlikely that the pot calcination process will ever be implemented commercially since it is inherently a batch process with limited scaleup potential and the product properties are less desirable. The fluidized bed concept is being implemented and an additional process step would provide substantial improvement of product properties through incorporation into an alumina-phosphate glass.

Thus, the potential exists for solidifying a wide range of high-level wastes in the phosphate system. The phosphate glass process is the model solidified product which will be used throughout the remainder of this dissertation. Typical properties are summarized in Table II. In all subsequent calculations it is assumed that the final solidified product results from the reprocessing of spent fuel from a reference LWR whose characteristics are summarized in Table III.

Photon and neutron release rates are tabulated in Table IV. Note that greater than 99% of the photons released are in the lowest four energy groups. The sources of neutrons are (α - n) reactions with light elements and spontaneous fission of heavy nuclides, principally 242Cm and 244Cm and to a lesser extent 238Pu and 242Pu. Spontaneous
Table II. Properties of typical phosphate glass product [70, p. 3.12]

<table>
<thead>
<tr>
<th>Glass composition:</th>
<th>Weight percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>Additives</td>
<td></td>
</tr>
<tr>
<td>Na₂O</td>
<td>7.48</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>6.20</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>61.79</td>
</tr>
<tr>
<td>Fission products and other waste</td>
<td>24.53</td>
</tr>
</tbody>
</table>

Product density: 2.9 g/cm³
Product volume: 76.0 liters/metric ton of U processed

Table III. Characteristics of reference LWR fuel [33, p. 4]

| Enrichment (²³⁵U):               | 3.3 %           |
| Average specific power:         | 30.0 MW/metric ton |
| Exposure:                       | 33,000 MWd/metric ton |

Waste:
Consists of all fission products plus actinides remaining after removal of 99.5% of the uranium and plutonium following a post-irradiation decay period of 150 days.

fission accounts for more than 98% of the neutrons so that the majority of these neutrons have energies greater than 1 Mev.

Thermal characteristics of the model waste are illustrated in Fig. 3 for the typical unit volume of 76 liters/metric ton of U processed and the lowest unit volume of 50 liters/metric ton of U processed. Short-lived fission products decay away rapidly and by the end of 10 years, the waste is decaying with the approximately 30 year half-life of the principal fission products ⁹⁰Sr and ¹³⁷Cs. This is
Table IV. Volumetric photon and neutron release rates for 76 liter/metric ton phosphate glass product resulting from reprocessing reference LWR fuel [33, p. 6]

<table>
<thead>
<tr>
<th>Time since reprocessing</th>
<th>5 years</th>
<th>10 years</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Photons ([\text{Y/(cm}^3 \cdot \text{sec)]:})</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0.30 Mev</td>
<td>4.80 \times 10^9</td>
<td>2.46 \times 10^9</td>
</tr>
<tr>
<td>0.63 Mev</td>
<td>9.47 \times 10^10</td>
<td>4.72 \times 10^{10}</td>
</tr>
<tr>
<td>1.10 Mev</td>
<td>3.78 \times 10^9</td>
<td>1.91 \times 10^9</td>
</tr>
<tr>
<td>1.55 Mev</td>
<td>1.15 \times 10^9</td>
<td>2.68 \times 10^8</td>
</tr>
<tr>
<td>1.99 Mev</td>
<td>1.02 \times 10^8</td>
<td>2.63 \times 10^6</td>
</tr>
<tr>
<td>2.38 Mev</td>
<td>4.55 \times 10^7</td>
<td>1.45 \times 10^6</td>
</tr>
<tr>
<td>2.75 Mev</td>
<td>1.49 \times 10^6</td>
<td>4.70 \times 10^4</td>
</tr>
<tr>
<td>3.25 Mev</td>
<td>2.68 \times 10^4</td>
<td>8.54 \times 10^2</td>
</tr>
<tr>
<td><strong>Neutrons ([n/(cm}^3 \cdot \text{sec)}]:)</strong></td>
<td>4.11 \times 10^3</td>
<td>3.40 \times 10^3</td>
</tr>
</tbody>
</table>

the basis of current AEC regulations of shipment to a Federal repository within 10 years. In terms of decrease in specific thermal power, very little is gained by interim storage beyond 10 years.
Fig. 3. Thermal power density as a function of time since reprocessing for phosphate glass high-level waste resulting from reference LWR fuel [33, p. 4].
III. BEDDED SALT DEPOSITS

A. The Hutchinson Salt Section at Lyons, Kansas

There are several objections to the proposed siting of a Federal repository at Lyons, Kansas; but these are based primarily upon geological and hydrological considerations external to the salt formation. In its study of alternative sites, the Kansas Geological Survey stated that the salt formation at Lyons fits the structural engineering criteria well [1, p. 89]. In addition the depth and thickness of the formation are adequate and salt quality is fair [1, p. 92]. Thus the Hutchinson salt section at Lyons is representative of the type of salt formation which would be suitable for storage. Since this dissertation deals with the local environment of the waste containers, information from the Lyons site will be used in all further considerations.

At the Lyons site the Hutchinson salt member consists of about 300 feet of beds of salt, shale, and anhydrite, approximately horizontal in attitude. Salt comprises about 60% of this sequence. The thickest and most persistent salt zone lies at a depth of about 1000 feet. This zone would be the logical choice for mining salt or storing a waste container. The general stratigraphy of this area is illustrated in Fig. 4 along with a schematic of the vertical position of various operations within the mine. The clay and shale laminae constitute the major impurity in the salt and are typical of most bedded salt deposits. It is proposed that the high-level storage area be excavated in a northeasterly direction from the location of the old mine workings at a level of about 20 feet above the old mine
Fig. 4. General stratigraphy of the Hutchinson salt member at Lyons, Kansas, near level proposed for waste storage and vertical location of various operations within the mine.
floor [33, p. 30]. The waste container would be placed in the bottom of an 18 foot hole drilled in the floor of the room, thus placing the waste within the best salt zone. The upper 8 feet of the insertion hole is filled with crushed salt for shielding purposes until the remainder of the room is filled with waste and finally backfilled.

B. Hydrology

There are two sources of water within the salt formation. Small inclusions of brine, ranging in size from a few millimeters down to microscopic, are distributed throughout the salt and constitute about 0.5 volume percent of the salt [33, p. 40]. The shale layers are known to contain more water than the salt, with estimates ranging up to 10 volume percent [52, p. 3].

The major source of moisture in the vicinity of the containers is the brine content of the salt. During Project Salt Vault it was discovered that these inclusions tend to migrate up a thermal gradient toward the heat source [16, pp. 164-172]. The mechanism for cavity migration is the diffusion of sodium and chloride ions from the hot to the cold side of the cavity due to the slightly greater solubility of NaCl at the higher temperatures. The results of Bradshaw for total expected brine inflow are widely quoted. He estimated total inflow of 2-10 liters over a period of 20-30 years after waste burial [15]. In fact these values are probably too low because of the calculated temperatures which were employed. On the basis of more conservative assumptions and more accurate temperature profiles, Jenks has recently
estimated that maximum total inflow would range from 30-50 liters over a period of 50 years and slowly taper off thereafter [48, p. 13]. The brine inflow rates are highest during the first few years when temperature gradients are the steepest, with as much as 50% of the total inflow occurring within 10 years.

Shale layers will release their water as steam if subjected to temperatures in excess of 100 °C [16, p. 164]. For this reason, waste containers will be placed so that they do not intersect a major shale layer.

C. The Local Environment

A typical waste cylinder configuration immediately after insertion and backfilling is presented in Fig. 5. Various aspects of the local environment are illustrated schematically. Salt temperatures in the vicinity of a waste container range from 150-300 °C depending on proximity to the container. Container wall temperatures range from 250-350 °C and waste temperatures are 300-500 °C.

The chemical composition of the rock salt in weight percent is expected to be nearly the same as samples taken from the formation at Hutchinson, Kansas, which is: 96.970% NaCl; 1.407% water insolubles assumed to be mostly CaSO₄; 0.875% CaSO₄; 0.670% H2O; 0.066% MgCl₂; and 0.034% CaCl₂ [16, p. 277]. Jenks has estimated that the composition of the brine within the salt is within the following ranges: Mg²⁺, 2.3-3 M; K⁺, 0.4 M; Na⁺, 2-1 M; Cl⁻, 7-7.5 M; and Br⁻, 0.05 M [48, p. 4]. The brine is slightly basic but at elevated
Fig. 5. Schematic representation of typical waste cylinder configuration and environmental features immediately after insertion and backfilling.
temperatures would become slightly acid due to the hydrolysis of MgCl₂.

From the standpoint of container integrity, the most significant chemical reaction is the hydrolysis of MgCl₂ to form HCl. Jenks has investigated the overall reactions [48] and states that they can be represented as,

\[ \text{MgCl}_2 + \text{H}_2\text{O} \rightleftharpoons \text{Mg(OH)Cl} + \text{HCl} \]  

(1)

and

\[ 2\text{Mg(OH)Cl} \rightleftharpoons \text{MgCl}_2 + \text{MgO} + \text{H}_2\text{O} \]  

(2)

Reaction (1) should go to completion to the right at temperatures above 180 °C. Complete hydrolysis through reaction (2) only occurs at temperatures above 520 °C. The extent of hydrolysis is also limited by the ability of the HCl to move away from the reaction site and the MgCl₂. Based on fragmentary data on equilibria in these reactions, Jenks has made a conservatively high estimate that about 50% of the MgCl₂ should eventually react to form HCl assuming that the HCl is free to move away from the MgCl₂.

The most recent measurements by Jenks indicate that the brine is 2.1 M in Mg²⁺ and the estimated specific gravity is 1.23 [36]. Assuming 50% hydrolysis, 1.05 moles of HCl are produced per liter yielding a solution which is 3.11 weight percent HCl. There are several reasons to believe that containers would never be exposed to an acid concentration even this high. To begin with, the estimates are conservatively high. Also, the reaction is limited by the ability of the HCl to move away from the MgCl₂ so that very little hydrolysis is expected within
the brine inclusions. When the brine moves into the region of crushed salt (void fraction < 0.3) around the container more significant hydrolysis begins. However, experience with Project Salt Vault indicates that as the brine enters the region around the container it moves upward in the vapor phase through the crushed salt into the region above the container until temperatures low enough for condensation are encountered. Container temperatures during waste storage will not be low enough to allow condensation around the containers. Thus the brine will move away from the containers before complete hydrolysis can occur. Finally, while performance of stainless steel type 304L test heaters during Project Salt Vault was quite poor, the observed corrosion was not indicative of such high acid concentrations. The corrosion rate of type 304, even at room temperature in weak HCl solutions, is widely quoted as greater than 50 mils/year (mpy). Heaters tested in the Lyons mine at 124–400 °C for 3–16 months exhibited a maximum pit depth of 0.4 mm (16 mils) [52].

Important radiolysis products include H₂, O₂ and possibly ClO₃⁻ and BrO₃⁻ [48]. These should not have a significant effect on container stability in the amounts anticipated.

Radiation levels are calculated by converting volumetric release rates tabulated in Table IV to equivalent surface fluxes at a waste surface, accounting for self shielding of the waste. Since additives comprise approximately 75 weight percent of the waste (Table II), it is assumed for shielding calculations that the waste consists entirely of the additives. For gamma ray calculations this is a conservative assumption, since all waste nuclides have atomic numbers
larger than the additives and hence are better gamma attenuators. Since the majority of the neutrons have energies in excess of 1 Mev, the removal cross section concept is used. The assumption that the waste is entirely additives is also conservative in this case since the removal cross section increases with atomic mass.

Since greater than 99% of the photons are within the lowest four energy groups, only these groups are included. Total gamma ray mass attenuation coefficients are tabulated in Table V. For atomic weights (A) greater than 10, the total removal cross section can be approximated by [3]

\[ \sigma_R = 0.141 \times 10^{-2}A^{2/3} \text{ cm}^2 \]  

(3)

Removal cross sections are given in Table VI.

As suggested by Fano et al. [39] the mass attenuation coefficient for each chemical compound is taken as the weighted average of the coefficients of the constituent elements so that for \( \text{P}_2\text{O}_5 \) for example

\[ \overline{\mu}_{\text{P}_2\text{O}_5} = (62\overline{\mu}_p + 80\overline{\mu}_O)/142 \]  

(4)

Then the total linear gamma ray attenuation coefficient for the waste is given by

\[ \mu = \rho \left( W_{\text{Na}_2\text{O}} \overline{\mu}_{\text{Na}_2\text{O}} + W_{\text{Fe}_2\text{O}_3} \overline{\mu}_{\text{Fe}_2\text{O}_3} \right) \]

where \( \rho \) is the density of the waste and \( W \) is the weight fraction of each chemical compound. Macroscopic removal cross sections were calculated in the usual way, for example.
Table V. Total gamma ray mass attenuation coefficients ($\bar{\mu}$) in cm$^2$/g

<table>
<thead>
<tr>
<th>Energy (MeV)</th>
<th>O</th>
<th>Na</th>
<th>P</th>
<th>Fe</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.3</td>
<td>0.107</td>
<td>0.102</td>
<td>0.106</td>
<td>0.106</td>
</tr>
<tr>
<td>0.63</td>
<td>0.0791</td>
<td>0.0756</td>
<td>0.0766</td>
<td>0.0747</td>
</tr>
<tr>
<td>1.10</td>
<td>0.0612</td>
<td>0.0586</td>
<td>0.0594</td>
<td>0.0573</td>
</tr>
<tr>
<td>1.55</td>
<td>0.0511</td>
<td>0.0489</td>
<td>0.0495</td>
<td>0.0479</td>
</tr>
</tbody>
</table>

Table VI. Total neutron removal cross sections ($\sigma_R$) as a function of atomic mass

<table>
<thead>
<tr>
<th>Material</th>
<th>Atomic mass</th>
<th>$\sigma_R$ (cm$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>O</td>
<td>16</td>
<td>$0.895 \times 10^{-24}$</td>
</tr>
<tr>
<td>Na</td>
<td>23</td>
<td>$1.140 \times 10^{-24}$</td>
</tr>
<tr>
<td>P</td>
<td>31</td>
<td>$1.391 \times 10^{-24}$</td>
</tr>
<tr>
<td>Fe</td>
<td>56</td>
<td>$2.054 \times 10^{-26}$</td>
</tr>
</tbody>
</table>

\[
(\Sigma_R)_{P_2O_5} = (W_{P_2O_5} \rho N_A/142)[2(\sigma_R)_P + 5(\sigma_R)_O]
\]  

where $N_A$ is the Avogadro number. Then the total macroscopic removal cross section for the waste is

\[
\Sigma_R = (\Sigma_R)_{Na_2O} + (\Sigma_R)_{Fe_2O_3} + (\Sigma_R)_{P_2O_5}
\]

For a point on the principal axis of a right circular cylinder of radius $R_o$ and height $h$, Rockwell [85, p. 404] has given the following expression as an upper limit to the flux
\[
\phi = \left( \frac{S_V}{4\mu} \right) [G(\mu h_1, b_5) + G(\mu h_2, b_5) + G(\mu h_1, b_6) + G(\mu h_2, b_6)]
\]
(8)

where

- \( S_V \) is the volumetric source,
- \( \mu \) is the attenuation coefficient (use \( \Sigma_R \) for neutrons),
- \( G \) is a tabulated function \([85, p. 367]\),
- \( h_1 \) is the distance from one end,
- \( h_2 \) is the distance from the other end,
- \( b_5 = \mu (R_0 + d) \),
- \( b_6 = \mu \sqrt{R_0^2 - d^2} \), and
- \( d \) is the distance from the centerline.

The maximum surface flux is at a point equidistant from each end so that \( h_1 = h_2 = h/2 \), \( d = R_0 \), \( b_5 = 2\mu R_0 \), and \( b_6 = 0 \). Since \( G(\mu h, 0) = 0 \), the expression for the maximum surface flux is

\[
\phi = \left( \frac{S_V}{2\mu} \right) [G(\mu h/2, 2\mu R_0)]
\]
(9)

Maximum gamma ray and neutron surface fluxes for a typical waste cylinder, 6 in. in diameter and 8 ft in active length, are given in Table VII. Values for the volumetric source are taken from Table IV.
Table VII. Maximum gamma ray and fast neutron fluxes at the surface of a waste cylinder, 6 in. in diameter and 8 ft in active length (reference LWR fuel, 76 liters/metric ton)

<table>
<thead>
<tr>
<th>Gamma rays:</th>
<th>$\mu$ (cm$^{-1}$)</th>
<th>$G$</th>
<th>$\phi$ [Y/(cm$^2$ · sec)]</th>
<th>$\phi$ [Y/(cm$^2$ · sec)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E$(MeV)</td>
<td></td>
<td></td>
<td>5 years</td>
<td>10 years</td>
</tr>
<tr>
<td>0.30</td>
<td>0.3081</td>
<td>0.995</td>
<td>$7.751 \times 10^9$</td>
<td>$3.972 \times 10^9$</td>
</tr>
<tr>
<td>0.63</td>
<td>0.2253</td>
<td>0.984</td>
<td>$2.068 \times 10^{11}$</td>
<td>$1.031 \times 10^{11}$</td>
</tr>
<tr>
<td>1.10</td>
<td>0.1562</td>
<td>0.950</td>
<td>$1.149 \times 10^{10}$</td>
<td>$5.808 \times 10^9$</td>
</tr>
<tr>
<td>1.55</td>
<td>0.1439</td>
<td>0.935</td>
<td>$3.736 \times 10^9$</td>
<td>$8.707 \times 10^8$</td>
</tr>
<tr>
<td>Fast neutrons:</td>
<td>$\Sigma_R$ (cm$^{-1}$)</td>
<td>$G$</td>
<td>$\phi$ [n/(cm$^2$ · sec)]</td>
<td>$\phi$ [n/(cm$^2$ · sec)]</td>
</tr>
<tr>
<td>0.0881</td>
<td>0.822</td>
<td></td>
<td>$1.917 \times 10^4$</td>
<td>$1.586 \times 10^4$</td>
</tr>
</tbody>
</table>
IV. CONTAINMENT DESIGN CRITERIA

A. Temperature Limits

The maximum allowable heat generation rate per unit area of mine is limited by the materials involved. The maximum stable temperature for various solidified wastes ranges from 500-900 °C. In the phosphate glass waste, devitrification occurs at about 500 °C. However, the results of Paige indicate that complete devitrification has no significant effect on leaching rates [75].

Laboratory studies have shown that salt samples from the Lyons mine fracture with considerable violence and release their moisture as steam when heated to around 285-320 °C [16, p. 6]. This is characteristic of bedded salt although fracture temperatures may vary from 240-400 °C. Higher temperatures and temperature gradients also increase the rate of brine migration toward the heat source.

Salt starts to behave quite plastically at temperatures around 300 °C so that elevated temperatures and thermal-stress transfer effects could have a deleterious effect on mine stability during operation. Also extensive heating of the surface, fresh water aquifers near the surface, and areas beyond the property and mineral rights of the facility should be avoided.

Heat transfer calculations indicate that salt fracture, brine migration, and mine stability considerations provide the limiting temperature criteria. Temperature criteria established for the repository are: (a) the maximum temperature 8 in. (20.32 cm) from the wall of the hottest container should not exceed 250 °C; and
(b) the maximum salt temperature midway between two containers in the center of a room should not exceed 200 °C [33, p. 38]. If these two criteria are met, temperature limitations based on other considerations should also be satisfied.

B. Diffusion Processes

Since the time required for isolation of high-level wastes is very long, certain otherwise trivial physical processes might result in dispersal of radioactive nuclides. Loss of positive control over the radioisotopes through widespread migration and access to backfilled rooms and tunnels or to major shale partings is undesirable from a safety point of view and would make any recovery attempt nearly impossible. Any local migration near the waste containers tends to compromise the economic aspects and complicate the engineering aspects of a recovery operation.

Study of diffusion in the mine is complicated by the numerous processes that can be occurring simultaneously. Diffusion may take place along many paths: (a) inside the crystalline lattice, (b) along grain boundaries, (c) through intraparticle voids in the rock salt, (d) along pore surfaces, (e) through interparticle voids in the crushed salt, or (f) along fractures in the rock salt. Chemical reactions or vaporization-condensation may be taking place and the moving species may be diffusing in either the solid, liquid, or gaseous states. When diffusion is known to occur by several mechanisms, it is common to make use of an effective diffusivity in the transport equations. The
range of effective diffusivities for transport through intra- or inter-
particle voids or along surfaces is about $10^{-3}-10^{-9}$ cm$^2$/sec while that
for crystalline or grain boundary diffusion is usually lower, about
$10^{-6}-10^{-20}$ cm$^2$/sec [49, p. 27].

Representative values for crystalline and grain boundary diffusion
in NaCl are presented in Table VIII. Experimentally, the diffusion
coefficient is usually found to fit an Arrhenius relation of the
form

$$D = D_0 \exp \left( \frac{-Q_o}{kT} \right)$$

(10)

where $Q_o$ is the activation energy, $k$ is the Boltzmann constant, and $T$ is
absolute temperature. The results of F. Beniere and P. Chemla [5]
indicate that for single crystal NaCl in the extrinsic, structure-
sensitive region ($T < 550$ °C), $D_0$ increases and $Q_o$ decreases with in-
creasing impurity concentration. Thus diffusivities within the salt
mine would be slightly higher than the values in Table VIII due to
higher impurity concentrations. On the other hand, temperatures within
the salt in the mine will be lower than those in Table VIII, which
would tend to counter this effect. The results of F. Beniere, M. Beniere,
and M. Chemla [6] indicate that activation energies are nearly inde-
dendent of ionic radius in the extrinsic region and that values of $D_0$
do not decrease with a corresponding increase in the ionic radius of
the diffusing species. Thus one cannot conclude that fission products
and transuranium elements will exhibit lower diffusion coefficients
than sodium in NaCl due to increased ionic radius.
Table VIII. Representative diffusivities for crystalline and grain boundary diffusion in NaCl

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Medium</th>
<th>$D_{cm^2/sec}$</th>
<th>$T_{°C}$</th>
<th>Method</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{22}\text{Na}^+$</td>
<td>High purity Harshaw single crystal NaCl</td>
<td>$0.64 \times 10^{-12}$</td>
<td>390</td>
<td>Tracer — serial sectioning</td>
<td>5</td>
</tr>
<tr>
<td>$^{22}\text{Na}^+$</td>
<td>Less pure Harshaw single crystal NaCl</td>
<td>$1.81 \times 10^{-12}$</td>
<td>387</td>
<td>Tracer — serial sectioning</td>
<td>5</td>
</tr>
<tr>
<td>$^{22}\text{Na}^+$</td>
<td>High purity Harshaw single crystal NaCl</td>
<td>$3.60 \times 10^{-12}$</td>
<td>450</td>
<td>Tracer — serial sectioning</td>
<td>6</td>
</tr>
<tr>
<td>$^{86}\text{Rb}^+$</td>
<td>High purity Harshaw single crystal NaCl</td>
<td>$3.38 \times 10^{-12}$</td>
<td>450</td>
<td>Tracer — serial sectioning</td>
<td>6</td>
</tr>
<tr>
<td>$^{137}\text{Cs}^+$</td>
<td>High purity Harshaw single crystal NaCl</td>
<td>$1.23 \times 10^{-11}$</td>
<td>450</td>
<td>Tracer — serial sectioning</td>
<td>6</td>
</tr>
<tr>
<td>$^{24}\text{Na}^+$</td>
<td>High purity Harshaw single crystal NaCl</td>
<td>$1.00 \times 10^{-12}$</td>
<td>350</td>
<td>Tracer — serial sectioning</td>
<td>65</td>
</tr>
<tr>
<td>$^{22}\text{Na}^+$</td>
<td>High purity Harshaw bicrystals with 45° [100] symmetric tilt grain boundary</td>
<td>$8.67 \times 10^{-12}$</td>
<td>350</td>
<td>Autoradiographic analysis</td>
<td>83</td>
</tr>
</tbody>
</table>
Diffusivities for transport through voids or along surfaces vary over a wide range. Using a gamma ray spectrometer scanning system, Lakey has reported diffusivities for the migration of $^{137}$Cs in granular high-level calcined wastes which range from $10^{-7}$ cm$^2$/sec at 900 ºC to $10^{-3}$ cm$^2$/sec at 1200 ºC [55]. This granular product of the fluidized bed calcination process has a void fraction similar to the crushed salt backfill. Barrer has noted diffusivities for surface diffusion of Cs on tungsten which range from $1.5 \times 10^{-6}$ cm$^2$/sec at 227 ºC to $8 \times 10^{-6}$ cm$^2$/sec at 427 ºC [4, p. 361] and for Na on tungsten from $5.0 \times 10^{-4}$ cm$^2$/sec at 227 ºC to $1.28 \times 10^{-3}$ cm$^2$/sec at 282 ºC [4, p. 366]. At ORNL lanthanum oxide was used as a simulant for plutonium to measure surface transport on salt crystals [2]. Electron microprobe analysis revealed a diffusion-like concentration profile with an apparent diffusivity of $10^{-10}$ cm$^2$/sec. It was noted that this was several orders of magnitude less than that expected on the basis of a literature survey and further more detailed examination with the microprobe indicated that the lanthanum oxide had spread considerably further over the crystal than previously indicated. These results were later corroborated by analyses based on Auger electron emission and it was suggested that the behavior may have been an artifact of the deposition process.

A satisfactory estimate of the depth to which a given substance will have penetrated by diffusion into a given medium during a time $t$ can be obtained by using the root mean square displacement, which in one dimension is

$$\sqrt{\Delta x^2} = \sqrt{2Dt}$$

(11)
The derivation is discussed by Jost [49, p. 25]. Values of the root mean square displacement for a range of diffusivities and times are given in Table IX.

It is concluded from these calculations that crystalline and grain boundary diffusion do not pose a serious safety problem and would be at most a nuisance in a recovery operation. Given a container failure, however, diffusion through pore volumes in crushed salt and along surfaces provided by cracks or fractures in the rock salt could constitute a safety problem and would complicate a recovery operation. Thus, waste containers should be selected which can maintain their integrity until the salt has plastic flowed to reconsolidate the crushed salt backfill and seal other surface diffusion paths.

C. Migration Assisted by Moisture

If water is permitted to contact the waste, leaching of radionuclides ensues and the movement of these radionuclides is assisted by movement of the water in the liquid or vapor phase. Any moisture which moves into the region around the containers will vaporize and move upward through the crushed salt, following the 100 °C temperature front. Experience with liquids in the Carey Salt Co. mine at Hutchinson, Kansas, indicated that shale layers, disturbed by excavation, can also provide a suitable hydraulic connection for liquid phase transport [37, p. 40]. Liquid contained in one cavity leaked into an adjacent empty cavity. Thus significant horizontal permeability can be associated with shale bands due to cracks and voids within the
Table IX. Root mean square displacement \((\sqrt{\Delta x^2} = \sqrt{2Dt})\) in cm

<table>
<thead>
<tr>
<th>Time</th>
<th>5 years</th>
<th>10 years</th>
<th>30 years</th>
<th>10^2 years</th>
<th>10^3 years</th>
<th>10^6 years</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystalline and grain boundary:</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(^{22}\text{Na}^+) (D = 1.81 \times 10^{-12} \text{ cm}^2/\text{sec})</td>
<td>0.0139</td>
<td>0.0338</td>
<td>0.0585</td>
<td>0.1069</td>
<td>0.3380</td>
<td>10.69</td>
</tr>
<tr>
<td>(^{24}\text{Na}^+) (D = 1.00 \times 10^{-12} \text{ cm}^2/\text{sec})</td>
<td>0.0178</td>
<td>0.0251</td>
<td>0.0435</td>
<td>0.0795</td>
<td>0.2510</td>
<td>7.95</td>
</tr>
<tr>
<td>(^{86}\text{Rb}^+) (D = 3.28 \times 10^{-12} \text{ cm}^2/\text{sec})</td>
<td>0.0322</td>
<td>0.0455</td>
<td>0.0789</td>
<td>0.1440</td>
<td>0.4550</td>
<td>14.40</td>
</tr>
<tr>
<td>(^{137}\text{Cs}^+) (D = 1.23 \times 10^{-11} \text{ cm}^2/\text{sec})</td>
<td>0.0623</td>
<td>0.0882</td>
<td>0.1530</td>
<td>0.2790</td>
<td>0.8820</td>
<td>27.90</td>
</tr>
<tr>
<td>Pore and surface:</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>D = 10^{-3} \text{ cm}^2/\text{sec}</td>
<td>562.10</td>
<td>795.00</td>
<td>1377.00</td>
<td>2514.00</td>
<td>7950.00</td>
<td>2.514 \times 10^5</td>
</tr>
<tr>
<td>D = 10^{-5} \text{ cm}^2/\text{sec}</td>
<td>56.21</td>
<td>79.50</td>
<td>137.70</td>
<td>251.40</td>
<td>795.00</td>
<td>2.514 \times 10^4</td>
</tr>
<tr>
<td>D = 10^{-7} \text{ cm}^2/\text{sec}</td>
<td>5.62</td>
<td>7.95</td>
<td>13.77</td>
<td>25.14</td>
<td>79.50</td>
<td>2.514 \times 10^3</td>
</tr>
<tr>
<td>D = 10^{-9} \text{ cm}^2/\text{sec}</td>
<td>0.56</td>
<td>0.80</td>
<td>1.38</td>
<td>2.51</td>
<td>7.95</td>
<td>2.514 \times 10^2</td>
</tr>
</tbody>
</table>
shale itself or due to partings which develop between the shale and salt.

Again the moisture of immediate concern is the brine content of the salt. Within 30-50 years after waste insertion, the horizontal temperature distribution at the level of the waste containers will have leveled off, signaling the end of significant brine inflow. Thus the problem of radionuclide transport via brine can be avoided by maintaining container integrity beyond the time of significant brine inflow to the point when the salt has plastic flowed to reconsolidate the crushed salt and seal cracks and partings developed during excavation.

The problem of moisture associated with shale layers can be avoided by proper placement of containers within the salt formation. The proposed storage level at Lyons avoids significant shale layers. Contact with fresh water aquifers through old holes and wells which penetrate the salt formation must be prevented by locating and plugging these holes. The problem of loss of integrity of the salt formation and the problem of contact with water associated with solution mining and salt water disposal operations can be avoided by proper site selection based on regional geology and hydrology as outlined by the Kansas Geological Survey [1].

D. Time Required for Closure of the Salt Mine

It is necessary to obtain an estimate of the time required for reconsolidation of the crushed salt backfill within the rooms and for
the sealing of other migration paths since this determines the lifetime of the waste container. Schneider [86, p. 395] has estimated that "a few decades" are required while Culler [33, p. 47] has estimated 65 to 100 years. No calculations or other basis for these estimates were presented in either case.

When waste is inserted into the floor of a room the salt begins to heat and expand. During tests in Project Salt Vault in which heaters were inserted into the floor of the mine, an accelerated rate of vertical strain was observed in the pillars. Floor uplift was also noted along with an increased amount of roof sag. These effects are due to the increased plasticity of salt at the elevated temperature and to the stresses within the salt which are a superposition of pre-existing overburden and mining stresses and thermal stresses.

Qualitative experiments in Project Salt Vault indicated that crushed salt around the test heaters began to recompact after heating started [16, p. 188]. Very little recompaction was noted in the region immediately above the heaters. An attempt was made to monitor the rate of closure of the holes drilled to insert the heaters into the floor, but was unsuccessful. A maximum hole closure of 0.5 in. for holes originally 8-10 in. in diameter was estimated over the test period of about 2 years [16, p. 209].

The bulk of the closure in the backfilled rooms is due to the deformation of the pillars at elevated temperatures under the influence of the overburden load. For deformation within the plastic region, Poisson's ratio is about 0.5 so that pillar salt displaced in the vertical
direction flows into the room. Then pillar volume is conserved so that

Initial Pillar Volume = Final Pillar Volume \hspace{1cm} (12)

or

\[ (H)(L)(WP) = (H - \Delta H)(L + \Delta W)(WP + \Delta W) \hspace{1cm} (13) \]

where

- \( H \) is the original pillar height,
- \( L \) is the original pillar length,
- \( WP \) is the original pillar width,
- \( \Delta H \) is the change in pillar height, and
- \( \Delta W \) is the change in pillar width.

The rooms and pillars have the same length and height. Since the length of a pillar (300 ft) is much greater than its width (30-50 ft), end effects are neglected. Then for complete reconsolidation to occur, the entire volume of the room must be accounted for as follows

Room Volume = Backfill + Ceiling Drop + Pillar Expansion \hspace{1cm} (14)

\[ (H)(L)(WR) = (BFILL)(H)(L)(WR) + (\Delta H)(L)(WR) + (H - \Delta H)(L)(\Delta W) \hspace{1cm} (15) \]

where

- \( WR \) is the original room width and
- BFILL is the fractional crushed salt backfill or one minus the void fraction of the crushed salt.

Values for the required \( \Delta H \) resulting from the solution of the simultaneous equations (13) and (15) would be most accurate for a pillar
with shale layers at the top and bottom. The shale restricts flow of salt from the pillar into the floor and roof of the room. Ratios of $\Delta W/\Delta H$ predicted by the solution of equations (13) and (15) range from 2-3.5. Because it is assumed that all of the displaced pillar salt flows into the room, the predicted $\Delta H$ is the lowest physically possible and the resultant closure time is the shortest.

However, measurements of pillar deformation at ambient temperature in the Hutchinson and Lyons mines indicate that $\Delta W/\Delta H$ ratios are almost always unity or slightly below [16, pp. 30-37]. The results are attributed to the flow of salt from the pillar into the floor and roof of the room, producing a nearly uniform vertical convergence all the way across the room. This conclusion was verified by later model pillar studies [16, pp. 280-281]. However the effect may not be as pronounced at elevated temperatures due to decreased pillar spall. If $\Delta W = \Delta H$, then the required $\Delta H$ can be determined directly from equation (15) accounting for the total room volume. This results in a high value for $\Delta H$ and the longest predicted closure time.

Once a range of values for the required $\Delta H$ is determined, a range of times required for reconsolidation can be predicted by the use of an appropriate strain rate equation for rock salt pillars. Lomenick has performed extensive laboratory pillar model experiments on rock salt from the Lyons mine [16, pp. 257-290]. Models were fabricated to simulate pillar, roof, and floor conditions that would exist in the mine itself. In accordance with the results of earlier investigators, the size of the pillar models with respect to grain size of the rock salt and the amount of salt above and below the pillars was selected.
so as to be representative of actual mine conditions and to yield reproducible results. Tests on cylindrical pillars with a width to height ratio of 4 at loads of 2,000-10,000 psi, temperatures of 22.5-200 °C, strains up to 0.30 in./in., and times up to 1,000 hours yielded the following strain rate equation

\[ \dot{e} = 0.39 \times 10^{-37} T^{9.5} \sigma^{3.0} t^{-0.70} \]  

(16)

where

\( \dot{e} \) is the strain rate or vertical shortening of a pillar (in. in. \(^{-1}\) hr\(^{-1}\)).

\( T \) is the absolute temperature (°K),

\( \sigma \) is the average pillar stress (psi), and

\( t \) is the time (hr).

The strain rate depends strongly on temperature and stress and decreases with increasing time.

The use of this equation in predicting time required for closure should yield conservatively long estimates for several reasons. The model pillars used did not contain shale partings. Tests on other model pillars containing simulated shale partings indicate that the partings make the pillar weaker and accelerate deformation. Rectangular pillar models were also found to be weaker than their cylindrical counterparts.

In tests of model pillars with width to height ratio less than 4, it was found that pillars with \( WP/H = 3 \) deformed at a rate of approximately 1.43 times that for the pillar with \( WP/H = 4 \), while pillars with \( WP/H = 2 \) deformed at a rate of approximately 1.73 times that for
the pillar with WP/H = 4. This represents an approximately linear increase in strain rate with decreasing WP/H within the range of WP/H from 2 to 4. These acceleration factors were taken into account for the pillar geometries considered.

Laboratory studies have indicated that in salt mines, pillars should have width to height ratios of at least 2 to avoid the possibility of sudden catastrophic failure, even under shock loading [14, p. 25], so that

\[ \frac{WP}{H} \geq 2 \]  

But the room height must be at least 15 ft to provide clearance for machinery so that

\[ WP \geq 30 \text{ ft} \]  

In bedded salt, a good rule of thumb for maximum room width is 50 ft so that

\[ WR \leq 50 \text{ ft} \]  

It is desirable to have a fractional extraction (EXT) greater than 0.5 to make good use of available mine area so that

\[ \text{EXT} = \frac{WR}{WP + WR} \geq 0.5 \]  

or

\[ WR \geq WP \]  

Combining equations (18), (19), and (21) results in reasonable limits on pillar and room widths as follows

\[ 30 \text{ ft} \leq WR \leq 50 \text{ ft} \]
The void fraction of the crushed salt used to backfill the rooms will range from about 0.27 for a mixture of fine and medium crushed salt to about 0.37 for medium crushed salt [48, p. 26]. An average value of 0.32 is used in these calculations resulting in a BFILL value of 0.68.

Heroy has calculated a value of 730 psi for the overburden load in the Lyons mine [45, p. 2]. Then the average pillar stress is given by

\[ \sigma = \frac{730}{(1 - \text{EXT})} \text{ psi} \]  

A representative temperature distribution for the pillar centerline is given in Fig. 6. Temperature values for times beyond 45 years after waste insertion are not available since computer costs are prohibitively expensive. The temperature exhibits a very broad maximum, decreasing very slowly after 45 years, so that the temperature may be assumed constant beyond 45 years at the value calculated for time of 45 years. This should be a reasonable assumption for times on the order of 100 years.

Predicted ranges of time required for mine closure for various room and pillar widths are presented in Table X. The computer program (CLOS) and the numerical methods employed are discussed in Appendix A. For each geometry, estimates of the required \( \Delta H/H \) are made for the case where pillar volume is conserved \( (\Delta W \neq \Delta H) \) and the case where the pillar expands into the room, roof, and floor \( (\Delta W = \Delta H) \). Equation (16)
Fig. 6. Representative temperature at pillar centerline as a function of time since waste insertion [33, p. 43].
Table X. Prediction of range of times required for reconsolidation of crushed salt in a backfilled room for several extraction patterns.

H = 15 ft
L = 300 ft
BFILL = 0.68
Overburden load = 730 psi
Temperature distribution from Fig. 6

<table>
<thead>
<tr>
<th>WP (ft)</th>
<th>WR (ft)</th>
<th>EXT</th>
<th>σ (psi)</th>
<th>ΔW (ft)</th>
<th>ΔH (ft)</th>
<th>ΔW/ΔH</th>
<th>ΔH/H</th>
<th>Time (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>30</td>
<td>0.500</td>
<td>1460</td>
<td>5.45</td>
<td>2.53</td>
<td>2.15</td>
<td>0.169</td>
<td>57</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.47</td>
<td>3.47</td>
<td>1.00</td>
<td>0.231</td>
<td>84</td>
</tr>
<tr>
<td>30</td>
<td>40</td>
<td>0.571</td>
<td>1703</td>
<td>6.35</td>
<td>2.88</td>
<td>2.20</td>
<td>0.192</td>
<td>41</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.75</td>
<td>3.75</td>
<td>1.00</td>
<td>0.250</td>
<td>53</td>
</tr>
<tr>
<td>40</td>
<td>40</td>
<td>0.500</td>
<td>1460</td>
<td>7.16</td>
<td>2.57</td>
<td>2.79</td>
<td>0.172</td>
<td>66</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.75</td>
<td>3.75</td>
<td>1.00</td>
<td>0.250</td>
<td>111</td>
</tr>
<tr>
<td>30</td>
<td>50</td>
<td>0.625</td>
<td>1947</td>
<td>7.04</td>
<td>3.13</td>
<td>2.25</td>
<td>0.209</td>
<td>31</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.93</td>
<td>3.93</td>
<td>1.00</td>
<td>0.262</td>
<td>38</td>
</tr>
<tr>
<td>40</td>
<td>50</td>
<td>0.556</td>
<td>1642</td>
<td>8.86</td>
<td>2.84</td>
<td>2.84</td>
<td>0.189</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>3.93</td>
<td>3.93</td>
<td>1.00</td>
<td>0.262</td>
<td>72</td>
</tr>
<tr>
<td>50</td>
<td>50</td>
<td>0.500</td>
<td>1460</td>
<td>8.82</td>
<td>2.61</td>
<td>3.38</td>
<td>0.174</td>
<td>85</td>
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<td></td>
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<td>3.93</td>
<td>3.93</td>
<td>1.00</td>
<td>0.262</td>
<td>159</td>
</tr>
</tbody>
</table>

is then integrated to find the time corresponding to the required ΔH/H.

For a given temperature distribution, the time required for closure is largely a function of the extraction pattern since WP and WR determine the required ΔH/H and the average pillar stress. For constant pillar width, the required ΔH/H increases as the room width increases.
since there is a greater room volume to be filled. However this does not result in an increased closure time because an increased room width means a higher fractional extraction and hence a higher average pillar stress. It is noted that the increased pillar stress predominates and required closure times decrease.

For constant room width, the required $\Delta H/H$ decreases with increasing pillar width in the case where pillar volume is conserved ($\Delta W \neq \Delta H$) and remains constant in the case where the pillar volume is not conserved ($\Delta W = \Delta H$). In the former case, increased pillar size implies that a smaller amount of vertical deformation is required to cause an equivalent amount of salt to flow into the room. In the latter case, the required $\Delta H/H$ is independent of the pillar size since $\Delta W = \Delta H$. In both cases, however, the effects of decreasing pillar stress due to increased pillar size and decreasing strain rate due to increased WP/H predominate and required closure times increase. The rate of increase in closure time is lower in the case where pillar volume is conserved since the decrease in $\Delta H/H$ opposes the effects of decreasing pillar stress and strain rate.

Choosing an extraction pattern is largely a tradeoff between the desire for mine stability during operation and the desire for quick reconsolidation thereafter. The current ORNL concept calls for a mine lifetime of the order of 25 years [33, p. 32]. Therefore extraction patterns leading to closure times in the range of 50-85 years would be reasonable. The calculations indicate that this is feasible for the case of WP = 30 ft and WR = 30 ft and the case of WP = 40 ft and WR = 50 ft. There are other combinations within the range of
suggested dimensions which would also satisfy the above requirement. Thus, to provide continuous containment of the waste, container lifetimes of the order of 50-85 years are required, depending on the extraction pattern.
V. MATERIALS FOR CONTAINMENT

A. Corrosion

Fontana [41, Ch. 3] has classified corrosion into eight forms based on the appearance of the corroded metal. Of these eight forms, uniform or general attack, pitting, intergranular corrosion, and stress corrosion cracking are significant in the storage of high-level wastes in salt. Uniform attack is most accurately reported in mils per year (mpy). A general rule of thumb is that the wall thickness should be twice the thickness that would give the required life. Thus for a nominal wall thickness of 250 mils, corrosion rates yielding the desired lifetime of 50-85 years would range from about 2.5 down to 1.5 mpy. Pitting is an extremely localized form of attack resulting in holes in the metal. In general the chlorides of the oxidizing metal ions (CuCl$_2$, FeCl$_3$) are the most aggressive pitters with the nonoxidizing metal ions (MgCl$_2$, NaCl, CaCl$_2$) causing pitting to a much lesser degree. Where long lifetimes are required, materials which exhibit pitting in the environment under consideration should be avoided. Intergranular corrosion is a form of localized attack at and adjacent to the grain boundary with relatively little corrosion of the grains. It may be caused by impurities or by enrichment or depletion of alloying elements in the grain boundary area. Since the alloy in question loses strength and may disintegrate, alloys which may become susceptible to intergranular corrosion during processing or storage should be avoided. Stress corrosion cracking is the spontaneous failure of a metal resulting from the combined effects of tensile stress and a specific
corrosive medium. Materials which exhibit stress corrosion cracking in the mine environment should not be used for waste containers.

Container history during processing and solidification must be taken into account when evaluating possible later performance in the mine. More severe service conditions with respect to corrosion are experienced when the final storage container also serves as the processing vessel due to the higher temperatures involved as in the case of pot calcination or the in-pot melting variation of spray solidification. Other factors being equal, melts are more corrosive than are calcines, probably because melt fluidity allows diffusion of reactants to the wall and of products away from the wall more readily than is true with the calcine. The severely corrosive environment is offset somewhat by short process times of less than 100 hours. It should be noted that air oxidation of mild steels above 500 °C and of stainless steels above 800 °C may be orders of magnitude above waste corrosion effects and this is the primary basis used for materials selection in solidification processes [87, p. 8.20]. In the Waste Solidification Engineering Prototypes (WSEP) where all processes except the fluidized bed process have undergone engineering scale tests, container sizes and heat content are restricted so that the maximum container wall temperature after processing is 427 °C when cooled by natural air convection [87, p. 8.3].

ORNL tested austenitic stainless steels type 347, 310, and 304L for the pot calcination processing vessel and final container [22, p. 74]. Types 347 and 310 were slightly but consistently inferior to type 304L.
Corrosion of 304L is less than 100 mpy during processing and negligible thereafter [87, p. 5.18].

At Idaho, corrosion of storage vessels by fluidized bed calcine maintained below the maximum stable temperature has been found to be negligible and a straight chrome stainless steel type 405 was selected for the large storage bins [56, p. VI-3]. These have performed successfully. At the Midwest Fuel Recovery Plant, General Electric will use a "corrosion-resistant steel" [42, p. V-15-1].

For storage of the spray melter product, mild steels are adequate for wall temperatures less than 600 °C and 300 series stainless steels are adequate for wall temperatures less than 900 °C [87, p. 6.16]. In the in-pot melting variation, the process is temperature limited (T < 950 °C) because of potential corrosion of the pot. This limitation appears to be reasonable and type 304L is adequate [11, p. 3.27].

Standard coupon samples of types 304L and 310 stainless steel and of mild steel have been exposed to phosphate glass melts [37, pp. 8.20-8.21]. Again the limiting factor appears to be oxidation and mild steels are adequate for wall temperatures less than 500 °C and the stainless steels for up to 800 °C.

In the WSEP, pot wall corrosion has been below detection limits [67, p. 82]; and corrosion is not a problem in the storage of the fluidized bed calcine. Assuming a reasonable degree of process control (temperature and composition), corrosion with respect to the solidified product should be minimal.

However, there is a very serious potential problem with regard to long term storage which has been overlooked. The austenitic stainless
steels become sensitized or susceptible to intergranular corrosion when heated in approximately the temperature range 400-800 °C. For example a procedure to intentionally sensitize is to heat at 650 °C for 1 hr. The almost universally accepted theory for intergranular corrosion in austenitic stainless steels is depletion of Cr in the grain boundary area due to carbide precipitation. While 304L is suitable for welding, it is not recommended for extended service within the sensitization range. The results of Binder et al. [8] indicate that even 304L would become sensitized if heated in the range 600-850 °C for several hours. For complete immunity carbon content must be below 0.015-0.020% or carbon contents as high as 0.03% are permissible if a stabilizer is added. Sensitization is of most concern in the pot calcination and in-pot melting variation of spray solidification since containers are heated to about 850-950 °C and allowed to cool slowly through the sensitization range. A post-processing heat treatment of 950-1150 °C to redissolve the carbides, followed by quenching, would not be practical due to waste instability. Thus the possibility of sensitization to intergranular corrosion during processing must be taken into account when selecting containers for long term storage.

It is well known that moist chlorides at high temperatures represent a very severe service environment for most materials. However, prior to Project Salt Vault, it was thought that the salt mine environment would be relatively dry. Laboratory tests were carried out at ORNL to determine the suitability of various metals for use in Project Salt Vault [23]. The test environment consisted of ground natural salt (or reagent grade salt plus contaminants present in
Kansas rock salt) in air. Welded coupons along with the chemicals were sealed in quartz tubes and heated at the test temperatures of 200-500 °C for 30 days. Stainless steels 304L, 310, and 347, Hastelloy C, Haynes 25, Titanium 45A, carbon steel, Inconel, Inor-8, Monel, nickel, Nionel, and type 1100 aluminum were tested. Carbon steel exhibited the highest corrosion rates (1.0-3.8 mpy) and the most extensive pitting. Corrosion rates for all other metals were consistently less than 1 mpy. Only Hastelloy C, Titanium 45A and nickel exhibited effective pitting resistance. It was concluded from this testing program that type 304L stainless steel was the most economical and satisfactory material and that it should be used for all critical components where contact with salt could cause corrosion problems. This decision was made in spite of the fact that 304L exhibited pitting in a large number of test cases.

In preliminary high temperature heat experiments conducted in the Hutchinson mine, several thermocouples failed due to scaling and intergranular penetration of the type 304 sheath [16, pp. 40-41]. Type 304L heater cans also exhibited scaling and intergranular penetration when heated within the sensitization range for 33 days.

Welded specimens of austenitic stainless steel were attached to pots of simulated waste and buried in the floor of the Lyons mine [23, p. 30]. They were heated to an average temperature of 575 °C for 31 days. Corrosion rates for types 304, 304L, 316, and 347 ranged from 0.45-1.3 mils/month (5.4-15.6 mpy). All of the samples exhibited weld attack, type 316 exhibited deep pits and intergranular corrosion, and type 347 exhibited stress corrosion cracking.
During Project Salt Vault, cylindrical test heaters, fabricated from carbon steel and stainless steel type 304L, were placed in holes in the Lyons mine which were backfilled with crushed salt in order to simulate actual waste storage conditions. Temperatures ranged from 124-400 °C over test periods of 3-16 months. An examination of the test heaters was carried out by Kegley and Empson [52]. Since more moisture (brine inflow) was encountered than had been expected, the carbon steel outperformed the type 304L. This is not surprising in a moist chloride environment.

Carbon steel heaters exhibited the typical rust scale and irregularly corroded surfaces. There was no evidence of penetration. However, an unheated carbon steel pressure conduit pipe above one of the heaters was corroded very severely. Even though carbon steel did not exhibit catastrophic failure, it is unlikely that carbon steel containers would maintain their integrity beyond a few years in an actual waste storage situation.

The most severe corrosion of type 304L test heaters occurred in the unheated sections. Transgranular cracks, characteristic of stress corrosion, penetrated the wall completely in several instances. Extensive pitting was evident in both heated and unheated sections with a maximum reported depth of 0.4 mm (16 mils). One specimen exhibited a localized form of grain boundary attack and another had a cracked weld. Severe cracking and pitting were attributed to water condensate containing chloride ion around the unheated sections. However, severe pitting also occurred in the heated sections and transgranular cracks penetrating halfway through the wall were found in the middle of a
type 304L heater which was continuously maintained at 150-200 °C for about 350 days. These cracks cannot be attributed to condensate formation and thus imply that a chloride-stream atmosphere is sufficient to cause stress corrosion failure in type 304L. It appears that type 304L containers could not be counted on to maintain their integrity beyond a year after emplacement.

It is clear, then, that carbon steel, type 304L, and the austenitic stainless steels in general will not yield lifetimes as required to restrict radionuclide migration. In addition, the extreme difficulty encountered in removing the test heaters from their holes due to container failure and recompaction of salt around the containers [16, p. 130] is an indication of the problems which could be expected in recovering an actual waste container which has failed.

B. Methods and Materials to Overcome the Corrosion Problem

Several methods have been considered in the present study in an attempt to overcome these corrosion problems. Alteration of the environment was the first method considered. High temperatures, chlorides, and oxygen cannot be avoided in this application. Inhibitors might prove effective for a short time but reliance on inhibitors is questionable, especially for extended periods. Generally, inhibitors lose their effectiveness rapidly as the temperature and concentration increase.

The use of a drying agent is a second alternative which could reduce the environment to that which was originally anticipated, a
relatively dry salt. As a desiccant, quick-line (CaO) is particularly attractive since it has been used to a great extent in brine purification to remove Mg and Fe salts [51, p. 203]. Soluble Mg and Fe salts are precipitated as insoluble hydroxides yielding CaCl₂ which is itself often used as a desiccant. Again, however, a review of the literature on drying agents indicates that this mechanism could not be relied upon beyond a few years. In addition, desiccants lose their effectiveness rapidly as the temperature increases to the level considered here. The presence of a drying agent might also interfere with the recrystallization of the crushed salt around the containers.

At ORNL some consideration was given to the use of protective coatings, but no tests were ever carried out [36]. The main drawback of protective coatings is that they must be perfect to provide protection. Considerable remote handling is involved in transporting waste containers and the potential for coating damage would be high. Kaufmann has pointed out that abrasion is a major materials problem in salt mines [51, p. 484].

With these alternatives eliminated, the present study was extended to determine if there are any materials commonly available in tubular form which would provide sufficient corrosion resistance to serve as container materials or as secondary barriers at the time of insertion into the mine. With regard to metals, it appears that satisfactorily low overall corrosion rates can be achieved, with catastrophic failure due to pitting and stress corrosion cracking being the main limitation.

Tuthill and Schillmoller have written a summary of guidelines used in the selection of materials for use in marine atmospheres [92].
In quiet seawater, only titanium and nickel-chromium-high molybdenum alloys such as Hastelloy C were immune to pitting. This is in agreement with earlier tests at ORNL [23, p. 29] in a salt environment. Several excellent review articles on pitting have been published [44, 53, 90]. All are in agreement that chloride environments at high temperatures lead to pitting of most metals. As a class, the stainless steel alloys are more susceptible to damage by pitting than any other group of metals or alloys. Alloying additions of nickel and especially chromium and molybdenum are effective in increasing pitting resistance.

Logan [63, pp. 5-7] has given a long list of environments in which metals and alloys have been reported to undergo stress corrosion cracking. Austenitic stainless steels, aluminum alloys, and titanium alloys would be expected to stress corrosion crack in the waste storage environment. In Ni-Cr alloys, additions of Ni have been found to be most effective in preventing stress corrosion. Lee and Uhlig [58] and Copson [30] have reported that above 45% Ni these alloys become essentially immune to stress corrosion cracking as tested in boiling 42% MgCl\(_2\) (154 °C). Additions of Co and B have also been found to be beneficial [93]. Staehle et al. [89] have conducted extensive tests on the effect of alloy composition on stress corrosion cracking in Fe-Cr-Ni base alloys. They verified the deleterious effect of the group VA elements (N, P, As, Sb, Bi) and concluded that of the unfavorable alloying additions Mo had the least effect. It should be pointed out that they were able to induce stress corrosion failure of 0.38 mm wire specimens which contained greater than 50% Ni and small additions of Mo (1.5 atomic %). These specimens were stressed
to 90% of yield strength in boiling 45% MgCl₂ (154 °C) and failed after about 15,000 min (250 hr). While these results indicate that nickel-chromium-high molybdenum alloys such as Hastelloy C may not be completely immune to stress corrosion cracking in a chloride environment, the test conditions were certainly much more severe than would be expected in the mine. In fact, Leonard [60] has reported that stressed specimens of Hastelloy C and its low carbon form C-276 exhibited no cracks after 1000 hr in boiling 42% MgCl₂. Stainless steel type 304 cracked within 2 hr in the same test.

The approximate chemical composition of Hastelloy C is: Ni, 54%; Mo, 16%; Cr, 15.5%; Fe, 5.5%; W, 4%; Co, 2.5% maximum; C, 0.08% maximum; and others, 3% maximum. Its high Mo content along with Ni and Cr yield excellent pitting resistance, while its high Ni content along with a moderate addition of Co reduce susceptibility to stress corrosion cracking. This alloy should also exhibit suitably low overall corrosion rates. Hastelloy is the fuel container for SNAP-21 and as part of the materials evaluation program was exposed to hot seawater vapor (200 °C) for 200 days [54]. The maximum observed corrosion rate was $7 \times 10^{-4}$ mpy and no pitting was reported. In a more closely related application, Hastelloy C was tested for possible use as a heat exchanger tube in the first effect of a brine evaporation plant [91]. Three inch square specimens with one corner bent at 90° to cause an obstruction to flow and a stressed condition were exposed at 225 °F (107 °C) to a standard brine containing MgCl₂, CaCl₂, and NaCl in solution and to a highly alkaline brine containing Na₂SO₄, Na₂CO₃, NaOH, and NaCl in solution. Exposure to the alkaline brine gave no
indication of any corrosion. A weight loss of 0.1 mg dm$^{-2}$ day$^{-1}$ (0.016 mpy) was reported. Standard brine imparted a high polish to the coupon and a weight loss of 5 mg dm$^{-2}$ day$^{-1}$ (0.8 mpy) was reported. Kaufmann noted that Hastelloy C has given excellent service in salt plants with brines at all temperatures, with pH from 2.5-12.0 [51, p. 489].

Leonard investigated the thermal stability of Hastelloy Alloys C and C-276 [59]. Alloy C-276 is a low carbon (0.02% maximum) form of Hastelloy C. Under certain severe conditions, Alloy C has occasionally exhibited intergranular attack in the weld heat-affected zone due to precipitation of second phases at the grain boundaries. Time-temperature-transformation curves indicate that grain boundary precipitates can form in Alloy C-276 when it is exposed to temperatures of 650-1100 °C but that times required to form these precipitates are at least 30 times longer than for Alloy C. Thus Alloy C-276 can be used in the as-welded condition. However Alloy C-276 would not be recommended for use in the pot calcination or in-pot melting variation of spray solidification. In these cases it could be considered for use as a secondary container at the time of insertion into the mine.

In the phosphate glass, spray melter, and fluidized bed processes Hastelloy C-276 would perform quite well due to its high oxidation resistance up to 1050 °C. Also its low overall corrosion rates and resistance to catastrophic failure would yield the desired lifetime in a waste storage situation. Alloy C-276 is commonly available in tubular form and exhibits good formability, weldability, and machinability.
The other class of materials considered for service as containers or secondary barriers are the ceramics. These will be discussed in the following order: glasses, technical ceramics, and concretes.

Of the glasses, it would be desirable to use a corrosion resistant borosilicate glass; but these glasses are not recommended for normal service above about 250 °C and also exhibit poor thermal stress resistance [31]. A suitable glass for higher temperatures is the 96% silica glass which can be used up to 900 °C and has excellent thermal stress resistance and good chemical stability. However, communications with A. Flood of Corning Glass Works indicate that even this glass would not be practical for use as a waste container [40].

First, there is a problem in matching cements to the low thermal expansion coefficient \(8 \times 10^{-7} \text{ in. in.}^{-1} \text{ °C}^{-1}\) of 96% silica glass. Second, the dimensions required for waste containers are beyond drawing capabilities and thus would require hand blown pieces with a maximum length of about 2 ft. The pieces would then require chemical treatment to leach out non-silica constituents. The pieces would have to be joined using a cement seal to form a waste container of reasonable length. Such joints are notably weak in shear. Moreover, the total materials and fabrication costs would be prohibitively expensive and the final product would be very fragile.

Of the technical ceramics, high alumina ceramics have become the principal industrial ceramic materials since they as a class have characteristics of hardness, dimensional stability, strength at elevated temperatures, virtual chemical inertness, and oxidation resistance up to their melting point. In addition they are more
readily available and more attractive economically. Gitzen has published a comprehensive survey on alumina as a ceramic material [43]. Alumina ceramics are comparatively inert to all acids except those containing the fluoride ion. Communication with D. Briggs of the Research Division of Coors Porcelain Company indicates that a high Al₂O₃ content is desirable since corrosive agents in the mine would tend to attack any second phase (predominantly silica) which is present [18]. In this regard Coors AD-998 (99.8% Al₂O₃) would be quite suitable since it is readily available as cast in tubular form and contains essentially no second phase.

The bulk of chemical stability experiments with alumina have been published in the German literature and deal primarily with resistance to inorganic acids. Winzer reported that a 30 x 35 mm Al₂O₃ crucible lost no more than 0.02% by weight when exposed to boiling concentrated HCl [97]. Results of Dawihl and Klingler indicate that sintered alumina bodies containing 3% silicates or 0.2% magnesia were far more resistant to HCl than titanium, cast silicon, or Cr-Ni steels [34]. The high alumina ceramic containing 0.2% MgO was virtually inert in boiling 20% HCl, while that containing the silicates showed only slightly higher weight loss. Thus AD-998 should perform quite well as a storage container, yielding lifetimes beyond those required for salt reconsolidation.

Concretes are of interest because, if suitable, they would provide a relatively inexpensive form of protection for waste containers within the mine. Aluminous concretes, because of their low permeability and sulphate resistance, have been proposed for use in plugging holes
and wells near the salt formation to prevent ingress of water. Aluminous concrete is noted for its resistance to chemical environments similar to that expected during waste storage, that is, seawater, chlorides, sulphates, solutions containing $\text{Mg}^{2+}$ ion, and dilute acidity [7]. For this reason an aluminous concrete with a high thermal conductivity aggregate such as SiC has been mentioned as a possible means of protecting waste containers.

Information in the literature suggests that it might be possible to select an aluminous concrete which would provide the chemical stability and low permeability within the temperature range of 250-350 °C which is required in this application. However several reservations must be expressed.

Aluminous cement relies on three factors for its chemical stability: the absence of calcium hydroxide, the presence of aluminum hydroxide, and the chemical stability of the aluminates. However the compounds $\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 10\text{H}_2\text{O}$ and $2\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 8\text{H}_2\text{O}$ produced at ordinary temperatures are metastable and convert to the cubic compound $3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 6\text{H}_2\text{O}$ at temperatures above 50-100 °C [57]. This conversion leads to increased porosity and an accompanying loss of strength and lower chemical stability. It has been found that lower water to cement ratios can be used to counter the adverse effect of increased porosity upon conversion. Robson has noted that the use of water to cement ratios less than about 0.4 results in a porosity of less than 7% in the fully converted concrete, which is equivalent to the porosity in a normally cured aluminous concrete with a water to cement ratio of about 0.60 [84, p. 108]. However, refractory aluminous
concretes placed by vibration with the lowest water to cement ratios exhibit less spalling resistance because of their less porous internal structure [84, p. 220]. Budnikov has reported that additions of 25% insoluble anhydrite (CaSO₄) to aluminous cements can avoid the unfavorable effects of conversion [19]. The anhydrite reacts with aluminates to form stable calcium sulfoaluminate before the mass has set and the volume has stabilized. This anhydrite aluminous cement retains the characteristics of strength, low permeability, and chemical stability of the unconverted aluminous cement.

Eusner and Shapland [38] tested the permeability of carbon brick, mullite, super-duty fireclay, high-duty fireclay, and refractory concretes made with aluminous cements. The rate of gas flow through a 2 in. cube was measured in three directions. Of the refractories tested, the refractory concretes were the most porous (26-40%) but the least permeable (0.2-43 centidarcys). The mean permeability for 85 refractory castable samples was 4.5 centidarcys. The lowest permeability for a particular refractory concrete was obtained at the trowel-spading consistency. The low values of permeability persist to a temperature of about 850 °C where the permeability begins to increase markedly. This data suggests that permeability is not directly related to porosity in refractory castables.

Thus it may be possible through proper choice of mix proportions, the use of additives, or special surface treatments to obtain an aluminous concrete which would be suitable for use in this application. Many variables are involved, however, and no prediction of container lifetimes provided by encasement in concrete can be made at this point.
Considerable laboratory and in situ testing with different trial batches would be necessary. An aluminous cement with a SiC aggregate is included as an example in the heat transfer and economic considerations presented here because it does show some promise and as a means of illustrating the economic incentive for developing a less expensive form of container protection such as a concrete.

C. Mechanical and Physical Properties

Typical mechanical and physical properties of the materials under consideration are given in Table XI. Strength requirements for containers in a waste storage situation are not high. The crushed salt backfill around the containers is more than adequate to accommodate the thermal expansion of the salt as it is heated initially. A moderate amount of strength is desirable and this should be no problem for the materials considered here.

Steady state temperature calculations indicate thermal gradients of at most 2-3 °C/cm will exist across the walls of waste containers during storage so that thermal stresses will not present a problem. Robson has noted that in practical use refractory aluminous concretes show a characteristically good resistance to thermal shock which has also been verified in laboratory heating and quenching tests [84, p. 219]. As noted earlier, however, this depends on the porosity of the concrete. AD-998 has only fair thermal shock resistance. This should not present a problem in this application since the only time a large temperature change will be encountered is when the waste is initially inserted.
Table XI. Typical mechanical and physical properties of materials for use in waste storage (values are for room temperature unless otherwise noted)

<table>
<thead>
<tr>
<th>Property</th>
<th>Units</th>
<th>Hastelloy C-276 [20]</th>
<th>Al₂O₃ (AD-998) [29]</th>
<th>Aluminous concrete</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melting point</td>
<td>°C</td>
<td>1323-1371</td>
<td>&gt; 1900</td>
<td></td>
</tr>
<tr>
<td>Density</td>
<td>g/cm³</td>
<td>8.90</td>
<td>3.82</td>
<td>2.38</td>
</tr>
<tr>
<td>Coefficient of thermal expansion</td>
<td>10⁻⁶/°C</td>
<td>15.9 (24-871 °C)</td>
<td>3.0 (25-300 °C)</td>
<td>Linear change (+ 0.19%) (104-871 °C)</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>W/(m·°C)</td>
<td>13.0 (204 °C)</td>
<td>29.4 (25 °C)</td>
<td>2.101 (456 °C)a</td>
</tr>
<tr>
<td></td>
<td></td>
<td>21.0 (649 °C)</td>
<td>23.1 (100 °C)</td>
<td>2.353 (747 °C)a</td>
</tr>
<tr>
<td>Compressive strength</td>
<td>10³ psi</td>
<td>--</td>
<td>&gt; 300</td>
<td>6.3a</td>
</tr>
<tr>
<td>Tensile strength</td>
<td>10³ psi</td>
<td>115.6</td>
<td>38</td>
<td>0.35-0.45b</td>
</tr>
<tr>
<td>Modulus of elasticity</td>
<td>10⁶ psi</td>
<td>29.8</td>
<td>50</td>
<td>6.5-7.5b</td>
</tr>
<tr>
<td>Poisson's ratio</td>
<td></td>
<td>0.30</td>
<td>0.22</td>
<td>0.20b</td>
</tr>
</tbody>
</table>

\(^a\)Concrete resulting from 1 part Lumnite to 3 parts SiC (8 mesh-dust), 5 gallon/sack water to cement ratio, 24 hr moist cure + 24 hr in lab air + 18 hr at 220 °F (104 °C) [12].

\(^b\)Typical values for aluminous concrete [84].
into the container. At this time it would be a simple matter and relatively inexpensive to use a secondary heater to gradually bring the container up to waste temperature.

Because of the brittleness, general lack of toughness, and susceptibility to thermal shock of ceramics, they are more suited for use as secondary containers at the time of insertion into the mine where a minimal amount of handling and transport is required. Thus AD-998 and aluminous concrete will only be considered for use as secondary containers. Hastelloy C-276 will be considered for use as the sole container from time of solidification on.

D. Radiation Stability

While the radiation levels near the containers are relatively high, they are certainly not as high as those encountered in reactor service. Even assuming that the radiation level remained constant at its 10 year level as calculated for the model waste and presented in Table VII, the total exposure to a container in 100 years would be $2.33 \times 10^{20}$ Mev/cm$^2$ for gamma radiation and $5.0 \times 10^{13}$ fast neutrons/cm$^2$.

Nickel and nickel-base alloys such as Hastelloy C have shown potential as structural and containment materials in nuclear reactors. Changes in properties of Hastelloy C and other nickel base alloys are usually reported only after exposures of $10^{18}$-$10^{21}$ neutrons/cm$^2$ and even then changes are not extreme [88]. Likewise gamma ray exposure at these levels would not be significant.
The threshold of damage in ceramic materials of moderate stability appears to occur at about $10^{17}$ neutrons/cm$^2$, although property changes are small for integrated fluxes up to $10^{18}$ neutrons/cm$^2$ [13]. Irradiation at $3 \times 10^{18}$ neutrons/cm$^2$ causes no permanent effects on the properties of aluminum oxide. Gamma-induced defects are usually annealed out at 200-400 °C [43, p. 99]. Therefore AD-998 should be very stable in this environment with regard to radiation damage.

No data is available on the performance of aluminous concretes under irradiation. However Al$_2$O$_3$ and SiC are stable under irradiation at these levels and experience with other concretes in shielding applications indicates that radiation levels expected during waste storage should not be a problem.

E. Summary of Containment Systems

In summary there are three materials which show promise for providing extended container lifetimes as required to prevent radionuclide migration and allow for reasonable recovery. Three systems will be analyzed from the viewpoint of heat transfer and economics. A horizontal cross section of each containment system as it would appear shortly after insertion and backfilling is presented in Fig. 7.

The first is the use of a Hastelloy C-276 container beginning at the point of solidification and continuing through insertion into the mine. The second involves the use of mild or stainless steel as the initial container after solidification followed by encapsulation into a secondary alumina AD-998 container at the time of insertion into
HASTELLOY C-276

WASTE 0-3 in.
C-276 3-3.28 in.
CRUSHED SALT 3.28-5.28 in.
ROCK SALT 5.28 in.

CAST ALUMINA AD-998

WASTE 0-3 in.
STEEL 3-3.28 in.
GAP 3.28-3.5 in.
AD-998 3.5-3.75 in.
CRUSHED SALT 3.75-5.75 in.
ROCK SALT 5.75 in.

ALUMINOUS CONCRETE

WASTE 0-3 in.
STEEL 3-3.28 in.
GAP 3.28-3.5 in.
ALUMINOUS CONCRETE 3.5-7.5 in.
CRUSHED SALT 7.5-9.5 in.
ROCK SALT 9.5-

Fig. 7. Horizontal cross section of containment systems after insertion and backfilling.
the mine. The third scheme also involves an initial mild or stainless steel container, but a secondary container of aluminous concrete with a SiC aggregate.

Hastelloy C-276 and steel containers are 6 in. pipe with a nominal 0.28 in. wall thickness. Secondary AD-998 containers have a 7 in. inner diameter with 0.25 in. wall thickness. It is generally recommended that metal in concrete exposed to seawater should have a minimum cover of 3-4 in. so that a secondary aluminous concrete container with a 7 in. inner diameter and 4 in. wall thickness was chosen for this application. Outer containers are surrounded by a 2 in. layer of crushed salt backfill and then rock salt.
VI. HEAT TRANSFER ANALYSIS

A. Previously Published Results

In order to make the most efficient use of mine space, it is desirable to achieve the highest heat generation rate per unit area of mine which is consistent with design temperature limitations. Bradshaw et al. [14] have calculated spacings required for various waste heat contents based on an analytic solution of the time dependent heat transfer equation. The model used was a finite line source in the center of an infinitely long homogeneous salt cylinder with perfectly insulated boundaries. For the line source equivalent to a 6 in. diameter, 10 ft long waste cylinder, with waste aged ten years at time of burial and power densities of 1.14 to 10.35 W/liter, the corresponding center to center spacings required to keep salt temperatures below 200 °C ranged from about 4 to 13 ft. It was found that for wastes with 30 year half-lives, the time required to reach peak temperatures near the waste was of the order of 20-30 years. Wastes aged less than 5 years prior to insertion in the salt required a few thousand hours to reach peak temperatures.

Recently published results at ORNL by Cheverton and Turner involve the use of finite difference techniques [21]. Methods of formulation and solution of the difference equations have not been published by ORNL. A two dimensional model with the waste treated as a slab source in \( x-z \) geometry is presented. A cylindrical geometry two dimensional model with all waste containers treated together as a lumped source is also presented and temperature profiles outside the salt formation are
given. Of more interest here is a three dimensional model which involves two lines of equally spaced containers buried in a room. Fixed temperature boundaries are assumed at a distance of a few hundred feet above and below the waste. It is presumed that symmetric boundary conditions are used at the vertical planes through the center of the room and through the center of the pillar. One set of temperature profiles as a function of space and time is presented for the case of a 10 year old calcined waste with 2,000 W per 6 in. diameter container and 22 ft center to center spacing. It is shown that the temperature rise 8 in. from a container does not exceed 365 °F and the temperature rise midway between two containers does not exceed 328 °F. Since ambient mine temperature at the level of the waste is about 73 °F, the maximum temperatures are 438 °F (226 °C) and 401 °F (205 °C), respectively. Temperatures in the vicinity of the waste peak within 20-30 years.

B. Selection of Heat Transfer Model

In this investigation, a comparison of the three containment systems based on allowable heat generation rate per unit area of mine is sought. The heat content per container and the corresponding required container spacing determine the number of containers and the mine area required to store a given amount of high-level waste. This comparison requires the solution of the heat conduction equation for a very complicated geometry and combination of materials; and hence the use of finite difference techniques is desirable. Care must be taken
in selecting the heat transfer model so as to balance computer requirements against the accuracy of the desired results.

In this case the general heat conduction equation is

\[- \nabla \cdot (K \nabla T) + \rho c \frac{\partial T}{\partial t} = Q(t)\]  \hspace{1cm} (25)

where

\[T\] is the temperature,
\[K\] is the thermal conductivity,
\[\rho\] is the density,
\[c\] is the specific heat, and
\[Q\] is the volumetric heat release rate.

This type of problem is usually referred to as a propagation or marching problem in which mixed boundary conditions are specified for the spatial variables and the solution marches out in time as governed by these boundary conditions and the initial conditions.

There are two concerns in the use of finite difference techniques. The first is convergence, that is, whether or not the difference solution approaches the differential solution within a finite number of iterations. The second is stability, that is, whether or not any errors which are introduced into the difference solution tend to die out as the iteration proceeds. Explicit finite difference methods for the solution of parabolic equations such as (25) have a stability criterion usually expressed as

\[\frac{\Delta t}{\Delta x^2} \leq 0.5\]  \hspace{1cm} (26)

where \(\Delta t\) is the time increment and \(\Delta x\) is the spatial increment. This requires the use of small time steps. This restriction can be overcome
by the use of more complicated implicit techniques such as the Crank-Nicholson method which has unrestricted stability. However, even with this method it is suggested that $\Delta t/\Delta x^2$ be less than about 4 in order to reduce truncation errors.

For a comparison of the various containment systems, the maximum temperatures in the vicinity of a waste container are required and the detail of the materials involved must be taken into account. Since times of 20-30 years are required for these temperatures to reach a maximum, the restrictions on $\Delta t$ mentioned above mean that a very large amount of computer time would be required to solve equation (25).

Since only the maximum temperatures for the hottest container are desired, the application of the steady state heat conduction equation is possible, that is

$$- \overline{\nabla} \cdot \mathbf{K} \overline{\nabla}T = Q \quad (27)$$

For this case, the heat generation rate is not constant since the radioactive nuclides within the waste decay with a specified half-life. However, for a waste aged 10 years at time of insertion into the mine, the thermal power decays very slowly (approximately 30 year half-life) as illustrated in Fig. 3. Thus for the typical case of a waste aged 10 years, the use of a constant $Q$ equal to that at time of insertion into the mine in conjunction with equation (27) will yield an appropriately conservative estimate of the required container spacing and provide a sound basis for comparison of the various containment systems.
The problem is thus reduced to the solution of the familiar elliptic equation, similar in form to the neutron diffusion equation. This type of problem is referred to as an equilibrium or jury problem and can be solved by the use of iterative techniques. Wachspress has written an extensive treatise on the iterative solution of elliptic systems [96]. When the field of a boundary value problem depends continuously upon boundary conditions, the system is stable. Thus for a properly posed problem, the concern is the choice of an iterative technique which yields the desired rapid rate of convergence.

The minimum room width considered here is 30 ft. It is recommended that waste containers be placed at least 4 ft from the walls of the room to avoid excessive pillar heating. Thus for center to center spacings of less than 24 ft, even the minimum size room could accommodate at least three rows of containers equally spaced on triangular centers. This is equivalent to a hexagonal lattice and would make the best use of the available floor area for a given spacing. Thus the hottest container is located in the center of the room at the center of a hexagon formed by six equally spaced containers. If all the containers are assumed to have equal heat content and are placed in the room at approximately the same time, it can be assumed that no heat flows across the vertical boundaries of the hexagon formed by the perpendicular bisectors of lines drawn from the central container to the six surrounding containers. Then, as is commonly done in the solution of the neutron diffusion equation for heterogeneous reactors, this perfectly insulated hexagonal boundary can be approximated by a circle which reduces the problem to cylindrical geometry consistent
with the container geometry. The equivalent cylindrical unit cell has a radius which is 0.525 times the center to center spacing in a hexagonal lattice.

The problem can now be visualized as a waste container heat source in the center of a long cylinder, formed as if a large cylindrical core were extracted from the entire formation by drilling at the surface. A horizontal cross section of the various containment systems has been presented in Fig. 7. A vertical cross section of the model is presented in Fig. 8. The model results from the superposition of the detail of container materials and the backfilled hole and room onto the stratigraphy of the overall geologic formation. Symmetric boundary conditions (zero temperature gradient, Neumann condition) are imposed at the centerline and surface of the cylinder. Lumped source calculations performed at ORNL [21] indicate that formation temperatures 200-300 ft above and below the waste are not significantly affected during the time required for mine level temperatures to peak. Thus a constant temperature boundary condition (Dirichlet condition) is imposed at a distance of about 100 m above and below the waste. Wachspress has stated that the imposition of Dirichlet or Neumann boundary conditions for an elliptic system with a closed boundary yields a properly posed problem and a unique stable solution [96, pp. 29-31].

At the Lyons site, the geologic formation temperature has been found to vary approximately linearly as a function of depth over certain regions as illustrated in Fig. 9 [21, p. 13]. The most recent estimate of the geothermal heat flux based on experimental
Fig. 8. Vertical cross section of cylindrical unit cell used in heat transfer analysis.
CALCULATED THERMAL CONDUCTIVITIES (K) BASED ON GEOTHERMAL HEAT FLUX OF 0.02 Btu hr⁻¹ ft² = 0.063 W m⁻².

Fig. 9. Geologic formation temperatures prior to waste insertion. Temperatures from [21, p. 13].
data at the Lyons site is \(2.0 \times 10^{-2} \text{ Btu hr}^{-1} \text{ ft}^{-2}\) \((0.063 \text{ W m}^{-2})\) [21, p. 4]. Thus the effective thermal conductivities for each region are easily calculated from the Fourier heat conduction equation and are also given in Fig. 9.

Thermal conductivities as a function of temperature for the various materials involved are presented in Fig. 10. Birch and Clark have measured the thermal conductivity of single natural halite (NaCl) crystals and found that it decreases with increasing temperature as shown in Fig. 10 [9]. These measured values have been found to correspond well with samples from the Hutchinson formation [16, p. 3]. Thus, these lower values for salt were used instead of the higher value calculated for the salt and shale region as shown in Fig. 9. The thermal conductivity of crushed salt was assumed constant at a value used in previous ORNL calculations. Initial calculations with this model indicate that the results are not sensitive to small changes in thermal conductivity such as the adjustment for a change in temperature of the order of 50-100 °C. This is in agreement with earlier ORNL results [21, p. 24]. However, in the calculations presented here, care was taken to adjust thermal conductivities for materials in various regions to within at least 50 °C.

It is assumed that all heat generated by the decay of radioactive nuclides is released within the waste. Contact resistance between the waste and its container is neglected. With regard to the crushed salt backfill around the containers, experimental results indicate that contact will always exist at the container-crushed salt and crushed salt-rock salt interfaces and that the contact resistances
Fig. 10. Thermal conductivity as a function of temperature for materials used in waste storage systems.

Material:

- △ Hastelloy alloy C-276 [20]
- □ Cast alumina AD-998 [29]
- ○ Halite [9]
- ◊ Aluminous concrete [12]
- ▽ Phosphate glass waste [70, p. 6.5]
- ___ Crushed salt [33, p. 61]
- † Dry air [46, p. 577]
are small compared to the resistance of the 2 in. crushed salt annulus [32]. There are, however, two void regions which are significant. First, the containers considered here are 6 in. pipe, 10 ft in length which have a nominal active height (filled waste level) of about 8 ft. Second, for the cases of AD-998 and aluminous concrete used as secondary barriers, there is a gap between the primary steel container and the secondary container.

For the space at the top of the waste, the contribution from radiation heat transfer is negligible. However, since the density of air decreases with increasing temperature, when air is contained between two horizontal surfaces in which the upper surface is cooler than the lower one, colder air layers (more dense) are situated above warmer layers leading to an unstable situation and significant convection currents. Jakob [47] correlated the experimental results of a number of investigators and has shown that the apparent thermal conductivity \( (K_c) \) due to conduction and convection can be related to the actual thermal conductivity of the gaseous medium \( (K) \) by means of a modified Grashof number \( (N_{Gr})_L \), where

\[
(N_{Gr})_L = \frac{g \beta}{\nu^2} \frac{L^3}{\Delta T}
\]

(28)

where

g is the acceleration of gravity,
\( \beta \) is the volume coefficient of thermal expansion of the medium,
\( \nu \) is the kinematic viscosity,
\( L \) is the thickness of the horizontal layer, and
\( \Delta T \) is the temperature drop across the layer.
For values of $\left(N_{Gr}\right)_o$ greater than about $4 \times 10^5$ the apparent and actual thermal conductivities are related by

$$\frac{K_c}{K} = 0.068\left(N_{Gr}\right)_o^{1/3}$$

(29)

In this case $\left(N_{Gr}\right)_o$ is of the order $10^8$; and so for each case considered, an estimate of the apparent thermal conductivity of the space at the top of the waste was made using the properties of air and equation (29). For each case, the estimate of $K_c$ was adjusted for each computer run based on values obtained in the previous runs.

For the data reported here, a comparison of the estimated $K_c$ used in the computer run and the actual $K_c$ based on calculated results is given in Table XII.

Preliminary calculations for the gap between the primary and secondary containers indicated that the contribution from convection would be negligible but that the contribution from radiative heat transfer would be about the same order of magnitude as that from conduction and hence should not be neglected. The results of Graaf and Held [46, p. 391] indicate that convection is negligible in a vertical air layer for Grashof numbers less than 7,000. In the current calculations the largest temperature drop across this gap was 44 °C for the AD-998 secondary container. For this temperature difference and the properties of air taken at 316 °C, the Grashof number is less than 7,000 as long as the gap spacing is less than 2.87 cm. This confirms preliminary estimates that convection is negligible.
Table XII. Comparison of estimated and actual values of $K_c$ based on equation (29) and properties of air at 204 °C

<table>
<thead>
<tr>
<th>Container system</th>
<th>$Q$ W/liter</th>
<th>$\Delta T$ °C</th>
<th>$\frac{K_c}{W\ m^{-1}\ °C^{-1}}$</th>
<th>Estimated</th>
<th>Actual</th>
</tr>
</thead>
<tbody>
<tr>
<td>C-276</td>
<td>14.21</td>
<td>40</td>
<td>1.322</td>
<td>$\sim 46$</td>
<td>1.385</td>
</tr>
<tr>
<td>AD-998</td>
<td>14.21</td>
<td>40</td>
<td>1.322</td>
<td>$\sim 50$</td>
<td>1.424</td>
</tr>
<tr>
<td>Aluminous concrete</td>
<td>14.21</td>
<td>40</td>
<td>1.322</td>
<td>$\sim 45$</td>
<td>1.375</td>
</tr>
<tr>
<td>AD-998</td>
<td>21.60</td>
<td>60</td>
<td>1.514</td>
<td>$\sim 70$</td>
<td>1.593</td>
</tr>
</tbody>
</table>

In order to take radiation heat transfer into account in the finite difference scheme, it is necessary to define an effective radiation contribution to the thermal conductivity

$$K_r = F_A F_e \sigma_s (T_1^4 - T_2^4) (\Delta r/\Delta T)$$

(30)

where

- $F_A$ is the area or view factor,
- $F_e$ is the emissivity factor,
- $\sigma_s$ is the Stefan-Boltzmann constant,
- $T_1$ is the temperature of the inner surface,
- $T_2$ is the temperature of the outer surface,
- $\Delta T$ is $T_1 - T_2$, and
- $\Delta r$ is the gap thickness.

In fact, this is appropriate in the difference scheme employed since the gap region is treated as one mesh region and the temperature gradient is approximated by $\Delta T/\Delta r$. For a thin gap the concentric
cylinders are essentially finite parallel planes so that the view factor is unity and

\[ F_e = \left[ \left( \frac{1}{\varepsilon_1} \right) + \left( \frac{1}{\varepsilon_2} \right) - 1 \right]^{-1} \]  

(31)

where \( \varepsilon_1 \) and \( \varepsilon_2 \) are the emissivities of the inner and outer surfaces. Values for total emissivity were taken from McAdams [66] as follows:

- steel primary container, \( \varepsilon_1 = 0.40 \)
- alumina, \( \varepsilon_2 = 0.30 \)
- concrete, \( \varepsilon_2 = 0.63 \)

For each case the value of \( K_r \) was adjusted for each computer run based on results of previous runs. For the data reported here, a comparison of estimated and actual \( K_r \) values is given in Table XIII.

Calculations with the Hastelloy C-276 system indicated that the presence of the Hastelloy had no effect because its thermal conductivity is so much higher than the other materials involved. Since the steel primary containers used in the AD-998 and aluminous concrete systems have thermal conductivities even higher than C-276, the steel containers were neglected in the dual container systems. The space at the top of the waste and the primary-secondary container gap were accounted for as discussed above.

C. Temperature Profiles

The formulation of the finite difference equations appropriate for the physical model presented in Fig. 8, the iterative technique employed, and the computer program (HEAT) are discussed in Appendix B. The steady state heat conduction equation (27) is integrated over each mesh volume in \( r-z \) geometry. By the use of Gauss' theorem (the divergence
Table XIII. Comparison of estimated and actual values for $K_r$ based on equation (30)

<table>
<thead>
<tr>
<th>Container system</th>
<th>$Q$ W/liter</th>
<th>Estimated $K_r$ W m$^{-1}$°C$^{-1}$</th>
<th>Actual $K_r$ W m$^{-1}$°C$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>AD-998</td>
<td>14.21</td>
<td>0.040</td>
<td>0.040</td>
</tr>
<tr>
<td>Aluminous concrete</td>
<td>14.21</td>
<td>0.060</td>
<td>0.065</td>
</tr>
<tr>
<td>AD-998</td>
<td>21.60</td>
<td>0.043</td>
<td>0.049</td>
</tr>
</tbody>
</table>

Theorem), the volume integrals are converted to surface integrals which are approximated by finite differences. This yields a set of five point difference equations which can be put into a convenient matrix form and solved iteratively by means of an alternating direction implicit technique.

Calculations were performed for each containment system for the case of a 76 liter/metric ton phosphate waste buried at 10 years, that is, $Q = 14.21$ W/liter as shown in Fig. 3. For the cast alumina AD-998 system, a calculation was also performed for a 50 liter/metric ton phosphate waste buried at 10 years, that is, $Q = 21.60$ W/liter. For each case the calculation procedure was as follows:

1. choose an estimated allowable radius $R$ for the cylindrical unit cell;
2. estimate temperatures and physical constants;
3. execute computer program HEAT;
4. examine calculated temperatures to determine if physical constants correspond to the actual temperature distribution.
and if the temperatures correspond to the design temperature limitations.

This procedure was repeated several times for each case to determine the minimum allowable R consistent with temperature limitations.

The vertical position of the maximum radial temperature profile was approximately at the center of the waste container at a depth of 302.67 m. This position is about 30 cm (1 ft) above the center of the active waste section (only the lower 8 ft of the container is filled with waste). The occurrence of the maximum slightly above the midplane of the waste is reasonable due to the insulating effect of the backfilled room. Key temperatures on the maximum radial profile are given in Table XIV along with the minimum allowable R and the corresponding thermal power per unit area of mine (\(Q_M\)). The waste volume for all containers considered here is 45.45 liters so that

\[
Q_M = \frac{45.45 \times Q}{\pi R^2} \text{ W m}^{-2} \tag{32}
\]

for \(Q\) in W/liter and R in meters.

In agreement with results reported by ORNL [21], it is noted that the 200 °C temperature midway between two containers is the limiting criterion in every case. For the case of \(Q = 14.21\) W/liter, the AD-998 system yields the lowest R and highest \(Q_M\). The aluminous concrete system is second best followed by the C-276 system. The equivalent center to center spacing in a hexagonal lattice is 1.9 R and ranges from 4.94 m (16.2 ft) for AD-998 to 5.70 m (18.7 ft) for C-276.

These values are reasonable for the model used and appear to be slightly conservative when compared to earlier ORNL results as expected.
Table XIV. Key temperatures, minimum allowable $R$, and corresponding thermal power per unit area of mine ($Q_m$) for each containment system.

<table>
<thead>
<tr>
<th>Containment system</th>
<th>$Q$ (W/liter)</th>
<th>$R$ (cm)</th>
<th>$Q_m$ (W/m²)</th>
<th>Temperature, °C ($Z = 302.67$ m)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Waste centerline $r = 0$</td>
</tr>
<tr>
<td>C-276</td>
<td>14.21</td>
<td>300</td>
<td>22.84</td>
<td>317</td>
</tr>
<tr>
<td>AD-998</td>
<td>14.21</td>
<td>260</td>
<td>30.41</td>
<td>330</td>
</tr>
<tr>
<td>Aluminous concrete</td>
<td>14.21</td>
<td>290</td>
<td>24.44</td>
<td>334</td>
</tr>
<tr>
<td>AD-998</td>
<td>21.60</td>
<td>310</td>
<td>32.52</td>
<td>388</td>
</tr>
</tbody>
</table>
The difference between the C-276 system and AD-998 system is due mainly to the insulation provided by the primary-secondary container gap. The insulation causes temperatures within the waste to rise and lowers the temperatures outside the waste, thus allowing a lower minimum value of R.

It is also noted that the aluminous concrete system results are quite similar to the C-276 results even though the radiation contribution in the primary-secondary container gap is only 50% higher for aluminous concrete as compared to AD-998 (Table XIII). The reason for this is that the aluminous concrete container has a thickness of 10 cm compared to a wall thickness of about 0.6 cm for the AD-998 and C-276 containers. This increased container thickness moves the crushed salt annulus further away from the waste and reduces its effect as an insulator. This can be illustrated by considering the resistance concept for the case of radial heat flow in cylindrical geometry. The resistance of an annular region of length $L_1$, inner radius $a$, outer radius $b$, and thermal conductivity $K$ is given by

$$R_W = \frac{\ln(b/a)}{2\pi L_1 K}$$

The crushed salt annulus has the same thickness $(b-a)$ of 5.08 cm (2 in.) for all systems. But as "a" is increased from about 9.5 cm in the C-276 or AD-998 systems to 19 cm in the aluminous concrete system, $\ln(b/a)$ decreases from 0.43 to 0.24, thus decreasing the effective resistance of the crushed salt annulus by about 50%. The decrease in insulation provided by the crushed salt counters the effect of
insulation provided by the primary-secondary container gap in the aluminous concrete system.

Therefore the main factor in the determination of R for each system is the resistance to heat flow provided by regions of relatively low conductivity. As shown above, this resistance depends on the location as well as the effective conductivity of the insulating region. There is a limit to the amount of insulation which can be allowed in each case because of the undesirable effect of higher waste temperatures.

Calculations were also carried out for the AD-998 system for the case of \( Q = 21.60 \text{ W/liter} \). This result indicates that as the heat content per container is increased, the required mine area does not increase correspondingly as is illustrated by the higher value for \( Q_m \). This can be attributed to the increased area available for vertical heat transfer and the steeper temperature gradients produced by the higher heat flux. This has important economic implications as will be discussed in the next section.

Maximum radial temperature profiles for the various systems are presented in Figs. 11-13. A typical vertical temperature profile is presented in Fig. 14 for the AD-998 system with \( Q = 14.21 \text{ W/liter} \) at a radial position midway between two containers.
Fig. 11. Maximum radial temperature profile for Hastelloy C-276 system.
Fig. 12. Maximum radial temperature profiles for cast alumina AD-998 system.
Fig. 13. Maximum radial temperature profile for aluminous concrete system.
Fig. 14. Vertical temperature profile for cast alumina AD-998 system for radial distance corresponding to the position midway between two containers.
VII. COST ANALYSIS

A. Previous Cost Estimates for High-Level Waste Storage

Several cost estimates for various solidified high-level waste storage schemes have been developed at ORNL. Perona and coworkers have studied the effects of fission product removal on the cost of final waste disposal [80]. They estimated that the cost for management of wastes from which 90 to 99% of all fission products had been removed would range from 70 to 80% of those for management of waste from which no fission products had been removed. There would have to be a very large and diverse market for fission products to pay the cost of fission product separation and packaging; even then the cost of waste disposal would not be significantly reduced.

Perona and coworkers also have compared costs for storage of solidified high-level waste in surface concrete vaults, granite formations, and salt formations [78]. They concluded that storage in concrete vaults would cost five to seven times as much as storage in salt, and storage in granite would cost about twice as much as in salt.

Bradshaw and coworkers have conducted a detailed analysis of the cost of storage in salt [14]. Present-valued costs ranged from 0.003 mills/kwhr (electrical) for wastes aged 30 years at burial to 0.019 mills/kwhr (electrical) for wastes aged one year. For 6 in. diameter, 10 ft long containers buried at age 10 years, the present-valued cost ranged from 0.0056 to 0.0075 mills/kwhr (electrical). In another ORNL study [74, p. 6-37] present-valued costs for storage
in salt ranged from 0.001 to 0.011 mills/kwhr (electrical). For a 6 in. diameter container of waste aged 10 years at burial, the present-valued cost was estimated as 0.0033 mills/kwhr (electrical).

B. Costs of Mining and Waste Handling

In this investigation, costs of mining and waste handling are based on a 1969 study at ORNL [14]. The ORNL study is based on a mine which is one square mile in area and lies at a depth of 1000 ft below the surface. The mine is excavated by the room and pillar method. Rooms are 15 ft high, 50 ft wide, and 300 ft long. The salt extraction ratio is 0.5 so that the pillars have the same dimensions as the rooms. A one-square-mile mine can accommodate 552 such rooms for a net floor area of $8.28 \times 10^6 \text{ ft}^2$ ($7.69 \times 10^5 \text{ m}^2$). The concept of waste insertion and backfilling is the same as that presented here.

The costs are broken down into two categories, those which depend on the number of waste containers handled per year and those which depend on the life of the mine. Total annual cost as a function of the number of containers per year is given in Fig. 15. Costs depending on the number of containers include waste transporters, hole drillers, radiation monitors, personnel, and overhead and maintenance of the waste handling facilities and equipment. Personnel include superintendents, secretaries, health physicists, equipment operators, and maintenance men. Stepwise increases in cost are experienced whenever the number of containers reaches a level such that the number of waste transporters, hole drillers, or personnel must be increased.
Fig. 15. Total annual cost for items which are a function of the number of containers per year [14, p. 46].
Total annual cost for items which depend on the life of the mine is given in Fig. 16. These items include initial mining costs, annual mining costs (including shafts and machinery), topside waste handling facility, waste shaft, mine-level waste handling facility, purchase of land, and deactivation of the mine.

C. Cost of Materials

The cost of the final container is usually not included in the economic study of a waste storage concept. However, in this case, the container is closely related to the suitability of salt as a storage medium. For the AD-998 and aluminous concrete systems, the cost of the initial steel container is not included. In the case of Hastelloy C-276 system, the savings gained by eliminating the steel container is deducted from the cost of the Hastelloy C-276 to arrive at the net cost attributable to the storage concept.

The current cost of 6 in. Hastelloy C-276 pipe is $195/ft [61]. A recent estimate of the cost of 6 in. type 304L stainless steel pipe is $18.17/ft for 10 ft lengths purchased in quantities of 500 pieces [98]. Thus the net cost of Hastelloy C-276 containers which is attributable to the storage concept is $176.83/ft or $1,768.30 per container. The cost of end closure is negligible.

The cost of a 7 in. cast alumina AD-998 tube with a 0.25 in. wall ranges from $12.50 to $13.00 per inch in lengths up to 4 ft [72]. To form a 10 ft container several smaller pieces would have to be joined together. As noted earlier, such joints are weak in shear
Fig. 16. Total annual cost for items which are a function of the life of the mine [14, p. 54].
and would require some sort of mechanical assist. Communication with F. Miner of Coors Porcelain Company [72] indicates that the ends of shorter tubes can be precision ground to reduce the outside diameter and cemented together, using an O-ring fitted over the ground portion to provide the necessary mechanical assist. The cost of each joint would be about $400. Miner estimates that the total cost for each container, including fabrication and joints, for this application would be under $2,500.

Aluminous cement is available in the United States under the tradename Lumnite from the Universal Atlas Cement Division of United States Steel. It has a typical composition: \( \text{Al}_2\text{O}_3 + \text{TiO}_2, 46\% \) (\( \text{TiO}_2 < 2\% \)); \( \text{CaO}, 35\%; \text{SiO}_2, 10\%; \) and \( \text{Fe}_2\text{O}_3, 8\% \) [12]. The cost of Lumnite cement is about $4/bag where a bag is 94 lb or 1 ft\(^3\). The cost of type 12/30 RA SiC grain available from The Carborundum Company is about $0.345/lb [82]. This grain has a particle density of 3.20 g/cm\(^3\) (200 lb/ft\(^3\)). Assuming that the finished concrete is fully compacted, that a water to cement ratio of 5 gallons/bag is used, and that the ratio of cement to aggregate is 1/3, the cost of the finished concrete would be $45.15/ft\(^3\). Then a container having a 7 in. inside diameter with 4 in. walls and 10 ft in length would require about 17.5 ft\(^3\) of concrete for a total cost of $790 per container. It is believed that this estimate is sufficiently conservative so that the costs of labor and forms can be neglected.
D. Cost Comparison of Containment Systems

The following cost estimates are based on a 20,000 MW (electrical) nuclear economy that, annually, generates $1.753 \times 10^{11}$ kwhr of electricity and produces 670.34 metric tons of spent fuel which has been irradiated to 33,000 MWd/metric ton at an average specific power of 30.0 MW/metric ton with a thermal efficiency of 33% (reference LWR fuel, Table III). After reprocessing at 150 days, the waste from this fuel has a thermal power of 20,100 W/metric ton of fuel processed, which decays to 1,080 W/metric ton of fuel processed after 10 years [33, p. 4]. Thus a total thermal power of

$$670.34 \, \frac{\text{metric ton}}{\text{year}} \times 1,080 \, \frac{\text{W}}{\text{metric ton}}$$

must be inserted into the mine after 10 years.

For a waste container with active volume $V$ (liters) and power density $Q$ (W/liter) at time of insertion into the mine, the required number of containers per year is

$$N_c = \frac{(7.24 \times 10^5 \, \text{W/yr})}{(Q \cdot V)} \quad (35)$$

The mine area available for waste storage is $7.69 \times 10^5$ m$^2$. Therefore, for a containment system with allowable heat generation rate per unit area of mine $Q_m$ (W/m$^2$), the life of the mine is

$$t_m = Q_m \cdot \frac{7.69 \times 10^5 \, \text{m}^2}{7.24 \times 10^5 \, \text{W/yr}} \quad (36)$$
The required number of containers per year and mine lifetime for each case considered in the heat transfer analysis are summarized in Table XV.

A summary of total cost calculations for each system is presented in Table XVI. Inflation is neglected and all costs are reduced to present value assuming a conservative return on investment of 4% over 10 years (present value factor = 0.6756).

A summary of present-valued annual costs and the equivalent amount to be charged to electrical generation is presented in Table XVII. Mining costs range from 0.004960-0.005767 mills/kwhr (electrical) as compared to a range of 0.0033-0.0075 mills/kwhr (electrical) estimated for similar size containers in previous ORNL studies. Mining costs for all systems run about $10^6/year. Thus the variation in minimum allowable R for each system does not have a large effect on the cost of space in the mine.

The cost of containers is quite significant. For C-276 and AD-998 with Q = 14.21 W/liter, the cost of final storage is approximately tripled because of the use of the more expensive containers. The effect of container cost in the aluminous concrete system is less, resulting in only about a 60% increase in final storage cost since the containers are less expensive. For the AD-998 system with Q = 21.6 W/liter, the cost of final storage is increased by about 150%. This effect is less than for the AD-998 case with Q = 14.21 W/liter since fewer containers are required.

The two cases in the AD-998 system indicate that a considerable amount of money can be saved by using a higher waste power density
Table XV. Required number of containers per year \((N_C)\) and mine life \((t_M)\) for each containment system

<table>
<thead>
<tr>
<th>Containment system</th>
<th>(Q^a) (W/liter)</th>
<th>(N_C)</th>
<th>(Q_M^b) (W/m(^2))</th>
<th>(t_M) (years)</th>
</tr>
</thead>
<tbody>
<tr>
<td>C-276</td>
<td>14.21</td>
<td>1,121</td>
<td>22.84</td>
<td>24.26</td>
</tr>
<tr>
<td>AD-998</td>
<td>14.21</td>
<td>1,121</td>
<td>30.41</td>
<td>32.30</td>
</tr>
<tr>
<td>Aluminous concrete</td>
<td>14.21</td>
<td>1,121</td>
<td>24.44</td>
<td>25.96</td>
</tr>
<tr>
<td>AD-998</td>
<td>21.60</td>
<td>737</td>
<td>32.52</td>
<td>34.54</td>
</tr>
</tbody>
</table>

\(^a\) Waste volume per container = 45.45 liters.

\(^b\) From Table XIV.

since the cost of containers has more impact on final storage charges than the cost of mine space. It is also expected that the use of slightly larger containers would result in reduced costs. For instance increasing the inner diameter of the waste container from 6 in. to 8 in. would increase the available volume by about 70% while the cost of the container would not be increased significantly. Thus the container cost per unit volume of stored waste would decrease. These two points illustrate the fact that the cases considered here are not optimized with respect to cost.

The total cost of spent fuel processing including final waste storage is currently estimated at about 0.2 mills/kwhr (electrical) [74, p. 2-16]. The total storage cost for the concept considered here would range from 4.6-8.5% of this estimated total processing charge. This does not appear to be unreasonable.
Table XVI. Summary of total cost calculation for each containment system as related to waste thermal power density (Q)

<table>
<thead>
<tr>
<th></th>
<th>C-276</th>
<th>AD-998</th>
<th>Aluminous concrete</th>
<th>AD-998</th>
</tr>
</thead>
<tbody>
<tr>
<td>Q(W/liter)</td>
<td>14.21</td>
<td>14.21</td>
<td>14.21</td>
<td>21.60</td>
</tr>
<tr>
<td>Nc</td>
<td>1,121</td>
<td>1,121</td>
<td>1,121</td>
<td>737</td>
</tr>
<tr>
<td>tM (years)</td>
<td>24.26</td>
<td>32.30</td>
<td>25.96</td>
<td>34.54</td>
</tr>
<tr>
<td>Unit cost of container</td>
<td>$1,768</td>
<td>$2,500</td>
<td>$790</td>
<td>$2,500</td>
</tr>
<tr>
<td>1. Annual cost of containers</td>
<td>$1.982 x 10^6</td>
<td>$2.803 x 10^6</td>
<td>$8.856 x 10^5</td>
<td>$1.843 x 10^6</td>
</tr>
<tr>
<td>2. Present value of containers (4%, 10 yr)</td>
<td>$1.982 x 10^6 \text{a}</td>
<td>$1.894 x 10^6</td>
<td>$5.983 x 10^5</td>
<td>$1.245 x 10^6</td>
</tr>
<tr>
<td>3. Annual mining cost depending on Nc (Fig. 15)</td>
<td>$1.973 x 10^5</td>
<td>$1.973 x 10^5</td>
<td>$1.973 x 10^5</td>
<td>$1.870 x 10^5</td>
</tr>
<tr>
<td>4. Annual mining cost depending on tM (Fig. 16)</td>
<td>$1.300 x 10^6</td>
<td>$1.100 x 10^6</td>
<td>$1.300 x 10^6</td>
<td>$1.100 x 10^6</td>
</tr>
<tr>
<td>5. Total annual mining cost (3 + 4)</td>
<td>$1.497 x 10^6</td>
<td>$1.297 x 10^6</td>
<td>$1.497 x 10^6</td>
<td>$1.287 x 10^6</td>
</tr>
<tr>
<td>6. Present value of total mining costs (4%, 10 yr)</td>
<td>$1.011 x 10^6</td>
<td>$8.762 x 10^5</td>
<td>$1.011 x 10^6</td>
<td>$8.695 x 10^5</td>
</tr>
</tbody>
</table>
7. Total annual storage costs  
\[(1 + 5)\]  
\[3.479 \times 10^6\] \[4.100 \times 10^6\] \[2.383 \times 10^6\] \[3.130 \times 10^6\]

8. Present value of total annual storage costs \((2 + 6)\)  
\[2.993 \times 10^6\] \[2.770 \times 10^6\] \[1.609 \times 10^6\] \[2.115 \times 10^6\]

9. Present value cost mills/kwhr (electrical)  
0.01707 0.01580 0.009179 0.01207

*Present value factor of C-276 containers is unity since investment must be made at time of processing.*
Table XVII. Summary of present-valued annual costs for each containment system

<table>
<thead>
<tr>
<th>Q (W/liter)</th>
<th>C-276</th>
<th>AD-998</th>
<th>Aluminous concrete</th>
<th>AD-998</th>
</tr>
</thead>
<tbody>
<tr>
<td>Containers:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[mills/kwhr (electrical)]</td>
<td>$1.982 \times 10^6$</td>
<td>$1.894 \times 10^6$</td>
<td>$5.983 \times 10^5$</td>
<td>$1.245 \times 10^6$</td>
</tr>
<tr>
<td>(% of total cost)</td>
<td>[0.01131]</td>
<td>[0.01080]</td>
<td>[0.003413]</td>
<td>[0.007102]</td>
</tr>
<tr>
<td>Mining:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[mills/kwhr (electrical)]</td>
<td>$1.011 \times 10^6$</td>
<td>$8.762 \times 10^5$</td>
<td>$1.011 \times 10^6$</td>
<td>$8.695 \times 10^5$</td>
</tr>
<tr>
<td>(% of total cost)</td>
<td>[0.005767]</td>
<td>[0.004998]</td>
<td>[0.005767]</td>
<td>[0.004960]</td>
</tr>
<tr>
<td>Total annual storage costs:</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>[mills/kwhr (electrical)]</td>
<td>$2.993 \times 10^6$</td>
<td>$2.770 \times 10^6$</td>
<td>$1.609 \times 10^6$</td>
<td>$2.115 \times 10^6$</td>
</tr>
<tr>
<td>(% of total cost)</td>
<td>[0.01707]</td>
<td>[0.01580]</td>
<td>[0.009179]</td>
<td>[0.01207]</td>
</tr>
</tbody>
</table>
Also, the use of more reliable containers results in a final storage charge which is about 2-3 times the cost of storage in salt using steel containers. At the same time, costs for surface storage in concrete vaults are estimated at 5-7 times the cost of storage in salt using steel containers [78]. Thus the concept considered here, which provides adequate waste containment from the standpoint of safety and recoverability, is more expensive than the current ORNL concept yet still provides a considerable economic advantage over surface storage in concrete vaults.
VIII. CONCLUSIONS

The selection of containment systems in this investigation is based on the principle of continuous positive control and localization of radioactive nuclides. The application of this principle results in containment systems which minimize the possibility of widespread radionuclide migration and allow for recovery of the high-level waste at a reasonable cost and within a reasonable time.

A glass solidified waste product was chosen as the model waste because the leaching rate of glass products is three to four orders of magnitude lower than for a calcine. The inadvertent contact of liquid with a calcined waste during transportation or storage would result almost immediately in a high-level liquid waste, while at the same time the low leaching rate of a glass waste would allow sufficient time for safe recovery of the glass from the deleterious environment. In particular, the most highly developed glass forming process for solidification of high-level wastes is the phosphate system and the capability of incorporating a fluidized bed calcine product into a good quality alumina-phosphate glass with low leachability has been demonstrated by Paige (Fig. 2). In addition the higher thermal conductivity of the glass product permits higher waste thermal power densities.

The investigation of diffusion processes within the mine leads to the conclusion that crystalline and grain boundary diffusion of radionuclides in salt is not significant since the root mean square displacement is less than 1 cm in the 1,000 years necessary for fission product
decay and less than 20 cm in the 500,000 years necessary for decay of the transuranium elements. However diffusion along surfaces or through voids in crushed salt would lead to an undesirable dispersal of radionuclides which would seriously hinder any recovery attempt. Migration of radionuclides assisted by brine in the liquid or vapor phase would permit even more extensive radionuclide dispersal since it is known that the brine accretion will move upward in the vapor phase into the backfilled room. Thus it is concluded that the salt surrounding the waste cannot serve effectively as a container until such a time as the brine inflow has become negligible (30-50 years) and the plastic flow of salt has reconsolidated the crushed salt backfill in the rooms and insertion holes and sealed other possible migration paths such as shale partings introduced during mining.

Calculations of the time required to effect this reconsolidation, based on deformation of the salt pillars at elevated temperature under the influence of the overburden load, indicate that a mine with a lifetime of 25 years could be excavated in such a way as to allow reconsolidation within 50-85 years. Thus it is concluded that waste containers should be able to maintain their integrity for 50-85 years, depending on the extraction pattern, until the salt can serve effectively as the container.

An investigation of various metals and ceramics was conducted in order to select container materials which would yield the desired lifetime in the storage environment. Cast alumina AD-99S containers should maintain their integrity beyond the required time due to excellent chemical stability and resistance to even concentrated HCl at elevated temperatures. Hastelloy alloy C-276 has exhibited a suitably
low overall corrosion rate in environments similar to that encountered during waste storage and it adequately resists pitting, stress corrosion cracking, and intergranular attack. Aluminous concrete shows some promise for this application but the temperature requirements of 250-350 °C are stringent. Testing would have to be carried out to determine if, through proper choice of mix proportions, use of additives, or special surface treatments, an aluminous concrete with a balance between porosity required for strength and low permeability required for container protection could be obtained. An aluminous concrete was included as an example in the heat transfer and economic analysis in order to illustrate the economic incentive for developing an aluminous or other type of concrete for this application. Because of the brittleness, general lack of toughness, and susceptibility to thermal shock of the ceramics, AD-998 and aluminous concrete are suggested for use as secondary containers at the time of insertion into the mine.

A heat transfer analysis of the three containment systems was carried out to determine the minimum allowable spacing within the mine, consistent with design temperature limitations, for a typical case of a phosphate glass waste aged 10 years. This analysis indicates that the main factors in determining the minimum allowable spacing are the effective conductivity and location of relatively insulating regions such as the crushed salt backfill and the primary-secondary container gap. In this regard the AD-998 system requires the least amount of mine space for a given waste power density while the C-276 and aluminous concrete systems are similar in space requirements. Although the purpose of the heat transfer analysis has been to compare the various
containment systems with respect to required mine space for a typical waste power density of 14.21 W/liter, a calculation for the AD-998 system with a waste power density of 21.60 W/liter points out that space requirements do not increase proportionately with an increased waste power density. Thus it is expected that the waste power density could be varied within a certain range to determine which value yields the most efficient use of mine space.

The economic analysis indicates that the cost of containers for the systems considered here is more expensive than mine space except in the case of aluminous concrete. The cost of mining and waste handling is estimated at about $10^6/\text{year}$ for all systems. The cost of containers ranged from about 60% of the mining costs in the case of aluminous concrete to about twice the mining costs in the case of C-276 and AD-998 at a waste power density of 14.21 W/liter. Thus the containment systems considered here result in a final storage charge which is about 2-3 times the cost of the ORNL concept of storage in salt using steel containers.

Economic considerations amplify the desirable effect of increased waste power density in the AD-998 system since a savings of over $0.5 \text{ million/year}$ is obtained due to the fewer number of containers required to store an equivalent amount of waste. Even for the relatively expensive aluminous concrete (SiC aggregate) used as an example here, the cost of final storage is at least $1 \text{ million less annually}$ than for the C-276 and AD-998 systems at a waste power density of 14.21 W/liter. Thus there is a considerable economic incentive for
developing a less expensive form of container protection such as a concrete.

Comparing individual systems at a waste power density of 14.21 W/liter, the aluminous concrete storage system has the lowest final storage charge of 0.0092 mills/kwhr (electrical) because of lower material cost. The AD-998 system (0.016 mills/kwhr) is slightly less expensive than the C-276 system (0.017 mills/kwhr) due to less required mine space and the fact that C-276 containers must be purchased at the time of reprocessing. It is concluded that at this time the cast alumina AD-998 system offers the most reliability for the required investment.

In conclusion, the current ORNL concept of storage in salt using steel containers would lead to loss of positive control and localization of the high-level waste product within a few years due to container failure. In this concept there is a gap of at least 40 years between the time of container failure (1-10 years) and the time of reconsolidation of the formation (50-85 years). Because of the porosity of the crushed salt backfill and the brine movement, this period is the most crucial from the point of view of radionuclide migration. In contrast, the systems considered in this investigation provide for continuous containment of the high-level waste, initially by metal or ceramic containers and subsequently by the reconsolidated salt formation. The increased reliability of these containment systems over the use of steel containers alone and the associated implications for a realistic recovery operation will contribute to establishing greater confidence in the concept of storage of high-level wastes in bedded
salt. Thus a storage site which is marginally acceptable using the current concept of steel containers due to geological and hydrological uncertainties, could be acceptable using the containment systems presented here. In addition, the cost of the improved containment concept presented here is about 2-3 times the cost of storage in salt using steel containers alone, while the cost of surface storage in concrete vaults has been estimated at 5-7 times the cost of storage in salt using steel containers. Therefore the use of Hastelloy C-276 primary containers or cast alumina AD-998 secondary containers offers a reasonable alternative to the use of steel containers alone, yet maintains an economic advantage over engineered surface storage.
IX. RECOMMENDATIONS FOR FUTURE STUDY

It is recommended that testing of the cast alumina AD-998 and Hastelloy C-276 containment systems be initiated immediately at the Lyons, Kansas, site. Using the experience gained in Project Salt Vault, simulated waste containers with electrical heaters should be tested for corrosion resistance under waste storage conditions. Pressurization of the containers during these experiments can be used to give an indication of the integrity of welds or other seals. It is necessary that these tests be given the highest priority at this time in order to provide a firm basis for the comparison of the use of more reliable containment systems in salt with the concept of engineered surface storage to be developed at Hanford by the year 1975.

The economic incentive for developing a concrete suitable for this application has been pointed out. Testing of trial batches is necessary to determine the effect of mix proportions, choice of aggregate and aggregate size, additives, placement techniques, and special surface treatments on permeability and resistance to chemical attack, thermal stresses, and spalling within the required temperature range.

Also, laboratory studies on crushed salt reconsolidation should be conducted to determine the effect of the grade of crushed salt and the presence of shale or other impurities on the homogeneity of the reconsolidated salt. This investigation would determine whether or not it is necessary to use finely crushed salt or to remove impurities from the crushed salt prior to backfilling.
The materials testing programs outlined above can be initiated at the Lyons, Kansas, site during the 1-2 year period required for selection of a site for the demonstration Federal repository in salt. When the site of the repository is determined, priority should be given to determining salt characteristics such as content and composition of brine which bear directly on problems of container integrity. Several rooms should be reserved for container integrity tests during the initial mining phases and continuing through the period of waste insertion into the mine.

If waste is to be stored in salt using steel containers alone, it is desirable to minimize the space requirements in the mine for a given waste. However, it has been pointed out here that since the cost of containers is more than the cost of mine space for systems such as Hastelloy C-276 and cast alumina AD-998, minimization of mine space may not result in the most economical overall storage configuration. Thus it is suggested that a heat transfer analysis be undertaken to determine the best combination of waste power density and container size so as to minimize the combined costs of containers, mining, and waste handling.
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XI. ACKNOWLEDGMENTS

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Finally, the author expresses deep appreciation to his wife, Nancy, for her understanding and helpfulness during this investigation.
XII. APPENDIX A: MINE CLOSURE CALCULATIONS - PROGRAM CLOS

To determine the time required for the closure of a room based on pillar deformation, it is necessary to calculate the required strain $\frac{\Delta h}{h}$ from equations (13) and (15) and then to integrate the strain rate equation (16) to solve for the required time. In the case where pillar volume is not conserved ($\Delta w = \Delta h$), the required $\Delta h$ may be obtained by direct solution of equation (15) which accounts for the total room volume. In the case where pillar volume is conserved ($\Delta w \neq \Delta h$), the required $\Delta h$ must be calculated by solving the simultaneous nonlinear equations (13) and (15) which account for the pillar volume and room volume, respectively.

The simultaneous equations are solved using Newton's method. The function $f_o(\Delta w, \Delta h)$ corresponding to equation (13) is defined as

$$f_o(\Delta w, \Delta h) = (H)(L)(WP) - (H - \Delta h)(L + \Delta w)(WP + \Delta w) \quad (A-1)$$

and the function $g_o(\Delta w, \Delta h)$ corresponding to equation (15) is defined as

$$g_o(\Delta w, \Delta h) = (H)(L)(WR) - (BFILL)(H)(L)(WR) - (\Delta h)(L)(WR) - (H - \Delta h)(L)(\Delta w) \quad (A-2)$$

When the functions $f_o$ and $g_o$ are zero, equations (13) and (15) are satisfied. The functions $f_o$ and $g_o$ are expanded in a Taylor series about the trial solution point ($\Delta w_1, \Delta h_1$). Assuming that the trial solution point is sufficiently close to the actual solution so that
higher order terms can be neglected, the linear system resulting from
the Taylor series expansion can be solved directly to yield the iteration
algorithms
\begin{align}
\Delta W_{i+1} &= \Delta W_i - \left[ f \frac{\partial \sigma_o}{\partial \Delta H} - g_o \frac{\partial \sigma_o}{\partial \Delta W} \right] \Delta W_i \Delta H_i \quad (A-3) \\
\Delta H_{i+1} &= \Delta H_i - \left[ f \frac{\partial \sigma_o}{\partial \Delta W} - g_o \frac{\partial \sigma_o}{\partial \Delta H} \right] \Delta W_i \Delta H_i \quad (A-4)
\end{align}

This algorithm converges very rapidly for the case considered here.

Since a functional expression is not available for the pillar
centerline temperature as a function of time as presented in Fig. 5,
it is necessary to integrate the strain rate equation (16) numerically.
This is accomplished using Simpson's rule at half year intervals.
The strain rate is a smooth function of time so that the results should
be numerically accurate to at least the fourth power of the time
interval or 0.0625 years. The integration is carried out in one year
steps using the three point formula
\begin{align}
\int_{t_{i-1}}^{t_{i+1}} \ddot{\epsilon} dt &\approx \frac{\Delta t}{3} \left[ \ddot{\epsilon}(t_{i-1}) + 4\ddot{\epsilon}(t_i) + \ddot{\epsilon}(t_{i+1}) \right] \quad (A-5)
\end{align}

where \( \Delta t = 0.5 \) years and \( t_i \) are the times at half year intervals.
After each step the integral is compared to the required \( \Delta H/H \) and if
it equals or exceeds \( \Delta H/H \), the calculation is terminated. Thus
predicted times are given to the nearest whole year.
Program CLOS was written to carry out the calculations discussed above. A program listing and sample output are given on the following pages.
C
C *************************************************************************
C
C CLOS IS A FORTRAN IV PROGRAM WHICH CALCULATES:
C 1. PILLAR DEFORMATION REQUIRED TO EFFECT COMPLETE CLOSURE OF A ROOM
C 2. TIME REQUIRED FOR THIS PILLAR DEFORMATION
C
C INPUT DATA:
C TEMP  TEMPERATURE DISTRIBUTION AS A FUNCTION OF TIME
C       IN HALF YEAR INTERVALS
C H    INITIAL PILLAR OR ROCM HEIGHT
C L    INITIAL PILLAR OR ROCM LENGTH
C WP   INITIAL PILLAR WIDTH
C WR   INITIAL ROCM WIDTH
C LOAD  OVERBURDEN LOAD
C BFILL FRACTIONAL CRUSHED SALT BACKFILL
C DELH1 ESTIMATE OF REQUIRED CHANGE IN PILLAR HEIGHT
C DELW1 ESTIMATE OF REQUIRED CHANGE IN PILLAR WIDTH
C ERR  ERROR LIMIT IN ITERATIVE SOLUTION FOR DELH AND DELW
C GECM COEFFICIENT ACCOUNTING FOR GEOMETRY OF PILLAR
C
C SIGNIFICANT PROGRAM VARIABLES:
C F    EQUATION ACCOUNTING FOR PILLAR VOLUME
C G    EQUATION ACCOUNTING FOR ROCM VOLUME
C CLOS REQUIRED FRACTIONAL PILLAR DEFORMATION
C EXT  FRACTIONAL SALT EXTRACTION
C SIG  AVERAGE PILLAR STRESS
C TIME TIME REQUIRED FOR CLOSURE
C
C *************************************************************************
C
REAL L,LCAD
DIMENSION TEMP(90)
READ(5,1)(TEMP(I),I=1,90)
1 FFORMAT(16F5.1)
2 READ(5,3,END=11)H,L,WP,WR,LOAD,BFILL,DELH1,DELW1,ERR,GECM
3 FFORMAT(4F5.1,4F10.2,F10.5,F10.2)

ITER=0
ERRH=1.0
ERRW=1.0
IF (DELH1 .NE. DELH1) GO TO 4

C C DELH1=DELH1 IS THE CASE WHERE PILLAR VOLUME IS NOT CONSERVED, SOLVE
C DIRECTLY FOR REQUIRED CHANGE IN HEIGHT, ACCOUNTING FOR ROOM VOLUME
C
C ERRH=0.0
ERRW=0.0
F=0.0
G=0.0
DELH1=0.5*(((WR+H)-(((WR+H)**2)+(4.0*(BFILL-1.0)*H*WR))*0.5))
DELW1=DELH1
GO TO 5

C C IN THIS CASE PILLAR VOLUME IS CONSERVED, SOLVE SIMULTANEOUS EQUATIONS
C ACCOUNTING FOR ROOM AND PILLAR VOLUME USING NEWTON'S METHOD
C
4 F=(H*(L+WP)*DELW1)-(L*WP*DELH1)-(L+WP)*DELH1*DELW1)+(1(H*DELW1*DELH1)-(DELH1*DELW1))
G=((BFILL-1.0)*H*WR)+(WR*DELH1)+(H*DELW1)-(DELW1*DELH1)
IF (ERRH.LT.ERRR.AND.ERRW.LT.ERRR.OR.ITER.GT.50) GO TO 5
DFDELH=((-L*WP)+((L+WP)*DELW1)+(DELW1*DELH1))
DFDELW=(H*(L+WP))-(L+WP)*DELH1)+(2.0*H*DELW1)-(2.0*DELH1*DELW1)
DGDELH=WR-DELW1
DGDELW=1-DELH1
DEN=(DFDELW*DGDELH)-(DGDELW*DFDELH)
DELW2=DELW1+((G*DFDELH)-(F*DGDELH))/DEN)
DELH2=DELH1+((F*DGDELW)-(G*DFDELW))/DEN)
ITER=ITER+1
ERRH=ABS(DELH2-DELH1)
ERRW=ABS(DELH2-DELW1)
DELW1=DELW2
DELH1=DELH2
GO TO 4

5 CLCS=(DELH1/H)
ORIGINAL ROOM DIMENSIONS — FEET

115X, "EIGHT = ',F7.2,5X,’LENGTH = ',F7.2,5X,’WIDTH = ',F7.2,///

ORIGINAL PILLAR DIMENSIONS — FEET

115X, "EIGHT = ',F7.2,5X,’LENGTH = ',F7.2,5X,’WIDTH = ',F7.2,///

CHANGE IN PILLAR DIMENSIONS — FEET

115X, "DELTA HEIGHT = ',F7.2,5X,’DELTA WIDTH = ',F7.2,///

FRACTIONAL BACKFILL = ',F5.2,///

NUMBER OF ITERATIONS = ',I4,///

ERRH = ',F8.5,5X,’ERRW = ',F8.5,5X,’F = ',F8.5,5X,’G = ',F8.5,///

REQUIRED FRACTIONAL CLOSURE = ',F8.5,///

CALCULATE TIME REQUIRED FOR THIS FRACTIONAL CHANGE IN PILLAR HEIGHT

BY INTEGRATING STRAIN RATE EQUATION USING SIMPSON'S RULE

EXT=xR/(WP+WR)
SIG=LCAD/(1.0-EXT)
COEF=CECM*(0.33E-37)*(3765.8**0.3)*(SIG**3)
TINT=(COEF/0.3)*(TEMP(I)**9.5)
T=0.0
CC 7 I=2,88,2
T=T+1.0
IF (TINT.LT.CLCS) GO TO 7
TIME=T
TMAX=TEMP(I)
GO TO 9
7 TINT=TINT + (COEF*0.5)/3.0)*(((TEMP(I)**9.5)/(T**0.7)) +
14.0*(((TEMP(I+1)**9.5)/(T+0.5)**0.7)) +
11((TEMP(I+2)**9.5)/(T+1.0)**0.7))
TIME=T+1.0
TMAX=TEMP(I)
IF (TINT.GE.CLCS) GO TO 9
EXP=10.0/C/3.0
CCNST=(COEF*(TEMP(I)**9.5))/0.3
TIME=((CLOS-TINT+CONST*45.0**0.3))/CONST**EXP
9 WRITE(6,10)EXT,LCAD,SIG,TMAX,TIME
10 FORMAT(20X, '*** TIME REQUIRED FOR CLOSURE ***', ///
115X, 'FRACTIONAL EXTRACTION = ', F5.3, 'OVERBURDEN LOAD (PSI) = ', F10.2,
115X, 'AVERAGE PILLAR STRESS (PSI) = ', F12.5,
115X, 'MAXIMUM TEMPERATURE (ABSOLUTE) = ', F8.2,
115X, 'TIME REQUIRED FOR CLOSURE (YEARS) = ', F10.3

GC TG 2

11 STOP
END
ORIGINAL ROOM DIMENSIONS -- FEET

HEIGHT = 15.00  LENGTH = 300.00  WIDTH = 30.00

ORIGINAL PILLAR DIMENSIONS -- FEET

HEIGHT = 15.00  LENGTH = 300.00  WIDTH = 30.00

CHANGE IN PILLAR DIMENSIONS -- FEET

DELTA HEIGHT = 2.53  DELTA WIDTH = 5.45

FRACTIONAL BACKFILL = 0.68

NUMBER OF ITERATIONS = 3

ERRH = 0.00007  ERRW = 0.00003  F = 0.00154  G = -0.00000

REQUIRED FRACTIONAL CLOSURE = 0.16893

TIME REQUIRED FOR CLOSURE

FRACTIONAL EXTRACTION = 0.500  OVERBURDEN LOAD (PSI) = 730.00

AVERAGE PILLAR STRESS (PSI) = 1460.00000

MAXIMUM TEMPERATURE (ABSOLUTE) = 443.20

TIME REQUIRED FOR CLOSURE (YEARS) = 56.969
XIII. APPENDIX B: HEAT TRANSFER CALCULATIONS - PROGRAM HEAT

In this section a five point finite difference approximation to the steady state heat conduction equation with boundary conditions as specified in the physical model (Fig. 8) is developed. The grid scheme is illustrated in Fig. 17 for a typical interior point \( T_{ij} \). This grid represents a cut in the R-Z plane of a volume element obtained by rotating this section one radian about the \( R_1 = 0 \) axis. Mesh spacings \( G_i \) and \( H_j \) are variable. Each region has associated with it a volumetric heat source \( Q_{ij} \) and a thermal conductivity \( K_{ij} \).

The finite difference equations are formulated by integrating the steady state heat conduction equation (17) over the volume elements \( V_1, V_2, V_3, \) and \( V_4 \) so that

\[
\iiint_{V_1+V_2+V_3+V_4} -\nabla \cdot K \nabla T \, dV = \iiint_{V_1+V_2+V_3+V_4} Q \, dV \tag{B-1}
\]

where \( Q \) may be zero for a non-source region. Considering only the left hand side of equation (B-1) the volume integral may be reduced to an integral over the surface of each volume element using Gauss' theorem. Note that the contribution for the surfaces in the R-Z plane are zero since the model exhibits axial symmetry and the temperature gradient in the direction normal to any R-Z plane is zero. Also the surface contributions for the interfaces between volume elements cancel since the heat flux is continuous at the interface. Therefore
Fig. 17. Grid scheme used in formulating finite difference approximation to steady state heat conduction equation in cylindrical geometry.
\[ \int \int \int_{V_1+V_2+V_3+V_4} - \nabla \cdot K \vec{V} dV = \int \int \int_{\alpha_1} K_{i-1,j} \vec{V} \cdot d\vec{\alpha}_1 \]

\[ - \int \int \int_{\alpha_2} K_{i-1,j} \vec{V} \cdot d\vec{\alpha}_2 - \int \int \int_{\alpha_3} K_{i-1,j-1} \vec{V} \cdot d\vec{\alpha}_3 \]

\[ - \int \int \int_{\alpha_4} K_{i-1,j-1} \vec{V} \cdot d\vec{\alpha}_4 - \int \int \int_{\alpha_5} K_{i,j-1} \vec{V} \cdot d\vec{\alpha}_5 \]

\[ - \int \int \int_{\alpha_6} K_{i,j-1} \vec{V} \cdot d\vec{\alpha}_6 - \int \int \int_{\alpha_7} K_{i,j} \vec{V} \cdot d\vec{\alpha}_7 \]

\[ - \int \int \int_{\alpha_8} K_{i,j} \vec{V} \cdot d\vec{\alpha}_8 \]  

where the \( \alpha_i \) are the surface areas depicted in Fig. 17 and the \( d\vec{\alpha}_i \) are unit vectors normal to the surface. Each of the surface integrals on the right hand side represents the component of temperature gradient normal to the surface times the surface area. Therefore the surface integrals can be approximated by the temperature difference divided by the distance between temperature points times the surface area or

\[ \int \int \int_{V_1+V_2+V_3+V_4} - \nabla \cdot K \vec{V} dV = \]

\[ K_{i-1,j} \frac{T_{i,j} - T_{i,j+1}}{H_j} \alpha_1 + K_{i-1,j} \frac{T_{i,j} - T_{i-1,j}}{G_{i-1}} \alpha_2 \]

\[ + K_{i-1,j-1} \frac{T_{i,j} - T_{i-1,j-1}}{G_{i-1}} \alpha_3 + K_{i-1,j-1} \frac{T_{i,j} - T_{i,j-1}}{H_{j-1}} \alpha_4 \]

\[ + K_{i,j-1} \frac{T_{i,j} - T_{i,j-1}}{H_{j-1}} \alpha_5 + K_{i,j-1} \frac{T_{i,j} - T_{i,j+1}}{G_i} \alpha_6 \]
\[ + K_{ij} \frac{T_{ij} - T_{i+1,j}}{G_i} \alpha_7 + K_{ij} \frac{T_{ij} - T_{i,j+1}}{H_j} \alpha_8 \]  

where

\[ \alpha_1 = \left( R_j + \frac{H_j}{2} \right) \frac{G_{i-1}}{2} \]  
\[ \alpha_2 = \left( \frac{R_j H_i}{2} + \frac{H_i^2}{8} \right) \]  
\[ \alpha_3 = \left( \frac{R_j H_i}{2} - \frac{H_i^2}{8} \right) \]  
\[ \alpha_4 = \left( R_j - \frac{H_j}{2} \right) \frac{G_{i-1}}{2} \]  
\[ \alpha_5 = \left( R_j - \frac{H_j}{2} \right) \frac{G_i}{2} \]  
\[ \alpha_6 = \left( \frac{R_j H_i}{2} - \frac{H_i^2}{8} \right) \]  
\[ \alpha_7 = \left( \frac{R_j H_i}{2} + \frac{H_i^2}{8} \right) \]  
\[ \alpha_8 = \left( R_j + \frac{H_i}{2} \right) \frac{G_i}{2} \]  

The right hand side of equation (B-1) is given by

\[ \int \int \int \int_{V_1 + V_2 + V_3 + V_4} Q dV = Q_{i-1,j} V_1 + Q_{i-1,j-1} V_2 + Q_{i,j-1} V_3 + Q_{i,j} V_4 \]  

where

\[ V_1 = \left( \frac{R_j H_i}{2} + \frac{H_i^2}{8} \right) \frac{G_{i-1}}{2} \]
\[ V_2 = \left( \frac{R_{j-1}}{2} - \frac{H_{j-1}^2}{8} \right) G_{j-1} \]  
(B-14)

\[ V_3 = \left( \frac{R_{j-1}}{2} - \frac{H_{j-1}^2}{8} \right) G_j \]  
(B-15)

\[ V_4 = \left( \frac{R_{j-1}}{2} + \frac{H_{j-1}^2}{8} \right) G_j \]  
(B-16)

Thus equation (B-1) has been reduced to a five point difference formula which can be expressed in the following convenient matrix form

\[ D_{ij}T_{i-1,j} + E_{ij}T_{i,j-1} + A_{ij}T_{ij} + B_{ij}T_{i,j+1} + C_{ij}T_{i+1,j} = S_{ij} \]  
(B-17)

where

\[ A_{ij} = -(B_{ij} + C_{ij} + D_{ij} + E_{ij}) \]  
(B-18)

\[ B_{ij} = -\frac{\left( R_j + \frac{H_i}{2} \right)}{2H_j} \left[ K_{i-1,j}G_{i-1} + K_{ij}G_i \right] \]  
(B-19)

\[ C_{ij} = -\frac{1}{G_i} \left[ K_{i,j-1} \left( \frac{R_iH_{j-1}}{2} - \frac{H_{j-1}^2}{8} \right) + K_{ij} \left( \frac{R_jH_i}{2} + \frac{H_i^2}{8} \right) \right] \]  
(B-20)

\[ D_{ij} = -\frac{1}{G_{i-1}} \left[ K_{i-1,j} \left( \frac{R_iH_{j-1}}{2} + \frac{H_{j-1}^2}{8} \right) + K_{i-1,j-1} \left( \frac{R_jH_i}{2} - \frac{H_i^2}{8} \right) \right] \]  
(B-21)

\[ E_{ij} = -\frac{\left( R_j - \frac{H_{i-1}}{2} \right)}{2H_{j-1}} \left[ K_{i-1,j-1}G_{i-1} + K_{i,j-1}G_j \right] \]  
(B-22)

\[ S_{ij} = Q_{i-1,j}V_1 + Q_{i-1,j-1}V_2 + Q_{i,j-1}V_3 + Q_{i,j}V_4 \]  
(B-23)

And now, since

\[ R_{j-1} + \frac{H_{j-1}}{2} = R_j - \frac{H_{j-1}}{2} \]  
(B-24)
it can be shown that

\[ D_{ij} = C_{i-1,j} \]  \hspace{1cm} (B-25)

and

\[ E_{ij} = B_{i,j-1} \]  \hspace{1cm} (B-26)

so that equation (B-17) reduces to

\[
C_{i-1,j}T_{i-1,j} + B_{i,j-1}T_{i-1,j-1} + A_{ij}T_{ij} \\
+ B_{ij}T_{i,j+1} + C_{ij}T_{i+1,j} = S_{ij} \]  \hspace{1cm} (B-27)

The case considered above is for a typical internal mesh point. The constant temperature boundary conditions must be imposed at the horizontal surfaces at the top \((i = 1)\) and bottom \((i = IZ)\) of the cylinder. Since all \(T_{1,j}\) and \(T_{IZ,j}\) are known constants, no equation such as (B-27) is required for these points. When considering the points \(T_{2,j}\), the constant contribution from the term \(C_{1,j}T_{1,j}\) can be added to the source term. When considering the points \(T_{IZ-1,j}\), the constant contribution from the term \(C_{IZ-1,j}T_{IZ,j}\) can be added to the source term. After this is carried out, all \(C_{1,j}\) and \(C_{IZ-1,j}\) are reduced to zero.

The symmetric boundary conditions must be imposed at the center \((j = I)\) and surface \((j = IR)\) of the cylinder. For all \(T_{i,1}\), the integration is carried out only over the volume elements \(V_1\) and \(V_4\). Since the temperature gradient is zero along the boundary \(j = 1\), the only surface contributions are from \(\alpha_1, \alpha_2, \alpha_7,\) and \(\alpha_8\). For all \(T_{i,IR}\), the volume integration is carried out only over \(V_2\) and \(V_3\), with
surface contributions arising only from $\alpha_3$, $\alpha_4$, $\alpha_5$, and $\alpha_6$. Since there is no temperature point at $T_{i,IR+1}$, all $B_{i,IR}$ are zero.

The matrix equation for the simple case of $IZ = 5$, $IR = 3$ is

$$
\begin{bmatrix}
A_{21} & B_{21} & 0 & C_{21} & 0 & 0 & 0 & 0 & 0 \\
B_{21} & A_{22} & B_{22} & 0 & C_{22} & 0 & 0 & 0 & 0 \\
0 & B_{22} & A_{23} & 0 & 0 & C_{23} & 0 & 0 & 0 \\
C_{21} & 0 & 0 & A_{31} & B_{31} & 0 & C_{31} & 0 & 0 \\
0 & C_{22} & 0 & B_{31} & A_{32} & B_{32} & 0 & C_{32} & 0 \\
0 & 0 & C_{23} & 0 & B_{32} & A_{33} & 0 & 0 & C_{33} \\
0 & 0 & 0 & C_{31} & 0 & 0 & A_{41} & B_{41} & 0 \\
0 & 0 & 0 & 0 & C_{32} & 0 & B_{41} & A_{42} & B_{42} \\
0 & 0 & 0 & 0 & 0 & C_{33} & 0 & B_{42} & 0
\end{bmatrix}
\begin{bmatrix}
T_{21} \\
T_{22} \\
T_{23} \\
T_{31} \\
T_{32} \\
T_{33} \\
T_{41} \\
T_{42} \\
T_{43}
\end{bmatrix}
= 
\begin{bmatrix}
S_{21} \\
S_{22} \\
S_{23} \\
S_{31} \\
S_{32} \\
S_{33} \\
S_{41} \\
S_{42} \\
S_{43}
\end{bmatrix}
$$

(B-28)

An alternating direction implicit (ADI) method can be used to solve a matrix equation of this form. One takes the first set of $IR$ equations and multiplies the off diagonal vector $[C_{2j}]$ times the temperature vector $[T_{3j}]$ and combines the resulting vector with the source vector $[S_{2j}]$ to form a dummy source vector $[S'_{2j}]$. The resulting system of $IR$ equations is then in tridiagonal form

$$
\begin{bmatrix}
A_{21} & B_{21} & 0 \\
B_{21} & A_{22} & B_{22} \\
0 & B_{22} & A_{23}
\end{bmatrix}
\begin{bmatrix}
T_{21} \\
T_{22} \\
T_{23}
\end{bmatrix}
= 
\begin{bmatrix}
S'_{21} \\
S'_{22} \\
S'_{23}
\end{bmatrix}
$$

(B-29)

and can be solved for $[T_{2j}]$ by Gaussian elimination. Moving to the next set of $IR$ equations the dummy source vector becomes

$$
[S'_{3j}] = [S_{3j}] - [C_{2j}][T_{2j}] - [C_{3j}][T_{1j}]
$$

(B-30)
and the tridiagonal form is solved again for $[T_{3j}]$. This procedure is repeated for the IZ-2 sets of IR equations.

Next, the procedure is repeated for the IR sets of IZ-2 equations where the dummy source vector is

$$[S'_{ij}] = [S_{ij}] = [B_{i,j-1}][T_{i,j-1}] - [B_{i,j}][T_{i,j+1}] \quad (B-31)$$

These two calculational procedures from alternating directions are called one iteration in the ADI method. The iterations are repeated until convergence is achieved.

Computer program HEAT was written to carry out the calculations discussed above. A listing of the program appears on the following pages.
HEAT is a FORTRAN IV program which calculates temperatures at mesh points of a five point finite difference approximation to the steady state heat conduction equation in cylindrical geometry.

Main features are:
1. Variable mesh spacing in vertical and radial directions.
2. Volumetric heat source and thermal conductivity may be specified for each separate mesh region.
3. Matrix equation is solved iteratively by alternating direction implicit method.

Input data:
- IZ: Number of horizontal mesh lines
- IR: Number of vertical mesh lines
- ITRIP: Maximum number of iterations
- CNV: Convergence criterion
- QS: Volumetric heat source (W/liter)
- IQ1: Upper horizontal source boundary
- IQ2: Lower horizontal source boundary
- IQ1: Inner vertical source boundary
- IQ2: Outer vertical source boundary
- T: Temperature (degrees centigrade)
- K: Thermal conductivity (W/(M*Degree centigrade))
- R: Radial position of vertical mesh line (cm)
- Z: Vertical position of horizontal mesh line (m)

Significant program variables:
- Q: Volumetric heat source (W/M**2)
- H: Radial mesh spacing (K)
- G: Vertical mesh spacing (M)
- A: Diagonal matrix element
- B: Closest off diagonal matrix element
- C: Farthest off diagonal matrix element
- S: Source vector
C
C ******************************************************************************
C
REAL K(57,20)
DIMENSION T(58,21),Q(57,20),R(21),Z(58),G(57),H(20)
COMMON A(58,21),B(58,21),C(58,21),S(58,21),IZ,IR
READ(5,1)IZ,IR,ITRP,CNV,QS,IQ1,IQ2,JQ1,JQ2
1 FORMAT(3I5,F10.5,F10.2,4I5)
READ(5,2)({T(I,J),J=1,IR},I=1,IZ)
2 FORMAT(16F5.0)
   IMAX=IZ-1
   JMAX=IR-1
   READ(5,3){K(I,J),J=1,JMAX},I=1,IMAX
3 FORMAT(10F8.3).
   READ(5,3){R(I),I=1,IR}
   READ(5,3){Z(I),I=1,IZ}
C
C ASSIGN SOURCE TO WASTE REGION
C
   DO 5 I=1,IMAX
       DO 5 J=1,JMAX
       Q(I,J)=0.0
   5   IF (I.EQ.IQ1) Q(I,J)=100.
       IF (J.EQ.JQ2) Q(I,J)=100.
   6   Q(I,J)=QS*1000.
C
C CALCULATE R AND Z MESH SPACINGS
C
   DO 7 I=2,IR
       R(I)=R(I)/100.
       N=I-1
       H(N)=R(I)-R(N)
   7   DO 8 I=2,IZ
       N=I-1
       G(N)=Z(I)-Z(N)
C
C CALCULATE MATRIX ELEMENTS FOR FIRST TEMPERATURE ROW, I=2
C
DO 9 J=1,IR
A(I,J)=0.0
B(I,J)=0.0
C(I,J)=0.0
9 S(I,J)=0.0
I=2
J=1
M=I-1.
GEOM1=(R(J)*H(J)/2.0)+(H(J)*2)/8.0)
B(I,J)=(K(I,J)*G(I,J))*((R(J)*H(J)/2.0)/(2.0*H(J)))
C(I,J)=GEOM1*(K(I,J)*G(I,J))
CONST=GEOM1*(K(I,J)*G(I,J))
A(I,J)=-B(I,J)+C(I,J)+CONST
S(I,J)=(0.5*GEOM1*(Q(I,J)+Q(I,J)*G(I,J)))+(CONST*T(I,J))
DO 10 J=2,JMAX
GEOM1=(R(J)*H(J)/2.0)+(H(J)*2)/8.0)
GEOM2=(R(J)*H(J-1)/2.0)+(H(J-1)*2)/8.0)
B(I,J)=(K(I,J)*G(I,J))*(R(J)*H(J)/2.0)/(2.0*H(J)))
C(I,J)=(GEOM2*(K(I,J-1)/G(I,J)))+(GEOM1*(K(I,J)*G(I,J))
CONST=(GEOM1*(K(I,J)*G(I,J)))+(GEOM2*(K(I,J-1)/G(I,J))
A(I,J)=-B(I,J)+C(I,J)+CONST+B(I,J-1))
10 S(I,J)=(0.5*GEOM1*(Q(I,J)+Q(I,J)*G(I,J)))+
1(GEOM2*(Q(I,J-1)*G(I,J)+Q(I,J)*G(I,J)))+CONST*T(I,J)
J=IR
GEOM2=(R(J)*H(J-1)/2.0)-(H(J)*2)/8.0)
B(I,J)=0.0
C(I,J)=GEOM2*(K(I,J-1)/G(I,J))
CONST=GEOM2*(K(I,J-1)/G(I-1))
A(I,J)=-C(I,J)+CONST+B(I,J-1))
S(I,J)=(0.5*GEOM2*(Q(I,J-1)*G(I-1)+Q(I,J-1)*G(I,J)))+CONST*T(I,J)
C
C CALCULATE MATRIX ELEMENTS FOR INTERIOR ROWS, I=3,IR-2
C
IMAX=IMAX-1
DO 12 I=3,IMAX
J=1
GEOM1 = \( (R(J) \times H(J) / 2.0) + ((H(J) \times 2) / 8.0) \)

\[
B(I, J) = (K(I - 1, J) \times G(I - 1) + K(I, J) \times G(I)) \times ((R(J) + H(J) / 2.0) / (2.0 \times H(J)))
\]

\[
C(I, J) = GEOM1 \times (K(I, J) / G(I))
\]

\[
A(I, J) = -\{B(I, J) + C(I, J) + C(I - 1, J)\}
\]

\[
S(I, J) = 0.5 \times GEOM1 \times (Q(I - 1, J) \times G(I - 1) + Q(I, J) \times G(I))
\]

DO 11 J = 2, JMAX

GEOM1 = \( (R(J) \times H(J) / 2.0) + ((H(J) \times 2) / 8.0) \)

\[
B(I, J) = (K(I - 1, J) \times G(I - 1) + K(I, J) \times G(I)) \times ((R(J) + H(J) / 2.0) / (2.0 \times H(J)))
\]

\[
C(I, J) = GEOM2 \times (K(I - 1, J - 1) / G(I)) + (GEOM1 \times (K(I, J) / G(I)))
\]

\[
A(I, J) = -\{B(I, J) + C(I, J) + C(I - 1, J)\}
\]

11 S(I, J) = 0.5 \times \{GEOM1 \times (Q(I - 1, J) \times G(I - 1) + Q(I, J) \times G(I))\} +

1 \{GEOM2 \times (Q(I - 1, J - 1) \times G(I - 1) + Q(I, J - 1) \times G(I))\}

J = IR

GEOM2 = \( (R(J) \times H(J - 1) / 2.0) - ((H(J - 1) \times 2) / 8.0) \)

\[
B(I, J) = 0.0
\]

\[
C(I, J) = GEOM2 \times (K(I, J - 1) / G(I))
\]

\[
A(I, J) = -\{C(I, J) + C(I - 1, J) + B(I, J - 1)\}
\]

12 S(I, J) = 0.5 \times GEOM2 \times (Q(I - 1, J - 1) \times G(I - 1) + Q(I, J - 1) \times G(I))

C C CALCULATE MATRIX ELEMENTS FOR LAST TEMPERATURE ROW, I = IZ - 1

C

I = IZ - 1

J = 1

GEOM1 = \( (R(J) \times H(J) / 2.0) + ((H(J) \times 2) / 8.0) \)

\[
B(I, J) = (K(I - 1, J) \times G(I - 1) + K(I, J) \times G(I)) \times ((R(J) + H(J) / 2.0) / (2.0 \times H(J)))
\]

\[
C(I, J) = 0.0
\]

\[
CONST = GEOM1 \times (K(I, J) / G(I))
\]

\[
A(I, J) = -\{B(I, J) + CONST + C(I - 1, J)\}
\]

\[
S(I, J) = 0.5 \times GEOM1 \times (Q(I - 1, J) \times G(I - 1) + Q(I, J) \times G(I)) + CONST \times T(I + 1, J)
\]

DO 13 J = 2, JMAX

GEOM1 = \( (R(J) \times H(J) / 2.0) + ((H(J) \times 2) / 8.0) \)

\[
B(I, J) = (K(I - 1, J) \times G(I - 1) + K(I, J) \times G(I)) \times ((R(J) + H(J) / 2.0) / (2.0 \times H(J)))
\]

\[
C(I, J) = 0.0
\]

\[
CONST = GEOM2 \times (K(I - 1, J - 1) / G(I)) + (GEOM1 \times (K(I, J) / G(I)))
\]

\[
A(I, J) = -\{B(I, J) + CONST + C(I - 1, J) + B(I, J - 1)\}
\]
13 S(I,J)=0.5*GEOM1*(Q(I-1,J)*G(I-1)+Q(I,J)*G(I))+
1*GEOM2*(Q(I-1,J-1)*G(I-1)+Q(I,J-1)*G(I)))+CONST*T(I+1,J)
J=IR
GEOM2=(R(J)*H(J-1)/2.0-((H(J-1)**2)/8.0)
B(I,J)=0.0
C(I,J)=0.0
CONST=GEOM2*(K(I,J-1)/G(I))
A(I,J)=-(CONST+C(I-1,J)+B(I,J-1))
S(I,J)=0.5*GEOM2*(Q(I-1,J-1)*G(I-1)+Q(I,J-1)*G(I)))+CONST*T(I+1,J)
WRITE(6,14)CNV,ITRIP
14 FORMAT('1X,15X,'CONVERGENCE CRITERIA =',F10.5,/, 
15X,'MAX NUMBER ITERATIONS =',I10,/,15X,'ITERATIONS',11X, 
1'1.0'),15X,9'RATIO',/) 
C  
C SOLVE MATRIX EQUATION USING ALTERNATING DIRECTION IMPLICIT METHOD  
C   
ITER=0
TNUM=0.0
15 CALL ADIMTD(T)
ITER=ITER+1
TDEN=0.0
DO 16 I=1,IZ
DO 16 J=1,IR
16 TDEN=TDEN+(T(I,J)**2)
TDEN = SQRT(TDEN)
RATIO=TNUM/TDEN
TNUM=TDEN
WRITE(6,17)ITER,TDEN,RATIO
17 FORMAT(17X,15X,10X,F15.5,10X,F10.5)
IF (ITER.EQ.ITRIP) GO TO 18
IF (ABS(RATIO-1.0)<CNV)18,15,18
18 WRITE(6,19)QS.
19 FORMAT('1X,26X,***** TEMPERATURE DISTRIBUTION IN DEGREES', 
1'CENTIGRADE *****',10X,'HEAT SOURCE (WATTS PER LITER) =', 
1F10.2,10X,'RADIAL DISTANCES IN CENTIMETERS FOR WHICH', 
1'TEMPERATURES WERE CALCULATED:',/) 
DO 24 I=2,IR
24 R(I) = R(I) * 100.
   WRITE(6,20)(R(I), I=1,IR)
20 FORMAT(13X, 'R = ', 4X, 7(F7.2, 3X), //, (20X, 7(F7.2, 3X), /))
   WRITE(6,21)
21 FORMAT(/, 10X, 'TEMPERATURES CORRESPONDING TO ABOVE RADIAL *
   1* DISTANCES AS A FUNCTION OF Z IN METERS: */)
   DO 22 I = 1, IZ
22 WRITE(6,23) 2(I), (T(I, J), J=1,IR)
23 FORMAT(10X, 'Z= ', F7.2, 1X, 7(F7.2, 3X), //, (20X, 7(F7.2, 3X), /))
STOP
END

C ALTERNATING DIRECTION IMPLICIT METHOD
C
SUBROUTINE ADIMTD(T)
  DIMENSION T(58,21),STORA(53),STORB(58),STORC(58),DUMS(58)
  COMMON A(58,21),B(58,21),C(58,21),S(58,21),IZ,IR

C SOLVE IZ-2 SETS OF IR EQUATIONS
C
I = 1
   DO 1 J = 1, IR
1  STORT(J) = T(I,J)
   I = I + 1
   DO 3 J = 1, IR
3  STORA(J) = A(I,J)
      STORB(J) = B(I,J)
      DUMS(J) = C(I,J) * T(I+1,J) + C(I-1,J) * STORT(J) + S(I,J)
      STORT(J) = T(I,J)
   CALL TRIDIG(STORA,STORB,DUMS,T,0,IR,I)
   IF (I.LT.IZ-1) GO TO 2

C SOLVE IR SETS OF IZ-2 EQUATIONS
C
J = 1
   IMAX = IZ - 1
DO 4 I=2,IMAX
N=I-1
STORA(N)=A(I,J)
STORC(N)=C(I,J)
DUMS(N)=B(I,J)*T(I,J)+S(I,J)
4 STOR(N)=T(I,J)
CALL TRIDIG(STORA,STORC,DUMS,T,1,IZ-2,J)
5 J=J+1
DO 6 I=2,IMAX
N=I-1
STORA(N)=A(I,J)
STORC(N)=C(I,J)
DUMS(N)=B(I,J)*T(I,J)+B(I,J-1)*STOR(N)+S(I,J)
6 STOR(N)=T(I,J)
CALL TRIDIG(STORA,STORC,DUMS,T,1,IZ-2,J)
IF (J.GT.IR-1) GO TO 5
J=IR
DO 12 I=2,IMAX
N=I-1
STORA(N)=A(I,J)
STORC(N)=C(I,J)
DUMS(N)=B(I,J-1)*STOR(N)+S(I,J)
12 CALL TRIDIG(STORA,STORC,DUMS,T,1,IZ-2,J)
RETURN

C SOLVE TRIDIAGONAL MATRIX BY USE OF GAUSSIAN ELIMINATION

SUBROUTINE TRIDIG(STORA,STORC,DUMS,T,M,MAX,L)
DIMENSION STORA(58),STORC(58),DUMS(58),T(58,21)
DIMENSION COEF1(58),COEF2(58)
COEF1(1)=(STORC(1)/STORA(1))
COEF2(1)=(DUMS(1)/STORA(1))
KMAX=MAX-1
DO 7 K=2,KMAX
7 COEF1(K)=(STORC(K))/STORA(K)+STORC(K-1)*COEF1(K-1))
DO 8 K=2,MAX
8  COEF2(K) = - (STORBC(K-1) * COEF2(K-1) + DUMS(K)) / (STORA(K) + STORBC(K-1) * COEF1(K-1))
IF (M.EQ.1) GO TO 10
T(L,MAX) = COEF2(MAX)
DO 9 K=1,KMAX
   N = MAX-K
9   T(L,N) = COEF1(N) * T(L,N+1) + COEF2(N)
   RETURN
10  N = MAX+1
    T(N,L) = COEF2(MAX)
    DO 11 K=1,KMAX
       N = MAX+1-K
11   T(N,L) = COEF1(N-1) * T(N+1,L) + COEF2(N-1)
   RETURN
END