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**Krypton-85 Disposal Program
Semiannual Report
August 15, 1977 - March 31, 1978**

Robert D. Klett, Editor

Sandia Laboratories

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ERF-45 DISPOSAL PROGRAM
SUMMARY REPORT
AUGUST 15, 1977 - AUGUST 1, 1978

Robert D. Lloyd, Editor
Scrubber Programs Division 4536
Sandia Laboratories
Livermore, NM 94550

ABSTRACT

This report summarizes the first 7.5 months of the krypton-85 disposal program. Included are task definitions, and initial progress in program, disposal system studies, SURE compatibility, augmented feed dissolution, material qualification, exterior canister compatibility, concrete liners for canisters, and geologic transport. Feasibility studies indicate that ERF-45 can be disposed of at a SURE facility or in near-surface geologic repositories.

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KRYPTON-85 DISPOSAL PROGRAM
SEMIANNUAL REPORT
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Introduction

This semiannual report covers the first 7.5 months of the Krypton-85 disposal program. The body of the report contains an overview of the program and a technical summary. Details of activities for this reporting period are in the appendices.

The overall Kr-85 isolation program, from processing to isolation, is shown in Figure 1. The disposal part of the program includes (1) system analyses to select the best combination of waste form, disposal process, and disposal medium; (2) compatibility assurance of the canister with the disposal medium; (3) selection of a disposal process or processes; (4) transportation, if needed; and (5) the design and operation of a repository. The disposal options given in Figure 1 also show up in the flow diagram (Figure 2). After separation, the Kr-85 can either be stored and eventually disposed of at the reprocessing or repackaging sites, or it can be shipped to a central repository. With either option, the krypton could be processed into a solid form, disposed of as a compressed gas in bottles, or injected into low-pressure geologic reservoirs.

Near-term objectives of the Kr-85 program were stated and responsibilities were assigned during two program review meetings.^{1,2} Objectives are:

1. Design a repository for defense Kr-85 or Kr-85/Xe mixtures. This includes krypton from the Hanford, Idaho Falls, and Savannah River reprocessing plants and any krypton that would be released by repackaging or selective reprocessing as part of the terminal storage program.
2. Keep development and capital costs low because of the relatively small quantities of Kr-85.
3. Return the Kr-85 to the HLW waste stream for disposal if possible.
4. Develop the disposal process with the potential for handling larger quantities of waste if commercial reprocessing is resumed.

Figure 1. Krypton-85 Isolation Program

Figure 2. Krypton-85 Flow Diagram

At that time Sandia's assigned responsibilities were to:

1. Conduct system studies of waste processing, disposal processes, and disposal media.
 - a. Select an above-ground or near-surface geologic disposal process for on-site or central disposal.
 - b. Conduct alternate disposal concept feasibility studies for commercial waste.
 - c. Provide input for the selection of a waste form by optimizing loads and configurations, computing number of disposal modules, and predicting total disposal costs for each waste form.
2. Work on a preliminary design for a shallow geologic repository.
3. Investigate transportation problems and needs.
4. Investigate integration of Kr-85 into the HLW or spent-fuel programs.
5. Develop a glass-ceramic liner for pressurized gas bottles as a barrier between the bottle and the Kr-85 decay product, rubidium.
6. Insure compatibility between the canister exterior and the disposal medium.

The overall technical approach for the development of a repository for Kr-85 is summarized below. Only the system studies and applied research apply to the near-term program.

System Studies

Parametric Sizing - Waste concentration, waste form, disposal media, and disposal process

Compatibility with Other Storage and Disposal Programs

Thermal, Mechanical, and Radiation Modeling

Disposal Concept Evaluation

Recommendations for Waste Form

Applied Research

Materials Compatibility

Geologic Transport of Waste

Configuration-Dependent Heat Transfer

Design

Design and Test Prototype

Design Repository or Disposal Module

Pilot Plant

Monitor Design, Construction, and Operation

Program Results

Progress to date on the Kr-85 disposal program is summarized below:

1. Initial activities were a literature search and contacts with related programs to find areas of technical exchange and to prevent duplication of effort. Many similarities were found between the Spent Unreprocessed Fuel (SURF) and Kr-85 disposal programs.
2. Although many existing system computer programs can be used for the Kr-85 program, several new thermal system programs were needed. These programs are being developed.
3. Sizing studies, conducted with various geologic disposal media ranging in heat dissipation ability from salt to dune sand, indicate that the geologic disposal of Kr-85 is feasible in either near-surface repositories or in deep-mined cavities.
4. SURF compatibility studies are in progress. No technical reasons were found that would prohibit disposing of Kr-85 at a SURF storage facility.
5. The disposal-concept evaluation is nearly complete. Much of the input data used in the evaluation came from a similar SURF study.^{3,4}
6. A complete material qualification program plan was developed, although only the canister exterior and ceramic liner are Sandia's responsibility. In-house laboratories were modified and metallurgy and ceramic tests were initiated.
7. Detailed gas bottle convection and SURF thermal, mechanical, and radiation modeling were initiated.
8. A geologic transport program plan including medium characterization, fluid flow, and sorption was written.
9. A geologic flow testing system is being developed in conjunction with other nuclear waste disposal programs and the in-situ oil shale program.

Summary of Appendices

Sizing Study for Disposal in Various Geologic Media (Appendix A) -- A preliminary study was conducted to assess the feasibility of geologic disposal of Kr-85. Closed-form approximate equations were used in the analysis, making it possible to investigate a large number of parameter combinations. No attempt was made to obtain design data. Canister size, gas pressure (or concentration for a solid waste form), canister wall thickness, and media properties were varied parametrically. Table I shows the peak surface temperatures of a column of stacked standard gas bottles in each of the geologic media. On the basis of allowable pressures in the ASME Pressure Vessel Code, all media, with the exception of dune sand, are suitable for storage of gas bottles with a room-temperature compression ratio of 36.3 (initial heating rate of 4.28 kW/m^3). Even dune sand could be used as a disposal medium with smaller-diameter cylinders or lower loadings. High thermal resistance between the canister and the medium would reduce the maximum allowable waste concentration.

TABLE I

Peak Surface Temperatures of Standard Gas Bottles
(Initial Heating Rate = 4.28 kW/m^2 ;
 30°C Initial Medium Temperature)

<u>Disposal Medium</u>	<u>Peak Canister Temperature ($^\circ\text{C}$)</u>
Salt	61
Mafic Igneous	78
Felsic Igneous	92
Limestone, Sandstone, Shale	111
Granitic Soil	137
Dune Sand	280

SURF Compatibility Studies (Appendices B and C) -- A review of the SURF program revealed many similarities with the Kr-85 disposal program. Potential SURF/Kr-85 interactions include:

1. Disposal of small quantities of Kr-85 at the SURF facility to minimize duplication of receiving, lag storage, handling, and emplacement hardware.
2. Disposal of Kr-85 from commercial waste at the SURF facility if reprocessing is resumed.
3. Use of the SURF/E-MAD field test setup for Kr-85 disposal tests.
4. Technical exchange. Many of the SURF analyses, repository and handling-equipment designs and tests, and safety studies are used for the Kr-85 program. Kr-85 system and geologic transport studies should be useful to the SURF program.

The use of SURF and/or HLW repositories for Kr-85 disposal has the potential of reducing costs, reducing the total number of disposal sites, and returning Kr-85 to the rest of the nuclear waste stream. The first part of the compatibility study between these disposal programs consisted of comparing volumetric heating rates, canister sizes, and allowable temperatures. Figure 3 compares the heating rates of two solid Kr-85 waste forms to two packaging arrangements of 3-year-old spent fuel. Fuel rods are packaged 4.60 MTHM/m^3 and fuel bundles are packaged 1.38 MTHM/m^3 . The heating rates are very similar for the first 20 yr, during which time peak canister temperatures are reached for all proposed disposal geometries. Kr-85 canisters cool more quickly and affect a smaller volume of the disposal medium than spent fuel. Parametric thermal analyses show that waste form/canister size combinations can be found that are compatible with SURF and HLW facilities by using annular waste configurations with solid Kr-85 waste and adjusting fill pressures with gaseous waste.

More detailed stress and thermal analyses were made based on a modified SURF canister in a dry well disposal module (Figure 4). The canister is 169.65 in. long x 14 in. diameter with a capacity of 385 L. The only modifications needed for solid Kr-85 waste would be the addition of an annular sleeve. The top closure would be changed and a fill valve would be added for gaseous

6.0. If the dry well is placed in Hanford soil, the peak allowable gas pressure would be 590 psig and peak master temperature would be 240°C. This corresponds to a fill pressure at 25°C of 100 psig and a compression factor of 24 from standard conditions.

Figure 4. SURF Dry Well Configuration

Based on existing data,^{5,6} a preliminary study was made to compare two solid waste forms in a SURF dry well. The results in Table II show canister loadings and the expected numbers of disposal modules required for several scenarios. The higher allowable concentrations of Kr-85 in sputtered metal are due to a higher loading ratio, higher thermal conductivity, and higher allowable temperature.

TABLE II
Kr-85 in SURF Canisters

Parameter	Waste Form	
	Zeolite	Amorphous Sputtered Metal
Initial Heating Rate/Canister (kW)	0.19	2.25
Initial Radiation/Canister (MCi)	0.13	1.50
Canister/Yr (no commercial reprocessing)	10.	1.
Canisters/Yr (1500-Tonne reprocessing plant)	119.	11.
Canisters by Year 2000 (full reprocessing)	13,500.	1,200.

Heat Dissipation (Appendix D) -- The use of heat pipes was suggested to augment heat transfer from geologic repositories, either to the atmosphere from shallow modules or to surrounding rock from deep repositories. A review of current technology indicated that heat pipe applications for nuclear waste disposal are very limited because of their relatively short lifetimes, cost, and sealing problems. Using a heat pipe to remove heat from a cluster of canisters in a dry well was found to be less effective than a single canister with heat removal by conduction through the soil.^{3,4}

Material Qualification (Appendices E and F) -- A complete development program for Kr-85 containers would include the following steps:

1. Definition of functions and constraints.
2. Definition of internal and external environments.
3. Review of existing technology to find out what would prohibit the use of some materials and would suggest other materials for testing.
4. Laboratory tests.
5. Analytical modeling of test results to produce design data and interpret test results.
6. Trade-off analyses of material and fabrication costs with functional requirements.

7. Verification tests including surveillance, nondestructive, and demonstration tests to verify models.
8. Final design.

The program in Appendix E covers items 3, 4, 5, and part of 7. It is a complete materials qualification program plan and does not exclude work that has been or will be conducted by other contractors. Since there are several testing philosophies in these areas, some overlap in test programs is suggested. The purpose of this program is to yield design data and fabrication techniques as well as select structural materials and coatings.

Surveillance facilities were examined to determine their suitability for testing pressurized 100-L bottles. A modular steady-pressure design similar to those used at Sandia Laboratories, Livermore is recommended. Each bottle has its own secondary container and pressure monitor connected to an alarm panel. This type of facility has a low initial capital cost and can be enlarged as the need arises.

Materials Research and Development (Appendices G, H, and I) -- Several laboratory modifications were made to handle rubidium, to facilitate testing under geologic disposal environments, and for the application of coatings.

Most of the external metallurgy tests completed to date were in conjunction with other nuclear test programs. In-situ corrosion tests of several metal coupons are being conducted in salt, water, and helium. Preliminary laboratory tests indicate that stainless steels, Inconel 600, and titanium could be used as canister materials in salt; 1018 carbon steel is marginal, and aluminum is not good.

Samples of several glasses are being exposed to rubidium but results are not yet available.

Geologic Transport (Appendix J) -- A program plan was laid out for geologic transport of radionuclides from a leaking canister. It included (1) hydrologic flow characteristics, heat transfer, and soil characterization, mineralogical and chemical measurements from candidate disposal sites; (2) response of media from the disposal sites to heat and radiation; and (3) krypton migration studies. Initial migration studies would be parametric, followed by site-specific modeling. Sorption will also be studied.

Although krypton is an inert gas, it may be sorbed to a limited degree by the disposal medium. Based on information in the literature, an extremely simplified model of a leak 20 ft below the surface was used to predict sorption and retention in soil pores. The soil column could retain the krypton from two 50-L bottles pressurized to 500 psig.

Geologic Flow Testing (Appendix K) -- Flow characteristics of geologic repositories such as transmissivities, locations, and effective hydraulic radii of cracks; and location, thickness, porosity, and effective permeability of porous layers are essential for transport analyses and safety assessments. These properties cannot be measured in a laboratory because geologic materials are not homogeneous and flow paths are disturbed when samples are removed from a formation. A new in-situ flow-measuring system is being developed to fulfill these needs. Flow is established between two wells and a tracer is injected into the input well. A string of detectors in the outlet well measures times of arrival, intensities, and changes in pulse shape of the tracer at various locations in the well. The flow was modeled for cracks and porous regions and data-reduction equations were derived. The system was used with the first generation of field-test hardware at Oak Ridge, TN, in the Conasauga shale heater experiment and for evaluating oil shale rubblization.

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APPENDIX A

Sizing Study of Krypton-85 Canisters in Various Geologic Media (Robert D. Klett, 4536)

A preliminary sensitivity study of Kr-85 geologic disposal was conducted to investigate the feasibility of burying gas cylinders filled with Kr-85 in various geologic media. The reference configuration for this study was standard gas cylinders buried in cased drilled holes in a geologic formation, with heat transferred from the cylinders to the formation through a fluid such as water. Each cylinder had a 23-cm OD, 0.5-cm wall thickness, and was 1.32 m long, including a 17-cm valve. The cylinders were loaded for a total initial heating rate of 187 W/cylinder, which is equivalent to 4.28 kW/m^3 with a decay half-life of 10.73 yr. The primary purpose of the study was to predict the maximum surface temperature of buried reference cylinders.

The analysis was conducted with the aid of computer program TMAX. This program uses a closed-form approximate solution to predict the maximum surface temperatures of cylinders containing exponentially decaying heat sources and the time after burial that the maximum surface temperatures are reached. Run time is very short and all input variables can be varied over wide ranges making this program well-suited for system and sensitivity studies. The accuracy of the program was checked against CINDA finite-difference models and agreement was within 5% for conditions similar to this study. The program also prints out a parameter (δT) which is an indication of the accuracy for each set of input variables.

In addition to the reference case, this analysis includes 6 geologic media, 5 volumetric heating rates, and 10 canister sizes for the purpose of defining the limiting conditions for this concept and the sensitivity of the concept to each variable. The results of the study are given in the enclosed computer printouts and in Figure A-1. Each page of the computer output is for a different group of geologic media that are listed in order of decreasing ability to absorb and transmit heat. The first two columns of the output are typical values for heat capacity (ρC_p) and thermal conductivity of the media group. Granitic soil and dune sand bracket the heat dissipation properties of most soils. The third column of the output is the initial heating rate in kW/m^3 . The fourth column is the decay half-life of the waste. The fifth and sixth columns are the inner and outer radii of the cylinders. In addition to the reference cylinder, several other standard Schedule 40 pipe sizes are included. Schedule 20 and 100, size 12 pipes are also included to show the effect of wall thickness. The following listing correlates pipe sizes with inner and outer radii:

<u>Pipe Size</u>	<u>Schedule</u>	<u>IR (m)</u>	<u>OR (m)</u>
6	40	.07793	.08414
8	40	.1014	.1095
Reference	Reference	.1100	.1150
10	40	.1273	.1365
12	20	.1556	.1619
12	40	.1516	.1619
12	100	.1405	.1619
14	40	.1667	.1778
16	40	.1905	.2032
18	40	.2143	.2286

Computed values are shown on the right side of the output at the end of this appendix. TIME is years after burial needed for the surface of the container to reach peak temperature. TR is the peak surface temperature if initial rock temperature is 30°C. The LAMT columns indicate that the accuracy was good over all conditions selected for this study. The figure at the end of this appendix is a graphical representation of the effects of initial volumetric heating rate (pressure) and canister size on peak temperature.

Results of this study show that the disposal medium surrounding the reference container must have heat dissipation properties at least as good as felsic igneous rock if water is used to transmit heat from the container to the medium. Peak temperatures exceed 100°C for other media, higher heating rates, or larger diameters. If radiant-heat transfer and gas conduction are used between the container and the medium, many other combinations may be usable, including storage in soils.

This was far from being a complete system study and no attempt was made to optimize any part of the disposal concept. Since Kr-85 is a gas, may be pressurized, has a relatively short half-life, and has a low sorption coefficient, the isolation methods being developed for solidified waste may not be applicable. It will probably be necessary to develop an integrated program to find the best combination of waste form, containers, disposal process, and disposal medium for safe, economical isolation of radioactive gases.

Figure A-1. Peak Rock/Canister Interface Temperatures With Kf-85 Waste in Schedule-40 Pipe Canisters and Felsic Igneous Rock, $T_i = 30^\circ\text{C}$

[illegible]

TI = 30.3 (DEG C)

References

$$V_2 = 31.2 \text{ (0.63 : 0.9)}$$
[illegible]

LYMESTONE, GYPSUM, SANDSTONE

VI = 10.01060 G

CMR(KJ/MSD)	CMR(KJ/MSD)	CMR(KJ/MSD)	ML(40)	ML(40)	ML(40)	TIME(00)	TIME(00)	TIME(00)
2.400F+03	1.700F+02	2.140F+03	1.373F+01	7.730F+02	4.414F+02	1.714F+03	5.170F+01	1.195E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.814E-01	1.893F+01	1.403F+03	6.500F+01	1.166F-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.180F+01	1.153E-01	1.821E+03	7.072F+01	1.179F-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.273F+01	1.365F+01	1.943E+03	4.234E+01	1.272E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.350E-01	1.613E-01	1.362E+03	1.043E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.516F+01	1.613E-01	2.140F+03	1.011E+02	1.263E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.405F+01	1.613E-01	1.962E+03	9.111F+01	1.267E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.667E-01	1.773E-01	2.034F+03	1.140E+02	1.236E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.305F+01	2.332F+01	2.067E+03	1.359E+02	1.337E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	2.143E-01	2.246E-01	2.126E+03	1.534E+02	1.375E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	7.730F+02	4.414E-02	1.708E+03	7.319F+01	1.105E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.814E-01	1.893E-01	1.403E+03	1.000E+02	1.156E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.180F+01	1.150E-01	1.821E+03	1.114E+02	1.178E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.273E-01	1.355E-01	1.943E+03	1.347E+02	1.222E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.355E-01	1.613E-01	1.362E+03	1.799E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.516E-01	1.613E-01	1.962E+03	1.723E+02	1.259E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.405F+01	1.613E-01	1.932E+03	1.522E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.667E-01	1.773E-01	2.034F+03	1.908E+02	1.236E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.305E-01	2.332E-01	2.067E+03	2.618F+02	1.337E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	2.143E-01	2.246E-01	2.126E+03	2.809E+02	1.375E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	7.730F+02	4.414E-02	1.708E+03	1.164E+02	1.105E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.814E-01	1.893E-01	1.403E+03	1.708E+02	1.156E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.180E-01	1.150E-01	1.821E+03	1.929F+02	1.179E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.273E-01	1.355E-01	1.943E+03	2.354E+02	1.222E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.355E-01	1.613E-01	1.362E+03	3.290E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.516E-01	1.613E-01	1.962E+03	3.146E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.405E-01	1.613E-01	1.932E+03	2.744E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.667E-01	1.773E-01	2.034F+03	3.659E+02	1.236E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.305E-01	2.332E-01	2.067E+03	4.535F+02	1.337E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	2.143E-01	2.246E-01	2.126E+03	5.491E+02	1.375E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	7.730F+02	4.414E-02	1.708E+03	1.536E+02	1.105E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.814E-01	1.893E-01	1.403E+03	2.402E+02	1.156E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.180E-01	1.150E-01	1.821E+03	2.743E+02	1.179E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.273E-01	1.355E-01	1.943E+03	3.441E+02	1.222E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.355E-01	1.613E-01	1.362E+03	4.791E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.516E-01	1.613E-01	1.962E+03	4.659E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.405E-01	1.613E-01	1.932E+03	4.664E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.667E-01	1.773E-01	2.034F+03	5.333E+02	1.236E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.305E-01	2.332E-01	2.067E+03	6.453E+02	1.337E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	2.143E-01	2.246E-01	2.126E+03	8.047E+02	1.375E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	7.730F+02	4.414E-02	1.708E+03	2.624E+02	1.105E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.814E-01	1.893E-01	1.403E+03	3.133E+02	1.166E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.180E-01	1.150E-01	1.821E+03	3.553E+02	1.179E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.273E-01	1.355E-01	1.943E+03	4.447E+02	1.222E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.355E-01	1.613E-01	1.362E+03	6.236F+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.516E-01	1.613E-01	1.962E+03	5.942E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.405E-01	1.613E-01	1.932E+03	5.149E+02	1.269E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.667E-01	1.773E-01	2.034F+03	7.013E+02	1.236E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	1.305E-01	2.332E-01	2.067E+03	4.771F+02	1.337E-01
2.400F+03	1.700F+02	2.140F+03	1.373F+01	2.143E-01	2.246E-01	2.126E+03	1.004E+03	1.375E-01

Reference

GRANITIC SOIL

T1 = 30.0000 G

COMPONENT	COMPONENT	COMPONENT	COMPONENT	COMPONENT	COMPONENT	COMPONENT	COMPONENT	COMPONENT
2.100E+03	2.100E-04	2.143E+03	1.673E+01	7.736E-02	4.414E-02	1.673E+07	5.033E+31	1.122E-01
2.100E+03	2.100E-04	2.140E+03	1.673E+01	1.014E-01	1.195E-01	1.432E+07	7.553E+31	1.135E-01
2.100E+03	2.100E-04	2.140E+03	1.673E+01	1.100E-01	1.153E-01	1.451E+07	9.333E+31	1.197E-01
2.100E+03	2.100E-04	2.140E+03	1.673E+01	1.273E-01	1.165E-01	1.371E+07	3.033E+31	1.262E-01
2.100E+03	2.100E-04	2.141E+03	1.673E+01	1.553E-01	1.019E-01	1.337E+07	1.280E+32	1.231E-01
2.100E+03	2.100E-04	2.143E+03	1.673E+01	1.514E-01	1.013E-01	1.337E+07	1.230E+32	1.231E-01
2.100E+03	2.100E-04	2.143E+03	1.673E+01	1.405E-01	1.013E-01	1.337E+07	1.093E+32	1.291E-01
2.100E+03	2.100E-04	2.143E+03	1.673E+01	1.667E-01	1.774E-01	2.043E+07	1.353E+32	1.313E-01
2.100E+03	2.100E-04	2.143E+03	1.673E+01	1.905E-01	2.332E-01	2.136E+07	1.643E+32	1.352E-01
2.100E+03	2.100E-04	2.143E+03	1.673E+01	2.143E-01	2.246E-01	2.167E+07	1.934E+32	1.431E-01
2.100E+03	2.100E-04	2.200E+03	1.673E+01	7.736E-02	4.414E-02	1.355E+07	9.661E+31	1.122E-01
2.100E+03	2.100E-04	2.200E+03	1.673E+01	1.014E-01	1.095E-01	1.432E+07	1.217E+32	1.135E-01
2.100E+03	2.100E-04	2.200E+03	1.673E+01	1.100E-01	1.153E-01	1.451E+07	1.366E+32	1.197E-01
2.100E+03	2.100E-04	2.201E+03	1.673E+01	1.273E-01	1.165E-01	1.371E+07	1.470E+32	1.262E-01
2.100E+03	2.100E-04	2.201E+03	1.673E+01	1.553E-01	1.019E-01	1.337E+07	2.240E+32	1.231E-01
2.100E+03	2.100E-04	2.201E+03	1.673E+01	1.514E-01	1.013E-01	1.337E+07	2.160E+32	1.231E-01
2.100E+03	2.100E-04	2.202E+03	1.673E+01	1.405E-01	1.013E-01	1.337E+07	1.890E+32	1.291E-01
2.100E+03	2.100E-04	2.202E+03	1.673E+01	1.667E-01	1.774E-01	2.043E+07	2.495E+32	1.313E-01
2.100E+03	2.100E-04	2.202E+03	1.673E+01	1.905E-01	2.332E-01	2.136E+07	3.065E+32	1.352E-01
2.100E+03	2.100E-04	2.203E+03	1.673E+01	2.143E-01	2.246E-01	2.167E+07	3.449E+32	1.431E-01
2.100E+03	2.100E-04	2.567E+03	1.673E+01	7.736E-02	4.414E-02	1.355E+07	1.322E+32	1.122E-01
2.100E+03	2.100E-04	2.567E+03	1.673E+01	1.014E-01	1.095E-01	1.432E+07	2.133E+32	1.135E-01
2.100E+03	2.100E-04	2.567E+03	1.673E+01	1.100E-01	1.153E-01	1.451E+07	2.432E+32	1.197E-01
2.100E+03	2.100E-04	2.567E+03	1.673E+01	1.273E-01	1.165E-01	1.371E+07	3.033E+32	1.262E-01
2.100E+03	2.100E-04	2.567E+03	1.673E+01	1.553E-01	1.019E-01	1.337E+07	4.219E+32	1.231E-01
2.100E+03	2.100E-04	2.567E+03	1.673E+01	1.514E-01	1.013E-01	1.337E+07	4.020E+32	1.231E-01
2.100E+03	2.100E-04	2.568E+03	1.673E+01	1.405E-01	1.013E-01	1.337E+07	3.495E+32	1.291E-01
2.100E+03	2.100E-04	2.568E+03	1.673E+01	1.667E-01	1.774E-01	2.043E+07	4.590E+32	1.313E-01
2.100E+03	2.100E-04	2.568E+03	1.673E+01	1.905E-01	2.332E-01	2.136E+07	5.311E+32	1.352E-01
2.100E+03	2.100E-04	2.569E+03	1.673E+01	2.143E-01	2.246E-01	2.167E+07	7.075E+32	1.431E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	7.736E-02	4.414E-02	1.355E+07	1.998E+32	1.122E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.014E-01	1.095E-01	1.432E+07	1.853E+32	1.135E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.100E-01	1.153E-01	1.451E+07	3.449E+32	1.197E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.273E-01	1.165E-01	1.371E+07	1.811E+32	1.262E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.553E-01	1.019E-01	1.337E+07	1.853E+32	1.231E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.514E-01	1.013E-01	1.337E+07	1.811E+32	1.231E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.405E-01	1.013E-01	1.337E+07	1.643E+32	1.291E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.667E-01	1.774E-01	2.043E+07	4.403E+32	1.352E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.905E-01	2.332E-01	2.136E+07	6.173E+32	1.262E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	2.143E-01	2.246E-01	2.167E+07	5.801E+32	1.291E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.014E-01	1.095E-01	1.432E+07	5.091E+32	1.231E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.100E-01	1.153E-01	1.451E+07	6.034E+32	1.313E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.273E-01	1.165E-01	1.371E+07	6.530E+32	1.362E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.553E-01	1.019E-01	1.337E+07	1.046E+33	1.431E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.514E-01	1.013E-01	1.337E+07	2.564E+32	1.122E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.405E-01	1.013E-01	1.337E+07	3.967E+32	1.135E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.667E-01	1.774E-01	2.043E+07	4.665E+32	1.197E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.905E-01	2.332E-01	2.136E+07	5.774E+32	1.262E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	2.143E-01	2.246E-01	2.167E+07	6.141E+32	1.291E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.014E-01	1.095E-01	1.432E+07	7.741E+32	1.231E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.100E-01	1.153E-01	1.451E+07	9.031E+32	1.313E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.273E-01	1.165E-01	1.371E+07	1.046E+33	1.362E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.553E-01	1.019E-01	1.337E+07	1.141E+33	1.431E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.514E-01	1.013E-01	1.337E+07	1.141E+33	1.431E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.405E-01	1.013E-01	1.337E+07	1.141E+33	1.431E-01
2.100E+03	2.100E-04	1.264E+01	1.673E+01	1.667E-01	1.774E-01	2.043E+07	1.141E+33	1.431E-01

Reference

DUNF 5443

FI = 33.10000 00

CM(X)/Y(X)	Z(X)/Y(X)	DI(X)/Y(X)	LI(Y)	LI(Y)	LI(Y)	LI(Y)	LI(Y)	LI(Y)
1.750E+01	1.500E-04	2.147E+00	1.073E+01	7.750E-02	4.414E-02	1.972E+00	9.669E+01	1.212E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.014E-01	1.013E-01	1.733E+00	1.374E+02	1.246E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.100E-01	1.153E-01	2.112E+00	1.548E+02	1.380E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.273E-01	1.394E-01	2.194E+00	1.894E+02	1.554E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.354E-01	1.501E-01	2.173E+00	2.577E+02	1.412E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.516E-01	1.601E-01	2.146E+00	2.461E+02	1.412E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.405E-01	1.501E-01	2.197E+00	2.156E+02	1.412E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.407E-01	1.777E-01	2.295E+00	2.644E+02	1.445E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	1.105E-01	2.312E-01	2.311E+00	3.645E+02	1.435E-01
1.750E+01	1.500E-04	2.147E+00	1.073E+01	2.147E-01	2.246E-01	2.146E+00	4.201E+02	1.542E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	7.750E-02	4.414E-02	1.876E+00	1.633E+02	1.213E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.014E-01	1.013E-01	1.999E+00	2.447E+02	1.296E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.100E-01	1.153E-01	2.012E+00	2.797E+02	1.307E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.273E-01	1.394E-01	2.096E+00	3.645E+02	1.354E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.354E-01	1.501E-01	2.197E+00	4.853E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.516E-01	1.601E-01	2.197E+00	4.622E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.405E-01	1.501E-01	2.143E+00	4.013E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.407E-01	1.777E-01	2.295E+00	5.386E+02	1.445E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.105E-01	2.312E-01	2.311E+00	6.495E+02	1.435E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	2.147E-01	2.246E-01	1.976E+00	9.101E+02	1.472E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	7.750E-02	4.414E-02	1.943E+00	2.967E+02	1.246E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.014E-01	1.013E-01	2.012E+00	5.293E+02	1.307E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.100E-01	1.153E-01	2.096E+00	6.495E+02	1.354E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.273E-01	1.394E-01	2.197E+00	9.407E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.354E-01	1.501E-01	2.197E+00	9.407E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.516E-01	1.601E-01	2.143E+00	6.945E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.405E-01	1.601E-01	2.197E+00	7.725E+02	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.407E-01	1.777E-01	2.295E+00	1.648E+03	1.445E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.105E-01	2.312E-01	2.311E+00	1.380E+03	1.435E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.273E-01	2.246E-01	2.146E+00	1.590E+03	1.542E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.354E-01	2.246E-01	1.976E+00	4.301E+02	1.213E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.516E-01	1.601E-01	1.943E+00	6.746E+02	1.246E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.405E-01	1.501E-01	2.012E+00	7.798E+02	1.307E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.407E-01	1.777E-01	2.294E+00	9.895E+02	1.354E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.105E-01	1.601E-01	2.143E+00	1.390E+03	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.273E-01	1.601E-01	2.197E+00	1.327E+03	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.354E-01	1.601E-01	2.197E+00	1.146E+03	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.516E-01	1.777E-01	2.295E+00	1.556E+03	1.445E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.405E-01	2.312E-01	2.311E+00	1.574E+03	1.435E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.407E-01	2.246E-01	2.146E+00	2.373E+03	1.542E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.105E-01	2.312E-01	1.976E+00	5.635E+02	1.213E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.273E-01	2.246E-01	1.943E+00	6.945E+02	1.246E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.354E-01	2.246E-01	2.012E+00	8.094E+02	1.296E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.516E-01	2.312E-01	2.096E+00	1.023E+03	1.307E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.405E-01	2.312E-01	2.096E+00	1.330E+03	1.354E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.407E-01	2.246E-01	2.197E+00	1.651E+03	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.105E-01	2.312E-01	2.197E+00	1.759E+03	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.273E-01	2.246E-01	2.146E+00	1.515E+03	1.412E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.354E-01	2.246E-01	2.146E+00	2.065E+03	1.445E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.516E-01	2.312E-01	2.146E+00	2.540E+03	1.435E-01
1.750E+01	1.500E-04	4.267E+00	1.073E+01	1.405E-01	2.312E-01	2.146E+00	3.174E+03	1.542E-01

Reference

APPENDIX B

Compatibility of Kr-85 With SURF and High-Level Waste Repositories (Robert D. Klett, 4536)

The use of SURF and/or high level waste (HLW) repositories for Kr-85 disposal has the potential of reducing disposal costs, reducing the total number of disposal sites, and returning Kr-85 to the rest of the nuclear waste stream. Possible uses of SURF include:

1. Much of the SURF analyses, design, handling and shipping hardware, and safety studies could be used for Kr-85, because of the similarities between SURF and the disposal of Kr-85 fixed in solids.
2. The SURF field-test setup and handling equipment at the NTS Engine Maintenance Assembly and Disassembly Facility (E-MAD) could be used for a Kr-85 demonstration test.
3. SURF could possibly be used for HLW storage if commercial reprocessing begins. Such a facility could probably be used for Kr-85 disposal.
4. If a SURF facility is no longer needed after reprocessing begins, the facility could be used for Kr-85 after a cool-down period.
5. A SURF facility could probably store small quantities of Kr-85 from Hanford, Idaho Falls, and Savannah River.

The first part of the feasibility study to investigate the use of SURF and HLW repositories consisted of a comparison of volumetric heating rates, canister sizes, and allowable temperatures. Volumetric heating rates versus time after disposal are given in Figure B-1 for three solidified forms of Kr-85. B-1 Heating rates for HLW, spent fuel, and selectively reprocessed waste are given in Reference B-2. Figure B-2 shows the heating rates for single fuel bundles packaged in cylindrical containers $(1.38 \frac{\text{MT} \cdot \text{ft}^3}{\text{m}^3})$ and disassembled fuel rods packed in containers $(4.60 \frac{\text{MT} \cdot \text{ft}^3}{\text{m}^3})$. Heating rates for HLW are given in Figure B-3 for waste buried immediately after reprocessing and for waste aged 10 and 30 yr before disposal. The time after burial for any waste form to reach maximum temperature depends on the disposal medium, decay rate of the waste, and canister size. Parametric studies have shown that peak temperatures are reached in less than 5 yr for most configurations, waste forms, and disposal media. Comparison of the heating rates in Figures B-1 through B-3 during the first 5 yr shows that Kr-85 is nearly the same as spent fuel and HLW, indicating that disposal sites designed for the heating rates of the other waste forms should be compatible with Kr-85 waste. The major differences are that a Kr-85 repository would remain hot for a much shorter time and would heat a much smaller volume of the disposal medium.

Figure B-1. Heating Rate for Krypton-85 Buried 1 Yr After Reprocessing

Figure B-2. Heating Rate for Spent Fuel Buried 3 Yr After Discharge From Reactor

Figure B-3
Heating Rates for High-Level Waste 13.1 MTHM/m³

The disposal medium and canister materials define the maximum allowable temperatures of the canister/medium interface and the waste form defines the maximum centerline temperature. Maximum canister temperatures for SURF are predicted to be about 325°C. Maximum predicted interface temperatures are not as yet available. The maximum allowable centerline temperature for borosilicate HLW is between 600° and 800°C; allowable interface temperatures are usually governed by the disposal medium. Geochemical analyses of all candidate media are not completed, but the following listing summarizes the limiting temperatures used for analyses and preliminary design work:

Salt	250°C
Seabed	200°C
Shales	450-900°C
Limestones	500-800°C
Tuff	100-300°C
Mafic Igneous	1100°C
Felsic Igneous	1000°C
Soils	1000°C
Concrete	300-400°C

Temperature gradients can also be a critical parameter in brittle media. Maximum temperatures have not been established for Kr-85 fixed in solids, but leakage rates appear to be acceptable for zeolites at 150°C.^{B-1} Much higher temperatures are acceptable for sputtered metal. Reference B-3 shows that a 150°C temperature limit would not be exceeded in any rock with 10-in. diam containers filled with zeolite. The alluvium near the surface around E-MAD has an average thermal conductivity of 0.54 W/m °C placing it between dune sand and granitic soil. An interpolation between the last two tables in Reference B-3 shows a 9-in. cylinder of zeolite could be tested at E-MAD without exceeding 150°C. Cylindrical canisters from 12 to 24 in. in diameter have been considered for HLW and SURF, but only the 12-to 14-in. range meets the requirements for all geologic media. Zeolite containers 12 in. in diameter would remain below 138°C in salt and both igneous rocks.

If large-diameter Kr-85 containers would be more compatible with other waste-disposal facilities, an annular configuration could be used to reduce heating rates per unit length. Annular geometries also have the advantage of reducing the radial temperature gradient in the container more than they reduce the volume of waste they contain. The ratio of the volume of an annular cylinder to the volume of a solid cylinder is given by

$$\frac{V_{an}}{V_{cyl}} = 1 - \left(\frac{R_i}{R_o}\right)^2$$

and the ratio of the radial temperature gradients in annular and solid cylinders that contain heat-generating material is

$$\frac{\Delta T_{an}}{\Delta T_{cyl}} = 1 - \frac{R_i}{R_o} \left(1 - 2 \ln \frac{R_i}{R_o} \right).$$

These two equations are plotted in Figure B-4. An example shows that an annular cylinder with $R_i/R_o = 0.44$ would have only half the radial temperature rise of a solid cylinder but would contain 82% as much waste. This is an important consideration when leak rates from solids increase with temperature.

Figure B-4. Comparative Volumes and Maximum Radial Temperature Gradients of Solid and Annular Cylinders Filled With Heat-Generating Material

These preliminary analyses indicate that there should be no thermal incompatibilities between solid forms of Kr-85 waste and HLW repositories or SURF.

References

- B-1. D. A. Knecht, An Evaluation of Methods for Immobilizing Krypton-85, ICP-1125, July 1977.
- B-2. Memo, R. D. Klett to Distribution, "Reference Heating Rates for Spent Fuel and Reprocessed Waste," dated May 16, 1977.
- B-3. Appendix A of this report.

APPENDIX C

Maximum Allowable Pressures and Temperatures for Kr-85 Gas Stored in a SURF Canister (Robert D. Klett, 4536 and Donald B. Longcope, 1281)

Preliminary thermal and stress calculations were performed to determine the amount of Kr-85 gas that can safely be loaded in a modified SURF canister and stored in a dry well at Hanford, WA. The SURF canister is basically a cylinder 14 ft long by 14 in. OD, made of 304 stainless steel. It is constructed from Schedule 30 pipe (0.375-in. wall thickness), except for the top 8 in. where the support pins are located; this is Schedule 100 pipe. The lower end is closed with a Schedule 30 ellipsoidal pipe cap. The only change in the SURF canister used in this study was that the threaded closure at the top of the canister was replaced with a welded ellipsoidal Schedule 100 pipe cap and a valve.

A parametric thermal analysis was conducted to predict maximum canister temperatures as a function of initial volumetric heating rate. The calculations were made using a computer program based on a modified version of the MUFFI solution presented in Reference C-1 that includes the effects of the earth's surface and computes canister and waste temperatures. Maximum pressures were computed from heating rates and temperatures using heat generation and compressibility data. C-2 The peak temperature/pressure relationship is shown as a solid line in Figure C-1.

Figure C-1. Maximum Pressures and Temperatures of a Kr-85 Filled SURF Canister in a Dry Well at Hanford, WA

Maximum allowable pressure as a function of canister temperature was obtained by applying the rules of Reference C-3 to the specified pipe. These rules basically require a computation of the maximum tensile stress in the pipe according to the membrane theory of shells. Localized stresses near the ends were not considered, since it was assumed that if the canister were used as a pressure vessel the ends would be modified so the stresses there would be in balance with the membrane stresses away from the ends. The stainless steel considered is specified as SA 312, grade TP 304 L; tables in Reference C-3 account for the decrease in its strength (yield stress and ultimate tensile stress) with increasing temperature over the range from -20°F to 800°F. Data from References C-4 and C-5 were used for the temperature range from 800° to 1600°F. The maximum allowable internal pressure is plotted as the dashed line in Figure C-1. The intersection of the two curves is the design point. The peak allowable temperature is 240°C with a corresponding peak pressure of 590 psig. The fill pressure at 25°C would be 333 psig and the gas would be compressed by a factor of 24 from standard conditions.

This preliminary study did not include convection, variable thermal properties, or thermal stresses; the configuration was simplified slightly to permit the use of analytical solutions. Until a more detailed study is completed, a conservative peak pressure of 500 psig will be used for tests and analyses.

References

- C-1, D. W. Larson, D. K. Gartling, and H. C. Hardee, Thermal Response of Buried Radioactive Waste: Conduction Solutions, SAND75-0410, September 1975.
- C-2, A. B. Christensen, Physical Properties and Heat Transfer Characteristics of Materials for Krypton-85 Storage, KP-1128, September 1977.
- C-3, ASME Boiler and Pressure Vessel Code, Section VIII, Division 1.
- C-4, Structural Alloys Handbook, Vol. 2, Mechanical Properties Data Center, 1977 Edition, p. 304.
- C-5, Metals Handbook, Vol. 1, Metals Handbook Committee, American Society for Metals, 8th Edition, P. 503.

APPENDIX D

Heat Pipe Application Workshop (Robert D. Klett, 4536)

On October 20, 1977, a Heat Pipe Application Workshop was conducted at Los Alamos Scientific Laboratory to get researchers, suppliers, and government agencies together to discuss the state of the art, applications, ways to expand the use of heat pipes, and anticipated future requirements to help direct research. The first part of the workshop consisted of presentations by suppliers describing their products and research, followed by presentations by users describing their programs and heat-transfer problems that may have heat pipe applications. The rest of the time was spent in workshops discussing technical and management problems. Following is a brief summary of the meetings.

The speakers and companies or organizations they represented were

Introduction - Will Ranken - LASI.

Suppliers

Jim Taylor - Q Dot
Slater O'Hare - McDonnell Douglas
Ed Luedke - TRW
Frank Arcella - Westinghouse
Gabor Miskolczy - Thermoelectron
Walter Hienert - Dynatherm
Yale Eastman - Thermacore
Bob Haslett - Grumman
Al Basiulis - Hughes Aircraft
Ralph Vaughn - Ionics
Dale Walters - Sigma Research

Users

Mike Perlisweig - DOE (Cont RT) - Power Systems
Frank Bradburn - Idaho NEL - Industrial Conservation
Pat O'Riordan - DOE (NRA) - Spacecraft Power
Joe Joyce - NASA (LeRC) - Thermal Storage
Robert Klett - Sandia Laboratories - Nuclear Waste
John Bankston - Solar Heating and Cooling
Sam Ollendorf - NASA (Goddard) - Heat Recovery
Jim Morris - NASA (LeRC) - Thermionics

According to the suppliers and researchers, heat pipes have distinct advantages over other heat-transfer components in some applications. These are:

1. High equivalent thermal conductivity
2. Variable conductivity in zero gravity
3. Isothermal
4. Low thermal inertia
5. Reduced thermal stresses
6. Liquid freezing permissible in some designs
7. Smaller and lighter
8. Easily cleaned
9. Cheaper if designed into the system
10. Can be designed for a wide range of configurations and heat fluxes.

Suppliers felt that heat pipes could be used more in the following applications:

1. Heat flux amplification and deamplification
2. Thermal diode (only in the presence of a gravity field with heat flow in the direction opposite to gravitational force)
3. Temperature regulation
4. Multistream, crossflow, and superheat heat exchangers
5. Condensers
6. Recuperators
7. Configurations requiring curved heat transmission pipes (90 degrees limit when gravity is present)
8. Soil stabilization
9. Batch process heating
10. Where even-heat distribution is required.

All suppliers stressed that the entire system had to be designed for heat-pipe use and that add-ons or replacing other heat-transfer components with heat pipes is not usually economical or efficient. They also claim that heat pipes are used in many applications where they should not be used; that heat pipes designed for one set of operating conditions were often used in different conditions. Heat pipes are designed for specific heat-transfer conditions at each end, specific heat flux and temperature ranges, and chemical compatibility with an environment.

Currently, heat pipes are made to operate up to 700°C. Low-cost heat pipes usually last about 15 yr but the life can be extended to about 75 yr with more expensive structures and coatings. The longest heat pipe now being made is 75 ft but this is not a state-of-the-art limitation. Lengths over 125 ft could not be assembled in the factory because of transportation regulations.

Material research for heat pipes includes ceramic and refractory structures, refractory metal wicks and return troughs, and liquid metal and sulfur carrier fluids. Research goals include extending the operating life, increasing maximum temperatures to 1100°C, and operating at cryogenic temperatures. High transport wick and trough configurations are being investigated. McDonnell Douglas is doing a feasibility study of oil-well stabilization using heat pipes 1000 ft long.

The working-group tasks were to suggest what DOE could do to aid commercialization of heat-pipe technology and what could be done to stimulate industrial energy conservation (stressing heat pipes). The consensus was that DOE should

1. Sponsor information dissemination to educate project engineers about the capabilities and limitations of heat pipes.
2. Sponsor research and establish a center of excellence for heat pipes (IASI was suggested).
3. Sponsor a workshop with suppliers, researchers, and project engineers to exchange information on capabilities and specific design applications.

A suggested stimulus for industrial energy conservation was an energy audit for all new and existing plants, with tax credits as an incentive. The audit should cover all points where energy is used or lost, and all environments at each stage of the plant.

The potential uses of heat pipes in the nuclear waste disposal program are limited because of the short lifetime of heat pipes compared to the isolation requirements for nuclear waste. It is also desirable not to breach the repository unless it is necessary. There are some relatively short-term phases of nuclear waste storage that could utilize heat pipes. They might be used to remove heat from heavily shielded shipping containers or from storage structures to permit a higher packing density. Another application would be storage and disposal of short half-life isotopes such as Kr-85. A safety, cost, and land utilization study will be made to compare passive single-column geologic storage of gas bottles against clustered storage, with heat removal supplemented by heat pipes. If longer heat pipes are developed, they could be used to remove heat from thicker sections of low-pressure geologic reservoirs. Heat pipes would not be practical in salt because of corrosion problems.

APPENDIX E

Material Qualification Program Plan for Kr-85 Canisters

C. P. Ballard
J. W. Braithwaite
R. J. Egan
R. D. Klett
D. B. Longcope
J. W. Munford

Sandia Laboratories
November 11, 1977

This program plan outlines the research and development activities needed to qualify materials for Kr-85 storage and disposal containers. It includes internal and external environments, primary and secondary containers, closures, and (if needed) liners, overpacks, and coatings. Containers for short-term use include pressurized bottles for storage and vessels for transportation. Long-term disposal containers include casks for krypton fixed in solids or advanced pressure bottles. This program would yield design data and fabrication techniques as well as material selection information.

Metal Structures and Liners

The material qualification program for metal structures and liners for use in Kr-85 storage, shipping, and disposal bottles or canisters consists of the following steps:

1. Initial selection of several candidate alloys.
2. Reduction of the number of alloys to one prime candidate and one or two back-up materials.
3. Selection of processing and fabricating techniques that will not significantly degrade the material. These processing steps may include mechanical working (rolling, spinning, forging, and deep drawing), heat treatments, cleaning, welding, etc.
4. Complete testing of the final alloy selected and the production methods used to assure total compatibility in all phases and environments of production and service lifetimes.
5. Surveillance testing of prototype units

The basis for the alloy selections and specifics of the test and surveillance programs are outlined below. There are three primary material requirements for pressure vessels:

1. Assurance that the material has sufficient toughness to preclude sudden catastrophic failure from existing flaws under normal operating pressures.
2. Resistance to all possible exposure environments so that existing flaws will not grow (environmentally enhanced crack growth through stress-corrosion cracking, liquid-metal embrittlement, hydrogen embrittlement, caustic cracking, etc) to a critical size during the lifetime of the pressure vessel, and
3. High corrosion resistance in all exposure environments to preclude failure due to a variety of corrosive mechanisms (i.e., pitting, galvanic corrosion, general corrosion, etc).

The material qualification program is aimed at providing specific quantitative design data and to assure that the material will function properly in all the aforementioned areas.

Background

Experience in the design of high-reliability pressure vessels has indicated that the best way to guarantee against failures is to rely heavily on the methodology of fracture mechanics in all material evaluation programs. The fracture mechanics test technique involved will address the first two of the three material requirements outlined above. The reason intentionally flawed test specimens are selected rather than smooth specimens is that all forms of environmentally assisted cracking initiate from flaws which either existed prior to exposure to the environment or are generated through general corrosive attack. Many materials are very resistant to general corrosion and, as a consequence, appear immune to environmental cracking; however, if small flaws (cracks) are present, these same materials will rapidly fail under similar exposure conditions. There are numerous examples in the open literature and within our Laboratory where smooth-specimen testing has not only failed to correctly rank several materials with regard to environmental cracking susceptibility but has erroneously predicted immunity to environmental cracking. Since it is virtually impossible to fabricate any engineering structure that will not contain preexisting flaws of various sizes, the use of fracture-mechanics type specimens assures the most conservative approach to guarantee maximum safety and reliability.

These specimens and testing techniques allow us to determine threshold stress intensities for the alloys of interest. Stress intensities are a measure of the stresses at the tip of a crack and are proportional to flaw size, specimen geometry, and applied load. Values of the threshold stress intensity are then applied to the actual pressure vessels to calculate the maximum allowable flaw sizes that, at the design pressures, will assure us that crack growth will not occur. Nondestructive testing (NDT) techniques will then be used to guarantee that preexisting flaws will be less than this critical size and therefore will not grow.

Information generated from this testing program will yield a quantitative correlation between stresses, flaw sizes, failure time, crack growth rate, and environments which will be directly applicable through stress analysis and NDT techniques to a variety of pressure vessel designs. Data from smooth-specimen testing can have no such design applicability. Environments to be investigated include pressure, elevated temperature, radiation, and contact with rubidium and rubidium hydroxide.

The third material requirement is resistance to corrosion due to outside burial environments and impurities in the waste. The environments in which the materials must be inert consist of internal and external pressures, elevated temperatures, radiation flux, and corrosive agents. Some of the major external corrosive constituents which must be investigated are H_2O , O_2 , Cl^- , Cl_2 , CO_2 , H_2S , and SO_2 . Exact definition of the external environment is site-dependent and will need further investigation. The major internal corrosive agents are O_3 , H_2O , NO_x , and CO_x . Although a metal structural member will be required to ensure sufficient mechanical strength, it may be necessary to coat or overpack the canister to protect against the external environment and/or line the canister because of the internal environment.

Program Definition

The general outline for the physical metallurgy program, which depends heavily on preflawed specimens, is as follows:

1. Initial selection of candidate materials (for both gas bottles and related valves, tubing, etc) based on cost, fabricability, weldability, and known resistance to the exposure environments; e.g., Rb and caustic (RbOH).
2. Evaluation of an alternate bottle design based on liners which have high environmental resistance. Although the use of liners is common technology, they may increase unit cost for fabrication of each pressure vessel. However, liners may allow more design flexibility through the use of higher-strength and lower-cost structural materials, and decreased storage volume by allowing higher fill pressures.
3. Expansion of the test matrix to include weldments. If environmental degradation occurs, it will most likely be at welds where extensive metallurgical alterations exist. Also, degradation will usually be more severe in the weld area.
4. Candidate materials must be evaluated in caustic environments. Experience has shown that in any high-pressure gas system it is difficult to exclude water. Even stringent process controls may still allow small amounts of water which will rapidly react with rubidium to form the caustic (RbOH). Caustic environments severely degrade most standard gas bottle structure materials; e.g., 304SS and 4130. Thus it would seem inappropriate to exclude RbOH from the test matrix.

Specific test techniques will make extensive use of a variety of specimen geometries (i.e., compact tension, modified wedge, open loaded, etc) and load applications (increasing and constant load, and constant displacement) which will determine both threshold stress intensities and crack growth rates for the various combinations of materials and environments under consideration.

Test procedures are as follows. Figure E-1 shows a typical specimen geometry and the equation that relates stress intensity to specimen dimensions and test data. There are two basic approaches used to determine the threshold stress intensity for a given material-environment combination:

1. **Constant-Load Tests** - For these tests, six to eight specimens are loaded over a range of stress intensities in the environment of interest and the time to failure at each stress intensity is measured. The stress intensity at which no failure occurs is called the "threshold" (Figure E-2).
2. **Constant-Displacement Tests** - In this test a single specimen is loaded to a "high" stress intensity in the environment such that crack growth occurs. As the crack propagates, stress intensity decreases until crack growth ceases. The final crack length is used to calculate the stress intensity at which the crack no longer will grow. Typical data showing crack arrest for a constant-displacement test are given in Figure E-3.

Figure E-4 shows how test data are applied to a pressure vessel design. A graph obtained by stress analysis of pressure versus flaw size for a pressure vessel constructed of a material with fracture toughness (K_{IC}) and threshold stress intensity (K_{ISCC}) is shown, along with an explanation of its use.

In addition to these static or delayed-failure type tests, a series of dynamic tests is also planned. These tests measure the propensity for crack growth in the environments under various rates of load application. In this manner material response in the environments over a wide range of time scales will be determined. The test technique consists of loading the specimens to failure in the environments of interest while measuring the influence of loading rate on toughness, yield strength, ultimate strength, ductility, and fracture morphology.

Standard production practice for commercial pressure vessels and gas bottles generally involves a series of deep drawing operations; joints, when they are made, are circumferential welds. Welding will result in a significant increase in design flexibility; its use, as mentioned, will necessitate evaluation of weldments to assure satisfactory performance in the environments of interest. All the techniques previously discussed will be applied to weldments as well as to the parent material. Typical specimen configurations are depicted in Figure E-5, along with a discussion of test procedures and interpretation.

The chemical-metallurgy program would begin with a literature review to select the most economical material for most geologic environments. Heated pipelines and associated processing equipment have been buried for years and extensive corrosion data are available. Unfortunately, bedded salt and some rock formations proposed for waste disposal have not been extensively studied and, because of the great potential for corrosion, a laboratory study will have to be undertaken to allow proper material selection. The eventual material recommendation will be based on the following three studies:

1. Small Scale Laboratory Studies - These experiments will consist principally of electrochemical measurements that yield data concerning corrosion tendency and provide an accelerated method for determining corrosion rates.
2. Simulated Environment Laboratory Testing - Coupons of candidate materials will be placed in simulated environments for extended periods to verify and extend the results of the electrochemical testing of No. 1 above.
3. In-Situ Testing - Coupon assemblies will be designed and placed in-situ to verify the results of laboratory testing in close-to-actual conditions.

The types of metal corrosion which need to be evaluated in the laboratory and in-situ studies are (a) pitting corrosion, (b) general corrosion, (c) crevice corrosion, (d) galvanic coupling, and (e) enhanced corrosion due to heat sensitization during welding and due to radiation effects.

The primary stress-analysis task required in the development of containers for Kr-85 storage or disposal is the calculation of the stress intensity factor for the particular vessel geometry, fabrication process, material, expected crack size and shape, and loading conditions; i.e., storage pressure and wall temperature distribution. This calculated stress-intensity factor should be less than that determined from the companion fracture-mechanics testing program to assure that preexisting cracks will not grow. It will be necessary to estimate possible embrittlement-induced changes in material parameters entering the stress-intensity calculation. Some information on the effects of radiation embrittlement is available from experience in the nuclear-reactor industry. Also any effects of material removal by corrosion must be included in the stress analyses. The temperature at which a creep analysis of the gas bottle may become necessary should be determined, although a preliminary thermal analysis indicates low vessel-wall temperatures at which creep should not be significant.

Finally, a surveillance program is recommended which should consist of both continuous monitoring and periodic burst testing of prototype and full-scale gas bottles. Both continuous monitoring and burst testing should utilize pressure, temperature, strain, and acoustic emission data to maximize the usefulness of these tests. The initial fill environments of some gas bottles should be controlled to simulate various storage times (e.g., 1, 5, 10, 20 yr) by varying the temperature and the ratio of Kr-85/Rb. Because of the shortage of Kr-85, it may be necessary to simulate the internal environment using Rb, RbOH, an inert cover gas, external heating, and a γ

source if it is found that radiation damage is significant. All production techniques and procedures need to be duplicated in the full-size and scaled-down bottles. Most bottles should be production quality but some should be intentionally flawed to provide a quantitative correlation between laboratory tests and production bottles. It is anticipated that the surveillance tests could begin about 18 months after the qualification program begins and should precede the start of storage or disposal by at least 2 yr.

Glass-Ceramic Coatings and Liners

The objectives of this phase of the material qualification program are to evaluate glass and glass-ceramic coatings as corrosion-resistant barriers for Kr-85 pressure vessels and containers for immobilized Kr-85, to provide design data, and to define manufacturing procedures. The reaction of krypton and rubidium will be studied with regard to protection of the interior of the vessel. Water, brine, and attack by potentially corrosive geologic formations will be studied with regard to attack of the exterior.

Background

The chemical industry currently assures the reliable and continuously efficient operation of many reaction chambers by the use of protective glass-enamel coatings. The use of these coatings permits vessels to be made of relatively inexpensive mild steel. Lifetimes of glass-coated vessels of this sort are generally 15 yr or more, depending on usage. The cost of glass-coated vessels is comparable to that of stainless steel in most cases. Because the thermal expansion of the glass enamel must approximately match that of the steel, it contains a large concentration of alkali ions. This large concentration of alkali ions to some extent reduces the chemical durability of these glass coatings. An alternative type of coating, a glass-ceramic coating, may be more corrosion resistant. Glass-ceramic compositions have been formulated with expansion coefficients ranging from 0 to 200×10^{-7} cm/cm/°C and therefore can match most metals.

Glass ceramics are prepared by the controlled devitrification of selected glass compositions. In general the zero-porosity polycrystalline body that results is more resistant to chemical attack and more refractory than traditional glasses. The exact properties of a particular glass ceramic are dependent upon the type and amount of crystalline phases precipitated, as well as the volume and composition of any residual glass matrix. As a coating, the principal advantage of glass ceramics is that they may be applied to the alloy substrate by normal glass enameling techniques, then subsequently converted to a glass ceramic by controlled-heat treatment. In certain applications, these heat treatments have been designed to be compatible with superalloy strengthening processes.

Corrosion of metals by rubidium or other alkali metals occurs after the penetration of a surface oxide layer. Generally, the alkali metal does not wet an oxide; therefore it must penetrate cracks in the coating. This suggests that an appropriate way to protect the metal vessel would be to apply a pore-free, relatively thick, stable oxide coating. Most of the oxides used in preparation of silicate glasses are stable with regard to attack by rubidium; therefore, a coating of this sort would resist attack of rubidium metal and protect the underlying metal used for the containment vessel. Alternatively, the coating itself may act as a getter for rubidium with some glass constituents reacting with rubidium metal to form a lower-valence oxide and rubidium oxide. In this case, rubidium could be removed as a potential corrosive medium for attacking vessel walls by converting it chemically to the more stable oxide. Under certain storage conditions, the outside of the canister may be in contact with water, brine, clay, or shale at temperatures up to 200°C. Glass coatings may provide enhanced protection for relatively low-cost steel, as previously described for applications in the chemical-process industries.

Program Definition

Corrosion-resistant glass and glass-ceramic compositions which are compatible with steel alloys acceptable for use as vessels will be developed and evaluated. Glass-ceramic coatings based on barium aluminosilicate and magnesium aluminosilicate compositions currently being used to coat steel alloys will be evaluated along with commercial glass coatings currently used in the chemical industry. Test results can be used immediately for material development.

The coatings will be applied to coupons of candidate alloys and exposed to brine, hot water, and various aqueous mixtures of clay and ground shale. Sample surfaces will be characterized before and after isothermal exposures using optical and electron microscopy. Corrosion cells will be operated at atmospheric pressure in the 20° - 100°C range. Coating compositions which show promise will be subjected to long-term tests (> 6 months). Materials of interest for coating inside walls of the container will be evaluated with respect to resistance to rubidium attack. As Kr-85 decays to metallic rubidium, contact with vessel walls is inevitable. Rubidium is very highly reactive and is known to displace silicon from glass at temperatures above 300°C. The extent of reaction with multicomponent glasses must be evaluated in the temperature ranges experienced during storage. The compatibility of rubidium with candidate coatings will be evaluated by immersing samples of candidate coating materials in liquid rubidium for various times and temperatures. The extent of corrosion will be evaluated by measurement of weight loss/gain and microscopic examination of the reaction surface. The effects of the solution of krypton in glass coatings will also be examined. Krypton has a low but finite solubility in glass and the effects of radioactive decay are unknown. Atomistically dissolved Kr-85, which ultimately decays to rubidium, could behave quite differently in the coating than liquid rubidium in contact with the surface. Facilities are available for ion-implanting known quantities of krypton within the glass.

Because the technology for fabrication of glass-lined vessels is established, relatively few problems are anticipated in coating major surfaces of the containment vessels with these materials

on a commercial scale. Coating of closure and seal areas may be difficult and could require the use of a second glass composition. Experiments appropriate to this phase of the program will be conducted once potential closure techniques are established. Sandia will collaborate with appropriate industries to establish techniques for processing glass-ceramic coatings if these prove to be particularly resistant to attack by corrosive media.

An analysis will be made of the time-varying spatial temperature differences between coatings and the container walls so that thermal expansions of the composite can be matched. Thermal stress calculations will be made to locate potential cracking problems. Results of the stress analysis will be checked during the demonstration test of the selected container design.

There are similar corrosion problems in all nuclear waste disposal programs. Much of the research for Sandia's seabed, bedded salt (WIPP), and shale programs for the disposal of solid nuclear waste is applicable to the K-85 program. Coordination of all these programs reduces equipment requirements and provides a larger data base with a smaller total investment of time and money.

The stress intensity at the crack tip, K , is given by

$$K = \frac{PY\sqrt{A}}{BW}$$

where $Y = fct(A/W)$ = compliance factor

Figure E-1. Compact Tension Specimen Typical of the Type Used for Fracture Mechanical Testing

$K_{TH} = K_{sec}$: Threshold stress intensity below which crack growth will not occur.

t_{max} : Maximum test time at which no failures occur - typical values are 10^3 to 10^4 hr but may be less than 100 hr, depending upon the specific crack growth mechanisms and their associated kinetic processes

Figure E-2. Initial Stress Intensity vs Time to Failure for Constant Load Testing of Fatigue Precracked Specimens

A_i : Initial crack length

A_f : Final crack length at which subcritical crack growth ceases

T_i : Initiation time

T_f : Time to reach crack arrest

From the above data and specimen geometry one may calculate the stress intensity at which crack growth ceased - the threshold stress intensity.

Figure E-3. Crack Length vs Time for Constant Displacement Testing of Fatigue Precracked Specimens

- P_p = Proof Pressure; identifies maximum flaw size (A_p) which may exist in the vessel without exceeding P_p fracture toughness, K_c .
- P_o = Operating Pressure; at this pressure all flaws equal to or greater than A_o will grow to failure.
- P_1 = Operating Pressure Lower than P_o ; since the proof test has shown that no flaws exist in the bottle greater than A_p and the critical flaw size at P_1 is A_1 , we know subcritical crack growth will not occur - since $A_1 > A_p$.

Our test program will define the above parameters to preclude environmentally assisted crack growth. It should be noted that the above technique may not be applicable for some materials (depending on toughness and environmental susceptibility). However, the use of the threshold stress intensity concept (i. e., that stress intensity below which subcritical crack growth will not occur) remains valid.

Figure E-4. Schematic Depicting Relationship Between Pressure, Flaw Size, Fracture Toughness, and Threshold Stress Intensity for Pressure Vessels

Specimen A: In this orientation the fatigue precrack is parallel to the weld centerline. The offset distance is varied such that all the regions in the weldment (e.g., fusion zone, heat affected zone, etc) are fully evaluated.

Specimen B: This orientation results in a crack that initially propagates at an angle ($\sim 15^\circ$) to the weldment. If the crack "turns" at a particular region in the weld, this is indicative of decreased toughness and/or increased environmental susceptibility for that region.

In the above manner various weldments may be quantitatively evaluated and correctly ranked with regard to toughness and environmentally assisted cracking.

Figure E-5. Schematic Showing Typical Specimen Orientations for Toughness and Environmental Susceptibility Evaluation of Weldments

APPENDIX F

Surveillance Testing of Kr-85 Storage Bottles (Robert D. Klett, 5336)

On November 29, 1977 the Sandia Livermore surveillance facilities were visited to determine their suitability and availability for testing Kr-85 bottles and to obtain information useful for the design of a separate Kr-85 surveillance facility. Contacts were Bill Hoover, Jack O'Connor, and Anton West (8314) and Larry Brown (8142).

SLL uses four types of surveillance testing:

1. Steady pressure for nonradioactive bottles. Each bottle has its own secondary container and pressure monitor connected to an alarm panel.
2. Cyclic pressure for nonradioactive bottles. Pressure in the secondary container is monitored and the valve to that bottle is closed automatically if a leak is detected.
3. Steady pressure for small radioactive bottles. Bottles and secondary containers are kept in glove boxes. Monitoring consists of pressures and chemical detectors.
4. Short-duration tests in autoclaves. Tests are conducted in blast cells. Pressure is monitored.

The first three types depend on the secondary container to isolate the bottle contents in case of a leak and to stop fragments in case of a rupture. These facilities can be placed in any type of structure, can be moved from one location to another, and can be enlarged by adding modules. Periodically, bottles are removed from surveillance and a chemical analysis is made of the gas, the bottle is burst-tested, and material from the bottle is subjected to mechanical tests and analyzed with a scanning electron microscope.

Unfortunately, none of the SLL facilities can handle full-size Kr-85 bottles; they are not shielded sufficiently and cannot dissipate the heat generated by Kr-85. If Kr-85 bottles are surveillance tested, a separate facility for that purpose will probably have to be built. A modular design similar to the first SLL facility listed is recommended. Secondary containers with O-ring seals and a bolt ring can take up to 30,000 psi. Containers of this type should cost about \$350-\$400 each for scaled bottles (15 cm x 6 cm dia). The secondary containers are proof-tested with HE equivalents of ruptured bottles. A monitoring system similar to that used at SLL would cost about \$450 for each bottle including plumbing. It consists of a pressure gauge connected to the secondary container with high- and low-alarm settings. The lower alarm level indicates a leak and the high level indicates a ruptured bottle. In addition to the alarm, there is a light panel indicating normal

(green), leaking (yellow), or ruptured (red) conditions for each bottle. Location of the facility should be at a processing site where leaking bottles and bottles to be burst-tested can be emptied. The facility could be housed in a bunker to provide radiation shielding. Concrete compartments for each bottle should provide adequate internal shielding. The desired temperature profiles for bottles containing Kr-85 could be maintained using insulation or extended surfaces on the containers. Bottles containing only rubidium and RbOH would have to be heated.

APPENDIX G

Canister Exterior Corrosion Tests (Jeffrey W. Braithwaite, 5831)

In-Situ Experiments

As part of the corrosion program for identifying suitable materials for use in geologic environments, the corrosivity of several metal coupons was tested in-situ in a large salt-block heater experiment, in electrically heated field tests in shale at Oak Ridge, and in argillite at the Nevada Test Site.

The largest problem encountered so far in the material selection process is a definition of the actual environment to which the canister/instrumentation materials will be exposed. The complicating factor in salt is the migration of brine inclusions. Will the environment be hot, dry salt; hot, saturated brine; or some combination of the two? The answer is important because the corrosion rate of a given metal depends almost entirely on its surroundings. It was with this fundamental environmental definition question in mind that metal coupons were placed in the salt block.

Coupons were made from 14 candidate alloys, picked to represent a wide range of available metals. Included were (1) titanium, (2) titanium-6V-4Al, (3) Hastelloy C-276, (4) Inconel-600, (5) zirconium, (6) Zircaloy-2, (7) Zircaloy-4, (8) Carpenter 20-Cb-3 stainless steel, (9) Armco 22-13-5 stainless steel (10) 304 stainless steel, (11) 17-4 PH stainless steel, (12) 70-30 brass, (13) aluminum 6061, and (14) 1018 carbon steel. These alloys are listed in order of expected increasing corrosivity, according to a brine corrosion literature survey. Each coupon was machined to 2.54 cm per side by 0.64 cm thick. Because it seemed plausible that brine inclusions would not move far or fast enough to wet each coupon for the total length of the experiment, one surface of each coupon was mirror polished and photographed. This would at least allow a qualitative visual determination of any corrosion results. Finally, each coupon was weighed to the nearest 0.1 mg.

A 3.8-cm-diam hole was bored parallel to the heater hole and 7.5 cm from the outside edge of the heater itself. The hole bottom was at the vertical midplane of the salt block. The coupons were placed flat in the hole with approximately 1.5 cm of crushed salt between each two to prevent any corrosion-accelerating galvanic effects. Also, the coupons were ordered such that the most reactive were on the bottom. The measured thermal history at the bottom of the hole was as follows: 0 day: 25°C; 10 day: 61°C; 20 day: 78°C; 30 day: 103°C; 40 day: 138°C; 60 day: 137°C; 80 day: 138°C; 90 day: 120°C; and termination in 99 days: 49°C. A thermal gradient existed going up the hole, with the temperature at the top sample lagging the bottom by 30°-50°C.

Laboratory examination of all coupons confirmed expectations nothing corroded. All the coupons and the plain carbon-steel-shelled heater appeared exactly as they did 4 months earlier, except for the 70-30 brass, 1018 steel, and 304 stainless-steel coupons. Each of these had a thin, nonadherent, unweighable oxide film on the polished surface, indicating the presence of moisture at one time or another, but the absence of pitting on the SS 304 and aluminum samples showed that the moisture had only been temporary, most likely occurring before startup. The lack of significant corrosion on the heater shell was not unexpected. The temperature of the heater was above the boiling point of the brine; therefore, only if the overlying crushed salt could seal steam in at 576 psi (steam pressure at 250°C) could any rust-causing moisture condense. Steel must reach a temperature of over 500°C before air or steam surface oxidation becomes significant.

It is reasonable to question the applicability of these results to an actual salt environment. The unobservable rates of corrosion may be due to (1) not enough time being allowed for significant brine migration to occur, and/or (2) system perturbation caused by a bored hole backfilled with crushed salt. On the positive side, this experiment may have demonstrated that indeed the corrosion problem in bedded salt will not be very significant. Further in-situ experimentation is needed.

Candidate canister materials selected for in-situ corrosion testing were: 1018 plain carbon steel, 4130 high-strength low-alloy steel, Carpenter 20 stainless steel, Armco 22-13-5 stainless steel, Inconel 600, Hastelloy C-276, MP 35 N (a multiphase alloy), Zircaloy-2, titanium, and aluminum 2024. These metals were flush-mounted in special disks of 304 stainless steel which were then welded to the bottom of the heaters being used in field experiments near Oak Ridge and at NTS. Each coupon was electrically insulated to prevent galvanic corrosion.

Laboratory Experiments

A compatibility study was undertaken to aid in selection of canister and instrumentation material for nuclear waste disposal in bedded salt and the sea bed. A major requirement of the WIPP program is to guarantee retrievability for at least 30 yr. This requirement, coupled with the likely possibility of a highly corrosive environment, makes some form of compatibility study mandatory. The long-term stability of the canister becomes even more important if the canister is to be a major barrier or if fuel-rod assemblies are to be buried instead of reprocessed waste. The containment requirement for Kr-85 would be for as long as 50 to 100 yr.

The goal of this compatibility study is to select materials which (1) are compatible with the hot bedded-salt environment for desired lifetimes, (2) have the mechanical strength and thermal conductivity properties desirable for bedded-salt disposal, and (3) have minimal cost and minimal consumption of critically limited materials. A literature survey was made to find what had been

done and to avoid duplication of effort. It was evident that an extensive amount of brine-related corrosion studies had been done, but none had been conducted under conditions that might exist in the H.W. environment. Corrosion studies in most low-temperature, low-pressure brine environments have been well documented and need not be repeated until more stringent requirements and a better definition of the waste environment are decided upon. For these reasons a high-pressure, high-temperature (autoclave) laboratory was set up in which a deep burial environment can be simulated.

In conjunction with the in-situ experiments, laboratory experiments were completed to satisfy the following objectives:

1. To determine a reasonable test period for the experimental measurement of corrosion results.
2. To provide preliminary data for scoping and instrumentation requirements.

Experiments included;

Test A: Coupons of aluminum, brass, stainless steels, steel, Inconel 600, Hastelloy C-276, titanium, and zircaloy-2 were tested in salt chips (from 1-m salt-block machining) at 90°C and 1-atm pressure (67-day duration).

Test B: Coupons of SS 304, Inconel 600, aluminum 6061, and 1018 carbon steel were tested at 25°C and 1-atm pressure in solution "B" (67-day duration). Solution "B" is a saturated brine of salt from the WIPP site.

Test C: Coupons of SS 304, Inconel 600, and 1018 carbon steel were tested at 90°C and 1-atm pressure in solution "B" (67-day duration).

Results were:

Test A: All samples tested in the hot salt showed nil or negligible corrosion rates. In fact, polished surfaces were not even dulled. Some samples were coated with salt (probably from water initially present) but did not corrode.

Tests B and C: Measured weight losses resulted in the following:

Material	mm Penetration/Yr		Comments
	25°C	90°C	
Al-6061	2.1×10^{-3}	-	Pitted severely
304 SS	9.2×10^{-4}	5×10^{-3}	-
Inconel 600	1×10^{-3}	5×10^{-3}	-
1018 Steel	5.3×10^{-3}	Nil	-

The corrosion rates calculated and shown above are very low and are therefore extremely hard to measure accurately. This measurement problem can be solved by extending the time of testing so that a more measurable weight loss can be achieved, or by measuring initial rates electrochemically. For reference, a material is said to have excellent corrosion resistance if its overall corrosion or penetration rate is less than 5×10^{-2} mm/yr. Additionally, the seemingly anomalous results reported in the above table for 1018 steel corrosion are not uncommon. The lower rate at higher temperature is due to a significant reduction in the concentration of dissolved oxygen (the oxidant in this situation) as the temperature approaches 90°C.

Some preliminary data have been collected using the newly completed autoclave facility to simulate HLW environments. Investigated were the effects of temperature and brine composition on corrosion rate. Studies are ongoing in which a more expanded temperature range is being explored, along with the effects of dissolved oxygen concentration and hydrostatic pressure.

Electrochemical techniques are very efficient for determining corrosion rates. A charge-transfer rate-controlled process (such as the corrosion of steel in deaerated solutions) can be mathematically described by the Butler-Volmer equation:

$$i = i_o \left[e^{\alpha F(E-E_o)/RT} - e^{-(1-\alpha) F(E-E_o)/RT} \right]$$

where

- i = measured current density
- i_o = exchange or corrosion current density
- α = transmission coefficient
- E = applied potential
- E_o = rest potential

This equation can be linearized at low E values and the corrosion current calculated from the slope of i versus E plots. Or, i_o can be calculated from the intercept of a $\ln(i)$ versus E plot at high E values. Normally data (i vs E) are collected by sweeping the voltage and measuring the current density in an electrochemical cell. These data are plotted and the result is referred to as a polarization curve. Rates calculated from polarization measurements have the disadvantage of being only initial rate determinations (typically, corrosion rates decrease with time) and the corrosion rate must be controlled by a charge transfer process for the measurement to be meaningful.

Initial corrosion rates calculated from electrochemical data are shown in Table G-1 for a plain carbon steel corroding in Brine "B".

TABLE G-I

Effect of Temperature on the Initial Corrosion Rate
of 1018 Steel in Brine "B"

<u>Temperature (°C)</u>	<u>Rate (mm/yr)</u>
200	2.9
81	0.5
24	0.1

If the cathodic reduction of H^+ controls the rate, then from the Butler-Volmer equation derivation,

$$i_o = zFa_{H^+} k_c^o e^{-(Q+(1-\alpha)FE_o)/RT}$$

where

z = number of electrons transferred in the reaction

F = Faraday constant

a_{H^+} = hydrogen ion activity

k_c^o = chemical reaction rate constant

Q = Arrhenius activation energy

A normal Arrhenius plot ($\ln(\text{rate})$ vs $1/T$) yields an apparent activation energy of 5.1 kcal/mole, which is actually a combination of the chemical activation energy and a bias due to the electrical potential difference on the system, E_o .

The temperature effect on quenched brine pH is shown in Table G-II. The decrease in pH at higher temperature is probably due to the hydrolysis of magnesium chloride at higher temperatures. This reaction is:

The K_{sp} for $Mg(OH)_2$ decreases with increasing temperature. This drop in pH is important because cathodic reactions often control the corrosion rate on steels and in deaerated solutions the cathodic reaction is the production of H_2 from H^+ or H_3O^+ . The quenched pH of seawater is also shown. The pH decrease of seawater is much greater than brine because of the higher Mg^{+2} concentration in seawater (1.2 g/L vs 10 ppm).

TABLE G-II
Temperature Effect on Quenched Solution pH

<u>Heated Temperature (°C)</u>	<u>Seawater pH Measured at 25°C</u>	<u>Brine "B" pH Measured at 25°C</u>
25	8.1	7.2
100	8.6	7.6
200	5.5	6.6
270	3.3	6.3

Corrosion rates at 200°C, determined by weight loss after 28 days for five alloys, are shown in Table G-III. It is worth noting that the corrosion rates are not particularly high. For example, if uniform penetration of the 20-Cb-3 alloy occurred for 1000 yr at the brine rate shown (not a very valid extrapolation at this point, however) the total reduction in thickness would be 20 mm or about 4/5 in. Also, a comparison of the rates for 1018 steel, shown in Tables G-I and G-III, reveals how significantly the uniform corrosion rate can decrease with time.

TABLE G-III
Typical Corrosion Rates at 200°C in Deaerated Brine "B"
and Molal Salt (P = 1000 psi)

<u>Alloy</u>	<u>Brine "B" (mm/yr)</u>	<u>Crushed Salt +100% RH (mm/yr)</u>
1018 steel	0.080	0.152
3046 stainless steel	0.005	0.004
20-Cb-3 stainless steel	0.020	0.004
Titanium (Ticode 12)	0.018	0.002
Inconel 600	0.006	0.004

APPENDIX II

Glass-Ceramic Coatings for Kr-85 Canisters (Helen K. Baca, 5845)

A program to evaluate the use of glass or glass-ceramic coatings as corrosion-resistant linings for Kr-85 pressure vessels has been defined. The background for the use of these coatings, and the program definition, are given in Reference II-1. Equipment necessary for applying coatings of candidate compositions to metal (Reference II-2) has been purchased and is operational. Progress has been made on assembling a system to contain and recycle the liquid rubidium to be used in glass-corrosion experiments. A container that can be backfilled with argon will hold the corrosive liquid and suspended glass samples in a glove box. To avoid possible corrosion of the samples or the container by contaminants in the rubidium, a purifying system is necessary. The formation of rubidium oxides will be prevented by removing oxygen with a zirconium getter. A 24-hr heating cycle is necessary to remove traces of oxygen. After removal of oxygen, the liquid rubidium will be passed through a distillation system before being reused for corrosion experiments. Recycling the rubidium allows a large ratio of corrosive liquid volume to glass surface area to be maintained and should improve the accuracy of results.

Construction of a system to allow immersion of glass and glass ceramics in liquid rubidium at elevated temperatures has been completed. Samples of several commercial glasses are presently being exposed to rubidium. Corrosion of these glasses will be evaluated by optical and electron microscopy, microprobe analysis, and weight-change measurements. Results from these tests will provide information for the development of glass or glass-ceramic compositions suitable for coatings. One candidate compositions have been identified, coatings of glass ceramics on metal coupons will be prepared for further study.

References

- II-1. Appendix E of this report.
- II-2. Appendix I of this report.

APPENDIX I

Laboratory Upgrading
(R. J. Eagon, 583)
R. H. Nilson, 1262
J. W. Munford, 5832)

Heat Transfer

Laboratory equipment has been purchased to facilitate bench-scale experiments in the areas of heat transfer and fluid flow. An oscilloscope has been added to an existing laser anemometer which will be used to study natural convection in enclosures. Laboratory space and basic hardware have been acquired for use in heat-transfer experiments and bench-scale simulation of Kr-85 disposal. This equipment may also be useful in future investigations of the flow of Kr-85 through porous earth following a hypothetical loss of container integrity.

Physical Metallurgy

The major emphasis in laboratory improvements was aimed at material evaluation in aggressive reaction environments. With these objectives, modifications of existing equipment and new equipment consisted of:

1. The Mechanical Test System (MTS) equipment used for specimen testing was expanded (a new frame was added) to increase test capabilities. The MTS is a closed-loop servo-hydraulic test machine which greatly facilitates the testing of fracture-mechanics type specimens.
2. Additional controllers, gages, and sensors were added to allow more quantitative control and monitoring of testing environments.
3. Equipment was added to improve data collection and reduction techniques.

This equipment was designed and modified to facilitate the testing program outlined in Appendix E. It is suitable for a wide variety of material evaluation programs and will be available to support the Kr-85 program.

Ceramics

The following equipment additions or modifications in the ceramics laboratory were for evaluating glass-ceramic coatings for Kr-85 storage containers.

DSC Cell

The DSC cell is an attachment for a DuPont Thermal Analyzer that is used to determine transitions in glass-ceramic systems. It will be used to study the annealing point and crystallization of glass-ceramic coatings which we will be developing for use on the inside and outside of krypton containment vessels.

Enamel-Spraying System

This system uses a unique vapor carrier to suspend a particulate glass or glass ceramic for transport onto a metal surface. The advantage of this device is that pore-free coatings can be sprayed on surfaces with irregular shapes, such as a canister or parts of valves that might be used in a canister. Initially, we will use it for coating coupons to evaluate the corrosion resistance of various glass ceramics to rubidium as well as to brine which may attack the outside of the canister.

Furnace, Controller, and Thermocouple Indicator

These devices will be used to construct a furnace appropriate for fusing enamel coatings onto metal coupons and containers. The furnace is a unique design made by Research Inc. that uses focused-infrared energy to heat the enamel. This design was chosen because it is potentially applicable to scale-up of large containers or, alternatively, it can be made into a device which could be used to fuse a coating on the inside of a container by focusing the infrared energy in a desired area.

Hardness Tester

Previous studies have suggested that the strength, abrasion resistance, and corrosion resistance of glass ceramics and glasses are a function of composition. This device will be used to study variations in composition on properties germane to the use of glasses or glass ceramics as coatings for the krypton container.

APPENDIX J

Program Plan for Kr-85 Transport in Geologic Media (James L. Krumhals, 5337)

Introduction

A number of options have been suggested for the disposal of gaseous radioactive isotopes. Although present programmatic emphasis is directed toward evaluation of the SURF site, similar sites involving the shallow burial of Kr-85 may also eventually be constructed at Savannah River and at Idaho Falls. It has also been suggested that consideration be given to storage in an HLW repository at considerable depth, or perhaps to direct injection into a porous sandstone lens encased in a relatively impermeable shale formation. The purpose of this memo is to summarize a methodology that will be applicable to assessing the rate of krypton migration from a shallow repository. It should be noted that the isolation time for Kr-85 is about 100 yr and that it is likely the galleries of a deep repository will remain open for this period. Since there would be but a few meters of rock separating a krypton canister from an opened gallery, it follows that an understanding of krypton migration at low pressures will be useful in this context. Radionuclide migration studies in ocean sediments are underway that extend into the hydrothermal regime. These studies would be applicable to seabed disposal of Kr-85.

Summary of Existing Data

The major factor to be considered in analyzing the consequences of a slow leak is that krypton is an inert gas and, as such, its migration will not be retarded by the formation of immobile compounds. It may, however, be sorbed to a limited degree. Fanale and Cannon^{J-1} noted that the natural krypton content of shales may be explained in terms of sorption characteristics measured on two shale samples in pure krypton at 0°C and by consideration of the partial pressure of krypton in the earth's atmosphere. From their data one may infer that for fine-grained materials, about $0.1 \text{ cm}^3 \text{ STP/g}$ of krypton would be sorbed at a partial krypton pressure of 0.1 atm. It also appears that over the pressure range 10^{-5} to 10^{-1} atm, a tenfold decrease in krypton partial pressure would bring about a tenfold decrease in the amount of krypton sorbed per gram of soil. Their measurements further indicate that the amount of sorbed krypton does not appear significantly affected by the nature of the fine-grained material. That is, at 0° and 25°C an ashfall tuff ($52 \text{ m}^2/\text{g}$) and a shale ($10 \text{ m}^2/\text{g}$) sorb almost identical amounts of krypton, while at -77°C samples of Wyoming Montmorillonite ($36 \text{ m}^2/\text{g}$), 60-mesh crushed limonite ($16 \text{ m}^2/\text{g}$), and 60-mesh crushed basalt ($4 \text{ m}^2/\text{g}$) behaved almost identically. J-1, J-2

From this data a crude model may be constructed to assess the ability of a soil profile to provide a barrier against krypton migration. Assume the soil to have a density of 2.0 g/cm^3 and a porosity of 15%. The depth to the leaky canister is taken as 6.1 m and the partial pressure of krypton in a sphere of this radius is assumed to be 0.1 atm. It follows that within this sphere about 8 kg of krypton is sorbed onto the soil and another 0.5 kg is retained in the soil pores. In other words, the soil could retain approximately 2.27 m^3 (std) of krypton gas. One may also inquire as to the consequences of flooding the pore space in the soil. Henry's Law constants for krypton solubility in both fresh and seawater are tabulated^{J-3} and indicate that only about 1/20th of the krypton initially in the pores could be retained in solution. The great weakness of this model is, of course, the initial assumption that krypton sorption is unaffected by various other gases in the atmosphere. Indeed, the data of Phinney^{J-4} suggests that the values cited by Fanale and Cannon^{J-2} lie in the upper range of krypton contents in shale, and that values 1/50th as great may be appropriate in other instances. It is this matter that needs particular attention in evaluation of any particular site.

General Program Plan

To do a credible site evaluation, the following activities should be carried out:

Site Characterization

1. General geologic and hydrologic framework, particularly as it pertains to ground water recharge and migration, and the vertical and lateral extent of horizons in the soil profile, if any.
2. Textural properties of the soil: density, porosity, permeability, moisture content, specific surface area, and particle size determination.
3. Mineralogy and chemistry of each unit noted; proportions of the major minerals, major element content, nature of amorphous materials, presence of organic materials, and the composition of groundwater or the nature of materials easily leached from the soil by rainwater in the event that an arid region is under consideration.

Response to Heat and Radiation

1. Mineralogic changes.
2. Composition of evolved gases.
3. Textural changes, particularly those that affect surfaces of individual mineral grains or amorphous materials.

³ Activities would be carried out by research or test groups at Sandia or the University of New Mexico.

4. Radiolysis of groundwater, or intermittent flood water, particularly the rate at which any H_2 generated recombines in the soil. (If krypton escapes, the rubidium generated will recombine almost instantaneously to form H_2 gas as well.)
5. Amount of energy stored as radiation damage in soil minerals, if any.
6. Effects of heat, radiation, and the geologic environment on cement or other construction materials used in the immediate proximity to the canister.

Krypton Migration Studies

1. Modeling, initially of a slow leak (i.e., point source term) in which the following are varied parametrically:
 - a. Leak rate
 - b. Depth of burial
 - c. Soil permeability
 - d. Soil porosity
 - e. Krypton sorption characteristics

The half-life of Kr-85 is to be incorporated into the model; the desired output is the amount of krypton arriving at the surface as a function of time.

A second generation of models to incorporate the additional site-specific variables of:

- a. A soil with a stratified permeability
 - b. Displacement or transport due to groundwater movement
 - c. A sorption coefficient which varies with both distance and depth; i.e., in response to both temperature and stratigraphy
 - d. Transport due to thermally induced fluid (air, water) motion
 - e. Leaks in several canisters, or multiple leaks in a single canister
 - f. Kinetic effects of krypton sorption
 - g. Pumping effects arising from changes in atmospheric pressure.
2. Sorption parameter determination, as a function of temperature, soil type, and radiation dose. Determinations need to be made at realistic partial pressures of the common atmospheric gases, associated reprocessing gases (Xenon in particular), and heat- or radiation-induced gases from the soil. The consequences of saturating a previously dry soil need to be assessed, and its needs to be determined whether sorption in a radiation field is significantly different from that observed without the continuous generation of charge imbalances on surfaces of silicate mineral grains. The kinetics of the sorption processes require attention. The rubidium arising from krypton decay may also have an effect on krypton sorption as it should have a rather pronounced effect on the surface chemistry of the soil materials. Finally, consideration should be given to whether a particular mineral is responsible for the most of the sorption.

Specific Activities for FY78

Even with a substantial amount of help from other groups at Sandia, the entire program just presented could not be initiated in FY78. Furthermore, from a system standpoint, the most significant data that could be generated in the available time would be "ballpark" figures on the sorption characteristics of krypton in Hanford soil. A number of approaches are listed in order of increasing complexity.

1. Preparation of Kr-air and Kr-Xe-air mixtures in which gas ratios are known very exactly. If these mixtures are mixed with soil in a high soil:air ratio, it may be possible to gauge the amount of sorbed krypton from the amount of krypton missing from gas samples. A second nonsorbed gas, such as helium, could be added to account for the air in the pores of the soil. As an alternate, before the spiked air is introduced, the sample could be momentarily pumped to a low enough pressure that effects of dilution would be less than expected from krypton sorption.
2. Modification of a surface area analyzer to operate at room temperature, or higher, and at pressures of almost 1.0 atm.
3. *Preform column experiments using the amount of krypton traversed through a long filled pipe and again compared to the amount of a nonsorbed gas such as helium.* Initially a Kr-85 tracer could be injected and then swept through the pipes with nonradioactive krypton. Distribution of the tracer might give an idea as to the sorptive characteristics of the sediments.

Each proposed method has advantages and disadvantages. The first procedure could use a pressure vessel already under development for use in the GIFF facility, and would allow for making measurements over a large temperature range. The second method has the advantage of measuring pressure decreases as they happen; kinetic studies would be possible and the process of withdrawing a sample for analysis on a second instrument would be eliminated. It should also be noted that we already have surface area set up using krypton; therefore conventional low-temperature, low-pressure krypton sorption measurements are available. The third alternative has the advantage that a transport phenomenon is being measured directly.

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APPENDIX K

Geologic Flow Testing (R. D. Klett, 4536)

Nomenclature

f	= Fanning friction factor
g	= Gravitational constant
K	= Hydraulic conductivity
L	= Length
N	= Frictional resistance, $\frac{fL}{R_H}$
P	= Pressure
Q	= Volumetric flow rate
q	= Volumetric flow rate/unit hole length
r_H	= Hydraulic radius
r	= Radius
S	= Half the distance between holes
t	= Time
V	= Velocity
ϵ	= Porosity
μ	= Viscosity
ρ	= Mass density
τ	= Transmissivity
ϕ	= Velocity potential

Introduction

Safety assessments of geological Kr-85 disposal concepts and disposal sites will require an analysis of Kr-85 transport through the geologic media in the event of leaking containers and predictions of ground and/or surface water flow through the repository. Flow tests will be needed to characterize specific sites, to provide input data for modeling, and to evaluate concepts and analytical models. The ability to measure fluid-flow characteristics of geologic formations is either helpful or essential for gas, oil, and water extraction, geothermal energy utilization, in-situ coal gasification, and in-situ oil shale retorting, as well as nuclear waste isolation. Flow characteristics include formation transmissivity, locations and effective hydraulic radii of cracks and location, thickness, porosity, and effective permeability of porous layers. These properties

cannot be measured in the laboratory because (1) crack patterns are disturbed during sample acquisition; (2) stresses are relieved when the samples are removed from the formation; (3) the formations are not homogeneous; and (4) flow paths in geologic formations are not continuous. Groundwater characteristics that must be measured in-situ include depths and thicknesses of aquifers and the direction and velocity of groundwater movement. A survey of existing geologic flow measuring techniques was made, followed by the design of a new geologic flow test system and the derivation of flow equations for data reduction.

Available Techniques

The following measurement techniques are in common usage and the equipment needed for the tests is available from drilling suppliers.

1. Flow nets are used to map streamlines and equipotential lines for steady groundwater flow in confined or unconfined aquifers. The nets are constructed using potentiometric surfaces generated by measuring the water level in nonproducing wells. As few as three wells are needed to define groundwater flow at a specific location using methods given in References K-1, K-2, and K-3. Direction of flow can be read directly from the flow nets. If permeability is known, the flow rate can be computed; if porosity is also known, velocities can be computed. Aquifers with anisotropic permeability can be handled using the transforms $x' = x/\tau_x$ and $y' = y/\tau_y$.
2. Pump-down and recharge of wells can be used to obtain qualitative information such as communication between wells below given levels, and relative water contents. Some quantitative data on groundwater flow and permeability can be obtained using methods defined in References K-2 and K-3.
3. Permeability of a formation can be measured with gas-flow methods. Some methods locate and define the orientation of fissures in impermeable rock. Gas-flow tests are limited to wells that can be pumped dry. The formation must be blown dry using pressures higher than the test and hydrostatic pressures. Any capillary water remaining in the formation results in erroneously low permeability measurements. In tight formations, a continuity test is advisable before more complex tests are started. One well is pressurized and continuity is measured either with a flow meter at constant inlet pressure or with a pressure gage measuring bleed-down time with no flow. If there is flow out of the input well, communication to other wells is measured with flow meters on the outlet wells. Flow paths from a well to the surface can be found by pumping a gas tracer such as SF_6 into a well, capping all surrounding wells, and traversing the surface with a detector (sniffer).

Of the two gas-flow permeability tests now in use, the radioactive Kr-85 tracer is the quickest and easiest to use, but only qualitative data are generated. The test procedure is as follows. Air flow is established between two wells and a spike or square wave of Kr-85 is introduced at the top of the inlet well. A gamma radiation sensor is placed in the mouth of the outlet well; the times and intensities of the radiation are recorded. A spike input appears as one or more skewed Gaussian curves on the output. Each crack between the wells is represented by a curve on the output. The amplitude of the curve is proportional to the permeability of the fracture. The time for the tracer to travel from the source to the detector is dependent upon both the depth and permeability of the fracture. A square-wave input appears as a curve or sum of curves that are asymptotic to the input level.

The gas-flow logger is the other gas permeability test method. Steady flow is established between two wells and a flow-rate meter is placed in the bottom of the outlet well. The meter is slowly raised and a plot of flow rate versus depth is obtained. Uniform permeability is represented by a steady increase in flow rate. A step increase in the flow rate represents a horizontal crack and a ramp represents a vertical or angled crack. Quantitative permeability data can be obtained using the pressure drop between holes, the rate of flow change with depth, and techniques developed later in this report. Flow rates can be measured with hot-wire anemometers or turbine-type meters. Limitations of the gas-flow logger are that measurements cannot be made in low-permeability formations and the outlet well must have a uniform cross section.

4. Liquid flow tests are conducted using tracers introduced in the groundwater or in water pumped into a well. In addition to being readily detectable, the tracer must be nondegradable, not absorbed by the formation, not masked by existing minerals, not eaten by organisms, and nonpolluting. Therefore, the selection of the tracer is location- and formation-dependent. Common tracers are methanol and ethyl alcohols, acetone, MEK, formic acid, acetic acid, formaldehyde, acetaldehyde, sugar, sodium, potassium, lithium, phenols, iodine, EDTA, and nonbiodegradable detergents. Tracer detection is usually by chemical analysis but in some cases electrical conductivity measurements can be used. Natural groundwater movement is measured by putting a tracer in one well and measuring the time for the tracer to reach wells in the downstream direction. Permeability between holes can be measured by pumping the tracer into one well under pressure and measuring the travel time. Data-reduction techniques are the same as for low-pressure differential gases (incompressible flow). The advantage of liquid flow methods is that the formation does not have to be pumped dry.

5. Geophysical methods can be used to help understand regional hydrology by measuring infiltration capacity, locating water tables, and defining the stratigraphy of the formation. Techniques described in References K-2 and K-4 include magnetic, gravity, seismic, electrical resistivity, spontaneous electrical potential, core logs, caliper logs, radioactive logs, and infiltrometers.

Geologic Fluid-Flow Test System

This test system measures formation-flow properties between two or more drill holes. The down-hole flow system used to obtain quantitative data is shown schematically in Figure K-1. The system can operate with gas- or liquid-carrier fluids and either chemical or radioactive tracers and detectors. Air and water are the most common carrier fluids. Kr-85 is a typical gas radioactive tracer used with Gieger tube, semiconductor PIN, or scintillation detectors. Sulfur hexafluoride is a typical chemical gas tracer used with gas chromatograph detectors. Numerous tracers can be used in liquids. In operation, steady flow is established between wells as indicated by the flow panels which consist of meters to measure volumetric flow rate, temperature, and pressure. Then a short (approximately 1-s), square pulse of tracer is injected in the input well at the same time the detector recorders are started. The injector is electrically controlled and is a positive displacement type. The tracer travels to the output well through various flow paths and tracer intensity is measured as a function of time by the detector string. Typical output well traces are shown in Figure K-2. A pulse detected at a deep location is repeated at each higher detector but at progressively later times and reduced amplitude. Amplitude is reduced because of increased upward velocity as flow is added to the output well. The time of first arrival and the areas under the traces are used in conjunction with the flow rates, temperatures, and pressures measured at the flow panels, and well geometries to compute flow properties between wells.

Figure K-1. Down Hole Tracer System

Figure K-2. Typical Detector Traces and Formation Flow Paths They Represent

Data Reduction

The basic theory for flow between wells was developed in Reference K-5. Using those results, data-reduction techniques were derived to find crack sizes and friction factors in laminar and turbulent flow, and the permeability and porosity of porous media using the time of first arrival of a tracer at the detectors.

The equipotential and stream lines for the two-well test are shown in Figure K-3. From the velocity potential,

$$\phi = \frac{q}{4\pi} \ln \frac{(x-S)^2 + y^2}{(x+S)^2 + y^2} \quad (K-1)$$

the x component of velocity in a porous medium with porosity ϵ is

$$V_x = \frac{q}{2\pi\epsilon} \left[\frac{(x-S)}{(x-S)^2 + y^2} - \frac{(x+S)}{(x+S)^2 + y^2} \right] \quad (K-2)$$

Integrating Eq. 2 along the shortest flow path yields the time of first arrival of a tracer;

$$t_F = \frac{2\pi S^2 \epsilon}{q} \left[\frac{2}{3} - \left(\frac{r}{S} \right)^2 + \frac{1}{3} \left(\frac{r}{S} \right)^3 \right] \quad (K-3)$$

Figure K-3. Equipotential and Streamlines for Flow Between Two Wells

The pressure drop between the two wells through a porous medium is (Reference K-2):

$$\Delta P = \frac{96\mu}{2\pi K} \ln \frac{(2S - r)^2}{r^2} \quad (K-4)$$

Combining Eqs. K-3 and K-4,

$$t_F = \frac{8\mu S^2 \epsilon}{\Delta P K} \left[\frac{2}{3} - \left(\frac{r}{S}\right)^2 + \frac{1}{3} \left(\frac{r}{S}\right)^3 \right] \ln \frac{(2S - r)^2}{r^2} \quad (K-5)$$

for $r \ll S$

$$t_F \approx \frac{4\mu S^2 \epsilon}{3\Delta P K} \ln \frac{2S}{r} \quad (K-6)$$

From Eq. K-3,

$$\epsilon \approx \frac{3t_F q}{4\mu S^2} \quad (K-7)$$

The effective hydraulic conductivity (K) can be computed using Eq. 6 since all other variables are known or can be measured. In the case of laminar flow through a fracture, hydraulic conductivity can be expressed as

$$K_{lam} = \frac{R_H^2 \rho g}{2\mu} \quad (K-8)$$

Substituting Eq. (K-8) into Eq. (K-6) and eliminating ϵ ,

$$t_F \approx \frac{8\mu S^2}{3R_H^2 \Delta P} \ln \frac{2S}{r} \quad (K-9)$$

which allows the hydraulic radius (R_H) of the fracture to be computed.

In the case of turbulent flow through a fracture,

$$K = \frac{2gR_H}{fV} \quad (K-10)$$

Substituting Eq. K-10 into Eq. K-6, eliminating ϵ , and using

$$\bar{V} = \frac{2S}{t_F} \quad (K-11)$$

$$t_F \approx \left[\frac{4S^3 \rho f}{3R_H \Delta T} \ln \frac{2S}{r} \right]^{1/2} \quad (K-12)$$

The friction resistance of the fracture ($N = \frac{fI}{R_H}$) can be computed from Eq. K-12.

All the above equations were developed assuming slug flow. In the real case of boundary-layer flow, there will be a slight spreading of the ideal curve (Figure K-4). There is also a spreading of the curve in a porous region due to different flow times in the vertical pipes.

Figure K-4. Tracer Level in the Sink Well With a Spike Input and Slug Flow

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