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SYMPOSIUM D

SIXTH INTERNATIONAL SYMPOSIUM ON THE SCIENTIFIC BASIS FOR NUCLEAR WASTE MANAGEMENT

Chair

D.G. Brookins
University of New Mexico

November 1-4
Imperial Ballroom

Principal Symposium Support

Department of Energy, Office of Defense Waste
and Byproducts (G. Oertel)
Nuclear Regulatory Commission (K.S. Kim)

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SYMPOSIUM D

SIXTH INTERNATIONAL SYMPOSIUM ON THE
SCIENTIFIC BASIS FOR NUCLEAR WASTE MANAGEMENT

Chair:

D.G. Brookins, University of New Mexico

November 1-4, 1982

Session D1

HIGH LEVEL WASTE FORM PROCESSES AND PROPERTIES

Chair: W.A. Ross, Battelle Pacific Northwest Laboratories

R.D. Walton, U. S. Department of Energy

Monday Morning, November 1

- 8:25 Introductory Remarks, D.G. Brookins
- 8:30 D1.1 *The Scientific Basis for Long Term Prediction of Waste Form Performance Under Repository Conditions, J.E. Mendel, Pacific Northwest Laboratory
- 8:55 D1.2 *SYNROC: Properties, Production Technology and Disposal Strategy, A.E. Ringwood, Australian National University, Australia
- 9:20 D1.3 Determination of Crystal Site Occupancy in Synrock Using Analytical Electron Microscopy, J. Taft, J.C.H. Spence, Arizona State University, and D. Clarke, Rockwell International Science Center.
- 9:40 D1.4 New Single Phase Ceramic Waste Form: [NZP], R. Roy, L. Yang, E.R. Vance, J. Alamo, Pennsylvania State University
- 10:00 Break
- 10:20 D1.5 *Leaching Chemistry of Defense Borosilicate Glass, R.M. Wallace and G.G. Wicks, Savannah River National Laboratory
- 10:45 D1.6 *Preferential Dissolution Phenomena and Their Relation to Microstructural Inhomogeneities in Nuclear Waste Materials, D.R. Clarke, Rockwell International
- 11:10 D1.7 Morphology and Chemical Composition of a Surface Layer Formed Upon Leaching of Nuclear Waste Glass, W. Lutze and G. Malow, Hahn-Meitner Institut, FRG, and T.J. Headley, Sandia National Laboratories
- 11:30 D1.8 Glass Surface Area to Solution Volume Ratio and its Implications to Accelerated Leach Testing, L.R. Pederson, C.Q. Buckwalter, and G.L. McVay, Pacific Northwest Laboratory

Session D2

LEACHING STUDIES

Chair: G.L. McVay, Battelle Pacific Northwest LaboratoryL.A. Boatner, Oak Ridge National Laboratory

Monday Afternoon, November 1

- 1:30 D2.1 *Theory of Leaching of Alkali-Borosilicate Glass, D.E. Clark and L.L. Hench, University of Florida
- 1:55 D2.2 Product Leaching and Solid State Properties of Formula 127 Glass Prepared with Actual Radioactive Zirconia Calcine, B.A. Staples and H.S. Cole, Exxon Nuclear Idaho Company, Inc.

- 2:15 D2.3 Growth of Surface Layer During the Leaching of the Simulated Waste Glass and its Barrier Effects on the Leaching, K. Ishiguro, Power Reactor and Nuclear Fuel Development Corporation, Japan; N. Kawanishi, Toshiba Corporation, Japan; N. Sasaki, H. Nagaki, and M. Yamamoto, Power Reactor and Nuclear Fuel Development Corporation, Japan
- 2:35 D2.4 Effects of Solution Chemistry and Atmosphere on Leaching of Alkaliborosilicate Glass, H-P. Hermansson, H. Christensen, Studsvik Energiteknik, L. Werme, SKBF/KBS, Sweden; D.E. Clark, Univ. of Florida
- 2:55 Break
- 3:15 D2.5 *Basic Leaching Processes in Silicate Glass: The Effects of Solution pH and Ions in Solution on Gel Layers, B.C. Bunker, E.K. Beauchamp, and G.W. Arnold, Sandia National Laboratories
- 3:40 D2.6 The Dominant Mechanisms in Leaching of Composites of Cements with Radioactive Waste, M.W. Barnes and D.M. Roy, The Pennsylvania State University
- 4:00 D2.7 Leach Models for a Commercial Nuclear Waste Glass, W.L. Kuhn, R.D. Peters, Pacific Northwest Laboratory
- 4:20 D2.8 Comparison of the Leachability of Three TRU Cement Waste Forms, J.H. Westsik, Jr., W.A. Ross, F.P. Roberts, C.O. Harvey, Pacific Northwest Laboratory
- 4:40 D2.9 Extended Leach Studies of Actinide Doped SRL 131 Glass, J.K. Bates, D.J. Lam and M.J. Steindler, Argonne National Laboratory

Session D3

CHEMICAL INTERACTIONS IN GEOLOGIC SETTINGS

Chair: D. Isherwood, Lawrence Livermore National LaboratoryH.A. Wollenberg, Lawrence Berkeley Laboratory

Tuesday Morning, November 2

- 8:10 D3.1 *Suitability of Natural Geomedia for Consideration for Radwaste Storage, W.S. Fyfe, University of Western Ontario
- 8:35 D3.2 Evaporite Dissolution Relevant to the WIPP Site, Northern Delaware Basin, Southeastern New Mexico, S.J. Lambert, Sandia National Laboratories
- 8:55 D3.3 Natural Analogues: Alamosa River Monzonite Intrusive into Tuffaceous and Andesitic Rocks, D.G. Brookins, M.S. Abshian, University of New Mexico; L.R. Cohen and A.E. Williams, University of California-Riverside; H.A. Wollenberg and S. Flexser, Lawrence Berkeley Laboratories
- 9:15 D3.4 Geological and Petrological Considerations Relevant to the Disposal of Radioactive Wastes by Hydraulic Fracturing: An Example at the U.S. Department of Energy's Oak Ridge National Laboratory (ORNL), C.S. Haase, Oak Ridge National Laboratory
- 9:35 D3.5 Technetium Behavior in Sulfide and Ferrous Iron Bearing Media, S.Y. Lee and E.A. Bondietti, Oak Ridge National Laboratory

* Invited Talk

* Invited Talk

55 D3.6 Natural Analogues--A Way to Increase Confidence in Predictions
of Long Term Performance of Radioactive Waste Disposal, G.F. Birchard,
U.S. Nuclear Regulatory Commission

15 Break

Session D4
NUCLEAR WASTE MANAGEMENT OVERVIEWS

Chair: C. Sombret, Commissariat à l'Energie Atomique, France
T. Ishihara, Radioactive Waste Management Center, Japan
Tuesday Morning, November 2

35 D4.1 *NWTS Waste Package Design and Materials, Testing Status FY 82,
J.F. Kircher and D.J. Bradley, Office of Nuclear Waste Isolation

00 D4.2 *The French Waste Management Program, J.F. Lefevre, Commissariat
à l'Energie Atomique, France

25 D4.3 *United Kingdom Regulatory Procedures for Radioactive Wastes, F.S.
Feates and H.J. Richardg, Dept. of the Environment, United Kingdom

50 D4.4 *Some Contributions of Basic Inorganic Chemistry to Waste or
Spent Fuel Storage, I. Grenthe, The Royal Institute of Technology,
Sweden

Session D5 (Poster)

GEOLOGIC TOPICS, MODELLING, AND RELATED TOPICS
Chair: W.R. Daniels, Los Alamos National Laboratory
J.D. Tewhey, Jordan Gorrill Associates
Tuesday Afternoon, November 2, 2:00 - 4:00

D5.1 Hydrothermal Interactions of Cement or Mortar with Zeolites or
Montmorillonites, S. Komarneni and D.M. Roy, The Pennsylvania State
University

D5.2 K-Ca and K-Ar Dating of Sylvite From the Salado Formation, New
Mexico and Implications for the Stability of Evaporite Minerals, J.D.
Obradovich, M. Tatsumoto, U.S. Geological Survey, Denver; O.K. Manuel,
University of Missouri-Rolla; H. Mehnert, U.S. Geological Survey; and
M. Domenick and T. Wildman, Colorado School of Mines

D5.3 Radioactive Waste Isolation: Significance of Microcracks,
G. Simmons, and L. Caruso, Massachusetts Institute of Technology

D5.4 Radionuclide Migration into Natural Fracture Surfaces of
Granitic Rock, B. Torstenfeld, T. Eliasson, B. Allard, K. Andersson,
H. Högglund and U. Olofsson, Chalmers University of Technology, Sweden

D5.5 Salt Creep Design Consideration for Underground Nuclear Waste
Storage, W.T. Li and C.L. Wu, Bechtel National

D5.6 Megascopic Controls on Radionuclide Transport in Groundwater
Flow Systems, L. Smith, University of British Columbia, and F. Schwartz,
University of Alberta, Canada

D5.7 Effect of Channelling in a Porous Medium on Radionuclide Mig-
ration, A.K. Ray and S. Nair, University of Kentucky

D5.8 Performance Analysis of a Nuclear Waste Repository in a Basalt
Geology, R.G. Baca, M.S. Bensky and N.W. Kline, Rockwell Hanford Operation

ited Talk

D5.9 Statistical Site Characterization: Lessons for "Regulatory -
Geology", G.B. Baechter, Massachusetts Institute of Technology

D5.10 The Scepter Waste Package Subsystem Computer Model for
Performance Assessment, A. Brecher, A.D. Little, Inc., and J.F.
Pearson, Intera Environmental Consultants, Inc.

D5.11 Long-Term Thermomechanical and Thermohydrological Factors
Controlling the Optimal Design of a Nuclear Waste Repository, J.S.Y.
Wang, D.C. Mangold, and C.F. Tsang, Lawrence Berkeley Laboratory

D5.12 An Analysis of Waste Package Behavior for High-Level Waste,
M.S. Chu, S. Stuckwisch and M.D. Siegel, Sandia National Laboratories,
and J.E. Campbell, INTERA Environmental Consultants, Inc.

D5.13 The Formulation of an Integrated Physicochemical-Hydrologic
Model for Predicting Waste Nuclide Retardation in Geologic Media,
A.B. Muller, Sandia National Laboratories, D. Langmuir, Colorado
School of Mines, and L.E. Duda, Sandia National Laboratories

D5.14 Plutonium Speciation in Selected Ground Waters and Its Implica-
tions for Nuclear Waste Disposal, T.F. Rees, J.M. Cleveland, and K.L.
Nash, U.S. Geological Survey

D5.15 Studies of the Bonding of Cobalt on Feldspar Surfaces Using
X-Ray Photoelectron Spectroscopy [XPS], Scanning Electron Microscopy
[SEM], and Scanning Auger Microscopy, D.L. Perry, L. Tsao and K.A.
Gaugier, Lawrence Berkeley Laboratory

D5.16 Studies on Speciation of Americium, Technetium, and Neptunium
in Simulated Vitrified-Waste Leachates, G. Bidoglio, A. De Piano,
A. Chett, Commission of European Communities, Ispra Establishment

Session D6 (Poster)

LOW LEVEL AND TRANSURANIC WASTES, ENGINEERING
PROCESSES, AND RELATED TOPICS

Chair: C.W. Brown, Rockwell International - Rocky Flats
G.L. DePooter, Los Alamos National Laboratory
Tuesday Afternoon, November 2, 4:00 - 6:00 PM

D6.1 Releasing Behavior of Tritium From Neutron Irradiated Boro-
silicate Glass, I. Yamamoto, K. Kitagawa, Kobe Steel, Japan; H.
Atsumi, S. Yamanaka and M. Miyske, Osaka University, Japan

D6.2 Laboratory Investigations of Ground Water Interactions with
Buried Radioactive Sodium Wastes and Attendant Release and Migration
of Tritium in Adjacent Soil, G. Subbarawan, O.P. Steele, III, Rockwell
International; L.C. Witbeck, C.S. Abrams, Argonne National Laboratory

D6.3 The Functional Dependence of Leaching on the Surface Area to
Solution Volume Ratio, A.J. Machfeis, and C. Pascatore, University of
Illinois.

D6.4 Water Chemistry at Commercially Operated Low-Level Radioactive
Waste Disposal Sites, R.F. Pietrzak, R. Dayal, Brookhaven National
Laboratory, K.S. Czyscinski, Office of Nuclear Waste Isolation, and
A.J. Weiss, Brookhaven National Laboratory

D6.5 Leach Tests of Simulated Low-Level Transuranic Waste Forms Containing Transuranic Elements, J.M. Welch, C.W. Sill, and J.E. Flinn, EG&G Idaho, Inc.

D6.6 Formulation and Durability of Tailored Cementitious Hosts Applied to TRU Waste Generated at the Rocky Flats Plant, J.H. Kessler, G.C. Rogers, L.R. Dole and M.T. Morgan, Oak Ridge National Laboratory

D6.7 Low Level Radwaste Encapsulation in Portland Cement, M.C. Skriba and E.E. Smeltzer, Westinghouse Research and Development Center, and G.A. Cefola, Westinghouse Nuclear Energy Systems Division

D6.8 Removal of Actinides from Waste Solution by Ferrite Treatment, T.E. Boyd and R.L. Kochen, Rockwell International

D6.9 Theoretical Predictions for Continuous Slurry Feeding of a Glass Melter, K.R. Rouff, E.I. duPont de Nemours & Co.

D6.10 SYNROC Production Using a Fluid-Bed Calciner, F.J. Ackerman, J.Z. Grens, P. Peters and J.H. Campbell, Lawrence Livermore National Laboratory

D6.11 Efficient Particulate Scrubber for Radioactive Melter Off-Gas Applications, G.T. Wright, E.I. duPont de Nemours & Co.

D6.12 Electric Arc Furnace Melting of Simulated Transuranic Wastes, R.H. Nafziger and L.L. Oden, Albany Research Center

D6.13 Ra-226 Radioassay of Soil and Tailings, C.S. Sabau, D.R. Rayno, N.D. Kretz and P.W. Zelle, Argonne National Laboratory

D6.14 Analyses of Soils at Commercial Radioactive Waste Disposal Sites, P.L. Piciulo, C.E. Shea and R.E. Barletta, Brookhaven National Laboratory

D6.15 Process Characteristics for Immobilizing and Compacting Krypton-85, A.B. Christensen, J.A. DeDebbio, D.A. Knecht and J.E. Tanner, Exxon Nuclear Idaho Company, Inc.

Session D7

LOW LEVEL AND TRANSURANIC WASTE

Chair: G.L. DePoorter, Los Alamos National Laboratory
C.J. Northrup, Jr., Sandia National Laboratories
Wednesday Morning, November 3

D7.1 *The Oak Ridge National Laboratory Hydrofracture Process for the Disposal of Radioactive Waste, H.O. Weeren and E.W. McDaniel, Oak Ridge National Laboratory

D7.2 *An Evaluation of Alternative TRU Technologies, C. Bida and D.R. Mackenzie, Brookhaven National Laboratory

D7.3 Use of Poisoned Land/Inland Seas for Low Level Radioactive Waste Disposal, D.G. Brookings, H.A. Vogler, University of New Mexico, and J.J. Coheu, Science Applications, Inc.

D7.4 Behavior of Hazardous Contaminants in Vitrified Soil Produced by In Situ Vitrification, C.M. Mummerman, J.M. Rusin, and R.O. Lokken, Pacific Northwest Laboratory

9:30 D7.5 Ceramization of Inorganic Ion Exchangers Loaded with Nuclear Waste Into Red Clay Tiles, J. Lehto, University of Helsinki; O.J. Heinonen, Technical Research Centre of Finland, and J.K. Miettinen, University of Helsinki, Finland

9:50 D7.6 Geophysical Modeling of a Low-Level Nuclear Waste Disposal Site, K.A. Horton, L. Isaacson, and R.M. Morey, GEO-CENTERS, INC.

10:10 Break

Session D8

RADIATION EFFECTS

Chair: C.J. Northrup, Jr., Sandia National Laboratories
G.L. DePoorter, Los Alamos National Laboratory
Wednesday Morning, November 3

10:20 D8.1 *A Review of the Current Status of Radiation Effects in Solid Nuclear Waste Forms, W.J. Weber, Pacific Northwest Laboratory

10:45 D8.2 Mechanical Stability of a Cm-Doped Celsian Glass-Ceramic, J.L. Routbort, Argonne National Laboratory; P.J. Offermann, Hahn-Meitner Institute for Nuclear Research, FRG, and R.J. Matzke, Commission of the European Communities, Joint Research Center, FRG

11:05 D8.3 Ion-Implantation Damage in Silicate Glasses, C.W. Arnold, Sandia National Laboratories

11:25 D8.4 Radiolysis of Groundwater from HLW Stored in Copper Canisters, H. Christensen, Studsvik Energiteknik AB, Sweden, and E. Bjergbakke, Risø National Laboratory, Denmark

11:45 D8.5 Effect of Storage Temperature on Self-Irradiation Damage of ²³⁸Pu-Substituted Zirconolite, F.W. Clinard, Jr., Los Alamos National Laboratory; L.W. Hobbs, Massachusetts Institute of Technology; D.E. Peterson, D.L. Rohr and R.B. Roof, Los Alamos National Laboratory

Session D9

MODELLING

Chair: S.V. Topp, Savannah River National Laboratory
E.L.J. Rosinger, Atomic Energy Commission, Ltd., Canada
Wednesday Afternoon, November 3

2:00 D9.1 Approximations for Adapting Porous Media Radionuclide Transport Models to Analysis of Transport in Jointed Porous Rock, K.L. Erickson, Sandia National Laboratories

2:20 D9.2 Towards a Comprehensive Model of Chemical Transport in Porous Media, C.W. Miller, Lawrence Berkeley Laboratory

2:40 D9.3 Waste Classification Based on Total Hazard, L.E. Wickham and S.K. Rope, EG&G Idaho

3:00 D9.4 Compliance Assessment of Hypothetical Geologic Nuclear Waste Isolation Systems with the Draft EPA Standard, M.D. Siegel, M.S. Chu, R.E. Pepping, Sandia National Laboratories

* Invited Talk

D9.5 Modeling of Waste/Near Field Interactions for a Waste Repository in Bedded Salt: The Dynamic Network (DNET) Model, R.M. Cramwell, Sandia National Laboratories

D9.6 A Study of Coupled Thermomechanical, Thermohydrological and Hydromechanical Processes Associated with a Nuclear Waste Repository in a Fractured Rock Medium, C.F. Tsang, J. Noorishad and J.S.Y. Wang, Lawrence Berkeley Laboratory

Session D10 (Poster)

HIGH LEVEL WASTE FORM PROCESSES AND PROPERTIES,
LEACHING STUDIES, RADIATION EFFECTS

Chair: W.B. White, Pennsylvania State University
K. Kim, U.S. Nuclear Regulatory Commission
Wednesday Afternoon, November 3, 4:15-6:00 PM

D10.1 The Fixation of High-Level Solid Waste (HLSW) in a TiO_2 -Base Ceramic Matrix Provided with an Integrated Overpack, C. Bauer and G. Ondracek, Institut für Material- und Festkörperforschung, Germany

D10.2 Correlation Between Dynamic Leach Test Results and Geochemical Observations as a Basis for Repository-Relevant Release Rate Predictions, Aa. Barkatt, P.B. Macedo, W. Sousanpour, Al. Barkatt, M.A. Boroomand, P. Szoke and V.L. Rogers, The Catholic University of America

D10.3 Gamma-Zirconium Phosphate as a Cs-Waste Form for Partitioned Wastes, S. Komarneni and R. Roy, The Pennsylvania State University

D10.4 Strontium Leachability of Hydrofracture Grouts for Sludge-Slurries, M.T. Morgan, E.W. McDaniel, J.G. Moore, H.E. Devaney and L.R. Dole, Oak Ridge National Laboratory

D10.5 Alteration of Nuclear Waste Glass by Hydration, J.K. Bates and M.J. Steindler, Argonne National Laboratory

D10.6 Supernate Treatment Considerations for the Neutralized Waste at West Valley, G.B. Gockley, J.M. Pope, E.J. Lahoda, Westinghouse Research and Development Center

D10.7 Leach Resistance of the Borosilicate Glass VG98/12 Containing High Concentrations of Uranium, L. Kahl, M. Nesovic, J. Saidl, Institute of Nuclear Waste Management, and E. Mainka, Institute of Radiochemistry, Nuclear Research Center Karlsruhe, FRG

D10.8 Comparison of 200 Liter and 40 Milliliter Leach Tests, W.A. Ross, J.H. Westsik, Jr., F.P. Roberts, and C.O. Harvey, Pacific Northwest Laboratory

D10.9 Electrical Conductivity Measurements of Leachates for the Rapid Assessment of Wasteform Corrosion Resistance, B.C. Sales, M. Petek and L.A. Boatner, Oak Ridge National Laboratory

D10.10 Correlation of ^{137}Cs Leachability from Small-Scale (Laboratory) Samples to Large-Scale Waste Forms, N. Morcos, R. Dayal and A.J. Weiss, Brookhaven National Laboratory

D10.11 Long-Term Radioactivity Release From Solidified High-Level Waste Part II; Parametric Study of Waste Form Properties, Temperature and Time, F.K. Alténhef and W. Lutze, Hahn-Meitner Institut, FRG, and R.C. Ewing, University of New Mexico

D10.12 Radiolytic Gas Generation and Oxygen Depletion in Ion Exchange Materials, W.P. Capolupo and J.R. Sheff, University of Lowell

D10.13 Irradiation Effects in the Storage and Disposal of Radioactive Ion-Exchange Resins, K.J. Swyer, C.E. Dodge, R. Dayal and A.J. Weiss, Brookhaven National Laboratory

D10.14 Compatibility Relationships in the Fe-U-O-H System at 400°C: The Implications of the Ferric-Ferrous Buffer for the Immobilization of Uranium and Transuranic Elements, D.K. Smith, W.P. Freeborn and B.E. Scheetz, The Pennsylvania State University

D10.15 Internal Hydrogen Effects in Ti Code 12 Canister Overpack Material, N.R. Moody and S.L. Robinson, Sandia National Laboratories

D10.16 Corrosion of TiCode-12 in a Simulated Waste Isolation Pilot Project (WIPP) Brine, T.M. Ahn, B.S. Lee, J. Woodward, R.L. Sabatini and P. Soo, Brookhaven National Laboratory

D10.17 Hydrothermal Simulation of Ion Migration in Selected Argillaceous Sediments: Implication for Backfill Design, L. Bratton, B.E. Scheetz and W.B. White, The Pennsylvania State University

D10.18 Diffusion Measurements in Concrete and Compacted Bentonite, A. Muurinen, J. Rantanen, K. Uusheimo, O.J. Heinonen, Technical Research Centre of Finland, Finland

D10.19 Rheological and Physical Properties of Magnesium Oxide and Silica Fume-Modified Cement Mortars Cured at High Temperature, Z. Nakagawa, E.L. White and D.M. Roy, The Pennsylvania State University

D10.20 Dissolution and Reaction Studies of $CsZr_2(PO_4)_3$ and Zr Phosphates in Aqueous Media, E.R. Vance and F.J. Ahmad, The Pennsylvania State University

D10.21 Differential Scanning Calorimetry of Metamict Zirconolite, D.E. Peterson and F.W. Clinard, Los Alamos National Laboratory

Session D11

CANISTER AND BACKFILL

Chair: J.F. Kircher, Office of Nuclear Waste Isolation
T. Westermark, The Royal Institute of Technology, Sweden
Thursday Morning, November 4

8:15 D11.1 *Corrosion Resistant Canisters for Nuclear Waste Isolation, N.J. Magnani, Sandia National Laboratories

8:40 D11.2 The Canadian Container Development Program for Fuel Isolation, K. Nuttall, J.L. Crosthwaite, P.J. McKay, P.M. Mathew, Atomic Energy of Canada Limited, Canada; and B. Teper, P.Y.Y. Maak and M.D.C. Moles, Ontario Hydro, Canada

* Invited Talk

D11.3 Some Effects of Microstructure and Chemistry on Corrosion and Hydrogen Embrittlement of TiCode-12, J.A. Ruppen, R.B. Diegle, R.S. Glass and T.J. Headley, Sandia National Laboratories

D11.4 Investigations of Suitable Metallic Container Materials for HAW Solidification, V. Mirschinka, R. Odoj, St. Halaszovich, Institute for Chemical Technology, Germany, and K. Forch, U. Forch, Thyssen-Henrichshutte AG, Germany

D11.5 Laboratory Studies of Fluid Flow Through Borehole Seals, D.L. South and J.J.K. Daemen, University of Arizona

Break

D11.6 *Thermal Conductivity of Waste Forms and Geologic Media, R.O. Pohl, Cornell University

D11.7 Thermal Conductivity of Bentonite/Quartz High-Level Waste Package Backfill, M. Moss and M.A. Molecke, Sandia National Laboratories

D11.8 Experimental Investigation of Sodium Bentonite Stability in Hanford Basalt, M.I. Wood, Rockwell Hanford Operations

D11.9 Diffusion of Radionuclides in Brine-Saturated Backfill Barrier Materials, E.J. Nowak, Sandia National Laboratories

D11.10 Inertial Effects in Transport of Radioactive Waste Through Engineered Barriers, C.L. Carnahan, Lawrence Berkeley Laboratory

D1.1 THE SCIENTIFIC BASIS FOR LONG TERM PREDICTION OF WASTE FORM PERFORMANCE UNDER REPOSITORY CONDITIONS. John E. Mendel, Pacific Northwest Laboratory, P.O. Box 999 Richland, WA 99352.

The definition of repository conditions is different for each of the principal candidate geologic repository locations, bedded salt, basalt, and tuff, especially when the potential differences in waste package design are factored in, as they must be, since the engineered waste package can control the immediate environment around the waste form to a significant degree for a very long time. The changes in repository conditions that are projected with time will be described.

The aging, or time-dependent changes in the waste form due to radiation and thermal effects will also be described. Finally, the types of interactions that can occur between the aged waste form and its repository environment will be discussed and the techniques that are being used to study and predict the long-term consequences of these interactions will be reviewed. The role of test methods defined by the Materials Characterization Center will be presented.

D1.2 SYNROC: PROPERTIES, PRODUCTION TECHNOLOGY AND DISPOSAL STRATEGY. A.E. Ringwood, Research School of Earth Sciences, Australian National University, Canberra, A.C.T. 2600, Australia.

The comparative leaching performances of SYNROC and borosilicate glass in water at 90°C, 200°C and 300°C are reviewed. At 90°C, univalent and divalent elements (e.g. Cs, Ca, Sr, Ba) are 500-2000 times less leachable from SYNROC than from glass. For multivalent elements, e.g. Ti, Zr, U, Nd, SYNROC is about 10,000 times more resistant to leaching. SYNROC continues to display excellent resistance to leaching at higher temperatures whereas borosilicate glass rapidly disintegrates above 150°C. Results of leaching studies of individual SYNROC minerals are consistent with measurements on bulk SYNROC samples.

The effects of radiation damage on SYNROC minerals have also been investigated. The amount of alpha radiation which would be received during the decay of radwaste in SYNROC minerals to safe levels causes only minor structural changes in the zirconolite and perovskite phases which are the hosts for α -emitting actinide elements. Experimental studies demonstrate very low leachability of these minerals despite their exposure to enormous radiation doses.

The superior leach resistance of SYNROC at elevated temperatures provides considerable flexibility in strategies for subsequent intermediate storage and geological disposal and could yield large cost savings. These topics will be addressed in more detail. Important advances in production technology have recently been made in a joint research program conducted by the Australian Atomic Energy Commission and the Australian National University. A new technique of uniaxially hot-pressing SYNROC powder in collapsible stainless steel bellows to produce fully dense monoliths is now operating at full scale and will be described.

ted Talk

3
 DETERMINATION OF CRYSTAL SITE OCCUPANCY IN SYNROC USING ANALYTICAL ELECTRON MICROSCOPY, J. Taft[†], J.C.H. Spence, and D. Clarke* Department of Physics, Arizona State University, Tempe, AZ 85287 and *Rockwell International Science Center, 1049 Camino del Rio, Thousand Oaks, CA 91360

The rate of radiation damage from radioactive species in crystals (e.g. SYNROC) may depend on the crystallographic site of the emitting species. It is therefore of importance to determine the site occupancy of the radioactive elements. No established crystallographic technique is appropriate for such studies due to the small crystal grains and the many different phases present in SYNROC. By taking advantage of the fact that an electron beam (-100keV) can be focussed to a particular crystal site by electron diffraction, a new technique capable of locating small amounts of ions in small crystal volumes (100Å)³ has recently been developed. By monitoring a characteristic x-ray emission using an analytical electron microscope equipped with an energy dispersive x-ray spectrometer, the site occupancy of the different elements in perovskite, spinels and possibly other crystallites present in SYNROC can be located. Experimental results will be presented and the dependence of the rate of radiation damage on the crystal site of the radioactive species will be discussed.

4
 A SINGLE PHASE CERAMIC WASTE FORM: [NZP]. R. Roy, Liji Yang, E.R. Vance and J. Alamo, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

We have experimentally demonstrated that the components of typical commercial processing waste can be incorporated into an essentially single-phase ceramic, based on the [NZP] structure family. 20-40 wt.% of simulated PW-4b processing waste has been incorporated into an [NZP] phase (plus a little hazite on occasion). Crystal chemical modeling for each of the ions making up the PW-4b has been used to guide the experimental compositions studied.

Two different processing routes were studied for the production of the ceramics: one based on a batch processing from sol-gels; the other on the use of a preformed Zr-phosphate gel as a starting component. Various heat treatments, varying temperature and pO₂, have been carried out. A great advantage of these ceramics is that satisfactory consolidation could be achieved by sintering at temperatures as low as 900°C. Preliminary dissolution data on the ceramics under simulated repository wastes will be reported.

5
 LEACHING CHEMISTRY OF DEFENSE BOROSILICATE GLASS. R. M. Wallace and G. G. Wicks, I. du Pont de Nemours & Company, Savannah River Laboratory, Aiken, South Carolina 29808

A reference concept for long term management of Savannah River Plant (SRP) waste involves immobilization of the high level waste into borosilicate glass. A multi-phase program is in progress to assess the leachability of SRP glass. The primary objectives of this program are to measure and understand glass leaching as a function of many variables so that the long term behavior of SRP waste glass in a repository environment can be predicted.

Recent studies at the Savannah River Laboratory have shown that waste glass corrodes congruently (even after short periods of time) but only a fraction of the material originally in the glass enters the leachate. The remainder is deposited on the surface as a layer composed primarily of elements with insoluble hydroxides that contain adsorbed fission products and actinide elements. The surface layer is thought to inhibit further corrosion of glass. A theoretical model that involves reaction of water with glass followed by diffusion of the resulting soluble silicates through a developing surface layer was developed to describe leaching. This approach describes observed leaching behavior better than existing models.

Continuing work includes detailed studies of the early stages of glass leaching. Other experiments are aimed at determining the conditions under which surface layers are formed and their effect in protecting glass from further corrosion.

D1.6
 PREFERENTIAL DISSOLUTION PHENOMENA IN NUCLEAR WASTE MATERIALS. David R. Clarke, Rockwell International Science Center, Thousand Oaks, CA 91360.

In evaluating the long term (geologic time) dissolution behavior of candidate nuclear wasteforms extrapolation is made of elemental release rates measured in short-term leach tests that are typically days or months in duration. Whilst such methods can be expected to lead to lower bound estimates of the stability of the wasteform, refinements in predicting the overall dissolution require that all the major dissolution mechanisms be identified. In particular, preferential dissolution phenomena need to be identified. One method that has proved to be particularly valuable is the direct observation of dissolution using the thin foil samples in the transmission electron microscope. Such studies led to the demonstration that intergranular glass phases are preferentially leached out of crystalline waste materials. More recently the microstructural connectivity of the nepheline phase has been shown to dictate the penetration of leachant in defense waste modified Synroc-D causing regions of penetration that extend further into the wasteform than the uniform dissolution front. In addition a number of the detrimental aspects of cracks in both glass and ceramic wasteforms have been investigated by direct microscopy observations. In each case the extent of the preferential dissolution is related to the processing conditions under which the materials were fabricated. The presentation will describe the observations of preferential dissolution made to date and emphasize their connection to the fabrication conditions.

D1.7
 MORPHOLOGY AND CHEMICAL COMPOSITION OF A SURFACE LAYER FORMED UPON LEACHING OF NUCLEAR WASTE GLASS. Werner Lutze and Günter Malow, Hahn-Meitner Institut, 1,000 Berlin 39, Germany, and Tomas J. Headley, Sandia National Laboratories, Albuquerque, New Mexico, USA.

A borosilicate glass was leached at 165° and 200°C in water (S/V ratio of ~0.01) in a closed system. Microprobe and transmission electron microscopy were used to determine the chemical composition and the morphology of the surface layer formed as a function of time. The leachate was analyzed for all elements. The layer formation can be described in terms of solubility limits, i.e., precipitation of leached elements. Drastic changes in the layer's composition with time indicate

chemical activity of the precipitating layer. Crystals (50Å and larger) are formed after a few days within and on top of the layer. Sharp boundaries become visible between the layer and pristine glass and within the layer, separating it into regions with different morphologies and composition. This investigation is being carried out to provide evidence that leaching models using linear time law and/or constant diffusion coefficients may not be appropriate to characterize the leaching mechanism. A thorough investigation of the above processes with the help of TEM can provide the answer to: -whether a layer can protect the glass from further leaching, -whether the layer reaches a steady state of chemical composition, - whether internal stresses can lead to layer detachment.

INFLUENCE OF SURFACE AREA TO SOLUTION VOLUME RATIO AND ITS IMPLICATIONS TO ACCELERATED LEACHING TESTING. Larry R. Pederson, C.Q. Buckwalter, and G.L. McVay, Pacific Northwest Laboratory, PO Box 999, Richland, WA 99352.

Leaching of a complex simulated nuclear waste glass was significantly decreased with an increase in the glass surface area to solution volume ratio (SA/V). The decrease in leaching is attributed to approaching the limits of silicon solubility. Congruent leaching was never observed for this glass--unlike trends previously reported for some simple and complex silicate glasses. Roughened surfaces resulted in a decrease in leaching rate increase which may be explained in terms of the local surface area to solution volume ratio. Increasing the SA/V ratio may be a viable method of increasing the leaching rate as it pertains to solution concentration. This may in turn be used to model long-term behavior of leaching.

MECHANISMS OF LEACHING OF ALKALI-BOROSILICATE GLASS. D.E. Clark and L.L. Hench, Department of Materials Science and Engineering, University of Florida, Gainesville, FL 32601.

The alkali-borosilicate (ABS) system provides the basis for a wide variety of commercially important products among which are the nuclear waste glasses. Although a large number of corrosion investigations have been undertaken in the last five years, the leaching mechanisms of the ABS glasses have not been characterized as well as for the soda-lime-silicate (NCS) glasses commonly used for containers. It is well known that the leaching of the latter glasses involves both ion exchange and network dissolution processes resulting in the development of one or more types of surface films. In the present paper we compare the leaching behavior of the ABS and NCS glasses and discuss our current understanding of ABS leaching in terms of mechanisms, kinetics, surface film formation and hydrodynamics.

D2.2
PRODUCT LEACHING AND SOLID STATE PROPERTIES OF FORMULA 127 GLASS PREPARED WITH ACTUAL RADIOACTIVE ZIRCONIA CALCINE. B.A. Staples and H.S. Cole, Exxon Nuclear Idaho Co., Inc., Box 2800, Idaho Falls, ID 83402.

Formula 127 glass has been developed to immobilize zirconia type calcine which makes up the bulk of the solidified nuclear fuel reprocessing high level wastes being stored at the Idaho Chemical Processing Plant (ICPP). This borosilicate glass has been prepared with actual radioactive zirconia calcine and is being subjected to the MCC-1 and MCC-2 static leach tests, the Soxhlet leach test, and various solid state and analytical techniques to verify that its product characteristics are the same as those of Formula 127 glass prepared with nonradioactive simulated zirconia calcine. With respect to total mass loss by application of the MCC-1 tests, the glass prepared with actual radioactive calcine is similar to glass prepared with simulated nonradioactive calcine. Initial results from the application of these leaching tests to the radioactive glass indicate that only very small amounts of actinides and fission products are leached.

D2.3
GROWTH OF SURFACE LAYER DURING THE LEACHING OF THE SIMULATED WASTE GLASS AND ITS BARRIER EFFECTS ON THE LEACHING. K. Ishiguro, N. Kawanishi, N. Sasaki, H. Nagaki and M. Yamamoto, Power Reactor and Nuclear Fuel Development Corporation, Tokai-mura, Japan.

Growth of surface layer on simulated waste glass during the leaching and effect of layer formation on the leaching rate have been studied. Two different leaching methods in distilled water, static leaching and Soxhlet leaching, were used at a temperature of 100°C. Leaching rates were determined from the measurement of the weight loss of samples and the analysis of the leachants. Solid samples after leaching were analyzed by using Auger and ESCA. Enrichment of transition metal elements and rare earth elements were observed in the layer. The layer consisted of two or three sublayers which were distinguished by their different components. Iron, Nickel and zinc were more concentrated at the outer surface, on the other hand rare earth elements such as neodymium and lanthanum were segregated at the inner layer. Silicon, Boron and alkali were depleted in all sublayers. These profiles through the layer were common to the samples in the both type of leaching. Growth kinetics of the layer followed approximately linear relations. The layer in the static leaching grew more rapidly than that in the Soxhlet leaching. Comparison between the leaching rate and the thickness of the layer showed that layers did not work as effective diffusion barriers until the threshold thickness. The threshold of the layer at which the leaching rate decreased was about 3000 Å for the static leaching and about 8000 Å or over for the Soxhlet leaching.

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D2.4
EFFECTS OF SOLUTION CHEMISTRY AND ATMOSPHERE ON LEACHING OF ALKALI-BOROSILICATE GLASS. H-P. Hermansson, H. Christensen (Studsvik, Energiteknik AB, Sweden) and D.E. Clark (University of Florida, USA).

The leaching behavior of two alkali-borosilicate glasses containing 9 wt% simulated fission products and 1.6 wt% uranium oxide

been studied. Samples were exposed to one of eight types of leachants including doubly distilled water, simulated groundwater, brackish water, a brine solution, and solutions containing various concentrations of iron, aluminum or sodium maintained at either 40°C or 90°C for up to 182 days. The most aggressive leachants were the solutions containing sodium and simulated groundwater and silicate water. These solutions increased the extent of leaching by a factor of 2-3 over that for distilled water for one of the glasses. A partially protective surface film rich in cesium and potassium formed on the glass exposed to the brine solution.

In order to evaluate the effects of atmosphere on leaching, samples were also immersed in doubly distilled water over which relative concentrations of oxygen, nitrogen and carbon dioxide were varied. Increasing the carbon dioxide concentration from 0 to 100% resulted in a factor of 3 increase in the leaching rate.

The significance of these findings and their relevance to the design of a waste form/repository system will also be addressed.

D2.6 LEACHING PROCESSES IN SILICATE GLASS: THE EFFECTS OF SOLUTION CHEMISTRY AND IONS IN SOLUTION ON GEL LAYERS.* Bruce C. Bunker, E. K. Chapp and G. W. Arnold, Sandia National Laboratories, Albuquerque, NM, 87185.

The leaching of alkali cations from $(1-x)\text{Na}_2\text{O} \cdot x\text{K}_2\text{O} \cdot 3\text{SiO}_2$ glasses was investigated using pH stat titrations, solution analyses, and depth profiling by elastic recoil detection (ERD) and Rutherford backscattering (RBS). Leaching kinetics show two stage behavior, going from parabolic (as \sqrt{t}) in stage #1 to linear with time (stage #2). In the diffusion controlled stage, it is demonstrated that the interdiffusion of Na^+ and alkali cations via ion exchange is controlled by the structural and chemical properties of hydrated glass, which are shown to be a strong function of the effective pH and water content. In stage #2, alkali release is controlled by the rate at which water reacts at the interface between hydrated and unhydrated glass (the gel:glass interface). The hydrated glass does not serve as a diffusion barrier. Experiments were conducted to see how ions in solution (Pb^{2+} , Al^{3+} , Fe^{3+} , Cu^{2+} , and Ag^+) interact with the hydrated glass which forms during leaching. These investigations show that the interactions between ions in solution and hydrated glass are a strong function of the solution pH and ion concentrations, which control the acid-base chemistry of silica and the formation and solubility of cation hydrolysis products.

This work supported by the United States Department of Energy (DOE) under contract DE-AC04-76-DP00789.

D2.6

THE DOMINANT MECHANISMS IN LEACHING OF COMPOSITES OF CEMENTS WITH RADIOACTIVE WASTE. Mary W. Barnes and Della M. Roy, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

Because composites of radioactive waste and cement are being used for disposal, it is important to be able to predict their leaching behavior. Major reactions involved are dissolution of Ca_3SiO_5 , reaction of Ca_3SiO_5 to form calcium silicate hydrate (C-S-H), dissolution of silica, and reaction of dissolved species to form C-S-H. All four reactions are shown to occur; the dominant one was identified from free energy-activity calculations. Free energies of these reactions were calculated and used to predict solution activities in equilibrium with each reaction. Activities from analyzed leachate concentrations, both for cement alone and for pollicite plus cement, fit within the calculation accuracy, the activities for the reaction of dissolved silica and Ca^{2+} to form C-S-H.

The reaction of C-S-H is thus buffering the leachate even in the presence of a silicate waste form. It is producing pH's from 10.0-11.5, and the behavior of any potential waste form to be used in these composites must be considered in these alkaline conditions.

D2.7

LEACH MODELS FOR A COMMERCIAL NUCLEAR WASTE GLASS. W. L. Kuhn, R. D. Peters, Pacific Northwest Laboratory, P. O. Box 999, Richland, WA 99352.

A review of the leaching behavior of 76-68 glass shows that it cannot be explained in terms of solid state diffusion/dissolution, which has been the basis for several leach models. Instead, we present two models based on a dissolution rate impeded by surface processes: the accumulation of a protective layer of insoluble reaction products, and adsorption of reaction products on the surface. The resulting rate laws are identical and predict a change from linear to parabolic rate laws for soluble species, which is found to agree with the data over a range of temperatures. Incongruent release is attributed primarily to solubility effects. The relative merits of the models and their usefulness in modeling repository behavior are discussed.

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D2.8

COMPARISON OF THE LEACHABILITY OF THREE TRU CEMENT WASTE FORMS. J.H. Westsik, Jr., W.A. Ross, F.P. Roberts, C.O. Harvey, Pacific Northwest Laboratory, Richland, WA 99352.

Three waste forms--a cast hydraulic cement, a cold-pressed hydraulic cement and a FUEAP cement--were leach tested using the Materials Characterization Center MCC-1 procedure. Each waste form contained a reference waste composed of a process sludge and an incinerator ash and contained about 0.01 percent plutonium. The leachability of the three cements was evaluated in terms of the releases of Pu, Al, Ca, Na, and Si. Releases of these elements are dominated by the rapid increase of the leachate pH to greater than 11. No significant differences were observed in the leachability of the cast, cold-pressed, and FUEAP cements. Selection of one form over the others will have to be based on other criteria such as the process economics.

9
 ENDED LEACH STUDIES OF ACTINIDE DOPED SRL 131 GLASS. John K. Bates,
 Mel J. Lam, and Martin J. Steindler, Argonne National Laboratory, Argonne, IL
 39.

effects of subtle changes in glass composition on the leaching of SRL 131
 ense waste glass have been noted. Solution analyses have been correlated with
 face examination and for the base 131 frit the layer formation is tenuous,
 eriorating on drying, and alteration product coverage is ~5%. For modified 131
 t an adherent surface layer forms and after 390 days leaching alteration product
 erage is 100%. The leaching of modified frit containing the actinides U, ²³⁷Np,
²³⁹Pu, and ²⁴¹Am, together with the fission products ⁸⁹Sr, ¹³⁵Ba, ¹³⁷Cs, ¹⁴¹Ce,
¹⁵²Eu has been measured and leaching was correlated to surface effects using
 X-ray photoelectron spectroscopic technique. Leach rates of ²³⁹Pu and ²⁴¹Am
 ~400 times less than U and ²³⁷Np, while the order of fission product leaching
 Cs > Ba, Sr >> Ce, Eu.

1
 STABILITY OF NATURAL GEOMEDIA FOR CONSIDERATION FOR RADWASTE STORAGE. W.S. Fyfe,
 Department of Geology, University of Western Ontario, London, Canada, N6A 5B7.

choosing the optimum geological environment for radwaste disposal two factors
 perhaps dominant. First, the host rock volume should have the lowest possible
 dictable permeability for the next 5 million years. Second, where fluid flow
 curs, dispersion should be maximal. Both these considerations imply that we
 t understand the nature and history of fracture formation and porosity change,
 rticularly over the last 5 million years or so. There has been too little study
 this problem but common wisdom is that thinly bedded rocks have the most closely
 aced and low aperture fractures while massive and thick rock units have widely
 aced fractures with larger apertures. Thus thinly laminated rock units may pre-
 nt a fracture system for maximal flow dispersion and the minimal chance for
 astrophic flow via large aperture fractures.

need geologic evidence for time integrated fluid flux in appropriate rock types
 here, the study of "sensitive" minerals should be intensified. The alteration
 Fe-Mn-U oxides for example, can indicate fluxes of reduced or oxidized fluids.
 omineralization products with their unique levels of many trace elements (both
 tionic and anionic species) may be used to show trace metal dispersion, particu-
 larly if the original bio-mineral is metastable (e.g. amorphous SiO₂, aragonite,
 c.).

her geological considerations involve consideration of variables such as rock
 n-exchange capacity, crack sealing mechanisms, thermal stability of constituent
 nerals, regional stress fields, and regional hydrogeologic regimes on all
 ropriate scales. All the above must be related to the nature of waste
 terials, backfill materials and mining technology.

ere is great need for an intensified effort on geological aspects of disposal
 d geological evidence on the preservation of "sensitive" materials. Evidence
 "million year" stability of a region containing possible host rocks may be best
 nd from study of processes in recent cover rocks. There is no good reason to
 lieve that old rocks are better than young rocks for waste disposal.

is stressed that while a great emphasis is at present placed on laboratory
 udies of waste forms etc., too little effort is directed at providing field
 idence for the best host rock and geological environment.

D3.2

EVAPORITE DISSOLUTION RELEVANT TO THE WIPP SITE, NORTHERN DELAWARE BASIN, SOUTH-
 EASTERN NEW MEXICO.* Steven J. Lambert, Sandia National Laboratories, Albuquerque,
 NM 87185.

Evaluation of the threat of evaporite dissolution in the Delaware Basin to the
 Waste Isolation Pilot Plant (WIPP) site has been based on several assumptions con-
 cerning: (1) the original volume of evaporites; (2) the efficiency of the dissolu-
 tion model, dependent on climate, hydrologic regime, and accessibility of evaporites
 to undersaturated solutions, and (3) the time interval during which dissolution has
 occurred. Geological assumptions sometimes used to estimate the amount of halite
 removed by dissolution in the Castile and Salado Formations are invalid. The use of
 the brine-density flow model to account for various dissolution features in the
 Delaware Basin, while physically plausible, is inconsistent with geological data.
 Stable-isotope and major solute compositions of groundwaters below and within the
 evaporites are characteristic of brines which are not simple products of active dis-
 solution by meteoric water, but have resulted from a complex sequence of interac-
 tions involving a large rock/water ratio. Dissolution within the evaporite sequence
 leaves characteristic surficial and subsurface features, such as collapse, fracture,
 dissolution brines and gypsified zones. Geomorphic features and "brine reservoirs"
 cannot be attributed to dissolution without evidence of rock-water interaction
 characteristic of dissolution. Constraint of most evaporite dissolution in the
 Delaware Basin to the Quaternary yields on unrealistically high rate estimate, in
 view of the fact that dissolution has taken place episodically since the Permian.
 Geologically reasonable models for dissolution indicate that the threat of dissolu-
 tion to the WIPP in the next 250,000 years is not greater than it has been in the
 last 600,000 years of highly variable climate.

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D3.3

NATURAL ANALOGUES: ALAMOSA RIVER MONZONITE INTRUSIVE INTO TUFFACEOUS AND
 ANDESITIC ROCKS. D.C. Brookins, M.S. Abashian, Univ. New Mexico; L.H. Cohen
 and A.E. Williams, Univ. California-Riverside; H.A. Wollenberg and S. Flexser,
 Lawrence Berkeley Laboratories.

The Alamosa River monzonite stock intruded tuffaceous and andesitic rocks of the
 Summitville-Platoro caldera complex 29 MYBP. Extensive oxygen isotopic exchange
 between the stock and host rocks occurred during intrusion and at post-intrusion
 hydrothermal events (Williams, 1980). Convective circulation of waters has also
 been documented (ibid.). Recent studies of elemental behavior in the contact
 zones, and in the stock and intruded rocks removed from the contact, have been
 undertaken to test this convective system as analogue for elemental transport
 around a hypothetically breached radwaste canister. Samples were taken from
 three traverses from stock into tuff and andesite, using a field gamma spectrom-
 eter for monitoring measurements of K, U, Th. There is a pronounced chemical
 gradient for many elements between the monzonite and tuff, thus enhancing possible
 elemental exchange. Our studies show, however, that despite the evidence for
 oxygen isotopic exchange, other elements have not migrated except possibly in a
 ten-meter wide zone at the exact contact between monzonite and tuff. Data for K,
 Cs, Co, Cr, Ta, U, Th, V, Ti, Fe, Sc, Rb, Sr, Ba and the REE show that they have
 apparently been retained in their original host rock except for the ten-meter
 contact zone. The REE distribution patterns show rock-specific signatures not
 affected by the intrusion. Some quartz-diluted alteration zones in the ten-
 meter contact zone are slightly enriched in Cs and K, but other elements are
 unaffected even in this narrow zone. The results of the study suggest re-
 tention of radio- and fission-trace elements in their original hosts despite
 the high temperature, convective system operative during and post-emplacement.

4
GEOLOGICAL AND PETROLOGICAL CONSIDERATIONS RELEVANT TO THE DISPOSAL OF RADIOACTIVE WASTES BY HYDRAULIC FRACTURING: AN EXAMPLE AT THE U. S. DEPARTMENT OF ENERGY'S OAK RIDGE NATIONAL LABORATORY (ORNL). C. S. Haase, Environmental Sciences Division, Bldg. 3504, Oak Ridge National Laboratory, Oak Ridge, TN 37830

ORNL the Pumpkin Valley Shale (PVS) is used as a host formation for hydraulic fracturing waste disposal. Measurements on core samples of PVS and data from past hydraulic fracturing tests and operations indicate that the PVS has the very low permeabilities (10^{-1} to 10^{-4} millidarcies) required to make it a suitable host formation. To relate the empirically measured permeabilities to petrological properties of the PVS a comprehensive study of the formation was undertaken.

PVS is a lithologically complex, highly fractured and jointed interbedded mudstone and siltstone. It has experienced a complex diagenetic history that has resulted in the occlusion of primary and secondary porosity, and thus, produced a geology of low intrinsic permeability. Fracture permeability of the PVS is likewise greatly reduced by calcite filling of joints and the lack of long range continuity of joint sets. These factors explain the previously measured low permeability values. Furthermore, diagenetic patterns and general porosity trends can be related to specific different lithology types. The spatial distribution of geologies is a function of the depositional environment of the PVS. Documentation of this relationship between lithology distribution and porosity-permeability allows important host formation properties, such as permeability patterns, to be related to measurable and mappable geological and petrological parameters. It permits extrapolation of such patterns to little-studied portions of the formation. Such a situation better allows for the satisfactory operation and assessment of the hydraulic fracturing at ORNL.

5
TECHNETIUM BEHAVIOR IN SULFIDE AND FERROUS IRON BEARING MEDIA. Suk Y. Lee and Robert A. Bondietti, Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830

Technetate anion (TcO_4^-), a potentially mobile species in leachate from a leached radioactive waste repository, was removed from brine solution by precipitation with sulfide, ferrous iron, and ferrous iron sulfide at environmental pH's. The black precipitate obtained from the TcO_4^- -sulfide reaction was poorly crystallized technetium sulfide ($Tc_2S_7?$) and insoluble in both acid and alkaline solution in the absence of strong oxidants. Magnetite or possibly maghemite was the dominant mineral in the precipitate formed from the TcO_4^- -ferrous iron reaction. The reduced particle size and poor crystallinity of the iron oxide in the presence of TcO_4^- suggests that the technetium was incorporated into the oxide structure after reduction to a lower valence state.

Ferrous iron sulfide, an initial phase precipitating in the TcO_4^- -ferrous iron-sulfide reaction, effectively removed TcO_4^- from solution. Although ferrous sulfide quickly transformed to an iron hydroxide mineral possibly ferrocrocite, technetium was retained by the solid phase during structural transformation. The results suggest that ferrous iron and/or sulfide precipitating ground waters and minerals in host rock or backfill barriers would reduce the mobility of TcO_4^- by the formation of less soluble technetium-bearing iron minerals.

D3.6
NATURAL ANALOGUES--A WAY TO INCREASE CONFIDENCE IN PREDICTIONS OF LONG TERM PERFORMANCE OF RADIOACTIVE WASTE DISPOSAL, G. F. Birchard, U.S. Nuclear Regulatory Commission, Washington, DC 20555

No exact analogue to a radioactive waste disposal system exists but by studying natural analogues of the most important components or subsystems of a waste repository, confidence in long term predictions can be increased. The U.S. Nuclear Regulatory Commission is supporting research on igneous intrusions into proposed repository-type host rock (basalt, tuff, crystalline hardrock, and salt), on uranium ore body analogues, on actinide solubility as a function of natural complexing agents in closed basin lakes and on field migration of radionuclide species and low level waste and uranium mill tailings disposal sites. The NRC is studying analogues to ensure that important parameters which might be overlooked in laboratory tests and in modelling because of problems with scaling time, space and geologic complexity can be identified and their importance bounded.

D4.1
NMTS WASTE PACKAGE DESIGN AND MATERIALS TESTING STATUS: FY 82. J.F. Kircher and D.J. Bradley, Office of Nuclear Waste Isolation, Battelle Memorial Institute, 505 King Avenue, Columbus, OH 43201.

The National Waste Terminal Storage Program (NMTS) has been actively involved in materials testing and design efforts with respect to a nuclear waste package for use in geologic disposal. This effort has been aimed at addressing questions related to materials behavior in salt, basalt, and tuff geochemical environments.

This paper presents conceptual waste package designs for spent fuel and commercial high-level waste for these geologies as well as a synopsis of the materials testing programs that input to waste package design. Conceptual waste package designs will be described for repositories in salt, basalt, and tuff. These designs are site specific. Clay, or mixed clay-host rock, backfills are an integral part of the package design for basalt and tuff but not salt. The main structural/corrosion barrier is iron or steel in all cases with possibly a titanium alloy overpack. Single package, vertical borehole emplacement is considered for salt whereas multipackage, horizontal borehole emplacement is being considered for the other two geologies. These package concepts and the geochemical environment then dictate materials requirements and testing needs.

A general approach to testing waste package materials, both singly and in concert, has been developed and will be described.

With respect to studies on materials in support of waste package design, a good deal of progress has been made as of the end of FY 1982. This work has progressed from generic screening studies to tests performed in more site-specific environments. The major areas of study have covered;

- Glass, Spent Fuel, and TRU waste forms
- Metallic canister/overpack materials
- Bentonite/zeolite backfill materials.

major findings to date on materials testing in these major areas will be discussed.

In order to be able to describe behavior of the waste package with respect to performance criteria, predictive models, based on experimental and theoretical work, are being developed. The present status of component and waste package models will be presented.

2
FRENCH WASTE MANAGEMENT PROGRAM, Jean F. Lefevre, Directeur chargé des fluides et déchets, Commissariat à l'Énergie Atomique B.P.6, 92260 Fontenay aux Eaux, France.

To deal with the wastes from the French atomic power program, an extensive waste management plan has been set up. The technical options taken, the areas of future research and the institutional arrangements will be described. Wastes are immobilized in solid matrices ranging from cement, bitumen and thermosetting resins for HL and ML wastes to borosilicate glasses for HL liquid wastes. A shallow land disposal site is in operation for disposal of L and ML wastes at La Manche by the National Waste Management Agency (ANDRA). α -bearing wastes are kept in interim storage, awaiting the availability of a geologic disposal site by 1992. HL glasses are stored in steel containers at the production site in an engineered ventilated storage. A deep geological disposal demonstration site is hoped to be in operation before 1995. Research and development aims at an improvement in the efficiency and economics of confinement matrices, evaluation of barrier characteristics and modifications in order to optimize options and parameters, and the identification, construction and installing of deep geologic repositories.

3
UNITED KINGDOM REGULATORY PROCEDURES FOR RADIOACTIVE WASTES. F.S. Feates and J. Richards, Department of the Environment, Romney House, 43 Marsham Street, London SW1P 3PY, England

The UK policy for radioactive waste management is the responsibility of government ministers. In England the Department of the Environment is also responsible for authorisation of all radioactive waste disposal activities and has its own Inspectorate to ensure that government policy is respected. An independent Advisory Committee reports directly to the Secretary of State.

Low level wastes are currently disposed of by shallow burial or to the deep sea. Sites for the disposal of intermediate waste (with and without significant lived activity) are being sought as a matter of urgency and reprocessing (i.e. generating) wastes will be stored on the surface for at least fifty years. The rationale of this policy will be explained.

D4.4

SOME CONTRIBUTIONS OF BASIC-INORGANIC CHEMISTRY TO WASTE OR SPENT FUEL STORAGE. Ingmar Grenthe, Department of Inorganic Chemistry, Royal Institute of Technology, Stockholm, Sweden.

Radioactive products may be released from a nuclear waste repository to the biosphere through dissolution in and transport by ground water. The fundamental chemical phenomena involved are solubility, complex formation, redox and sorption (e.g. ion-exchange) processes. The quantitative modelling/predictions of the long time effects of the nuclear repositories usually involve data describing the equilibrium properties of the system as an important element. The models are no better than the quality of the data used, hence there is a need for a critical evaluation of the status of the thermodynamic data base currently used. The following topics will be addressed:

- the ionic strength dependence of equilibrium constants
- some recent equilibrium data in actinoid - $\text{CO}_2(\text{g})$ - H_2O systems
- comparisons between laboratory and field data

The importance of the redox-potential of ground water for the corrosion of metallic materials and the migration of radionuclides is well known. The quality of redox-data obtained in the field in comparison with laboratory measurements will be briefly discussed.

D5.1

HYDROTHERMAL INTERACTIONS OF CEMENT OR MORTAR WITH ZEOLITES OR MONTMORILLONITES. Sridhar Komarneni and Della M. Roy, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

Concretes, mortars, grouts, clays and/or zeolites are candidate borehole, shaft or tunnel plugging materials for any nuclear waste repository. Interactions between these plugging materials may take place under mild hydrothermal conditions during the life of a repository. Class H cement or mortar (PSU/WES mixture) was reacted with two montmorillonites or clinoptilolite or mordenite at 100° and 200°C for different periods under a confining pressure of 30 MPa. The solid reaction products were characterized by x-ray powder diffraction after the hydrothermal treatments. When zeolites were in contact (no intimate mixture) with Class H cement, they did not seem to alter but clinoptilolite altered to analcime and mordenite became poorly crystalline in the presence of mortar (containing NaCl) at both 100° and 200°C. When cement or mortar was intimately mixed with zeolites or montmorillonites and reacted hydrothermally, the reaction resulted in the formation of Al substituted tobermorite (11Å type) in all cases (this type of reaction is expected at the interface) at both 100 and 200°C. The mechanism of tobermorite formation includes the decomposition of zeolites or montmorillonites in the presence of the alkaline (pH = 12) cement or mortar and recrystallization to form Al substituted tobermorite. Cesium sorption measurements in 0.01N CaCl_2 on the reaction products revealed that selective Cs sorption increased in most cases, even though little or none of the original zeolites and montmorillonites remained in the products. For example, Cs sorption K_d (ml/g) increased from 80 in the untreated mortar + Ca montmorillonite mixture to 1700 in the interaction product which is Al substituted tobermorite. Thus, we discover here that Al substituted tobermorite has good selectivity for Cs.

D5.2
K-CA and K-AR DATING OF SYLVITE FROM THE SALADO FORMATION, NEW MEXICO AND IMPLICATIONS FOR THE STABILITY OF EVAPORITE MINERALS. J. D. Obradovich, M. Tatsumoto, U.S. Geological Survey, Denver, CO 80225, O. K. Manuel, University of Coilla, MO 65401, H. Mehnert, U.S. Geological Survey, Denver, CO 80225, M. Domenick and T. Wildman, Colorado School of Mines, Golden, CO 80401

Because of a number of favorable properties, salt in the Permian Delaware Basin in southeastern New Mexico would seem to provide a promising repository for the disposal of nuclear waste. Salt, however, is soluble and petrographic and other evidence suggest that much of the salt has been dissolved and removed. One point of view holds that this process took place in pre-Cretaceous time, whereas the other contends that it is a continuing process that has operated in very recent time. A resolution of the time of dissolution and recrystallization is of prime importance.

Prior radiometric dating supports the view that dissolution and recrystallization occurred in pre-Cretaceous time based on a mean age of 208 m.y. (K-Ar) on pure polyhalite and isochron ages of 214 m.y. and 126 m.y. (Rb-Sr) on mixtures of sylvite with halite, polyhalite, langbeinite, and anhydrite. An age of 250 m.y. (K-Ar) was obtained on langbeinite, and ages of 140 m.y. and 150 m.y. (K-Ar) on langbeinite-sylvite mixtures. The younger ages have been interpreted to be the result of partial loss of argon from sylvite by diffusion.

Age determinations (K-Ca, supported by K-Ar) on sylvite samples from the Salado formation give ages in the range 33-37 m.y. and 2-4 m.y. Contrary to the earlier work, dissolution and recrystallization have taken place since the Cretaceous and a relatively recent geologic time. Reservations regarding the long term stability of salt may be in order.

D5.3
RADIOACTIVE WASTE ISOLATION: SIGNIFICANCE OF MICROCRACKS. Gene Simmons and Louis Russo, Dept. of Earth and Planetary Sciences, M.I.T., Cambridge, MA 02139.

Microcracks in crystalline rocks contain significant information for the siting of high-level radioactive waste storage facilities. The open cracks in core control several physical properties commonly measured on the core, are often produced in drilling or subsequent handling of the core, and can be distinguished from the cracks that were open *in situ* on the basis of the pressures at which they close mechanically and by their general appearance. Microcracks that were partially open *in situ* and also affect physical properties can be recognized by the presence of mineral bridges 0.1 micron diameter that extend across the opening and are often disturbed by drilling and sample preparation. Previous migration of uranium (U) and rare earth elements (REE) through microcracks can be readily recognized.

In the Conway (NH) granite, U has been deposited in carbonate-sealed cracks and adsorbed on clays that resulted from extensive alteration of amphiboles in the vicinity of the cracks. Uraninite fills some microcracks in this granite. In the Sherman (WY) granite, microcracks contain U and an REE+U mineral. This granite is altered over a large area, U-bearing microcracks are widespread, and the U migration probably occurred when the rock was at pressures of a few kb and temperature in the range 150-500°C. This migration probably occurred in the Precambrian and the uranium is thought to have been immobilized since then.

These results suggest that altered plutonic igneous rocks that contain carbonate and clay minerals and few open microcracks may be the more desirable crystalline rock for siting of high-level rad waste facilities.

D5.4
RADIONUCLIDE MIGRATION INTO NATURAL FRACTURE SURFACES OF GRANITIC ROCK. B. Torstenfelt, T. Eliasson, B. Allard, K. Andersson, S. Höglund and U. Olofsson, Department of Nuclear Chemistry, Chalmers University of Technology, S-412 96 Göteborg, Sweden.

Fracture filling materials from water exposed natural fractures in granitic bedrock have been sampled from three different locations and characterized. Interactions with Cs, representing a non-hydrolyzed species, Am, representing a hydrolyzed species and TcO_4^- , representing an anionic species, were studied under the following conditions:

1. Batch-wise sorption studies on the natural fracture minerals as well as on pure minerals, considering the corresponding groundwater composition.
2. Batch-wise sorption studies on the major components of granite.
3. Uptake on macrosurfaces (ca 5 cm²) of samples taken from the natural fractures.
4. Radionuclide depth penetration through the coating of the fracture surfaces into micro fissures in the unaltered underlying granite.
5. Studies of the reducing capacity of the unaltered granite (TcO_4^- to Tc(IV)).

The possibility of radionuclide diffusion through the fracture surface coatings and penetration into micro fissures are discussed in particular.

D5.5
SALT CREEP DESIGN CONSIDERATION FOR UNDERGROUND NUCLEAR WASTE STORAGE. William T. Li and C. L. Wu, Bechtel National, Inc., P. O. Box 3965, San Francisco, CA 94119.

One of the methods of radioactive waste management receiving considerable attention today is the storage of radioactive waste in salt formations. The most significant mechanical property of the salt is that it creeps, therefore, in design of storage facilities in salt, the behavior of creep has to be carefully evaluated.

This report summarizes the creep consideration for the design of storage facilities in salt, describes the non-linear analysis method for evaluating the design adequacy, and presents computational results for the current storage design. The application of rock mechanics instrumentation to assure the appropriateness of the design are also discussed.

D5.6
MEGASCOPIC CONTROLS ON RADIONUCLIDE TRANSPORT IN GROUNDWATER FLOW SYSTEMS. Leslie Smith, Dept. Geological Sciences, Univ. of British Columbia, Vancouver, Canada and Frank Schwartz, Dept. of Geology, Univ. of Alberta, Edmonton, Canada.

The objective of this paper is to investigate how the gross geologic structure of a groundwater basin influences the reliability with which predictions of radionuclide

transport within a repository horizon can be made. A series of stochastic simulations are carried out to identify the sensitive of transport in the repository horizon to the hydraulic characteristics of surrounding stratigraphic units. Results show that in some cases, relatively large variations in breakthrough of radionuclides from the repository horizon can occur for what is a reasonable degree of uncertainty in specifying effective hydraulic conductivity values for surrounding units. Other cases are identified where transport predictions are not sensitive to perturbations in the hydraulic properties of certain units in the basin. Given physical and economic constraints in a site evaluation program, such analyses are valuable in providing guidelines for design of data collection networks.

5.7
EFFECT OF CHANNELLING IN A POROUS MEDIUM ON RADIONUCLIDE MIGRATION. A. K. Ray and J. Mair, Department of Chemical Engineering, University of Kentucky, Lexington, Kentucky, 40506-0046.

A model has been developed to study migration of radionuclides through a fractured porous medium. The porous medium is treated as an infinite medium containing multiple, parallel, unconnected channels; each channel is surrounded by a rock matrix. The two dimensional convective-diffusional equations with a radionuclide source have been solved for the system. At the channel/rock matrix interface, the concentration and flux were matched to obtain global concentration profiles. By means of a parametric study the effects of channel frequency and size distributions, and other major parameters are elucidated. Correlation technique for tracer test data with the model has also been discussed.

5.8
PERFORMANCE ANALYSIS OF A NUCLEAR WASTE REPOSITORY IN A BASALT GEOLOGY. R. G. Baca, S. Bensky, and N. W. Kline, Rockwell Hanford Operations, P. O. Box 800, Richland, Washington 99352.

A preliminary performance analysis that considered various release scenarios for a reference repository site in a basalt geology was conducted. Results from this long-term, post-closure performance analysis are presented and include groundwater pathlines, travel times, radiocontaminant concentrations, and fluxes. The predicted performance is compared with the technical criteria and regulations proposed by the federal agencies. This comparison gives a clear perspective on the waste isolation capability of deep basalt formations underlying the Hanford site in southeastern Washington.

5.9
STATISTICAL SITE CHARACTERIZATION: LESSONS FOR "REGULATORY GEOLOGY", Gregory B. Sacher, Associate Professor of Civil Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139.

Statistical methods and decision analysis have been applied to geological exploration since the late 1940's. Increasingly they have found use in geotechnical

engineering and underground construction, and they will most certainly be applied to characterizing repository sites. Beyond the obvious quantitative benefits of such methods, the search for logical guidance in exploration and site characterization has led to epistemological insights that may prove to be more valuable to regulatory decision making than mere numbers are. The principal results of site characterization are not physical records, but hypotheses on site geology and degrees of credibility attached to them. These results are necessarily subjective: Formal analysis leads to logical consistency among prior and posterior probabilities, and among degrees of support, but cannot alter the fundamental nature of inductive reasoning.

The paper highlights some of the lessons learned from statistical strategies in geological and geotechnical exploration, and draws conclusions on their meanings for repository risk analysis and licensing decisions. In repository risk analysis, which involves geological as well as engineering predictions, care must be taken to identify sources of evidence actually used and their statistical predictiveness. Improved means of incorporating professional opinion in these analyses will be necessary, and is being worked on by several groups and in several different areas of waste disposal.

5.10
THE SCEPTER WASTE PACKAGE SUBSYSTEM COMPUTER MODEL FOR PERFORMANCE ASSESSMENT. Aviva Brecher, Arthur D. Little, Inc., Acorn Park, Cambridge, MA 02140, and J. F. Pearson, Intera Environmental Consultants, Inc., 11999 Katy Freeway, Houston, TX 77079.

The Waste Package Performance Assessment (WAPPA) code has recently been completed, verified and documented within the SCEPTER program for ONWI. The code is intended to serve as a tool for evaluating both the relative and the absolute performance of waste package design concepts, for guiding or selecting a preliminary design amongst conceptual designs, and for support of licensing activities. This is a systems model of the gradual degradation of physical and retardation barriers comprising a nuclear waste package under repository isolation conditions in various geologic media. The model describes and predicts the temporal and spatial extent of loss of containment capability for each barrier and for the entire waste package, on timescales of up to 10^6 years. The output describes the residual state of integrity of waste package components as a function of time and quantifies heat and radionuclides fluxes to the repository. This presentation discusses the assumptions and limitations of the current waste package system model and details its logic structure and input/output specifications of interest to this users' community.

The model includes five distinct degradation process models (Radiation, Thermal, Mechanical, Corrosion, and Leaching) which are driven both internally by waste decay and externally by repository stress and fluids and are coupled via state variables and descriptors. The modeling approach to degradation adopted is barrier-integrated and process-sequential, allowing modification of the entire waste package by the five process models sequentially within each time step.

D5.11
LONG-TERM THERMOMECHANICAL AND THERMOHYDROLOGICAL FACTORS CONTROLLING THE OPTIMAL DESIGN OF A NUCLEAR WASTE REPOSITORY, J. S. Y. Wang, D. C. Mangold, and C. F. Tsang, Earth Sciences Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720.

Surface uplift and buoyancy flow are two of the major long-term, far-field perturbations to the geologic formations around a nuclear waste repository. The allowable surface uplift has been accepted in the literature as a criterion limiting the repository waste loading density. On the other hand, it has also been recognized in generic modeling studies that the buoyancy distortion of the ambient groundwater flow around a repository is a major mechanism for radionuclide transport. However, the buoyancy considerations have yet to be quantified for the purpose of optimal design of a radioactive waste repository. The present paper investigates both the thermo-mechanical and thermohydrological factors that may limit the waste loading design.

We suggest the possibility of using the buoyancy gradient when compared with the ambient regional (horizontal) gradient as a measurable thermohydrologic factor that controls the design of waste loading. Thus for a given repository we may require its design to be such that the thermomechanically induced surface uplift be below a given critical value and also the thermohydrologic buoyancy gradient be below the ambient hydraulic gradient times a given multiplicative factor.

This study indicates that the buoyancy gradient could in some cases become the main controlling factor. To decrease the buoyancy effects, much lower values of waste loading density should be considered in the design of the waste repository. The reliability of these results and their implications will be discussed in the paper.

D5.12
AN ANALYSIS OF WASTE PACKAGE BEHAVIOR FOR HIGH-LEVEL WASTE. Margaret S. Chu, James E. Campbell*, Stephen Stuckwisch and Malcolm D. Siegel. Sandia National Laboratories, Fuel Cycle Risk Analysis Division, Albuquerque, NM, 87185.
*INTERA Environmental Consultants, Inc., Lakewood, CO, 80215.

A sensitivity analysis for waste package behavior was performed to examine the relative importance of radionuclide containment period and release rate in comparison with the Draft EPA Standard, 40CFR191. A time and temperature dependent leach rate of boro-silicate glasses from experimental data was used to describe the release behavior of radionuclides from a waste matrix. Failure behavior of canisters was described by the sum of an early failure mode and corrosion failure (wear-out) mode. Realistic ranges of values were assigned to each parameter in the analysis. Integrated releases of radionuclides over 10,000 years were calculated for different canister design lives. Results were compared with the Draft EPA Standard.

D5.13
THE FORMULATION OF AN INTEGRATED PHYSICO-CHEMICAL-HYDROLOGIC MODEL FOR PREDICTING WASTE NUCLIDE RETARDATION IN GEOLOGIC MEDIA. A. B. Muller, Sandia National Laboratories, Albuquerque, NM., D. Langmuir, Colorado School of Mines, Golden, CO., and L. E. Duda, Sandia National Laboratories, Albuquerque, NM.

The inability of empirical single value elemental R_d values to model radionuclide retardation in natural ground-water systems has demonstrated the need for develop-

ing a comprehensive integrated phenomenological model of the physicochemical and hydrologic mechanisms involved in retardation. The model must account for: (1) radioactive decay, (2) chemical precipitation/dissolution of bulk phases, (3) chemical substitution reactions, (4) isotopic exchange reactions, (5) cation and anion exchange, (6) specific sorption, (7) diffusion into adjacent flow paths, (8) diffusion into fluid not involved in flow, (9) diffusion into the solid matrix and (10) ultrafiltration. The formulation of such a model, based on a reaction-path-simulation code with modifications for adsorption processes (by a surface-complexation site-binding model) and diffusional processes is proposed. The coupling of the physicochemical portion of this model to a ground-water transport code can be achieved by alternately iterating through the chemical and physical portions of the code during each time step. Sensitivity analysis used to determine dominant species, dominant retardation mechanisms and temporal and spatial homogeneities in the flow system will allow the integrated model to be reduced to a manageable form.

D5.14
PLUTONIUM SPECIATION IN SELECTED GROUND WATERS AND ITS IMPLICATIONS FOR NUCLEAR WASTE DISPOSAL. Terry F. Rees, Jess M. Cleveland, and Kenneth L. Nash, U.S. Geological Survey, Lakewood, CO 80225.

Plutonium speciation has been studied in ground waters associated with rock types proposed as possible host rocks in the U.S. Department of Energy Environmental Impact Statement: Management of Commercially Generated Radioactive Waste. In the presence of high concentrations of sulfate, essentially all of the plutonium is rendered insoluble, regardless of temperature and starting oxidation state. In waters with significant free-fluoride concentrations, plutonium remains in solution, presumably due to formation of soluble fluoro complexes. In waters containing both sulfate and fluoride, the results are less predictable, especially if di- or trivalent cations are present that might sequester the fluoride. Dissolved oxygen, ionic strength, particulate, and carbonate ion concentrations have little effect. These results have potential significance in establishing site-selection criteria for radioactive-waste repositories. All other factors being equal (geologic, hydrologic, engineering, economic and political), host rocks whose associated ground waters contain high concentrations of sulfate ion and low concentrations of fluoride ion should be preferable for location of waste repositories.

D5.15
STUDIES OF THE BONDING OF COBALT ON FELDSPAR SURFACES USING X-RAY PHOTOELECTRON SPECTROSCOPY (XPS), SCANNING ELECTRON MICROSCOPY (SEM), AND SCANNING AUGER MICROSCOPY. D. L. Perry, Leon Tsao, and Kenneth A. Gaugler, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720

The surface chemistry of K-feldspar resulting from its interaction with aqueous solutions of cobalt(II) and cobalt(III) has been studied using x-ray photoelectron spectroscopy, scanning electron microscopy, and scanning Auger microscopy. The feldspar was reacted under varying experimental conditions, including elevated temperatures, concentrations, and pH. The concentration of elements involved in the reaction system was determined. Depth profiling

to the mineral bulk was performed using argon ion sputtering in conjunction with x-ray photoelectron and scanning Auger microscopy; the variable angle take-off approach was also used to look at surface enriched species. X-ray photoelectron binding energies for cobalt, oxygen and other elements were used to determine the chemical species formed on the feldspar surfaces. Both the oxidation and electronic states of the cobalt on the feldspar surface were studied using binding energies and core level (2p) satellite structure.

3.16
STUDIES ON SPECIATION OF AMERICIUM, TECHNETIUM AND NEPTUNIUM IN SIMULATED VITRIFIED-WASTE LEACHATES. G. Bidoglio, A. De Plano and A. Chatt, Commission of the European Communities, Joint Research Centre, Ispra Establishment, Radiochemistry and Nuclear Chemistry Division, I-21020 Ispra (Varese), Italy.

The safety of long-term storage of vitrified HAW in geological repositories is being evaluated by designing both experiments and models. In order to determine the dominating form of radionuclides, speciation studies have been carried out. Solvent extraction methods have been developed to characterize Am(III) complexes with bicarbonate and carbonate ions at ground water concentration levels; $\text{Am}(\text{CO}_3)_2$ has been observed to be the most important complex while the formation of $\text{Am}(\text{OH})(\text{CO}_3)_2^-$ has been indicated, and their stability constants have been calculated. These results are in agreement with the observation of presence of anionic species in simulated vitrified-waste leachates and of their mobility through the glauconitic sand columns. Precipitation and liquid-liquid partition methods, and electromigration techniques have been employed to study speciation of Tc under oxic and anoxic conditions (about 0 ppb dissolved oxygen); it appears that Tc(IV) is not stabilized by complexation with constituents of ground water. Similar studies on Np are being carried out.

3.17
LEAKING BEHAVIOR OF TRITIUM FROM NEUTRON IRRADIATED BOROSILICATE GLASS Yoshimasa Yamamoto, Kazuo Kitagawa, Mechanical Engineering Research Laboratory, Kobe Steel, Ltd., 651 Kobe, Japan. Hisao Atsumi, Shinsuke Yamanaka and Masanobu Miyake, Department of Nuclear Engineering, Osaka Univ., 565 Osaka, Japan.

Iron containing materials such as borosilicate glass and boron carbide are widely used as neutron absorbing elements in LWRs and FBRs. In case treatment and/or disposal of spent neutron absorbing materials is considered, information on the behavior of tritium, which is produced by nuclear reactions of boron, is important from the viewpoint of reducing the release of radioactivity to the circumstances.

Specimens of borosilicate glass were irradiated in a materials testing reactor at a neutron fluence and temperature of 10^{19} - 10^{20} nvt and below 250°C respectively. Tritium released from the specimen was determined by means of a specially designed sampling system and liquid scintillation counter. The amount of tritium in the order of 10^{-8} STPcc can be determined according to its chemical form of tritiated water or tritiated hydrogen with the efficiency above 97%, which was confirmed by the preparatory examination using standard tritium gas.

Temperature and time dependence on the releasing behavior of tritium from the specimen was obtained in the temperature range of 200-700°C. The amount of tritium released as tritiated water increases with temperature for a fixed sampling period, while the amount released as tritiated hydrogen reaches a constant value above 300°C. The total amount of tritiated water from the specimen is much greater than that of tritiated hydrogen. The releasing behavior of tritium was analyzed by diffusion theory and surface reactions.

D6.2
LABORATORY INVESTIGATIONS OF GROUND WATER INTERACTIONS WITH BURIED RADIOACTIVE SODIUM WASTES AND ATTENDANT RELEASE AND MIGRATION OF TRITIUM IN ADJACENT SOIL, G. Subbaraman and O.P. Steele III, Rockwell International Energy Systems Group, Canoga Park, CA 91304; and L.C. Witbeck and C.S. Abrams, Argonne National Laboratory, Argonne-West, Idaho Falls, ID 83401.

Shallow land burial of sodium bearing low-level wastes requires an assessment of safety consequences relating to (a) ground water interactions with the residual sodium in the event of a breach of containment, and (b) release of tritium dissolved in sodium along with hydrogen, both of which will permeate through adjacent soil. This paper describes tests and evaluations being performed to establish a data-base on safety consequences of burying low-level Radioactive Sodium Wastes (RSW). This data-base can be utilized in establishing the amounts of sodium that may be tolerated in burying RSW and related criteria.

D6.3
THE FUNCTIONAL DEPENDENCE OF LEACHING ON THE SURFACE AREA TO SOLUTION VOLUME RATIO, Albert J. Machiels and Claudio Pescatore, University of Illinois, Nuclear Engineering Program, Urbana, IL 61801.

In leaching experiments, the ratio of the exposed surface area of a glass specimen to the volume of aqueous corrosion solution is an important parameter, the effect of which has been experimentally investigated in a number of studies. The results have strong implications about the durability of glass in a repository since considerable cracking of the waste monolith is to be expected. Although the experimental investigations have uncovered a range of behaviors, expected as well as unexpected, the data can usually be rationalized by looking at the functional dependence of the leach rate and/or cumulative leaching on the surface area to solution volume ratio.

The procedure is illustrated by looking at examples of leach testing under static conditions. The functional dependence of cumulative mass losses from a borosilicate glass is predicted by using a leach model taking into account the waste form, leachant, and interface properties. Results are presented and compared to existing data.

R CHEMISTRY AT COMMERCIALY OPERATED LOW-LEVEL RADIOACTIVE WASTE DISPOSAL SITES. R. F. Pietrzak, R. Dayal, K. S. Czyscinski,** and A. J. Weiss, Oak Ridge National Laboratory, Upton, NY 11973.

Leachates from the disposal trenches of low-level waste sites were analyzed for their inorganic, organic, and radionuclide contents. Since oxidation of the trench waters occur during their movement along the groundwater flow path, experiments were performed to measure the chemical and physical changes which occur in these waters upon oxidation. The temporal stability of organic complexes (EDTA, NTA, etc.) with radionuclides in anoxic trench waters was also studied. This information is important in the interpretation of field data and modeling calculations.

Work was carried out under the auspices of the U.S. Nuclear Regulatory Commission. Contact address: Office of Nuclear Waste Isolation, Battelle Project Management Division, 505 King Avenue, Columbus, OH.

TESTS OF SIMULATED LOW-LEVEL TRANSURANIC WASTE FORMS CONTAINING TRANSURANIC ISOTOPES. Jane M. Welch, Claude W. Sill, and John E. Flinn, EG&G Idaho, Inc., P.O. Box 1625, Idaho Falls, ID 83415

Simulated waste forms that might be produced by slagging pyrolysis incineration of low-level transuranic (TRU) wastes stored at the Idaho National Engineering Laboratory (INEL) have been fabricated containing the transuranic isotopes, ^{239}Pu , ^{241}Am , and ^{244}Cm at levels up to a microcurie per gram of each of the isotopes.

Leach tests were performed using frit and monoliths of average INEL TRU waste composition, portland cement monoliths made with frit as aggregate, and monoliths of vitrified INEL soil, and simulated Rocky Flats sludge.

Leach tests were performed at 90°, 70°, 40°, and 25°C in deionized water for up to 364 days. Leachates were analyzed for the TRU elements by alpha spectrometry.

Generalizations that can be made from the data are:

- 1) Frit in cement and vitrified sludge, the waste forms that produce leachates with the highest pHs (>11), have the lowest TRU leach rates, 10^{-4} g/m²·d at 90°C.
- 2) Neptunium has a higher leach rate than the other three TRU elements by as much as two orders of magnitude.
- 3) The TRU leach rates display little temperature dependence.

D6.6

FORMULATION AND DURABILITY OF TAILORED CEMENTITIOUS HOSTS APPLIED TO TRU WASTE GENERATED AT THE ROCKY FLATS PLANT. J. H. Kessler, G. C. Rogers, L. R. Dole, and M. T. Morgan, Oak Ridge National Laboratory, P. O. Box X, Bldg. 3508, Oak Ridge, TN 37830.

This paper discusses the development of cementitious grout formulations at the Oak Ridge National Laboratory which immobilize up to 50% of Rocky Flats Building 374 simulated startup TRU waste. Over 80 trial grout mixes were tested, while minimizing the water content and maximizing the waste loading. Also, the effects of water reducers and set regulators upon workability and set time were examined.

Durability and physical property measurements were performed on formulations which are compatible with standard processing equipment. The densities, porosities, compressive strengths, and leachabilities of these solids are reported. Compressive strengths of the samples were found to increase by as much as 69% after the samples had been exposed to leachants for 28 days at 90°C.

D6.7

LOW LEVEL RADWASTE ENCAPSULATION IN PORTLAND CEMENT. M. C. Skriba and E. E. Smeltzer¹, and G. A. Cefola², ¹Westinghouse Research and Development Center, 1310 Beulah Road, Pittsburgh, PA 15235, and ²Westinghouse Nuclear Energy Systems Division, Haymaker Road, Monroeville, PA 15230.

A new approach to radwaste encapsulation in Type I Portland cement has been demonstrated at plant scale using a patented high speed, high shear mixer to achieve solidification. Solutions and suspensions of up to 60% boric acid, 22% sodium sulfate, 40% ion exchange resin, and 10% oil have been produced as homogeneous solids with no free water. Crush strengths were generally high and the ion exchange mixes were stable under rewet conditions.

D6.8

REMOVAL OF ACTINIDES FROM WASTE SOLUTION BY FERRITE TREATMENT. Thomas E. Boyd and Robert L. Kochen, Rockwell International, Chemical Research, Rocky Flats Plant, Golden, Colorado 80401.

Using the ferrite waste treatment method, plutonium concentrations in simulated Rocky Flats Plant waste were lowered from 10^{-4} g/l to around 10^{-9} g/l in one treatment step. At the same time, only 3.0 grams of solids were generated per liter of waste solution treated, as compared with 15.7 g/l now produced in present Plant operations at Rocky Flats. The results of these studies on both simulated and actual Rocky Flats Plant waste will be discussed.

D6.9

THEORETICAL PREDICTIONS FOR CONTINUOUS SLURRY FEEDING OF A GLASS MELTER. K. R. Routh, E. I. duPont de Nemours, Savannah River Laboratory, Aiken, S.C. 29808.

Radioactive defense waste currently stored at the Savannah River Plant in liquid form is to be immobilized by incorporation into a borosilicate glass. The glass melter for this process will consist of a refractory lined tank enclosed in a water-cooled steel vessel and fed by an aqueous slurry of glass frit plus radioactive waste. As an aid to understanding the melting process and scaling data from small melters, conservation principles were used to develop equations for predicting the melt rate for a feedpile of arbitrary radius plus additional information regarding feedpile behavior and energy consumption.

* The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00001 with the U. S. Department of Energy.

D6.10

SYNROC PRODUCTION USING A FLUID-BED CALCINER.* F. Jay Ackerman, John Z. Grens, Pat Peters and John H. Campbell, Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94550.

SYNROC is a titanate-based ceramic material currently being developed for immobilization of high-level nuclear reactor wastes in solid form. The use of a fluid bed for SYNROC production will permit slurry drying, calcining and redox to be carried out in a single unit. We present results of SYNROC fluid-bed studies from two 10-cm diameter fluid-bed units; the Idaho Exxon internally-heated fluid-bed unit and the externally-heated unit constructed at Lawrence Livermore National Laboratory.

The beds have been operated over a range of temperature, feed rate, fluidizing rate and redox conditions. Our tests indicate that high density, uniform particle-size SYNROC powders can be produced which facilitate the densification step and give HIP or HUP parts with dense, well-developed phases and good leaching characteristics.

*Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under contract number W-7405-ENG-48 for presentation at the Sixth International Symposium on the Scientific Basis for Radioactive Waste Management, November 1-4, 1982.

D6.11

EFFICIENT PARTICULATE SCRUBBER FOR RADIOACTIVE MELTER OFF-GAS APPLICATIONS,* G. T. Wright, E. I. duPont de Nemours & Co., Savannah River Laboratory, Aiken, South Carolina, 29808

Operation of joule heated, continuous slurry fed melters have demonstrated that off-gas aerosols are generated by entrainment of feed slurry and vaporization of volatile species from the melt. Effective off-gas stream decon-

tamination for these aerosols can be obtained by utilizing a suitably designed and operated wet scrubber system. Results are presented for performance tests conducted with an air aspirating-type venturi scrubber processing a simulated melter off-gas aerosol. Mass overall removal efficiencies ranged from 99.5 to 99.8%. Details of the testing program and applications for melter off-gas system design are discussed.

D6.12

ELECTRIC ARC FURNACE MELTING OF SIMULATED TRANSURANIC WASTES. R. H. Nafziger and L. L. Oden, Albany Research Center, Bureau of Mines, P.O. Box 70, Albany, OR 97321.

As part of an interagency agreement between the Bureau of Mines, U.S. Department of the Interior, and the U.S. Department of Energy through its contractor, EG&G Idaho, Inc., the Bureau has conducted six melting tests at its Albany Research Center to assess the feasibility of melting transuranic wastes. The tests were conducted with simulated wastes in a conventional refractory-lined electric arc furnace. Charge materials included concrete, soil, metal, wood, sludge, cement, and polyethylene mixed in various proportions in both unburned and partially incinerated forms. The investigation showed that it is possible to melt these materials in a 1-ton conventional electric arc furnace provided that suitable fluxes are added to condition the highly acid slags. However, the electric arc furnace cannot be considered as an incinerator. The molten slags can be poured into 55-gallon steel barrels having a 1/4-inch steel plate on the bottom. The concrete and sludge materials require the most energy for melting. The highest electrode consumption occurred when the sludges were melted. A high alumina-chrome refractory is satisfactory for use in melting these wastes. All slags were tapped from the furnace satisfactorily. Off-gases and particulates from all of the tests were sampled and analyzed. Results show that the greatest amount of particulate matter in the offgas stream was obtained when sludges were melted, followed by incinerated wastes.

It is recommended that if a conventional electric arc furnace is used to melt this material that it 1) be fed slowly to prevent excessive fumes and flames, and 2) be melted continuously so that startup problems are minimized and the material can be melted more efficiently.

D6.13

RA-226 RADIOASSAY OF SOIL AND TAILINGS. Carmen S. Sabau, Donald R. Rayno, Norbert D. Kretz and Phillip W. Zelle, Argonne National Laboratory, 9700 S. Cass Ave., Argonne, IL 60439.

Studies of inactive uranium tailings piles have shown that quantities of tailings sands containing Ra-226 and other radionuclides may be dispersed by wind and water erosion, causing contamination of adjacent areas. To conduct an effective cleanup operation, it is necessary that boundaries of contamination be well defined. Data from surface gamma-ray surveys made under the Measurement/Monitoring Program, as part of the Uranium Mill Tailings Remedial Action (UMTRA), are first used to define a general outline of the contaminated area. Then, data from portable scintillometer surveys and from sealed-can gamma-ray analysis performed on soil samples are used to define more specifically the periphery of Ra-226 contamination. These field measurements are supported by radiochemical analysis of randomly selected samples. Because of its adaptability

to the widely varying chemical composition of the material in these samples, a complexometric leaching procedure using the complexing agent EDTA is used for the analysis of Ra-226. With this procedure, natural concentrations of Ra-226 in soil (~ 1 pCi/g) can be measured routinely, while the potential limit of detection is in the 0.1-0.5 pCi/g range. Details of the method, which includes leaching of radium followed by radon de-emanation, are described. Comparative data for various soil and tailings samples are presented.

D6.14
ANALYSES OF SOILS AT COMMERCIAL RADIOACTIVE WASTE DISPOSAL SITES.*
P. L. Piccolo, C. E. Shea, and R. E. Barletta, Brookhaven National Laboratory, Upton, NY 11973.

Brookhaven National Laboratory, in order to provide technical assistance to the NRC, has measured a number of physical and chemical characteristics of soils from two currently operating commercial radioactive waste disposal sites; one at Barnwell, SC, and the other near Richland, WA. Soil samples believed to be representative of the soil that will contact the buried waste were collected and analyzed. Earth resistivities (field measurements), from both sites, supply information to identify variations in subsurface material. Barnwell soil resistivities (laboratory measurements) range from 3.6×10^5 ohm-cm to 8.9×10^4 ohm-cm. Soil resistivities of the Hanford sample vary from 3.0×10^5 ohm-cm to 6.6×10^3 ohm-cm. The Barnwell and Hanford soil pH ranges from 4.8 to 5.4 and from 4.0 to 7.2, respectively. The pH of a 1:2 mixture of soil to 0.01 M CaCl₂ resulted in a pH for the Barnwell samples of 3.9 ± 0.1 and for the Hanford samples of 7.4 ± 0.2 . These values are comparable to the pH measurements of the water extract of the soils used for the analyses of soluble ion content of the soils. The exchange acidity of the soils was found to be approximately 7 mg-eq per 100 g of dry soil for clay material from Barnwell, whereas the Hanford soils showed an alkaline reaction. Aqueous extracts of saturated pastes were used to determine the concentrations of the following ions: Ca²⁺, Mg²⁺, K⁺, Na⁺, HCO₃⁻, SO₄⁼, and Cl⁻. The sulfide content of each of the soils was measured in a 1:2.5 mixture of soil to an antioxidant buffer solution. The concentrations of soluble ions found in the soils from both sites are consistent with the high resistivities.

*Work carried out under the auspices of the U.S. Nuclear Regulatory Commission.

D6.15
PROCESS CHARACTERISTICS FOR IMMOBILIZING AND COMPACTING KRYPTON-85, A.B. Christensen, J.A. DelDebbio, D.A. Knecht, and J.E. Tanner, Exxon Nuclear Idaho Co., Inc., PO Box 2800, Idaho Falls, ID 83401.

Previous work showed that up to 20 wt% of ⁸⁵Kr can be immobilized at high pressures (1000 atm) and temperatures (700°C) in commercial zeolite 5A. Further work has been carried out to consolidate the Kr containing zeolite to a hard dense waste form suitable for transport and final disposal. The test results showed that hot isostatic pressing of the zeolite with a glass binder at 600 to 700°C and 300 to 400 atm pressure yielded such a waste form, and did not decrease gas loading or increase gas leakage. The glass frit contained by weight 57% SiO₂, 17% B₂O₃, 15% Na₂O, 10%

Li₂O, and 1% CuO. Higher compaction pressure yielded a less friable waste form, but forced some of the encapsulated gas out of the zeolite. A full-scale conceptual design was developed for both Kr encapsulation and zeolite compaction, using a 16-L work zone hot isostatic press (HIP) at 300 cycles per year. The process exhibits simplicity of operation and product handling while producing a dense solid waste form containing up to 50 m³ STP Kr per m³ solid.

D7.1
THE OAK RIDGE NATIONAL LABORATORY HYDROFRACTURE PROCESS FOR THE DISPOSAL OF RADIOACTIVE WASTE.* H.O. Weeren and E.W. McDaniel, Oak Ridge National Laboratory, Oak Ridge, TN 37830.

The hydrofracture process is a method of waste disposal currently in use at Oak Ridge National Laboratory for the permanent disposal of locally generated radioactive waste solutions. In this process, the waste solution is mixed with a solids blend of cement and other additives; the resulting grout is then injected into an impermeable shale formation at a depth of 200 to 300 m (700 to 1000 ft). The grout sets a few hours after completion of the injection, fixing the radioactive waste in the shale formation.

The process has been in operational use since 1966 and has been used to dispose of more than 8×10^6 L (2×10^6 gal) of waste grout containing over 600,000 Ci of radionuclide activity. A description of the process and its operational history are given. Operational disposal costs on the order of 25¢/L (\$1/gal) are reported. A new disposal facility has been built, and the first operational injections were made in the summer of 1982.

*Research sponsored by Radioactive Waste Management Program, U.S. Department of Energy under contract W-7405-eng-26 with Union Carbide Corporation.

D7.2
AN EVALUATION OF ALTERNATIVE TRU TECHNOLOGIES. G. Bida and D. R. Mackenzie, Brookhaven National Laboratory, Upton, NY 11973.

As part of a NRC sponsored technical support program in the development of nuclear waste form criteria, three main areas of transuranic waste management have been evaluated: immobilization processes and waste forms proposed for ultimate geologic disposal of TRU waste; management of TRU waste by decontamination; and potential problems associated with gas generation by certain TRU wastes. The objectives of this evaluation were (1) to provide NRC with the information necessary for the licensed geologic disposal of commercial TRU waste, (2) to recommend appropriate courses of action that provide support for proposed regulations and criteria and lead to the safe isolation of commercial TRU waste, and (3) to identify research needed to reduce significant uncertainties. In the evaluation, TRU waste forms are reviewed in terms of their ability to meet the controlled radionuclide release rate criterion that the proposed NRC rule (10 CFR 60) places on the engineered system, in order that they could be evaluated as creditable engineered barriers. The two classes of metallic waste which are candidates for decontamination treatment are Zircaloy cladding hulls, and failed facilities and equipment. Decontamination methods are addressed with regard to their ability to remove contamination to a level below the 10 nCi/g TRU limit. Other important factors are the volume reduc-

on achieved and compatibility of the secondary waste streams with acceptable waste forms. Gas generation by combustible TRU wastes and cast hydraulic cement containing TRU isotopes is discussed and its potential for damage to a geologic repository is considered. Exclusion of combustible TRU waste from repositories is recommended. Conclusions are drawn about the suitability of various waste forms. Recommendations are made regarding further work needed in the development of specific TRU waste forms.

3. **USE OF POISONED LAND/INLAND SEAS FOR LOW LEVEL RADIOACTIVE WASTE DISPOSAL.** D.G. Locking, R.A. Vogler, Univ. New Mexico, Albuquerque, NM 87131, and J.J. Cohen, Science Applications, Inc., Pleasanton, CA 94556.

Successful retention of low level radioactive wastes (LLRW) requires isolation geomedial. Volumetrically, LLRW is large compared to other radioactive wastes, and areas to store these large volumes are necessary. Naturally occurring poisoned areas, including some saline lake-playa areas, have been investigated to test their suitability for LLRW disposal consideration. These natural areas are actually laboratories from which one can determine the consequences of the gradual accumulation of many elements, including potentially hazardous elements. Once these areas have been so studied, they can be explored for the consequences of storing low level radioactive wastes in them. In many parts of the arid western southwestern United States, playa-bolson areas have been poisoned by gradual accumulation of potentially dangerous elements such as Hg, As, Sb, U, Co, Ag, Fe, Zn, Pb, Be, W and others. The sources of these elements can sometimes be related to mining activities, in other instances the source is the surrounding rocks. Regardless of source, these elements are stored in the closed basin environments and often render the area unsuitable for plant and animal life. Since many of these playa-bolson areas are in very isolated areas and do not possess economic accumulations of the elements concentrated therein, their natural toxicity makes them attractive for possible LLRW siting. For these poisoned areas to be suitable for LLRW disposal, their closed system integrity must be maintained. The ratio of precipitation to evaporation is often only 1:10 or so, and the elements found in the closed basins increase in abundance as a function of the degree the rocks weathered. Study of transport mechanism for several elements in the playa-bolson environment has been undertaken in the Estancia Valley, NM in order to investigate possible violation of closed system conditions. The work is to investigate retention of major elements and all minor elements studied. These results suggest that the poisoned lands in the more arid parts of Nevada, Utah and other States should receive direct investigation for LLRW disposal.

BEHAVIOR OF HAZARDOUS CONTAMINANTS IN VITRIFIED SOIL PRODUCED BY IN SITU VITRIFICATION. C.L. Timmerman, J.M. Rusin, and R.O. Lokken, Pacific Northwest Laboratory, Richland, WA 99352.

Abstract has been withdrawn.

D7.5

CERAMIZATION OF INORGANIC ION EXCHANGERS LOADED WITH NUCLEAR WASTE INTO RED CLAY TILES. J. Lehto, O.J. Heinonen and J.K. Miettinen, University of Helsinki, Department of Radiochemistry, Unioninkatu 35, 00170 Helsinki 17, Finland, Technical Research Centre of Finland, Reactor Laboratory, Otakaari 3A, 02150 Espoo 15, Finland

A new method to ceramize inorganic ion exchangers loaded with nuclear waste has been developed. It is simpler and cheaper than the methods used previously e.g. hot pressing. The inorganic ion exchangers, sodium titanate and ZrO_2 , were turned into final ceramic waste form by mixing them with a Finnish red clay in weight ratio 1:4 at maximum. The tiles moulded from the wet, bakeable mixture were ceramized at 1020-1060 °C. The leaching rates of Sr, Cs and Co from the tiles determined by ISO-test were after six months of leaching 10^{-6} - 10^{-7} g/cm²/d, in decreasing order. Mechanically the tiles are very durable: flexural strengths were in the range of 25-45 meganewton per square meter.

D7.6

GEOPHYSICAL MODELING OF A LOW-LEVEL NUCLEAR WASTE DISPOSAL SITE. Keith A. Horton, Louis Isaacson, and Rexford M. Morey; GEO-CENTERS, INC., 320 Needham Street, Newton Upper Falls, MA 02164.

Geophysical measurements can, depending on the techniques employed, be used at low-level nuclear waste disposal sites to map the subsurface stratigraphy, identify interfaces, locate objects and targets, and aid in selecting locations for obtaining sample cores and placing monitoring wells. The geophysical techniques of ground penetrating radar (GPR) and electrical resistivity (ER) have been applied in a complementary manner at nuclear waste disposal sites. The data obtained from the field surveys have been integrated in developing geophysical models of these sites.

A two-layer electromagnetic geologic model has been developed that allows the prediction of radar attenuation and depth of penetration. Field tests conducted at a low-level nuclear waste disposal site show values consistent with the model.

D8.1

A REVIEW OF THE CURRENT STATUS OF RADIATION EFFECTS IN SOLID NUCLEAR WASTE FORMS. William J. Weber, Pacific Northwest Laboratory, Richland, WA 99352.

The effects of radiation on solid nuclear waste forms are important since measurable changes in volume, leach rate, stored energy, and microstructure occur which may significantly affect the containment of the radioactive waste. Since the radiation-induced changes in properties follow an exponential dependence on dose which may exhibit saturation effects in 10^3 years, an understanding of the long-term cumulative effects of radiation on the properties of solid nuclear waste forms is essential.

The general nature of radiation effects in solid nuclear waste forms and the simulation techniques applied to accelerated testing will be briefly discussed.

The primary objective will be to review, in the context of previous studies, recently reported data on radiation effects in both glass and ceramic nuclear waste forms and to identify those areas where further theoretical and experimental research is needed.

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D8.2
MECHANICAL STABILITY OF A Cm-DOPED CELSIAN GLASS-CERAMIC. J.L. Routbort*, P. Offermann*, and H.J. Matzke, Commission of the European Communities, Joint Research Center, Postfach 2266, D-7500 Karlsruhe, Federal Republic of Germany.

The Hertzian indentation technique has been used to measure the fracture properties of a celsian glass-ceramic (B1-3) developed for high-level waste storage. The load at which Hertzian ring cracks are formed by spherical glass indentors pressed into the surface of the sample and the crack radii, r are measured for indentors sizes, R of 1.5 to 7.0 mm. For indentor radii, $R \leq 5$ mm the critical load is proportional to R and intersects the origin, confirming Auerbach's law. Using measured values of $r/a = 1.05$, where a is the calculated contact radius, Poisson's ratio, $\nu = 0.23$, and Young's modulus, $E = 100$ GPa, the fracture toughness $K_{Ic} = 1.3 \pm 0.2 \text{ MNm}^{-3/2}$ can be calculated from the crack extension function integrated by Warren. For $R > 5$ mm flaw statistics cause a deviation from the ideal behavior of Auerbach's law.

The self-radiation damage caused by recoiling Pu atoms resulting from α -decay of a Cm-244 doped sample increases K_{Ic} at least 25%. This increase in fracture toughness which is useful in applications is probably caused by the increase in internal stresses as a result of the disordering due to radiation damage. The x-ray spectra are also broadened by the radiation damage. The internal stress and the fracture toughness can be reduced by annealing.

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D8.3
ION-IMPLANTATION DAMAGE IN SILICATE GLASSES*, G. W. Arnold, Sandia National Laboratories, Ion Implantation Physics Division 5112, Albuquerque, NM 87185 USA

Recent work by Headley, et al.(1) has shown that Pb-ion implantation is a valuable technique for simulating α -recoil structural damage in zirconolite, zircon and hollandite. Pb-ion simulation of α -decay damage was also used by Arnold, et al.(2) in leaching studies of Savannah River Plant (SRP) nuclear waste glass. In this work (2) a critical Pb-ion fluence ($\sim 2 \times 10^{20}$ keV/cm²) was observed to result in accelerated near-surface compositional changes with leaching. The present paper presents the results of a study of the damage induced by ion implantation in simpler silicate glasses. We have extended our previous work (2) to studies (using Rutherford back-scattering and elastic recoil detection) of Corning 7740 (Pyrex) and other borosilicate glasses and find similar results, i.e., a definite radiation damage effect on leaching characteristics. These results are discussed in terms of a model of ion-implantation/recoil nucleus damage which involves structural transformation of the glass at a critical energy deposition value.

- (1) Headley, T.J., Arnold, G.W. and Northrup, C.J.M., Proceedings of the 5th Int. Symposium on the Scientific Basis for Nuclear Waste Management, June 7-10, 1982, Berlin, W. Germany (to be published).
- (2) Arnold, G.W., Northrup, C.J.M and Bibler, N.E., Proceedings of the 5th Int. Symposium on the Scientific Basis for Nuclear Waste Management, June 7-10, 1982, Berlin, W. Germany (to be published).

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D8.4
RADIOLYSIS OF GROUNDWATER FROM HLW STORED IN COPPER CANISTERS. H. Christensen, Studsvik Energiteknik AB, S-611 82 Nyköping, Sweden and E. Bjergbakke, Risø National Laboratory, Dk-4000 Roskilde, Denmark.

Conservative calculations and computer calculations have been carried out on the radiolysis of water irradiated from High Level radioactive Waste (HLW). The water was either outside the copper canisters, of varying thickness, or had penetrated the canisters and even come into contact with the fuel pellets, and thus had been irradiated with α -radiation.

γ -irradiation of water in bentonite containing Fe(II) results in the production of hydrogen and Fe(III). For a 1 cm thick canister the hydrogen concentration in the water, $C(H_2)$, becomes constant (10^{-3} M) after a short irradiation time. Hydrogen diffuses out of the system at a fairly constant rate (7×10^{-3} mole/year) almost independent of the dose rate. An iron content in the bentonite (corresponding to 1% of Fe_2O_3) is consumed in 24000 years.

Hydrogen and Fe(III) are produced as a result of α -irradiation of water containing Fe(II) by the cylindrical surfaces of the fuel pellets. Hydrogen is produced with a constant G-value, 0.89, corresponding to a production rate of 1.4 mol/year at a dose rate of 11 rad/s. The diffusion rate is insignificant. However, in less than one year $C(H_2)$ will exceed the solubility limit, and hydrogen bubbles will be formed. 1% iron will be consumed in 0.8 years.

D8.5
EFFECT OF STORAGE TEMPERATURE ON SELF-IRRADIATION DAMAGE OF ²³⁸Pu-SUBSTITUTED ZIRCONOLITE. F. W. Clinard, Jr., L. W. Hobbs, D. E. Peterson, D. L. Rohr, and R. B. Roof, University of California, Los Alamos National Laboratory, Los Alamos, NM 87545

Pu-substituted cubic zirconolite ($CaPuTi_2O_7$) was fabricated by sintering in air at 1673 K. Samples were held in dilatometers at ambient temperature, 5/3 K, and 873 K for 100 days while accumulating a dose of 1.3×10^{25} alpha decays/m³ (equivalent to 3×10^4 yrs of SYNROC storage time). Swelling was strongly temperature-dependent, reaching to date 4.5, 2.0, and 0.5 vol.%, respectively. X-ray studies showed the ambient-temperature material to be close to the x-ray-amorphous condition, while elevated temperature samples were less affected. TEM examination after four days

showed strong crystallinity with a dense array of damage clusters, but after 100 days revealed an electron diffraction-amorphous matrix along with small domains that had retained their crystallinity.

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D9.1

APPROXIMATIONS FOR ADAPTING POROUS MEDIA RADIONUCLIDE TRANSPORT MODELS TO ANALYSIS OF TRANSPORT IN JOINTED POROUS ROCK* K. L. Erickson, Sandia National Laboratories, Albuquerque, NM 87185.

Recently completed radionuclide transport analyses define criteria which determine when transport in jointed porous rock can be approximated as transport through an equivalent porous medium whose porosity is defined by the joint aperture, frequency, and orientation. For systems satisfying those criteria, the analyses further define the expression which should be used to evaluate the retardation factor which appears in the equivalent porous media models. The transport analyses will be summarized and the resulting criteria and expression for the retardation factor given in terms of fundamental physical and chemical parameters. Those parameters which can be evaluated in the laboratory include the radionuclide diffusion coefficient in aqueous solution, the distribution coefficient for sorption equilibrium between pore water and solid phases of the porous rock matrix, and the average grain density and porosity of the matrix. Those parameters which must be evaluated from field data include the average fluid velocity and the joint aperture, frequency, and orientation. The sensitivity of the derived criteria and expression for the retardation factor with respect to each of the above parameters have been examined, and the implications which those sensitivities have for the type and accuracy of data obtained from laboratory and field experiments will be discussed. Finally, the application of the criteria and retardation factor to analysis of an actual system will be illustrated.

* This work supported by the U. S. Department of Energy under contract DE-AC0476DP00789.

D9.2

TOWARDS A COMPREHENSIVE MODEL OF CHEMICAL TRANSPORT IN POROUS MEDIA.
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A chemical transport model that includes dispersion/diffusion, convection, complexation, sorption, and precipitation has been written. The sorption process is modelled by an ion exchange mechanism except it is possible to consider the electric double layer in this process when it is important. Chemical equilibrium is assumed so mass action equations are written for each reaction. In addition a mass conservation equation is written for each species as well as a site constraint on the number of sorption sites. This set of equations are expressed in a Differential/Algebraic form and the equations are solved simultaneously. The solution procedure described is flexible enough that time dependent boundary conditions can be considered and that different chemical mechanisms as non-equilibrium kinetics can be included. The model is used to investigate the applicability of

a K_d model to describe sorption, to determine the importance of precipitation when a waste form as $SrCO_3$ comes in contact with the groundwater, the effect of including the electric double layer concept in sorption, and the sensitivity of the parameters as equilibrium coefficients and dispersion coefficients needed in chemical transport calculations of radionuclide transport through engineered barriers.

D9.3

WASTE CLASSIFICATION BASED ON TOTAL HAZARD. Lori E. Wickham and S. K. Rope, EG&G Idaho, Inc., P.O. Box 1625, Idaho Falls, Idaho 83415.

Some low-level radioactive waste represents both a chemical and radiological hazard, and it is desirable that management practices take this into account. This paper describes a waste classification system that could provide a basis for such management practices.

The classification system formulates the innate hazard of waste as the ratio of potential exposure or intake to some selected criteria. Potential exposure or intake relates two factors: (1) the nature and amount of toxic materials disposed, and (2) the available biological hazard posed when those materials leave the soil and enter the biological pathway potentially exposing humans. The system will provide guidance on the degree of isolation each waste material needs and how wastes could be separated for disposal. Examples of the systems use are provided.

D9.4

COMPLIANCE ASSESSMENTS OF HYPOTHETICAL GEOLOGICAL NUCLEAR WASTE ISOLATION SYSTEMS WITH THE DRAFT EPA STANDARD. Segel, M.D., Chu, M.S. and Pepping, R.E. Fuel Cycle Risk Analysis Division, Sandia National Laboratories, Albuquerque, NM, 87185.

Potential radionuclide releases from hypothetical nuclear waste repositories in bedded salt and welded tuff have been calculated and compared to the limits set by the EPA Draft Standard. The importance of several parameters and model assumptions to the estimated discharges has been evaluated. The areas that were examined include the source-term model, radionuclide solubilities and sorption, the description of the local hydrogeology and the simulation of contaminant transport in the presence of fracture flow and matrix diffusion. The uncertainties in geochemical and hydrogeological parameters were represented by assigning realistic probability distributions to these variables. The MWFT/DVM transport code was used to calculate the radionuclide discharge. Our calculations suggest the following conclusions for undisturbed hypothetical repositories: (1) sorption of radionuclides by zeolitized tuff is an effective barrier to the migration of actinides even in the absence of solubility constraints; (2) violations of the EPA Draft Standard are primarily due to discharge of ^{99}Tc and ^{14}C . Retardation due to matrix diffusion, however, may eliminate discharge of these nuclides for realistic ground water flow rates in welded tuff. Breaching of a waste repository by drilling in bedded salt formations will lead to large violations of the EPA Standard. The amount of radionuclide discharge is strongly dependent upon the fraction of the canisters that is accessed and upon mixing processes within the engineered facility.

19.5

MODELING OF WASTE/NEAR FIELD INTERACTIONS FOR A WASTE REPOSITORY IN BEDDED SALT: THE DYNAMIC NETWORK (DNET) MODEL. Robert M. Cranwell, Sandia National Laboratories, Fuel Cycle Risk Analysis Division, Albuquerque, NM, 87185.

The Fuel Cycle Risk Analysis Division of Sandia National Laboratories has been funded by the U.S. Nuclear Regulatory Commission to develop a methodology for use in assessing the long-term risk from the disposal of radioactive waste in deep geologic formations. As part of this program, the Dynamic Network (DNET) model was developed to investigate waste/near field interactions associated with the disposal of radioactive waste in bedded salt formations. The model is a quasi-two-dimensional network model and simulates processes such as fluid flow, heat transport, salt dissolution, salt creep, thermomechanical effects, and subsidence. The network flow model used in DNET is loosely based on a hypothetical flow system that serves as a reference site for the demonstration of the Sandia risk assessment methodology. Because of the processes considered in the DNET model, many of the media fluid and hydraulic properties are allowed to vary with time.

19.6

STUDY OF COUPLED THERMOMECHANICAL, THERMIDROLOGICAL AND HYDROMECHANICAL PROCESSES ASSOCIATED WITH A NUCLEAR WASTE REPOSITORY IN A FRACTURED ROCK MEDIUM. C. F. Tsang, Noorishad, and J. S. Y. Wang, Earth Sciences Division, Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720.

It has been recognized that coupled thermomechanical, thermohydrological and hydro-mechanical processes may play an important role in the behavior of the geological formation around a nuclear waste repository. Recently Lawrence Berkeley Laboratory has completed the development of a numerical code, ROCHAS, that calculates coupled heat transfer, fluid flow and mechanical deformation in a fractured porous medium. Such a code can be used not only to model a nuclear waste repository but also to give a fundamental understanding of these nonlinear coupled processes and to establish limits of validity of simpler, uncoupled models.

In this investigation, illustrative examples from the modeling studies of the code ROCHAS are presented. The thermomechanical simulations of a fractured rock mass show that the fractures can absorb the thermally induced rock deformations which have been observed in the granite experiment in Stripa, Sweden. The thermohydrologic simulations of the propagation of temperature and pressure perturbations along a fracture may be used to develop nonisothermal well testing techniques to study the fracture characteristics. The variations of the fracture aperture profiles in the hydromechanical simulations indicate that the rigid fracture assumptions used in most uncoupled models may not be a good representation of the fracture behavior. These and other details are discussed in the paper.

20.1

FIXATION OF HIGH-LEVEL SOLID WASTE (HLSW) IN A TiO_2 -BASE CERAMIC MATRIX PROVIDED WITH AN INTEGRATED OVERPACK. Christof Bauer and Gerhard Ondracek, Institut für Material- und Festkörperforschung, Kernforschungszentrum-Karlsruhe, Postfach 3640, D-7500 Karlsruhe 1

Several international research activities revealed the extraordinarily high leaching resistance of TiO_2 in water and brines at temperatures up to 623 K. The paper deals with the feasibility of producing a waste form mainly consisting of

Rutile in which the oxide and metal constituents of HLSW are locked up as particles with particle sizes < 1 mm. Bodies of this structure were produced by uniaxial hot pressing at 1223 K and 10 MPa. Due to the fact that a reducing atmosphere can be maintained during the hot pressing process, Ruthenium is included in the metallic state, thus totally avoiding volatility problems of this element which usually occur at high temperatures. Attempts were successful to densify simultaneously the TiO_2 -base ceramic matrix with the HLSW-particles and create a non-radioactive layer of TiO_2 or Ti. This would make a corrosion resistant overpack unnecessary. Leach rates of the TiO_2 -base matrix at 373 K in water were found to be below 10^{-8} g/(cm²day). Even a failure of the non-radioactive layer would temporarily result in Cs-leach rates not above 10^{-5} g/(cm²day) until the waste particles on the surface are dissolved, whereafter the leach rate would again decrease close to the value of Rutile.

D10.2

CORRELATION BETWEEN DYNAMIC LEACH TEST RESULTS AND GEOCHEMICAL OBSERVATIONS AS A BASIS FOR REPOSITORY-RELEVANT RELEASE RATE PREDICTIONS. Aaron Barkatt, P. B. Macedo, W. Sousanpour, Alisa Barkatt, M. A. Boroomand, P. Szoke and V. L. Rogers, Vitreous State Laboratory, The Catholic University of America, Washington, D.C. 20064.

Results of leach tests on various glassy and ceramic waste-forms as well as on rocks and minerals are found to agree with a general model. The salient features of the model are as follows:

- (1) Under slow-flow conditions the rates at which the waste-form loses material reflect the saturation levels of silica, alumina, etc.
- (2) The saturation levels of Si and Al are inter-related and controlled by a combined aluminosilicate alteration product.
- (3) The single most important parameter which determines the solubilities at a given temperature is the pH value.
- (4) Alkalis and other soluble species continue to leach away and change the pH even at very slow flow rates.

Extrapolations of leach test results based upon this model are found to agree with observed groundwater compositions, validating the applicability of this method for predicting release rates from waste-forms under geologic repository conditions.

D10.3

γ -ZIRCONIUM PHOSPHATE AS A Cs-WASTE FORM FOR PARTITIONED WASTES. Sridhar Komarneni and Rustum Roy, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

Since Cs and Sr have been so successfully separated and solidified at Hanford, the applicability of this strategy to future commercial wastes calls for an optimization of this waste form.

zirconium phosphate (γ -ZrP), γ -Zr(HPO₄)₂·2H₂O is an extremely selective cesium ion adsorbent for nuclear waste solutions. Because of its layer structure, the adsorbed Cs can be directly fixed in γ -ZrP by collapsing the layers upon heat treatment. Fixation of up to 96% of the adsorbed Cs was achieved by simple heat treatment to 350°C. Subsequent heat treatment of this γ -ZrP leads to a good radiophase with good leach resistance. Cs sorbed γ -ZrP can also be converted to CsZr₂(PO₄)₃ (NZP radiophase) by reacting with ammonium monobasic phosphate and zirconyl nitrate at about 900°C. Thus, γ -ZrP serves as a precursor to CsZr₂(PO₄)₃ radiophase which has comparable leach resistance to other phases under consideration for radio-Cs immobilization.

4
STRONTIUM LEACHABILITY OF HYDROFRACTURE GROUTS FOR SLUDGE-SLURRIES. M. T. Gorman, E. W. McDaniel, J. G. Moore, H. E. Devaney, and L. R. Dole, Oak Ridge National Laboratory*, Chemical Technology Division, Experimental Engineering Division, Cementitious Waste Form Development Group, P. O. Box X - Bldg. 3017, Oak Ridge, TN 37830.

This paper summarizes the results obtained from a series of experiments performed to determine the strontium leachability of cement-based sludge-slurry hydrofracture grouts.

The hydrofracture process has been used at ORNL since 1966 for the routine disposal of intermediate-level waste (ILW) solutions. In this process, cement

and other additives are mixed with a waste stream to form a grout, which is then injected into a shale bed at a pressure sufficient to cause fracture along the horizontal bedding planes. The injected grout soon hardens, fixing the radioisotopes between the layers of the massive Conasauga shale formation.

The results of applying the modified IAEA dynamic leach tests to hydrofracture grout specimens showed improved leach resistance (by a factor of 3 to 5) as the leaching time was increased from 28 to 91 d and a weak trend toward lower leachability as increased amounts of dry solids were added.

The moving boundary plus diffusion leach model fit the dynamic leach data successfully in most cases. An apparent diffusion coefficient of 5×10^{-12} cm²/s and a moving boundary coefficient of 1×10^{-7} s⁻¹ were obtained for one of the grout compositions when leached in distilled water.

*Operated by Union Carbide Corporation under contract W-7405-eng-26 with the U.S. Department of Energy.

5
HYDRATION OF NUCLEAR WASTE GLASS BY HYDRATION. John K. Bates and In J. Steindler, Argonne National Laboratory, Argonne, IL 60439.

The leaching of simulated nuclear waste glass by contact with a controlled temperature, humid atmosphere results in the formation of a double hydration layer penetrating the glass and in the formation of alteration products on the glass surface. The hydration process has been studied as a function of time, temperature, glass composition and water vapor pressure. A dual stage hydration rate was observed at

all temperatures studied and rate constants were determined at each temperature. An Arrhenius plot for the initial rate stage indicates the reaction mechanism does not change between the temperature limits of the experiment (120-240°C). This conclusion is supported by the sequence of mineral formation on the surface. This hydration process represents the practical maximum in SA/V and provides a means of accelerating aging reactions while simulating conditions that may exist in a nuclear waste repository.

D10.6

SUPERNATE TREATMENT CONSIDERATIONS FOR THE NEUTRALIZED WASTE AT WEST VALLEY. G. B. Gockley¹, J. M. Pope², and E. J. Lahoda¹, Westinghouse R&D Center, Pittsburgh, PA 15235 and ²West Valley Nuclear Service Company, Inc., West Valley, NY 14171.

The neutralized waste stored at the Western New York Nuclear Service Center in West Valley, New York, was produced during the operation of the Nuclear Fuel Service, Inc., commercial nuclear fuel reprocessing plant. This waste consists of a supernate layer and a settled sludge layer. The supernate is a highly concentrated salt solution (Na₂SO₄, NaNO₃, NaCl, and NaOH) containing dissolved cesium and strontium, as the primary radioactive components. The sludge is believed to be primarily iron and aluminum hydroxides and contains the bulk of the long-lived isotopes.

The supernate can be treated to remove the bulk of the activity and then be disposed of as low level waste. The following supernate treatment considerations have been evaluated on a laboratory scale using simulated West Valley waste:

- Organic ion exchange resins
- Inorganic ion exchange resins
- In-tank processing

These processes will be described and preliminary laboratory data will be presented.

This work was supported by U. S. Department of Energy.

D10.7

LEACH RESISTANCE OF THE BOROSILICATE GLASS VG98/12 CONTAINING HIGH CONCENTRATIONS OF URANIUM. L. Kahl, E. Mainka*, M. Nesovic and J. Saidl, Institute of Nuclear Waste Management, *Institute of Radiochemistry, Nuclear Research Center Karlsruhe, PO Box 3640, 7500 Karlsruhe, Federal Republic of Germany.

The borosilicate glass VG98/12 developed for the vitrification of HLLW has been loaded with up to 40 wt% UO₂. The leach resistance of the glass products formed was investigated in water, salt brine and Q brine at elevated temperatures and pressure as the most important criterion of their quality. The total weight loss in all leachants mentioned was fairly independent of the quantity of UO₂ in the glass products. Nevertheless, the highest specific leachability of uranium was found in the Q brine.

The average total leach rate values after 30 days leaching were in the range of 10^{-4} g·cm⁻²·d⁻¹ at 100 and 200°C respectively and at the normal pressure and in the range of up to 10^{-7} (10^{-5} for the Q brine) at 200°C and 100 bars.

10.8

COMPARISON OF 200 LITER AND 40 MILLILITER LEACH TESTS. Wayne A. Ross, Joseph Westsik, Jr., Frank P. Roberts, and Clark O. Harvey, Pacific Northwest Laboratory, P. O. Box 999, Richland, Washington 99352.

During the past year we have conducted a modified MCC-1 leach test on a 200 liter block of a cast cement waste form. The leach vessel was a 200 liter teflon-lined drum and contained 100 liters of deionized water. The results of this large scale leach test were compared with the results of standard MCC-1 tests (40 ml) on smaller samples of the same waste form. There were ratios of 100 in leachate volume and 150,000 in sample mass between the two tests. The surface area to volume ratio was the same in both tests. The cast cement samples for both tests contained plutonium-doped incinerator ash.

The leachates from these tests were analyzed for both plutonium and the matrix elements. Plutonium plateout was evaluated in the large-scale leach test. This evaluation has indicated that the majority of the plutonium leached from the samples is plated out onto vessel walls and little (3×10^{-12} M) remains in solution. The overall results of tests support the assumption that laboratory-scale results can be scaled up without major errors. However, the differences which are observed suggest that additional large prototype tests with other waste forms be performed.

10.9

ELECTRICAL CONDUCTIVITY MEASUREMENTS OF LEACHATES FOR THE RAPID ASSESSMENT OF WASTE FORM CORROSION RESISTANCE.* B. C. Sales, M. Petek, and L. A. Boatner, Solid State Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830.

Several candidate nuclear wasteforms (borosilicate glass, SYNROC, and monazite) were leached in distilled water at 90°C in a teflon conductivity cell. As each simulated wasteform corrodes, the release of mobile ions into the leachate causes an increase in the conductivity of the leaching solution. By measuring the change in solution conductivity as a function of time, the corrosion resistance of each wasteform can be qualitatively assessed. In addition to comparing the corrosion resistance of one wasteform relative to another, this technique is extremely useful in minimizing the corrosion resistance of a given wasteform. The method also represents a possible means of quality assurance during the production of an actual primary wasteform. The technique is simple, very sensitive, fast, and, with appropriate calibration, can be made semiquantitative. For each wasteform the relationship between conductivity and the species present in the leachate will be discussed.

*Research sponsored by the Division of Materials Sciences, Office of Basic Energy Sciences, U.S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

D10.10

CORRELATION OF ^{137}Cs LEACHABILITY FROM SMALL-SCALE (LABORATORY) SAMPLES TO LARGE-SCALE WASTE FORMS.* N. Morcos, R. Dayal, and A. J. Weiss, Brookhaven National Laboratory, Upton, NY 11973.

A study correlating the leachability of ^{137}Cs from small-scale to large-scale cement forms was performed. The waste forms consisted of (a) organic ion exchange resins incorporated in Portland I cement, with a waste-to-cement ratio of 0.6 and a water-to-cement ratio of 0.4 (as free water) and (b) boric acid waste (12% solution), incorporated in Portland III cement, with a waste-to-cement ratio of 0.7. ^{137}Cs was added to both waste types prior to solidification. The sample dimensions varied from 1 in. x 1 in. to 22 in. x 22 in. (diameter x height) in size. Leach data extending over a period of 260 days were obtained. A method based on semi-infinite plane source diffusion model was applied to analyze the leach data. An effective bulk diffusion coefficient was calculated from the leach data for both types of solidified waste. A derived mathematical expression allows prediction of the amount of ^{137}Cs leached from the forms as a function of leaching time and waste form dimensions. A reasonably good agreement between the experimental and calculated data is obtained.

*Work carried out under the auspices of the U.S. Nuclear Regulatory Commission.

D10.11

LONG-TERM RADIOACTIVITY RELEASE FROM SOLIDIFIED HIGH-LEVEL WASTE PART II; PARAMETRIC STUDY OF WASTE FORM PROPERTIES, TEMPERATURE AND TIME. Friedrich K. Altenhein and Werner Lutze, Mahn-Meitner-Institut, 1,000 Berlin 39, Germany, and Rodney C. Ewing, University of New Mexico, Albuquerque, New Mexico 87131, USA.

A computer code QTERM (methodology presented by Altenhein et al., MRS Symposium Proceedings, 1982, Berlin) has been used to calculate the total released activity as a function of waste form properties, specific geologic scenario and time. This paper presents the results of a parametric study of waste form properties (A =pre-exponential frequency factor; b =apparent activation energy; n =exponent of time), as well as initial temperature of the waste canister and canister life-time. Of the waste form parameters, n has the greatest affect on the total activity and may be varied as a function of time to simulate changes in mechanism. The variation in waste form parameters covers the range characteristic of most proposed waste forms (borosilicate glass and crystalline ceramics) and allows their direct comparison for specific conditions. There is considerable overlap in the performance of waste forms, depending on canister life-time and repository history. The long-term activity is most dependent on short-term events (less than 200 years). This suggests that the importance of the repository may be greatest immediately after emplacement of the waste, rather than as a long-term barrier to radionuclide migration.

D10.12
RADIOLYTIC GAS GENERATION AND OXYGEN DEPLETION IN ION EXCHANGE MATERIALS, Wayne P. Capolupo, M.S. and James R. Sheff, Ph.D., Department of Nuclear Engineering, University of Lowell, Lowell, Mass. 01854

The process of ion exchange is used extensively throughout numerous industries where water of extremely high purity is required.

Government and commercial nuclear power facilities are typical of this, and have applied ion exchange processes for the removal of radioactive ions from aqueous solutions.

When water is decontaminated in this fashion the ion exchange medium becomes saturated with the radioactive ions from solutions, and is then stored as radioactive waste in standard burial facilities. During this indefinite storage period, the chemical changes which occur in the ion exchange medium may result in oxygen depletion and gas generation within the storage vessel, and subsequent loss of integrity.

It is the object of this work to investigate the early phases of radiolytic oxygen depletion and gas evolution through experimental procedure and observation. The results of this work will then be analyzed with respect to applications for the storage of radioactive ion exchange waste.

D10.13
IRRADIATION EFFECTS IN THE STORAGE AND DISPOSAL OF RADIOACTIVE ION-EXCHANGE RESINS* K. J. Swyer, C. E. Dodge, R. Dayal, and A. J. Weiss, Brookhaven National Laboratory, Upton, NY 11973

Research is under way to characterize the effects of self-irradiation on radwastes which may be generated when organic ion-exchange media are used in water demineralization or decontamination operations at nuclear facilities. External factors (radiation dose rate, atmospheric and moisture conditions, resin chemical loading, synergetic interactions, etc.) affecting the relation between laboratory evaluations and field performance are emphasized. Initial experiments show no evidence for a dramatic radiation dose-rate effect on radiolytic gas yields or acid product formation when (fully swollen) sulfonic acid resins** are irradiated in a sealed air environment. At the same time, oxygen gas is removed from the environment of irradiated resins. Interaction between mild (1018) steel coupons and acidic species (sulfuric acid) produced in the irradiation induced decomposition of sulfonic acid resin results in irradiation enhanced corrosion. Corrosion rates depend on radiation dose rate, moisture content and resin chemical loading. In some cases, corrosion rates decrease with time, suggesting depletion of acidic species within the resin bed, or a synergistic interaction between resin and corrosion coupon. Implications of these and other results on evaluating field behavior of radwaste containing ion exchange media are discussed.

*Work carried out under the auspices of the U.S. Nuclear Regulatory Commission.
**Amberlite IRN-77, Rohm and Haas Company, Philadelphia, PA.

D10.14
COMPATIBILITY RELATIONSHIPS IN THE FE-U-O-H SYSTEM AT 400°C: THE IMPLICATIONS OF THE FERRIC-FERROUS BUFFER FOR THE IMMOBILIZATION OF URANIUM AND TRANSURANIC ELEMENTS. Deane K. Smith, W. Phelps Freeborn, and Barry E. Scheetz, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

The current strategy for the immobilization of nuclear wastes is based upon a system of multiply redundant barriers (DOE, 1980) for which geological containment is the final barrier to the migration of radionuclides to the biosphere. The ability of the repository host rock to buffer the oxygen fugacity of the pore waters is a critical concern for the evaluation of the far-field migration of selected elements; notably Tc, U and TRU elements. This buffer capacity of all proposed host rocks, with the exception of salt, is based upon the presence of both ferric and ferrous irons in the host rock phases.

The system that is reported upon herein was chosen because of the implication of the solubility of the hexavalent- versus the tetravalent-uranium species with regards to the terminal disposal of unprocessed commercial power reactor fuel elements. The early work of Evans and White (1964) addressed the phase relationship in this system but only at temperatures well above the designed repository conditions. More recently Freeborn et al. (1980) reported upon the potential phase relationships derived from thermochemical data.

The calculated compatibility relationships proposed by Freeborn et al. were experimentally verified by hydrothermally reacting appropriate mixtures of finely divided hematite, magnetite, UO_2 and/or γUO_3 in order to define the compatibility relationships. The compositions of phases in the UO_2 - UO_3 solid solution series were verified by means of internal standards ($Si a_0 = 5.43088\text{\AA}$ and $MgAl_2O_4 a_0 = 8.0831$).

D10.15
INTERNAL HYDROGEN EFFECTS IN TI CODE 12 CANNISTER OVERPACK MATERIAL*. N. R. Moody and S. L. Robinson, Sandia National Laboratories, Livermore, CA 94550.

The effects of internal hydrogen on the mechanical properties of Ti code 12 were studied by gas phase charging smooth bar tensile samples. Gas phase charging was performed below the β transus to achieve substantial hydrogen concentrations and eliminate damage which may result from cathodic charging. Tensile tests showed no change in yield strength with increasing hydrogen concentration whereas ductility decreased slightly. However, a change in fracture mode, from microvoid coalescence to microvoid coalescence and secondary cracking along grain boundaries, occurred as hydrogen concentration increased. This was accompanied by a change in the amount of retained β . The mechanical property results will be discussed in terms of the hydrogen-induced microstructural change and the resistance of Ti code 12 to hydrogen embrittlement.

*This work supported by U.S. Department of Energy, DOE, under contract number DE-AC04-76DP00789.

D10.16
CORROSION OF TiCode-12 IN A SIMULATED WASTE ISOLATION PILOT PROJECT (WIPP) BRINE.*
T.M. Ahn, B.S. Lee, J. Woodward, R.L. Sabatini and P. Soo, Brookhaven National Laboratory, Upton, NY 11973.

The corrosion behavior of TiCode-12 (Ti-0.3 Mo-0.8 Ni) high level waste container alloy has been studied for a simulated WIPP brine at temperatures up to 150°C. Crevice corrosion was identified as a potentially important failure mode for this material. Within a mechanical crevice, a thick oxide film was found and shown to be the anatase form of TiO₂, with smaller amounts of Ti₃O₅ also present. Acidic conditions were found to cause a breakdown of the passive oxide layer. Increasing temperature and solution aeration accelerate the corrosion rate. The incubation time for crevice corrosion has been calculated for various test conditions. In hydrogen embrittlement studies, it was found that hydrogen causes a significant decrease in the threshold stress intensity level in fracture mechanics samples. Hydride formation is thought to be responsible for crack initiation. Gamma irradiation studies were performed in WIPP brine and the potential for radiolysis products to induce failure in TiCode-12 has been addressed. Attention has also been given to methods for extrapolating short term uniform corrosion rate data to extended times, in order to predict container performance.

*This work was performed under the auspices of the United States Nuclear Regulatory Commission.

D10.17
HYDROTHERMAL SIMULATION OF ION MIGRATION IN SELECTED ARGILLACEOUS SEDIMENTS:
IMPLICATION FOR BACKFILL DESIGN. L. Bratton, B.E. Scheetz and W.B. White, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802

The current approach for the disposal of high-level nuclear waste calls for the total isolation in a deep mined geological repository. Intrusion of ground water into a breached repository appears to be the most likely method of dispersal of hazardous radionuclides to the biosphere. Consequently, the mechanisms of ion migration in the near-field environment through backfills and host rocks must be understood.

Two argillites were studied that spanned the chemical types from a reduced carbon-sulfide rich to an oxidized, slightly metamorphosed shale. These samples were hydrothermally reacted at saturated steam pressure at 200°C in a solution containing the major radionuclides of concern.

Both samples were characterized and found to contain reduced uranium in the form of UO₂.₂₅ with associated REE. The reduced shale exhibited the precipitation of uranium on the outer surfaces and effectively prevented penetration of Cs and Sr into the interior. The oxidized shale formed a precipitate of uranium on a surface some 500 μm within the sample. Cs and Sr had completely penetrated the sample in 7 days. Within experimental errors a mass balance between the partitioning of cations between the liquid and solids was achieved.

D10.18

DIFFUSION MEASUREMENTS IN CONCRETE AND COMPACTED BENTONITE. A. Muurinen, J. Rantanen, K. Uusheimo, O.J. Heinonen, Technical Research Centre of Finland, Reactor Laboratory, Otakaari 3 A, SF-02150 Espoo 15, Finland

A laboratory method for measurement of diffusion of radionuclides in concrete products and compacted bentonite has been developed. In this method a tracer is introduced through one end into the cylindrical sample closed in a tube and pre-equilibrated with water. After the introduction period of the tracer the tube is sealed hermetically and the concentration profiles of the radionuclides are measured periodically from the outside of the sample using a collimated detector. Diffusivities are calculated from the activity profiles by fitting the theoretical curve with the measured profile. The paper will give diffusion data for some radionuclides in some concrete products and compacted bentonite.

D10.19

RHEOLOGICAL AND PHYSICAL PROPERTIES OF MAGNESIUM OXIDE AND SILICA FUME-MODIFIED CEMENT MORTARS CURED AT HIGH TEMPERATURE. Zenbe-e Nakagawa, Elizabeth L. White and Della M. Roy, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

Placement characteristics and physical properties of cementitious mixes with various expansive agents and industrial by-products have been investigated up to curing temperatures of 250°C, which is the upper limit of an emplacement site. Quantitative viscometric measurements on modified class H cement mortars showed both how the rheological properties changed systematically and how the shape, magnitude and direction of the loop indicated the nature of any structural changes in the slurry being tested. Each of three chosen mixtures were altered in composition by the addition of 1.0, 2.5, and 5.0 percent of an expansive agent and also of a fine-grained quartz fume. Each mixture was required to have pseudo-thixotropic flow behavior, that is it should have a yield point, should be plastic or pseudo-plastic, and should be shear thinning with time. The best mixture cured at 250°C (81-24) with a compressive strength of >180 MPa had a low density and intrinsic permeability (<10⁻⁸ Darcys) and a viscosity of 1700 cP at a shear rate of ~55 sec⁻¹. The best mixture cured at 175°C (81-21) with a compressive strength of 240 MPa had a low density and intrinsic permeability (<10⁻⁸ Darcys) and a viscosity of >5000 cP at a shear rate of ~55 sec⁻¹. A third mixture (81-20) cured at 175 and 250°C had the highest compressive strength at 1.0 percent expansive agent at 175°C whereas each of the 250°C cured samples decreased in strength with time. The strength of the 175°C samples cured at 175°C for 28 days was 210 MPa; the viscosity was >3000 cP.

D10.20

DISSOLUTION AND REACTION STUDIES OF CsZr₂(PO₄)₃ AND Zr PHOSPHATES IN AQUEOUS MEDIA. E. R. Vance and F. J. Ahmad, Materials Research Laboratory, The Pennsylvania State University, University Park, PA 16802.

The dissolution of CsZr₂(PO₄)₃, a possible phase for the immobilization of radioactive Cs, was studied at 70°C in a variety of aqueous media. The time dependences of the Cs dissolution rates did not fit linear or square-root laws very well. The dissolution rate was a minimum at neutral pH. In the acid range, the dissolution rate varied approximately as [H⁺]^{0.3}. The variation of the dissolution rate in the

was a stronger function of pH. Under near-neutral conditions, there was not great enhancement of the Cs dissolution rate when NaCl or CaCl₂ were present at 0.1M level.

Reaction of water at 300°C on α-Zr phosphate gel and ZrP₂O₇ was also studied in respect of alteration products and the extraction into solution of the ions.

.21
DIFFERENTIAL SCANNING CALORIMETRY OF METAMICT ZIRCONOLITE. Dean E. Peterson and Frank W. Clinard, Jr., Los Alamos National Laboratory, Los Alamos, NM 87545.

Samples corresponding to the mineral zirconolite were prepared by cold pressing and sintering. Plutonium was substituted for the zirconium in order to characterize radiation damage effects. The energy stored in fully metamict samples was measured with a differential scanning calorimeter. The total energy of 6.6 ± 0.4 cal/g is released over the range 758-988 K. The activation energy of annealing of the damage is 0.45 ± 0.05 eV. The temperature dependence of the rate constant is described by

$$k_T = 1.880 \exp(-0.45/k_B T) \text{ s}^{-1}$$

where k_B and T are the Boltzmann's constant and temperature respectively. These results are applied to analysis of waste incorporation in SYNROC and correlated with analogous parameters for other waste forms.

1.1
CORROSION RESISTANT CANISTERS FOR NUCLEAR WASTE ISOLATION.* N. J. Magnani, Sandia National Laboratories, Albuquerque, NM 87185.

Corrosion resistant canisters are an important component of the engineered barriers in a nuclear waste disposal system. In addition to providing containment for the waste form during transportation and emplacement, a durable canister can eliminate hydrothermal interactions with the waste form if the canister survives the thermal history of the waste (up to 300 years).

The selection of a repository site and emplacement technique will affect canister design and could also impact material selection. While there are still many issues to be resolved, there are two different concepts being evaluated to provide durable canisters for waste disposal: (1) canisters fabricated out of extremely corrosion resistant materials such as Ti base or Ni base alloys, and (2) canisters fabricated out of less durable materials but designed with a corrosion allowance. Each of these approaches has some advantages and some special problems associated with it. These issues will be discussed.

Canisters could fail to meet these design objectives through a variety of failure processes. The more important of these will be discussed and examples of work underway to establish confidence that the processes will not occur will be presented.

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A U. S. DOE facility.

D11.2

THE CANADIAN CONTAINER DEVELOPMENT PROGRAM FOR FUEL ISOLATION. K. Nuttall, J.L. Crosthwaite, P.J. McKay, P.M. Mathew, Atomic Energy of Canada Limited, Pinawa, Manitoba, ROE 1L0, Canada; and B. Teper, P.Y.Y. Maak and M.D.C. Moles, Ontario Hydro, Toronto, Ontario, Canada.

Progress in the Canadian program to develop high integrity containment systems for the direct disposal of irradiated fuel in a deep, hard-rock vault is described. Possible containment targets are discussed and the disposal-vault conditions which influence container design and materials selection are reviewed. To date, most effort has been directed toward developing containers with a minimum design lifetime of about 500 years, based on a corrosion-resistant metal shell as the primary barrier to radionuclide release. Two design concepts are being studied, one in which a thick container shell completely supports the service stresses, and one in which an internal medium provides support for the outer shell. For the latter concept, low melting point metals, packed particulates and internal structural members are being assessed. The design details for various containers are described, together with prototype fabrication and testing programs that are in progress or planned. Recent advances in materials research are presented, with emphasis on evaluating the corrosion performance and the ease of fabrication and inspection of candidate container materials. Finally, the possibility of incorporating additional independent barriers to extend the containment lifetime, and of developing long-term containment systems based on ceramic or thick metal containers is discussed.

D11.3

SOME EFFECTS OF MICROSTRUCTURE AND CHEMISTRY ON CORROSION AND HYDROGEN EMBRITTLEMENT OF TiCode-12.* James A. Ruppen, R.B. Diegle, R.S. Glass and T.J. Headley, Sandia National Laboratories, Albuquerque, NM 87185.

An experimental program is underway to determine the suitability of TiCode-12 (Ti-0.8 Ni-0.3 Mo) as a corrosion resistant overpack/canister for high level waste containment. This paper addresses two modes of degradation, namely, sensitization and hydrogen embrittlement. Sensitization refers to an increase in uniform corrosion rate after application of heat treatments that produce certain microstructural changes in the metallurgical phases present. Electrochemical studies with various synthesized phases, and microstructural characterization have suggested that sensitization involves galvanic coupling between Ti₂Ni and the alloy matrix, with resultant shifts in corrosion potential. Possible effects of Mo and Fe are also considered.

Tensile tests were performed in air and brine with TiCode-12 that had been charged with hydrogen to various concentrations. An experimental alloy of composition Ti-0.9 Ni-0.4 Mo-0.4 Fe-0.25 O₂ exhibited the highest ultimate tensile strength, the result of high β phase and oxygen contents. Hydrogen concentrations to 130 ppmw did not cause embrittlement. At 220 ppmw H, however, a reduction in time to failure occurred and the fracture surface displayed intergranular cleavage-like secondary cracks. Tensile results obtained for H-charged (220 ppmw) specimens were similar in air and brine. This similarity supports a hydrogen embrittlement (hydride cracking) rather than anodic dissolution mechanism for stress corrosion cracking at cathodic overpotentials. These results will be discussed in terms of hydrogen solubility and stress-induced hydride formation.

*This work supported by the United States Department of Energy (DOE) under contract DE-AC04-76-DPO0789.

Good Paper

D11.4
 INVESTIGATIONS OF SUITABLE METALLIC CONTAINER MATERIALS FOR HAW SOLIDIFICATION.
 J. Mirschinka, R. Odoj, St. Halaszovich, Institute for Chemical Technology, KFA
 Jülich, Germany, and K. Forch, U. Forch, Thyssen-Henrichshütte AG, Hattingen,
 Germany.

Six alloys with different chemical compounds, such as a very low carbon steel, a boiler steel, a Fe-Cr-Ni steel, a Ni-Cr alloy, a Ni-Cr-Mo alloy and a Co-base alloy were investigated as candidate container materials for the in-can-melting FIPS process developed in Jülich. In addition to possessing acceptable creep-rupture strength, the materials must be compatible with the glass-melting conditions.

Not only the corrosion attack of the container by the glass melt was investigated but also the influence of the main alloying elements upon the composition, the structure and the homogeneity of the glass bloc.

Mainly the alloying element chromium, assuring a sufficient high temperature oxidation resistance as well as corrosion resistance against water or salt solutions, was found to cause a violent blistering in the glass matrix.

The VLC-steel was found to show least interactions with the glass melt, and the 1% Mo boiler steel gave satisfactory results too, both having sufficient high temperature mechanical properties as well.

D11.5
 LABORATORY STUDIES OF FLUID FLOW THROUGH BOREHOLE SEALS. David L. South and Jaak
 K. Daemen, Department of Mining and Geological Engineering, University of
 Arizona, Tucson, Arizona 85721.

Boreholes associated with a high level nuclear waste repository must be reliably sealed. Laboratory experiments have been performed in which flow through a sample of "intact" granite is compared with flow through the same sample with a portion of the rock replaced by a cement plug. Flow rates through the cement-plugged sample were lower than through "intact" rock. The sample is a cylinder of rock with holes drilled along the axis from the top and bottom, leaving a bridge of rock in the center. Flow rate through this cylinder is measured under a triaxial stress state corresponding to a depth of about 920 meters. The rock bridge is then cored from the sample and replaced with a cement plug.

Flow rates on the order of $1E-3$ to $5E-3$ cc/min were measured using gradients of 000 to 10,000. Under the initial triaxial stress conditions, corresponding to a depth of about 920 meters, flow through the plugged sample was about half the flow through the "intact" rock. Decreasing the triaxial stresses to values corresponding to a depth of about 625 meters increased the flow to roughly three-fourths the rate through "intact" rock; a further decrease in stresses to values corresponding to a depth of about 350 meters resulted in an increase in flow to about twice that of "intact" rock.

Three experiments have been performed using this granite/cement combination, each taking about four months. Measurements are made after steady state flow has been achieved. Further tests will use basalt and tuff rock types and clay as well as cement as a plug material.

D11.6
 THERMAL CONDUCTIVITY OF WASTE FORMS AND GEOLOGIC MEDIA. Robert O. Pohl, Laboratory
 of Atomic and Solid State Physics, Cornell University, Ithaca, NY 14853.

In order to predict the temperature rise in a nuclear waste repository, thermal conductivity and specific heat of the rock and of the waste forms must be known. In particular, causes for the variation of the thermal conductivity have to be understood, including the effects of ionizing radiation and of chemical alterations due to extended heating. Of great importance for safety assessments is the question whether a lower limit of the thermal conductivity exists for a material of a certain chemical composition, regardless of its disorder. These questions will be reviewed.

D11.7
 THERMAL CONDUCTIVITY OF BENTONITE/QUARTZ HIGH-LEVEL WASTE PACKAGE BACKFILL.* Marvin
 Moss and Martin A. Molecke, Sandia National Laboratories, Albuquerque, NM 87185.

High-level waste (HLW) package backfill must, in addition to its radionuclide-sorption and hydrologic-barrier functions, adequately conduct heat away from the waste, thereby preventing the development of excessive temperatures. Bentonite clay and quartz sand mixtures have been investigated and their thermal properties optimized. This study included the fabrication of bentonite and bentonite/quartz compacts by isostatic pressing, measurement of thermal conductivities from 298 to 573 K, development of a predictive model for thermal conduction, and analysis of the thermal impact on HLW package design.

Compacts of bentonite/quartz of 100/0, 70/30, 50/50 and 30/70 weight percent were made at room temperature under a pressure of 100 MPa (15 ksi). The thermal conductivity of the 70/30 compact rose from 1.20 W/m²K at 298 K to 1.32 W/m²K at 373 K. It then fell to 1.10 W/m²K upon further heating to 473 K, reflecting the loss of interlamellar water from the bentonite. The conductivity of the now-dehydrated compact was reproducible through several heating and cooling cycles, ranging from 1.15 W/m²K at 573 K to 1.03 W/m²K at 298 K. The other mixtures were qualitatively similar to the 70/30: the 100/0, 50/50 and 30/70 dehydrated compacts displayed conductivities of 0.59, 1.06 and 0.88 W/m²K, respectively, at 298 K. Measured densities ranged from 1.98 to 2.12 g/cc. Possible ways of increasing the thermal conductivity are being investigated.

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D11.8
 EXPERIMENTAL INVESTIGATION OF SODIUM BENTONITE STABILITY IN HANFORD BASALT.
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Sodium bentonite is a candidate material for the waste package backfill component in a basalt repository at the Hanford Site. Preliminary hydrothermal experiments have been conducted under near-field geochemical conditions expected to occur at the reference repository location in the Umatum flow of the Grande Ronde Basalt formation. Experiments have been conducted in the basalt/groundwater.

bentonite/groundwater, and basalt/bentonite/groundwater systems. The experiments have been conducted at 300°C using a simulated Grande Ronde groundwater, reference Umtanum basalt, and sodium bentonite. Key data generated by the experiments include experimental solution analyses as a function of time and preliminary solids analysis by scanning transmission electron microscopy and X-ray diffraction. Solution trends of the major aqueous species were similar in the three systems and are characterized by (1) the gradual reduction of the pH value from ~9.75 to a steady state value of ~6, (2) an initial rapid increase followed by a gradual decrease in silica concentration, and (3) a slight or negligible increase in sodium, sulfate, chloride, and fluoride concentrations. In the bentonite/water experiment small amounts (<1%) of an albite reaction product were observed. Conversely, the formation of illite, a common bentonite alteration product was not observed. These results indicate that sodium bentonite will remain sufficiently stable at 300°C under hydrothermal conditions in basalt to permit its use as a backfill material.

D11.9
 DIFFUSION OF RADIONUCLIDES IN BRINE-SATURATED BACKFILL BARRIER MATERIALS*
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Results of one-dimensional radionuclide diffusion experiments in closed columns of brine-saturated backfill materials are presented. Diffusion measurements specifically for the concentrated brines of a repository in salt are needed to provide an experimental basis for the development of realistic mass transport models. Distribution coefficients derived from diffusion measurements are compared with distribution coefficients derived from batch sorption measurements. For diffusion of $^{137}\text{Cs}^+$, $^{99}\text{TcO}_4^-$, and $^{152}\text{Eu}^{+3}$ in uncompacted mixtures of bentonite with other getters, the comparison shows good agreement between diffusion and sorption results. Average values of the distribution coefficients calculated from diffusion data were as follows:

19 ml/g for $^{137}\text{Cs}^+$ in 40 wt.% mordenite - 60 wt.% bentonite
 500 ml/g for $^{99}\text{TcO}_4^-$ in 70 wt.% charcoal - 30 wt.% bentonite
 3000 ml/g for $^{152}\text{Eu}^{+3}$ in 100 wt.% bentonite.

Additional diffusion data, experiments with apparatus for measuring diffusion in highly compacted bentonite, methods for data interpretation, and estimates of backfill barrier effectiveness are also presented.

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D11.10
 INERTIAL EFFECTS IN TRANSPORT OF RADIOACTIVE WASTE THROUGH ENGINEERED BARRIERS.
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Addition of an inertial term to the constitutive relation for mass flux leads to a mass transport equation which is hyperbolic and describes propagation of a distorted wave with a finite velocity. This approach eliminates the "instantaneous propagation paradox" inherent in parabolic transport equations based on Fick's law. Analytical solutions of the wave transport equation have been derived and have the properties that the leading edge of a solute front propagates at a finite, predictable velocity and is truncated by a step function which decreases in magnitude exponentially with time. The inertial effects on computed solute fronts are most evident near the leading edge, and have potential significance in the prediction of engineered barrier performance.