A REVIEW OF LEACHING TEST METHODS
AND THE LEACHABILITY OF VARIOUS
SOLID MEDIA CONTAINING
RADIOACTIVE WASTES

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

JULY 1973

Prepared for the U.S. Atomic Energy
Commission under Contract AT(45-1):1830

NOTICE

The report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY
operated by
BATTELLE
for the
U.S. ATOMIC ENERGY COMMISSION
Under Contract AT(45-1)-1830

Printed in the United States of America
Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy \$5.45; Microfiche \$0.95

3 3679 00062 2920

BNWL-1765
UC-70
Waste Disposal
and Processing

A REVIEW OF LEACHING TEST METHODS AND THE
LEACHABILITY OF VARIOUS SOLID MEDIA
CONTAINING RADIOACTIVE WASTES

Compiled by
J. E. Mendel
Nuclear Waste Technology Department

July 1973

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

ABSTRACT

A review of experimental techniques which have been employed to measure the leachability of solidified radioactive wastes in the United States and foreign countries is made. Factors which effect leachability are described. The need for two different kinds of leach tests is emphasized, (1) a rapid comparison test, and (2) a more comprehensive performance test, wherein the actual exposure environment is simulated.

CONTENTS

ABSTRACT	iii
LIST OF FIGURES.	vi
LIST OF TABLES	vi
I. INTRODUCTION	1
II. SUMMARY	2
III. REVIEW OF LEACH DATA.	4
A. Solidified Radioactive Waste Leach Data	4
B. Leachability of Industrial Glasses.	10
IV. MEASUREMENT OF LEACH RATES.	12
A. Presentation of Data	13
1. Methods of Expressing Leach Rates.	13
2. Unequal Leach Rates of Various Elements.	15
3. Precision of Leach Rate Data	17
B. Procedures Used for Leaching Solidified Radioactive Wastes.	18
1. English	18
2. French	18
3. German	21
4. Canadian	21
5. Indian	21
6. American	21
C. Industrial Glass Durability Tests	26
V. UTILIZATION OF LEACH DATA	29
VI. LEACH TEST STANDARDIZATION	33
A. Comparison of Proposed IAEA Method with Methods Presently in Use.	35
B. Comments Concerning Comparison Tests	38
C. Comments Concerning Performance Tests.	38
VII. REFERENCES	39

LIST OF FIGURES

1	Rate of Attack of Distilled Water on Industrial Glasses as a Function of Temperature	11
2	Cell for Leaching FINGAL Glasses	19
3	Schematic of Leaching Installation in French "Vulcain" Cell	20
4	Apparatus for Study of Leaching Kinetics at HMI	22
5	Canadian Solidified Radioactive Waste Leaching System	23
6	Leach Tests Used at Bhabha Atomic Research Centre, Bombay	24
7	Slow Leach Test Used at the Pacific Northwest Laboratory	25
8	Accelerated Leach Test Used at the Pacific Northwest Laboratory	27
9	Apparatus for Standard Leaching Test Proposed in 1957	34

LIST OF TABLES

1	Leach Rate Measurements on Solid Media for the Containment of Radioactive Wastes	5
2	Composition of Representative Industrial Glasses	10
3	Relative Leachability of Elements	16
4	Comparison of Present Leaching Methods with Proposed IAEA Standard Methods	36

A REVIEW OF LEACHING TEST METHODS AND THE
LEACHABILITY OF VARIOUS SOLID MEDIA
CONTAINING RADIOACTIVE WASTES

J. E. Mendel

I. INTRODUCTION

Knowledge of the behavior of solidified radioactive wastes when contacted with water is of fundamental importance, therefore some type of leach testing has been included in almost all investigations of techniques for solidifying radioactive wastes. Unfortunately the usefulness of much of the data has been decreased by lack of standardization. Many different types of leach tests have been utilized, and various kinds of units have been used to report the results. Thus obtaining information on the leaching of these solids by water, which at best involves an extremely complex process, has been made even more complex. In recognition of this problem, a subcommittee on Leach Rate Testing Methods was organized after the second meeting of the Waste Solidification Development Working Group* at Hanford in March 1973. The goals of the subcommittee are to further understanding of leaching mechanisms and measurements among waste solidification investigators in the United States, and to promote standardization of leach test methods. One purpose of this report, which is a review of leach test techniques and results, is to provide background material for the subcommittee. Results and recommendations from the subcommittee studies will be reported later.

---

* The Waste Solidification Development Working Group was organized by the AEC's Division of Waste Management and Transportation to promote information exchange on high level waste solidification. Members are from duPont, Oak Ridge National Laboratory, Allied Chemical Company, ARHCO and Battelle-Northwest and the corresponding AEC operations offices.

II. SUMMARY

Thirty-five references which describe leachability measurements on solidified radioactive wastes are reviewed. The references cover the period from 1956 to 1972 and include investigations from 10 countries. Emphasis is placed on studies of inorganic solid media suitable for the high temperatures and radiation fields associated with the incorporation of high level wastes, however a few studies of other solid media, suitable only for the incorporation of low and intermediate-level wastes, are included for comparison.

The available data indicate that the most nonleachable radioactive waste solids have a chemical durability that is probably at least equivalent to that of common window glass, however, more testing is required to establish this fact with certainty.

There are two principal uses to which the results of leachability measurements on solidified radioactive wastes are put:

Use I: Comparison of one method of waste insolubilization with another, both within a laboratory and between different laboratories.

Use II: Making estimates of the hazards arising from the contact of a solidified radioactive waste with water under given conditions during storage or shipping.

The two different end uses of the data influence greatly the experimental procedures required. The survey of the leach data on solidified radioactive wastes shows that the distinctions have not been realized in practice. In most cases the leach data available do not permit simple intercomparisons because of differences in the leaching procedures employed. Usually the data available on a specific waste solid are insufficient for an in-depth analysis of the hazards of a real-life situation in which the solid contacts water. Thus neither Use I nor Use II of the data are being completely satisfied.

A standard leach test, which can be used by all laboratories active in the study of waste solidification is required to satisfy the comparison

requirements of Use I. The recent proposal for a standard leach test by the IAEA (Hespe, 1971) recognizes this need, however the proposed IAEA procedure, which requires many weeks, is more tedious than necessary to satisfy the requirements of Use I. Accelerated leach tests of the type utilized in the glass industry require only a few hours and seem to have more promise than the IAEA procedure as a standard comparison test.

The application of leach data to Use II is very complex. For instance in the United States at least three quite different cases may be distinguished in which the leachability of solidified high level wastes can become potentially important.

Case A: Loss of waste container integrity in a destructive accident, such as a shipping accident.

Case B: Water leakage into a solidified waste container during storage of the container in a water basin.

Case C: Deterioration of waste container integrity over the long-term while in the Retrievable Solid Storage Facility (RSSF) or in the ultimate waste repository.

Case A is the situation which has been studied most extensively and it can be shown that low leach rates, which are within the range of practical achievement, reduce the hazards potential for specific examples of this case to a degree which is of fundamental importance. However, all situations involving potential contact with water must be analyzed. Temperature, flow rate of leachant, leachant composition, prehistory of the waste, radioisotope of concern, and other factors may differ significantly depending upon the situation being considered. It is improbable that the results of any single standard test procedure, such as the proposed IAEA standard test, can be extrapolated to all of the eventualities. Investigators obtaining data for Use II will probably continue to use techniques tailored to their specific requirements; in some instances elaborate facilities which differ from one another are already in use. Here it is important that all of the experimental details be reported fully and in more detail than is usually done, so that other investigators can assess their products under duplicate conditions if desired.

III. REVIEW OF LEACH DATA

A. Solidified Radioactive Waste Leach Data

The basic purpose in converting radioactive wastes to solid forms is to reduce the potential for contamination of man's environment. A principal mechanism for the spread of contamination is by dissolution of the radioactivity in the waters of the environment. For this reason nearly all studies on the solidification of radioactive wastes have included an assessment of the leachability of the resulting solid product.

The leach rate assessments should serve two functions, namely:

1. Comparison of the degree of insolubility obtained with different solid forms (the comparisons should be possible both within a given laboratory and between laboratories).
2. Assist in prediction of the performance of the solid forms when exposed to various environments.

Most of the leach rate data available only partially satisfy these functions. It is not surprising, considering the variety of solid forms which have been studied, and the many different workers who have made contributions, that a diversity of leaching techniques, and also of methods for expressing the leaching rates, are now to be found in the literature. This diversity is illustrated by the leach data in Table 1, which contains results from a representative cross section of radioactive waste solidification studies made during the last 16 years.

Because of the great variety of leaching techniques used, no attempt is made here to compare or rank the leachabilities of the various products shown in Table 1. However, several products have leach rates in the range of 10^{-6} to 10^{-7} grams of glass/cm²-day, which are 1 to 2 orders of magnitude lower than those of the best glasses produced in the fully radioactive Waste Solidification Engineering Prototype (WSEP) demonstrations. (Mendel 1972) It should be noted that none of the glasses with superior leachability contained the large quantities of fission products of the WSEP products (28 wt% fission product oxides in the best WSEP product).

TABLE 1. Leach Rate Measurements on Solid Media for the Containment of Radioactive Wastes

Identification of Solidified Waste	Approximate Composition, Wt%	Leach Rate	Leaching Technique	References
Fired Clay (Los Alamos, USA)	Bentonite-Montmorillonite mixture with dilute CaSO_4 waste solution (α activity)	0.001 to 0.018% activity removed, fired at 1000°C .	Soaked in sea water for 7 days. No further details.	Thomas, 1956
Fired Clay (ORNL, USA)	12 Al_2O_3 , 10 CaO , 18 Na_2O , 60 shale (^{90}Sr traced)	0.5% leached when fired at 850°C . 0.23% leached when fired at 1050°C .	Sample sealed in jar of tap water for one month at ambient temperature.	McVay, 1956
Ceramic Clinker (ORNL, USA)	45.57 SiO_2 , 16.17 Al_2O_3 , 5.20 Fe_2O_3 , 11.06 CaO , 2.88 MgO , 12.04 Na_2O , 2.37 K_2O , 0.23 other. (fired at 750°C)	1.0 mg/gm/ 4 weeks (density not given)	1-inch cube in 1000 ml H_2O at room temperature. 500 ml of leachate replaced with distilled H_2O weekly.	Hemphill, 1958
	(Same)	120 mg/gm/week (density not given)	Drops of distilled H_2O impinge upon 1-inch cube sample and fill sample chamber which drains by intermittent syphon action. Flow = 1.5 liters, room temperature.	
	(Same)	1.4 mg/gm/2 hr. (density not given)	1-inch cube in 1000 ml boiling distilled water	
Silicate Glasses (Canadian)	Lead borosilicate	6×10^{-6} gram glass/ cm^2/day	5 gm glass in alumina crucible placed in polythene bag with 20 ml water. Water changed daily.	Durham, 1958
	85 nepheline syenite, 15 CaO	2×10^{-7} gram glass/ cm^2/day		
	65 nepheline syenite, 35 CaO	5×10^{-6} gram glass/ cm^2/day		
Nepheline Syenite Glass (Canadian)	50.5 SiO_2 , 20.3 Al_2O_3 , 9.1 Na_2O , 4.2 K_2O , 14.9 CaO , 1 other. (melted at 1350°C)	5×10^{-5} gm of glass/ cm^2/day first day of leaching, 8×10^{-8} after 45 days, and still decreasing slowly.	Samples were blocks of glass weighing about 1950 grams with 500 cm^2 surface. Samples were immersed in distilled H_2O in a once-through all-polythene system. Flowrate 3 ml/min. Test duration 45 days. Temp. approximately 25°C .	Bancroft, 1958
Borosilicate Glass (English)	16-28 Al_2O_3 , 4-40 B_2O_3 , 13-49 CaO , 4-38 SiO_2	Leach factors range around 10^6 [leach factor units = $(\frac{\text{cm}}{\text{week}})^{-1}$]	Surface area of irregular-shaped samples determined by graphical technique. Seven leach procedures were used: procedures different in leachant (water, HNO_3 and NaOH), temperature (25°C , 80 - 90°C , and boiling), and apparatus (glass vs polythene)	Grover, 1960
Silicate Glasses (USA)	30.3 Al_2O_3 , 30 SiO_2 , 30 CaO , 9.3 ZrO_2 , 0.4 other (melted at 1400°C)	6.2×10^{-8} to 2.1×10^{-7} cm/week	Samples ground and sieved to lie between 40 and 60 mesh. Exposed to boiling distilled H_2O 6 to 28 weeks. Weight loss measured weekly.	Eliasson, 1961
	20.3 Al_2O_3 , 30 SiO_2 , 40 CaO , 9.3 ZrO_2 , 0.4 others (melted at 1350°C)	(same)		
Fiber Glass Soaked in Waste Solution and fused (French laboratory work)	14.7 Na_2O , 60.8 SiO_2 , 11.4 Al_2O_3 , 11.5 Fe_2O_3 , 0.8 CaO , 0.8 K_2O (melted at 1250°C)	6.6×10^{-7} gm of glass/ cm^2 -day after 3 weeks 2.4×10^{-7} gm of glass/ cm^2 -day after 8 weeks	Sea water, no other details given	Bonniaud, 1961

TABLE 1. (contd)

Identification of Solidified Waste	Approximate Composition, Wt%	Leach Rate	Leaching Technique	References
Ceramic Sponge (Coors Porcelain Company and LASL, USA)	80.5 SiO ₂ , 13.0 Al ₂ O ₃ , 6.5 R ₂ O (contained 26 wt% dried TBP-25 waste before firing at 1280°C)	Lost 0.003% of ⁹⁰ Sr and ¹³⁷ Cs activity in 14 days.	Distilled H ₂ O, room temperature, no other details given.	Kelly, 1961
Glass from Ferric Hydroxide Scavenging (Russian)	50 SiO ₂ , 10 Na ₂ O, 5 B ₂ O ₃ , 35 Fe ₂ O ₃ (spiked with ¹³⁷ Cs)	0.0135% total activity/day to 0.0048% total activity/day after 8 days. (Sufficient data is given to convert leach rates to 6.7x 10 ⁻⁶ g/cm ² -day initially, and 2.4x10 ⁻⁶ g/cm ² -day after 8 days).	3 gram sample with surface area of 60 cm ² contacted with 100 ml H ₂ O at 20°C. H ₂ O changed daily.	Brezhneva, 1963
Silicate Glass (Russian)	50 SiO ₂ , 15 Na ₂ O, 8.75 Fe ₂ O ₃ , 17.5 Na ₂ V ₂ O ₇ , 8.75 CaO: Melted 2 hr @ 1100°C Melted 6 hr @ 1100°C Melted 2 hr @ 1200°C	1.8x10 ⁻⁴ g/cm ² /2hr 3.0x10 ⁻⁵ g/cm ² /2hr 1.5x10 ⁻⁵ g/cm ² /2hr	Sample was broken up and fraction between No. 8 and No. 12 screens was exposed to H ₂ O at 90°C for 2 hours and reduction in weight determined. (Method of determining surface area of grains not given).	Golobanov, 1963
Phosphate Glass (ORNL, USA)	25.0 Al ₂ O ₃ , 18.6 Na ₂ O, 40.5 P ₂ O ₅ , 15.9 PbO, 0.06 Fe ₂ O ₃ , 0.01 RuO ₂ . (spiked with ¹³⁷ Cs)	2x10 ⁻⁶ to 1.5x 10 ⁻⁷ (rate still decreasing at 56 days when test was terminated)	Sample in form of button (weight not given) immersed in 500 ml distilled H ₂ O which was recirculated by pump at 100 ml/min., polyethylene apparatus. Water changed weekly.	Clark, 1964
Borosilicate Glass (Russian)	33 SiO ₂ , 10 B ₂ O ₃ , 10 Na ₂ O, 47 mixture of CaO, Fe ₂ O ₃ , Cr ₂ O ₃ and MgO	Damage depth, 5 - 30 [Angstroms?] in 300 min.	Sample contacted with distilled H ₂ O and electrical conductivity of solution measured as function of time. Damage depth calculated by "familiar" formula. (Russian reference)	Dubhovich, 1965
Borosilicate and Phosphate Glasses (Hanford, USA)	17 Al ₂ O ₃ , 12 Na ₂ O, 1 Fe ₂ O ₃ , 38 B ₂ O ₃ , 32 SiO ₂ , (full-level Hanford Redox plant waste), melted at 900°C.	9x10 ⁻³ cm/day based on ¹³⁷ Cs 4x10 ⁻⁵ cm/day based on ¹⁴⁴ Ce	Samples submerged in 100 ml distilled H ₂ O (not stirred) at room temperature in plastic container. Water changed daily. Rates plotted versus time and extrapolated back to zero.	Allemann, 1965
	27 Na ₂ O, 21 Fe ₂ O ₃ , 5 Al ₂ O ₃ , 47 P ₂ O ₅ (full level Hanford Purex plant waste), melted at 900°C.	2x10 ⁻² cm/day based on ¹³⁷ Cs 9x10 ⁻⁴ cm/day based on ¹⁴⁴ Ce		
Borosilicate Glass (Russian)	29 SiO ₂ , 5 B ₂ O ₃ , 5 Na ₂ O, 7 CaO, 18 SrO, 17 TiO ₂ , 6 Cr ₂ O ₃ , 3 Na ₂ O, 11 Fe ₂ O ₃ , plus many variations, this one of best. (Spiked with ⁹⁰ Sr).	2x10 ⁻⁶ g/cm ² /day after one week, 2x10 ⁻⁷ g/cm ² /day after ten weeks.	Distilled water, 20-30°C, volume did not normally exceed 10 cm ³ for each cm ² of sample surface. Water was changed weekly, and in a number of tests every day of the first week.	Kulichenko, 1966
Cement (Russian)	Portland cement and 100 g/l NaNO ₃ . Weight ratios, solution to cement of 1:1.2 to 1:2.3 (¹³⁷ Cs traced)	1x10 ⁻² to 1x10 ⁻³ g/cm ² -day based on geometrical surface area, but 1x10 ⁻⁵ to 1x10 ⁻⁶ based on real surface area measured by "thermal desorption" method.	Cubes, 3 cm on a side, or tablets 2.4 cm dia. and 1 cm high contacted with distilled H ₂ O with ratio of water volume (cm ³) to geometrical surface area (cm ²) not exceeding ten. Water changed daily for 7 days, then weekly. Tests at room temperature.	Kulichenko, 1966

TABLE 1. (contd)

Identification of Solidified Waste	Approximate Composition, Wt%	Leach Rate	Leaching Technique	References
Phosphate Glass (Hanford, USA)	70 P ₂ O ₅ , 15 Na ₂ O, 11 Fe ₂ O ₃ , 4 Al ₂ O ₃ (400 Cf/liters fission products)	1 mg/cm ² /day	4.0 hours at 25°C in 100 ml H ₂ O.	Upson, 1966
Phosphate Glass (Idaho Falls, USA)	25 Al ₂ O ₃ , 15.9 PbO, 40.5 P ₂ O ₅ , 18.5 Na ₂ O	1x10 ⁻⁵ gm of glass/cm ² -day during first week @ 25°C 1x10 ⁻⁶ gm of glass/cm ² -day after 7 weeks @ 25°C 2x10 ⁻⁴ gm of glass/cm ² -day @ 94°C, rate constant for 7 weeks	Samples weighing about 40 grams, surface area about 35 cm ² , in 500 ml distilled water at 25°C or 94°C, water recirculated at approx. 100 ml/min. H ₂ O changed weekly.	Paige, 1966
Basalt (Czechoslovakia)	36-50 SiO ₂ , 7-18 Al ₂ O ₃ , 9-17 Fe ₂ O ₃ , 10-20 CaO, 8-22 MgO, 5 Na ₂ O+K ₂ O (137Cs or 90Sr traced, melted at approx. 1250°C)	Expressed as diffusion coefficient cm ² /sec. Cs: 7x10 ⁻¹⁸ to 4.5x10 ⁻¹⁷ @ 30°C 2.1x10 ⁻¹⁷ to 1.4x10 ⁻¹⁶ @ 50°C. Sr: 1.3x10 ⁻¹⁶ to 4.4x10 ⁻¹⁶ @ 30°C 5.4x10 ⁻¹⁶ to 8.6x10 ⁻¹⁶ @ 50°C.	Samples contacted with distilled H ₂ O. Leach H ₂ O changed periodically. No other details given.	Ralkova, 1967
Phosphate Glass (Brookhaven National Laboratory, USA)	68 P ₂ O ₅ , 11 Fe ₂ O ₃ , 4 Na ₂ O, 17 simulated fission products	1x10 ⁻⁶ cm/week in water. 2.5x10 ⁻⁶ cm/week in dil. HNO ₃ (pH=4.0)	35 to 45 mesh sample, no other details given.	Tuthill, 1967
Borosilicate and Phosphate Glasses (English)	30-32 Na ₂ B ₄ O ₇ , 35-43 SiO ₂ , 25-35 waste oxides (12-17 simulated fission products).	1x10 ⁶ (cm/week) ⁻¹ @ 60°C 4x10 ⁶ (cm/week) ⁻¹ @ 30°C No difference between distilled H ₂ O and sea H ₂ O.	Sample crushed and -16 + 22 B.S. sieve fractions retained. 10 gram powder samples leached in automated once-through system, flow rate 300 ml/week. Samples in Teflon cells. Leach test duration generally 100 days.	Elliot, 1967
	47.7 Na ₂ O·P ₂ O ₅ , 5.3 SiO ₂ , 7.0 Al ₂ O ₃ , 40 waste oxides (19 simulated fission products).	10 ⁵ (cm/wk) ⁻¹ @ 60°C 10 ⁶ (cm/wk) ⁻¹ @ 30°C No difference between distilled H ₂ O and sea H ₂ O.		
Phosphate Ceramic (Hanford, USA)	A large number of compositions within the range: 8-31 Na, 2-31 Al, 6-11 Fe, 2-3 Ni+Cr, 47-57 P ₂ O ₅ , 5-26 simulated Fission products.	Cast from 1200°C: Predominantly between 0.28 and 6.7%wt loss, few compositions ranged to 27% wt loss. Slow cooled at rate of 1°C/min: 2.3 to 59.7% weight loss.	Approx. 1 gram powder sample (45-60 mesh particle size) in stainless steel "tea bag" immersed in stagnant 150 ml distilled at 95°C. Water changed daily for three days. Results reported as weight loss.	Barton, 1968
Thermite (National Lead, USA)	FeAl silicate (137Cs tracer)	1.5x10 ⁻⁴ to 0.1x10 ⁻⁴ g/cm ² /yr at 73°C (not ground) 1.7x10 ⁻⁴ to 0.1x10 ⁻⁴ g/cm ² /yr at 100°C (ground)	Approx. 4 g sample leached in 100 ml water. Water changed at 2 or 8 hr intervals, 18 hrs total leach max. Surface area measured on duplicate nonradioactive sample, 1.6±0.5x10 ⁴ cm/g	Spector, 1968

TABLE 1. (contd)

Identification of Solidified Waste	Approximate Composition, Wt%	Leach Rate	Leaching Technique	References
Basalt (East Germany)	38 SiO ₂ , 20 Al ₂ O ₃ , 5 TiO ₂ , 8 Fe ₂ O ₃ , 6 FeO, 14 CaO, 3 MgO, 2 K ₂ O, 4 Na ₂ O (137Cs and 90Sr tracer added to powder, melted at 1200°C). Many other basalts and clays included in the study.	0.03% Cs leached in 2nd contact, 0.02% Cs leached in 3rd contact, 0.39% Sr leached in 2nd contact, 0.24% Sr leached in 3rd contact.	Approx. 2 gram sample contacted with 3 successive 100 ml portions distilled water. Each contact 4 hrs at reflux. Results of 1st contact not given because of potential sample contamination. (Sample surface area not given)	Sachse, 1968
Vermiculite Sludge Glasses (India)	21.5 SiO ₂ , 10.6 Al ₂ O ₃ , 6.7 Fe ₂ O ₃ , 8.2 MgO, 11.2 CaO, 21.2 Na ₂ O, 10 CaF ₂ , 10 Ca ₃ (PO ₄) ₂ (melted at 1050°C), and many similar compositions.	10 ⁻⁸ to 10 ⁻⁵ cm/wk.	Samples ground & screened to -40+60 mesh size. 1 grain samples then exposed to boiling distilled H ₂ O by one of two techniques: A. Enclosed in 200 mesh stainless steel cloth "tea bag" hung in 100 ml H ₂ O, changed weekly. B. Water at 97-99°C returning from reflux condenser contacts sample. 100 ml H ₂ O in bottom vessel changed weekly.	Rastogi, 1969
Cement (English)	50 cement - 50 sludge solids	1x10 ⁻⁴ g cm ⁻² d ⁻¹ the 1st day, 1.4x10 ⁻⁵ g cm ⁻² d ⁻¹ after 64 days.	1320 g samples, 619 cm ² surface area, "tap" water as leachant at 25°C. Water changed daily for 7 days, then weekly.	Smith, 1969
Polyethylene (ORNL, USA)	40 NaBO ₂ - 60 DYL polyethylene	5x10 ⁻³ fraction of sodium leached (cm ² /g)-1 day ⁻¹ , initially to 2x10 ⁻⁴ fraction of sodium leached (cm ² /g)-1 day ⁻¹ after 3 weeks.	100-250 cm ³ sample in 500 ml distilled H ₂ O at ambient temperature. Water changed weekly.	Fitzgerald, 1970
Fluidized Bed Zirconia Calcine (Idaho Falls, USA)	16 Al, 17.7 Ca, 20.3 F, 2.2 NO ₃ ⁻ , 16.7 Zr, (remainder oxygen) As produced calcine, average operating temperature, approx. 500°C. Calcine heated to 1200°C (sintered), 95% of 137Cs removed by volatilization.	40% 137Cs leached in one hour. 58% 137Cs leached in 2000 hours 18% 137Cs leached in one hour 20% 137Cs leached in 400 hours.	Calcine supported on fritted glass, 500 ml H ₂ O recirculated over 10 g calcine continuously at 100 ml/min at 25°C. Fresh distilled H ₂ O used for each leach period, leach periods averaged about one week.	Wilding, 1969 Wilding, 1971
Thermalit (Hanford, USA)	48-56 SiO ₂ , 1.2-11.2 Fe ₂ O ₃ , 15-34.8 Al ₂ O ₃ , 2-9.1 Na ₂ O, 0.1-3.8 K ₂ O, 1.3-10.3 CaO, 2.4-6.3 CaO, 0.2-2.1 simulated FP oxides.	1.4-2.0x10 ⁻⁷ g/cm ² /day	Powder sample (250-420 micron particles) leached with recirculating deionized water.	Brownell, 1971
Phosphate Glass (Italian)	3 to 13 wt% simulated fission product oxides, ΣM ⁺ /P = 1.0	10 ⁻⁶ to 10 ⁻⁷ g/cm ² -day	Distilled H ₂ O, 25 to 95°C, no other details.	Bocola, 1971

TABLE 1. (contd)

Identification of Solidified Waste	Approximate Composition, Wt%	Leach Rate	Leaching Technique	References
Borosilicate (Germany)	13 Al ₂ O ₃ , 5 B ₂ O ₃ , 55 SiO ₂ , 12 Na ₂ O, 15 CaO and other somewhat similar compositions.	2.3% Na leached 1st 7 days.	Sample powdered and 0.040 to 0.063 mm particles used for leaching in once-through apparatus. Flow 175 ml/wk. Apparatus is Lucite, sample suspended on nylon sieve. Temperature can be controlled from 20 to 50°C. pH is maintained between 4 and 5.	Heimerl, 1971
Asphalt (Summary of Data from MoI and Marcoule)	20-60 dry solids in asphalt.	3.9×10^{-6} g/cm ² -d ⁻¹ average over periods of 201 - to 2048 days.	Ground water, ambient temperature. No details given.	Burns, 1971
WSEP Products (Hanford, USA)	Phosphate Glass: 63 P ₂ O ₅ , 7 Na ₂ O, 8 Fe ₂ O ₃ , 22 FP oxides (12 Ci/gram initially in 8-inch pot)	Air quenched, 2×10^{-6} g/cm ² -day initially, 6×10^{-7} g/cm ² -day after 40 weeks 15 months pot storage; 6×10^{-3} g/cm ² -day initially, 3×10^{-4} g/cm ² -day after 40 wks.	500 ml distilled water recirculated continuously at approx. 200 ml/min over 5-15 gram samples at ambient temperature. Fresh distilled water used each leach period, leach periods were 1 week initially, 4 weeks after 3 months.	Mendel, 1972
	Phosphate Ceramic: 46 P ₂ O ₅ , 8 Na ₂ O, 18 Fe ₂ O ₃ , 28 FP oxides. (13 Ci/gram initially in 8-inch pot).	Air quenched; 3.0×10^{-3} g/cm ² -day initially, 4.4×10^{-4} g/cm ² -day after 40 weeks. 19 months pot storage; 4×10^{-3} g/cm ² -day initially 6.4×10^{-4} g/cm ² -day after 12 weeks.		
	Borosilicate Glass: 22 SiO ₂ , 25 B ₂ O ₃ , 10 Na ₂ O, 11 ZnO, 2 CaO, 2 Al ₂ O ₃ , 28 FP oxides. (7 Ci/gram initially in 8-inch pot).	7 months pot storage; 5×10^{-5} g/cm ² -day initially, 8×10^{-6} g/cm ² -day after 40 weeks.		

B. Leachability of Industrial Glasses

It is of interest to compare the leachability characteristics of the glasses being tested for the incorporation of radioactive wastes with industrial glasses of proven durability. Data suitable for comparisons are surprisingly sparse, but the data shown in Figure 1 are sufficient to permit at least a limited comparison. The compositions of the five glasses represented in Figure 1 are shown in Table 2.

TABLE 2. Composition of Representative Industrial Glasses

	Composition, wt%				
	Soda-lime Window Glass	Lead-Alkali Silicate Electrical	Borosilicate Low Expansion (Pyrex)	High-Lead Silicate	Jena Geräte Glass
SiO_2	73	63	81	35	65
Al_2O_3	1	1	2	--	6
Na_2O	17	7	4	--	7
K_2O	--	7	--	7	--
B_2O_3	--	--	13	--	10
CaO	6	--	--	--	--
MgO	3	--	--	--	--
PbO	--	22	--	58	--
ZnO	--	--	--	--	12

Most of the measurements on industrial glasses have been made at elevated temperatures in order to obtain more rapid measurements of the attack on the very corrosion resistant materials. Extrapolation to room temperature permits comparison with radioactive waste solids data. It is evident that the best nonleachable radioactive waste solids have approximately the same chemical durability as common window glass. The best industrial glasses are at least a factor of 10 less leachable.

FIGURE 1. Rate of Attack of Distilled Water on Industrial Glasses as a Function of Temperature

IV. MEASUREMENT OF LEACH RATES

The rate of leaching of radioactive isotopes from solidified wastes is generally not constant but is dependent upon the duration of contact with water. The leaching rate is maximum during the initial contact with water. Then the leaching rate usually decreases and finally becomes almost constant. The magnitude of the decrease in leach rate and the time required for the leach rate to reach an essentially steady state value differ with different solid forms.

The solid form for which the most leaching information exists is glass. The chemical durability of glass has been studied extensively because this characteristic is of great importance in most commercial applications of glass. The large amount of work which has been done on this solid form establishes mainly that leaching is a very complex phenomena. A recent review states that "...very different mechanisms of corrosion must be involved in the chemical durability of different types of glass-" and "Even among [a] group of relatively closely related glass compositions one cannot derive a general time law." (pp. 1106-7, Weyl, 1967) The time law expression which has found most widespread application as an approximation of the leaching behavior of many glasses is:

$$Q = at^{1/2} + bt \quad (1)$$

where Q is the cumulative quantity leached and a and b are empirical constants. At short times leaching is predominately diffusion controlled and the amount leached varies with the square root of time, however, as leaching continues for long times the rate becomes linear with time. (Douglas, 1967; Luce, 1972) The approximate diffusion control of the leaching of many solids incorporating radioactive wastes has been established over the short-term. Data which establish the long-term leach behavior are scarcer.

A. Presentation of Data

1. Methods of Expressing Leach Rates

Many different methods have been used for expressing the leach rate of solidified radioactive wastes. (See Table 1.) In recent years, however, most workers have reported their results in units having the dimensions of g of solid/cm²-day, or in units which can be converted readily to these units. These units are obtained from the following formula:

$$\frac{(\text{activity of isotopes leached})/\text{day}}{(\text{initial activity of isotopes in solid}) \frac{\text{Sample area, cm}^2}{\text{Sample weight, g}}} \quad (2)$$

$$= \frac{\text{fraction leached/day}}{\text{cm}^2/\text{g}} = \text{g/cm}^2\text{-day} \quad (3)$$

Expression (2) can also be stated as follows:

$$\frac{(\text{activity of isotopes leached})}{(\text{initial specific activity of isotopes in solid})(\text{sample area, cm}^2)(\text{time, days})} \quad (4)$$

If the activity of the solid is given per unit weight the rate has the dimensions g/cm²-day, while if it is given per unit volume (cm³) the dimensions are cm/day. The recent IAEA proposal for a standard leach method recommends the use of the latter rate, i.e., an incremental leaching rate, R_n , which has the dimensions cm/day: (Hespe, 1971)

$$R_n = \frac{a_n/A_o}{(F/V)_{t_n}}$$

where a_n = radioactivity leached during the leachant renewal period, n

A_o = radioactivity initially present in specimen

F = exposed surface area of specimen (cm²)

V = volume of specimen (cm³)

t_n = duration (days) of leachant renewal period.

The English use the inverse of the above rate, except that time is expressed in weeks instead of days, to obtain a "leach factor" which has the dimensions of week/cm. (Elliot, 1967) The leach factor is related to the more common leach rate as follows:

$$\frac{(\text{density of solid, g/cm}^3)}{(\text{leach factor})(7)} = \text{g/cm}^2\text{-day} \quad (6)$$

Since the rate of leaching usually varies with time it is common practice to present leach data as a function of time, often in the form of a plot of incremental leach rates versus time. The other method of reporting leach test results which is recommended in the IAEA proposal is by means of a plot of the cumulative fraction of radioactivity leached from the sample as a function of the total time of leaching:

$$\frac{\sum a_n}{A_0} \bigg/ \frac{F}{V} \text{ versus } \sqrt{\sum t_n} \quad (7)$$

$$\frac{\sum a_n}{A_0} \text{ versus } \sqrt{\sum t_n} \quad (8)$$

The latter method is of particular value because the diffusion coefficient of the radioactive isotope which was analyzed to obtain the leach rate can be determined from the slope of the plot, if it is a straight line.

Leaching experiments with many types of solidified wastes, including silicate, borosilicate, and phosphate glasses and asphalts and polyethylenes containing waste solids, have shown that elution of ions often follows, or is reasonably approximated, by Fick's law of diffusion. When this is true a plot of $\frac{\sum a_n}{A_0}$ versus $\sqrt{\sum t_n}$ is a straight line and for the case of a semi-infinite slab the diffusion coefficient D is expressed by

$$D = \frac{\pi}{4} \left(\frac{V}{F} \right)^2 m^2 \quad (9)$$

Where m is the slope of the straight line from a plot of $\frac{\sum a_n}{A_0}$ versus $\sqrt{\sum t_n}$.

A principal value of utilizing the diffusion coefficient in studying leachability is that it is a reproducible constant, independent of exposed surface or effective volume, dependent only on the temperature, which can be used to extrapolate leaching behavior over long periods of time for solidified wastes of various geometries. (Thomas, 1958; Saidl, 1966) Expressions have been derived for the cumulative long-term release of radioactivity, assuming a diffusion-controlled mechanism, which take into account the effect of radioactive decay. (DeJonghe, 1968; Godbee, 1969; Bell, 1971) Usually long-term leaching reveals additional mechanisms are involved, as represented by the second term of Equation (1), which decreases the accuracy of projections based on a single diffusion coefficient. (pp. 352-353, Rálková, 1962)

Leaching of high-surface solids such as concretes and calcines is generally not diffusion controlled. (Godbee, 1969) Expression of the leaching of radioisotopes incorporated in concrete in the units of $\text{g/cm}^2\text{-day}$ usually yields relatively high values when the geometric surface area is used. Russian experiments have shown that when the true surface area exposed to the leaching solution was determined, and used in the calculation, the nominal leach rate was decreased by a factor of 8,000. (Kulichenko, 1966)

The leach rate of calcines is generally expressed as cumulative percent of the initial activity leached as a function of time. The use of these units implies that the absolute quantity of radioactivity leached is directly proportional to the volume of calcine. These units cannot be applied to the leaching of monolithic solids where the quantity leached does not increase in direct proportion to the volume, but in a manner which is dependent upon the geometry of the solid.

2. Unequal Leach Rates of Various Elements

The leaching of most solidified wastes is incongruent, that is, certain constituents leach faster than others, at least in the initial phases of leaching. In general elements of low valence and small ionic radius tend to leach most readily. (Elliot, 1967) Since the

leach rate, as usually measured on solidified wastes, is dependent upon the fraction of a specific isotope leached (Equation 3), it is obvious that different leach rates can be obtained for the same solid, depending upon which element is being followed. It is good practice to state which element was analyzed when leach data are given, for instance, "The leach rate was 2×10^{-5} g of glass/cm²-day based on ¹³⁷Cs behavior."

The results of several investigations indicate a similar relative leachability for most elements from many different forms of solidified wastes (Table 3).

TABLE 3. Relative Leachability of Elements

Solid Type	Relative Leachability	Reference
Silicate Glass	Cs > Sr > Fe > Zr	Eliassen, 1961
Asphalt	Cs > Sr > Ce	Burns, 1971, p. 591
Alumina Calcine	Cs,Sr > Ru,Ce,Al	Paige, July 1966
Lead Glass	Cs > Al > Pb,Sr,Ce	Paige, February 1966
Phosphate Ceramic	Na,Cs > Sr > Zr-Nb > Ru,Ce	Allemann, 1965
Borosilicate Glass	Cs,Na > Zr-Nb > Ru,Ce	Allemann, 1965
Borosilicate Glass	Na > Si > B > Cs > Sr > Ce,Tb	Elliot, 1967
Borosilicate Glass	Na > B > Si	Heimerl, 1971, p. 34
Phosphate Glass	Cs > Na,Sr > Ru > Ce	Mendel, 1972

In general cesium and strontium, the long-lived fission products of most biological concern, are among the most leachable elements. Comparison of cesium leach data with actual weight loss measurements made during leach tests of several solidified wastes indicated that cesium was preferentially leached over the bulk solid by a factor of 2 to 4. (Mendel, 1972) In contrast the leaching of cerium, an element which is among the most nonleachable in all the solid forms (Table 3), was a factor of about 1000 less than that determined by weight loss in both phosphate and borosilicate glasses. Allemann found that cerium was 2.5 magnitudes less leachable than cesium from a borosilicate glass. (Allemann, 1965)

Aside from cesium and strontium, the other elements of most biological concern in solidified wastes are the transuranics. Very little, if any, leaching studies of these elements have been made on glasses. By chemical analogy with the rare earths it may be assumed that the transuranics would also be relatively nonleachable. However, Grover has reported that, in Britain, "In some of the early work, leach factors for uranium were obtained similar to the values now obtained for cesium and strontium." (Grover, 1966) Plutonium was found to be highly resistant to leaching from fluid bed calcine, about 0.1 percent being leached while 60 percent of the cesium was leached. (Wilding, 1969)

3. Precision of Leach Rate Data

The solubility of most chemical compounds in water can be measured very accurately and, for a given temperature, can be expressed precisely by one number. In contrast the corrosion, or leaching, of the multi-component glasses, and other solids being considered for the incorporation of radioactive wastes, is a different, usually nonequilibrium, complex phenomenon, and the precision or reproducibility of experimental results is lower than that associated with solubility measurements on simple compounds under equilibrium conditions.

The magnitude of variation in duplicate leach measurements, or in measurements on duplicate samples, has been reported by many workers. (p. 73, Rastogi, 1969; p. 8, Bancroft, 1958; p. 4.33, Thompson, 1970; p. 17, Allemann, 1965; p. 5, Elliot, 1967) Variations reaching as high as a factor of 5 to 10 for single determinations are not uncommon, although generally somewhat better precision is obtained. Apparently only Rastogi has made a statistical error determination. He reported 50 percent of the leach rate values laid within 22 percent error. (p. 73, Rastogi, 1969)

It is probable that much of the poor precision in leach rate determination is due to poor precision in determining the actual surface area exposed to the leachate. This is particularly true when the geometrical surface area, which ignores variations in surface roughness and porosity, is utilized.

B. Procedures Used for Leaching Solidified Radioactive Wastes

Leach rate measurements have been made in equipment ranging from simple breakers to complex automated facilities. In the European countries, particularly, elaborate facilities have been constructed to measure the leachability of candidate solidified waste forms.

1. English

After considerable development a method in which powdered samples were leached was used to evaluate the English FINGAL glasses. A diagram of one of the leaching cells is shown in Figure 2. Eight of the cells were arranged in a circle in each leaching rig, and three such rigs were constructed. (Elliot, 1967) Each rig was surrounded by an enclosure in which the air temperature could be maintained constant to within $\pm 0.5^{\circ}\text{C}$ over the temperature range 30 to 60°C . Once-through flow of the leachate was provided by an automated rotating pipette which delivered a preset volume of fresh leachate (0.5 to 5 ml) to each cell every 20 minutes. The English studies used glasses containing tracer quantities of radioactivity.

2. French

The French are measuring the leachabilities of radioactive glasses in one of the cells of their VULCAIN facility at Marcoule, using a system which regulates leaching conditions and analyzes the leachate automatically under computerized control. (Jouan, 1971) A shielded VULCAIN cell encloses eight identical leach devices permitting simultaneous study of the leaching of eight different radioactive glass compositions, which are prepared in the large adjoining vitrification cell in the VULCAIN facility. A glove box, located at the back face of the leach cells, encloses all supplementary equipment necessary for automatically sampling, counting, and replacing leachate daily. Glass blocks, suspended in baskets supported in the leaching tanks, are subjected to a dynamic leaching process in which the blocks are alternately covered with water and drained free of water on about a 4-minute cycle. The system is shown schematically in Figure 3.

(From Elliot,
1967)

FIGURE 2. Cell for Leaching FINGAL Glasses

FIGURE 3. Schematic of Leaching Installation in French "Vulcain" Cell

3. German

In the German research on waste solidification at the Hahn-Meitner Institute the glasses are powdered and the fractions with particle sizes between 0.040 and 0.063 mm diameter are leached in a specially constructed leaching facility. The leaching facility consists of six leaching units (Figure 4). Each unit can hold eight glass samples for long-term leaching tests at controlled temperatures between 20 and 50°C ($\pm 0.5^\circ\text{C}$). (Heimerl, 1971) The glass powder samples are held on nylon screens in the bottom of lucite columns. The once-through leachate flow can be programmed to give a weekly throughput of 20 to 200 ml. Only nonradioactive glasses are leached in this facility.

4. Canadian

The pre-leaching of the radioactive glass blocks which were buried in the ground at Chalk River was done in five identical sets of leaching apparatus, one of which is diagramed in Figure 5. Each glass block was an approximate hemisphere 5 1/2 inches in diameter and 3 inches high and weighed about 1950 grams. (Bancroft, 1958) Four blocks were usually leached together in one apparatus. Once through flow of the leachant (3 ml/min) was used, with an overflow weir which maintained a water depth of 4 inches, sufficient to completely cover the glass. All equipment surfaces that contacted the leachant were polythene.

5. Indian

A large number of glasses were leached in apparatuses of the type shown in Figure 6 during an investigation conducted in India on the incorporation of radioactive wastes in glasses. (Rastogi, 1969) A leaching setup containing 12 each of apparatus A and apparatus B was utilized. The leachates were changed and sampled on a weekly basis. Most of the glasses leached contained small quantities of radioactivity.

6. American

The most highly radioactive samples have been leached in apparatuses of the type devised by Paige, shown in Figure 7. Samples of solidified wastes from the WSEP program at Pacific Northwest Laboratory containing

RATE OF LOWERING CONTROLS
LEACHANT FLOW RATE

ADJUSTED TO pH4
WITH HNO_3

FEED TANK MAINTAINED ABOVE 50°C
TO PREVENT GAS BUBBLE FORMATION
ON SAMPLE

WATER JACKET @ $50 \pm 1^\circ\text{C}$

PLEXIGLAS

POWDER SAMPLE SUPPORTED ON
NYLON SCREEN. GRAIN SIZE
USUALLY 0.004-0.006 cm

FIGURE 4. Apparatus for Study of Leaching Kinetics at HMI (Six Leaching Apparatuses were Constructed, Each Holding Eight Units as Shown)

(From Bancroft, 1958)

FIGURE 5. Canadian Solidified Radioactive Waste Leaching System

(From Rostogi,
1969)

FIGURE 6. Leach Tests Used at Bhabha Atomic Research Centre, Bombay

FIGURE 7. Slow Leach Test Used at the Pacific Northwest Laboratory

up to 150 Ci/gram have been leached in these apparatuses. The apparatus shown in Figure 6 is designed to leach one-piece samples up to about 1-inch across. Powdered glass or calcine can be leached also if the sample is held in a porous bag or supported on sintered glass. The ratio of leachant volume to sample volume is comparatively high in this leaching technique, being usually 100 or higher. Dynamic contact between leachant and solid is maintained by an airlift recirculator. The leachant is changed periodically, usually daily or weekly during the initial time period when the leach rates are maximum, and at less frequent intervals thereafter.

An "accelerated" leach test was utilized at Hanford for comparisons of many glass and ceramic formulations for the solidification of radioactive wastes. (Barton, 1965) In the "accelerated" leach test the sample was crushed and sieved and an approximately 1 gram sample of -45 +60 mesh particles was placed in a stainless steel "tea bag" and suspended in 150 ml distilled water at 95°C (Figure 8). The leach water was changed and the weight loss determined at 24-hour intervals.

The proceeding review of leaching procedures, while not totally comprehensive, does include most of the major techniques that have been used to date for detailed study of the leaching behavior of candidate materials for the encapsulation of high-level radioactive wastes. The diversity of the techniques is obvious. This in spite of the early recognition that standardization was extremely desirable to permit ready comparison of the leaching behavior of solids of differing compositions, and solids prepared in different laboratories.

C. Industrial Glass Durability Tests

Glass has been a major material of construction for a very long time. The chief characteristic that makes glass preferred over physically stronger or less brittle materials in many applications is its superior resistance to corrosion in aqueous and atmospheric environments. Methods of testing and rating the resistance to corrosion, or "chemical

FIGURE 8. Accelerated Leach Test Used at the Pacific Northwest Laboratory

durability," of glasses have therefore been of fundamental importance and many tests have been developed, particularly during the last 80 to 90 years, for the study of chemical durability.

The best test is actual service in the particular purpose for which an article is designed, however such a test is time consuming. Almost all industrial glass durability tests attempt to shorten the time required by one or more of the following techniques; use of drastic reagents, increasing the surface through the use of powdered samples, use of high temperatures, or use of sensitive methods to detect and measure the corrosive action. (p. 106, Morey, 1954) The ASTM standard test for "Resistance of Glass Containers to Chemical Attack" (ASTM Designation: [225-68]) subjects the samples to a temperature of 121°C for 30 minutes in an autoclave. The results are reported as the number of milliliters of 0.020 N H_2SO_4 required to neutralize the NaOH extracted from the glass during the high temperature treatment. A 10 gram sample of -40 +50 mesh powdered glass is contacted with 50 ml of distilled H_2O in the test. Alternative tests in which the glasses are not powdered, but serve as the containers for the leach water, are also possible in the ASTM 9225-68 procedure.

In continental European glass technology a somewhat similar standard test, originated in Germany, is used, the "test of hydrolytic resistance" (Standard test DIN 12111). In this test 1 gram samples of powdered glass (0.3 to 0.5 mm diameter particles) are leached for 1 hour at 98°C in 50 ml distilled water. The results are reported as the number of milliliters of 0.01 N HCl consumed in titrating the leach water to neutrality. (Volf, 1961) A powder method has also been adopted as the standard test for chemical durability by the English Society of Glass Technology.

The powder tests have a great advantage in that they give a measurable result quite quickly. The results can be used to divide glasses into different classes of corrosion resistance. This has some utility in identifying large differences between glasses, but it is dangerous to extrapolate behavior in different circumstances from the results of

such tests, since most glasses differ in the degree of their reactions to changes in temperature, leachate composition, etc. It is probably for this reason that, although "standard" tests have been put forth, an examination of the technical glass literature quickly shows that the experimental techniques in actual use to measure leachability are at least as diverse as those that have been used in studying solidified radioactive wastes. The investigators utilize test conditions and leaching techniques which fit their individual requirements.

V. UTILIZATION OF LEACH DATA

The techniques used for leach testing industrial glasses vary depending upon the uses to which the data are to be put. Most of the tests are "accelerated" tests, which permit data to be collected rapidly so that the effects of a specific variable, for instance, the concentration of a certain constituent, can be studied over a wide range within a practical time scale. These results permit comparisons to be made, but do not necessarily correlate directly to the behavior of the glasses in specific use situations. There is general agreement that the behavior of a glass for a specific use can be determined unambiguously only by a test which simulates the use conditions as closely as possible. (p. 19.15, Shreir, 1963; p. 115, Morey, 1954)

Leach data from solidified radioactive wastes must serve the same two general purposes as leach data from industrial glasses, namely:

- Use I. Comparison of one glass with another, both within a laboratory and between different laboratories.
- Use II. Estimation of the behavior of the glasses in actual use situations.

There is a difference in emphasis however. Most of the industrial leach data on glasses is collected for Use I. This is because a long history of practical experience with glasses preceded the application of the scientific method to their improvement, hence glasses which have proven satisfactory in a certain application can be used as basepoints

for Use I comparison tests, and Use II data are often unnecessary. Use II data assume a much greater importance in the study of glasses for the immobilization of radioactivity, however, since there is almost no practical experience in this area. It is true also that, because of the limited experience in this unique application, optimum glass formulations have not been developed for incorporating radioactive wastes in glasses, and there is much need for Use I comparison test data. The conclusion is that both accelerated test (Use I) and longer-term environmental test (Use II) data on solidified radioactive wastes are needed with equal urgency.

Data for Use I comparison tests must be obtained at carefully defined, easily reproducible, standard conditions. While the conditions should be realistic they do not have to correspond accurately to any real-life situation. On the other hand, Use II data, by definition, must reflect real-life situations so it is necessary to define what those situations may be.

The leachability of solids incorporating radioactive wastes becomes important in any event in which water reaches the solids, and performance or Use II type data, may be required for any of these circumstances. Since the overall waste management plan in the United States is carefully designed to prevent contact between waste solids and water, any contacts which may occur are not easy to define. However, some suppositions concerning the contact conditions must be made in order to determine what type(s) of leach data is most meaningful.

At least three quite different cases may be distinguished in which the leachability of solidified high level wastes potentially can become important:

Case A: Loss of waste container integrity in a destructive accident, such as a shipping accident or bombing.

Case B: Water leakage into solidified waste container during storage of the container in a water basin.

Case C: Deterioration of waste container integrity over long-term while in the RSSF or the ultimate waste repository.

Although not often specified it is probably safe to assume that, up to the present time, Case A has been of most concern to investigators, with Cases B and C receiving much less consideration. The goal has been to compare the degree of hazard protection which would be presented by the resistance to leaching of candidate solid forms in an undefined catastrophic accident situation. Case B probably presents the most likely situation in which water will contact solidified wastes in the United States, however, in this case, the containers will be under controlled surveillance within secondary containment and corrective measures can be instituted safely. The parameters of importance in Case C are very dependent upon the nature of the ultimate repository. Investigations must be tailored to the conditions in the locality of the repository. For instance, if bedded salt is used as the final repository, slow dispersion within the salt bed is accepted. (Nichols, 1971) The long-term integrity of the salt bed itself becomes the principal reliance, and finally, after sufficient time has elapsed, the concentration of radioactivity in the salt bed will resemble other natural geological formations. The Canadian tests of the burial of unclad solidified waste are an example of another variation of Case C wherein principal reliance is placed upon low leachability of the solidified wastes, and upon the ion exchange characteristics of the surrounding soil, to prevent the dispersion of radioactivity. (Merritt, 1967)

In assessing consequences of an accident situation, such as Case A, the primary concern is that the environment can be restored to as near original condition as possible with minimum radiation exposure to the population. Retrieval of all appreciable quantities of solidified wastes which are released is essential. However, retrieval will be complicated if the released waste contacts water. Water presents a path for dispersal of dangerous quantities of radioactivity before retrieval can be effected and consideration of this situation offers an opportunity to place in perspective the importance of producing solidified waste forms of low leachability.

A maximum permissible leach rate can be calculated based on the maximum permissible radioisotope concentration in the water contacting the waste: (Appendix B, Watson, 1958)

$$L = \frac{KV}{930 \phi C} \quad (10)$$

where,

L = leach rate, grams of solidified waste/cm²-day

K = mpc of isotope in grams/ft³ of water

V = water flow rate, volume of water (ft³) per volume of solid (ft³) per day

ϕ = specific surface area of solidified waste in ft²/ft³

C = radioisotope fraction in glass (i.e., gram isotope/gram solid).

The hazardous radioisotope that is most limiting during the first few decades after a high-level waste is solidified is strontium-90, with an mpc of 3×10^{-7} $\mu\text{Ci/ml}$ or 6×10^{-11} g/ft³ H₂O. The permissible leaching rate, based on Equation (10), becomes:

$$L = 6.4 \times 10^{-14} \frac{V}{\phi C} \quad (11)$$

As an example of the usage of Equation (11) consider an accident in which a ruptured container of solidified waste is immersed in a flowing stream of water.

EXAMPLE

$V = 2,000,000$ (assume small water course with flow of 70 ft³/sec, 3 ft³ of solidified waste in container).

$\phi = 10$ (assume rupture inside of container, considerable fracturing of solid)

$C = 3 \times 10^{-3}$ (assume 0.5 ft³ solid waste per 10⁴ MWd(th) fuel exposure, aged 10 years).

Permissible Leach Rate, $L = 4 \times 10^{-6}$ g of solid/cm²-day.

Leach rates in the range of that required in this example have been achieved, and surpassed, in some waste solidification studies. (See Table 1.) Although the assumptions upon which this analysis were made are arbitrary they are believed realistic enough to point up the very real value of obtaining a low leachability in solidified wastes.

VI. LEACH TEST STANDARDIZATION

Although the need for standardization of the leaching studies being performed on diverse forms of solidified radioactive waste has been recognized for a long time, few concrete proposals have been made.

Thomas at the first meeting held in the United States on the fixation of radioactivity in stable, solid media proposed a standard leaching test which had as its purpose the comparison of the relative merits of various solids. (Thomas, 1958) The test conditions did not bear a close relation to practical leaching conditions; instead the test was designed to be easy to carry out and to yield reproducible numerical results which would permit comparison of a wide variety of solids studied in many different parts of the world. The technique was based on the use of a Soxhlet extractor operated to maintain the temperature in the leaching thimble at about 70°C (Figure 9).

Except for the Indian investigations (Rastogi, 1969), in which a modified Soxhlet extractor was used for some of the leach tests, the procedure proposed by Thomas has not been used for the study of solidified radioactive wastes. A very similar procedure has been used at the Department of Glass Technology, University of Sheffield, for fundamental studies in the elucidation of reaction mechanisms between glass and water. (Rana, 1961)

Godbee, et al., at the Richland symposium in 1966 registered a plea that the easily misinterpreted term, $\text{g cm}^{-2}\text{day}^{-1}$, be replaced by fraction leached $\text{g cm}^{-3}\text{day}^{-1}$ [see Equation (3), page 12] as the standard method of reporting leach test results. (Godbee, 1966) Subsequent leach data published by ORNL have followed this suggestion, having the dimensions of, for example, fraction of sodium leached $(\text{cm}^2/\text{g})^{-1}\text{day}^{-1}$.

FIGURE 9. Apparatus for Standard Leaching Test Proposed in 1957

= Oxidation Extractor

As the result of discussions among several investigators attending the Richland symposium, suggestions for a standard leach method were forwarded to the IAEA. (Blanco, 1966) It was suggested that the sample should be supported on a polyethylene rack in a polyethylene vessel. A volume of leachant about 10 times that of the sample was suggested, with 50 ml leachant being a convenient volume. The standard leachant was to be distilled water at pH 6.5-7.0 containing 100 mg NaCl/liter. Both the polyethylene equipment and the NaCl in the leach water specifications were designed to minimize precipitation of radioactivity on equipment surfaces. It was specified that the tests should be carried out in a dynamic system with continuously recirculated water at $25 \pm 3^\circ\text{C}$.

The foregoing suggestions and many others were considered when the IAEA brought together a small group of experienced workers in Vienna in August 1969 to recommend methods for the standardization of the leach testing of immobilized radioactive waste solids. The result of these discussions was a suggested standard leach test. (Hespe, 1971)

A. Comparison of Proposed IAEA Method with Methods Presently in Use

The proposed IAEA test is designed to be comprehensive. Comparisons between diverse immobilizing materials (cement, asphalt, plastic, glass) are facilitated, and it is also possible to obtain information for "reasonably accurate" hazards evaluations of the solidified wastes-environment system. The same test procedure is used for both inter-comparisons and hazards evaluation; distilled water is used as leachant for intercomparisons, water from the disposal environment is used for the hazards evaluations. Details of the IAEA proposed standard leach method are compared with those of other leach methods which have seen widespread application in the leaching of solidified high-level waste products in Table 4. The comparison shows that while there is essential agreement in many of the details, differences also exist, principally in the techniques used for sample preparation and for contacting leachant with the sample. The differences arise mainly because the IAEA method incorporates many procedural details commonly used for low and intermediate level waste solids but not for high level waste solids.

TABLE 4. Comparison of Present Leaching Methods with Proposed IAEA Standard Methods

		IAEA Proposed Standard	English	French	German	Indian	American
Sample Preparation	Specimen Container	Canned in container made of inert material, top surface only exposed.					
	Sample Size	At least 2.5 cm high x 2.5 cm diam.		✓*			
	Thermal Conditioning (Glasses)	Cast in container and cool. Reheat to maximum expected storage temperature and hold 500 hr. Cool at 5-10°C/hr.					
Leach Conditions	Leachant	Distilled water.	✓	✓	✓	✓	✓
	pH of Leachant	Record.					
	Temperature	25°C ± 5°C.	✓	✓	✓		✓
	Test Geometry	At least 5 cm depth above sample, which is facing upward.		✓	✓	✓	✓
	Leachant: Sample Surface Ratio	Ratio of leachant Volume (cm ³)/sample surface (cm ²) ≤ 10 cm.					
	Leachant Agitation	None, stagnant.					
Sampling	Method	Decant and replace removed leachant with fresh.	✓	✓	✓	✓	✓
	Frequency	Daily for 1 week, monthly for 6 months, twice yearly thereafter.	✓	✓	✓	✓	✓
	Duration	As long as is considered necessary.	✓	✓	✓	✓	✓
Sample Analysis	Analyses	pH, gross α and β-γ activity, concentrations of isotopes of particular interest.	✓	✓	✓	✓	✓
Reporting	Presentation of Results	Plots of either: a. Cumulative fraction leached $\frac{\Sigma a_n/A_0}{F/V}$ vs Σt_n ** or $\Sigma a_n/A_0$ vs $\Sigma \sqrt{t_n}$ or b. Incremental leach-rate, R_n , as a function of time, $R_n = \frac{a_n/A_0}{(F/V)t_n}$	✓	✓	✓	✓	✓

* Substantial agreement with IAEA proposed standard method is denoted by checks (✓).

** a_n = radioactivity leached during the leachant renewal period, n

A_0 = radioactivity initially present in specimen

F = exposed surface area of specimen (cm²)

V = volume of specimen (cm³)

t_n = duration (days) of leachant renewal period

A thermally conditioned, canned sample is specified for the IAEA method. This exact method of sample preparation has not been followed in any of the leach tests on solids for the incorporation of high level wastes. Small canned samples were taken of the highly radioactive WSEP melt streams as they flowed from the continuous melter to the receiver pots, however, these samples were air quenched and received no further thermal conditioning before leach testing. Generally, however, leach tests have been performed on uncanned samples with the surfaces exposed to the leachant being either fractured surfaces (powder samples) or sawed surfaces. Elsewhere, in some cases the samples were cast in crucibles which were removed and much of the surface leached was that which had been in contact with the crucible. (Clark, 1964; Bancroft, 1958; Jouan, 1971)

The extended periods at elevated temperatures which high-level radioactive waste solids may experience can have very deleterious effects upon leachability and any comprehensive study of candidate solidified waste forms must incorporate an investigation of thermal effects. Although 500 hours at storage temperature, the time specified in the proposed IAEA standard method may be sufficient to determine overall effects of longer range storage at elevated temperatures this fact is not completely established and more investigations in this area are required.

Another principal difference between the IAEA method and other techniques is the use of stagnant water at a low volume to sample surface area ratio ($\leq 10 \text{ cm}^2$). Most techniques for high-level waste leaching utilize moving water, either in a once-through pass or recirculating mode. Changing the leach water frequently (daily for the first week) as is done in the proposed IAEA test is probably an adequate substitute. Investigators in England and Germany have shown that a flow rate of $25 \text{ cm}^3/\text{day}$ through systems in which the sample surface area was 200 cm^2 was necessary to prevent the obtaining of an erroneously low leach rate. (Elliot, 1967; Heimerl, 1971)

B. Comments Concerning Comparison Tests

There is often a need for a leaching technique which permits the rapid scouting of the resistance to leaching of new formulations for waste solidification. The IAEA procedure can be used, but it is time consuming, particularly if the glass is very nonleachable. A process which accelerates the leaching process seems more suitable for comparison testing.

C. Comments Concerning Performance Tests

Performance tests (tests to obtain data for Use II, in the nomenclature of this report) are most meaningful if performed on the actual high-level solids. Remote handling is required and sample preparation and the operations during leaching must be as simple as possible. For instance, the use of powdered samples introduces an undesirable complication. The most highly radioactive solid wastes which have been leach tested to date were those prepared in the WSEP program at the Pacific Northwest Laboratory and they were leached by the technique described on page 25. This technique has shown trouble-free remote operation both at the Pacific Northwest Laboratory and at ICPP, where it originated. It permits good flexibility in such important operating parameters as leachant volume to sample surface area, leachant residence time and leachant temperature, and it is anticipated that the procedure will continue to be used at the Pacific Northwest Laboratory.

VII. REFERENCES

Alleman, R. T. "Leaching Tests of Spray Calcined Melts," Quarterly Progress Report, Research and Development Activities, Fixation of Radioactive Residues, October-December 1964, A. M. Platt, Editor, USAEC Report HW-84603, Atomic Products Operation, Richland, Washington, pp. 16-18, January 1965.

Bancroft, A. R. and J. D. Gamble. Initiation of a Field Burial Test of the Disposal of Fission Products Incorporated into Glass, CRCE-808, Atomic Energy of Canada Limited, Chalk River, Ontario, November 1958.

Barton, G. B. Solidification of High Level Wastes-Part IV, Phosphate Melts for Fixation of Radioactive Residues from Purex Type Wastes - Three to Fifty Percent Fission Product Oxides, BNWL-80, Battelle-Northwest, Richland, Washington, July 1965.

Barton, G. B. and J. A. Weed. Solidification of High Level Wastes, Part V. A Factorial Study of the Effect of Varying the Concentration of the Components of Purex Type Waste on the Properties of Phosphate Solids, BNWL-544, Battelle-Northwest, Richland, Washington, January 1968.

Bell, M. J. An Analysis of the Diffusion of Radioactivity from Encapsulated Wastes, ORNL-TM-3232, Oak Ridge National Laboratory, February 1971.

Blanco, R. E. Letter to L. A. Keher, Scientific Secretary, Division of Health, Safety, and Waste Disposal, IAEA, Vienna, May 20, 1966.

Bocola, W. and A. Donato. Program Ester: Solidification of Liquid Wastes of High Activity Characteristics of Phosphate Glass Melts Prepared from Simulated Fission Products Solution, RT/PROT-(70)40. Comitato Nazionale per l'Energia Nucleare, Rome, January 4, 1971 (In Italian) NSA:26:2149.

Bonniand, R. "Apparatus for Carrying Out Vitrification in a Hot Cell," TID-7613, pp. 280-299, 1961.

Brezhneva, N. E., Y. N. Golovanov, S. N. Oziraner, A. A. Eremin and V. N. Rozanova. "Solidification of Radioactive Waste Solutions," in Treatment and Storage of High-Level Radioactive Wastes, Proc. Symp. Vienna, 8-12 October 1962, pp. 441-464, IAEA 1963. AEC-tr 6171

Brownell, L. E., R. E. Isaacson, M. J. Kupfer and W. W. Schulz. The Thermalt Process for Immobilization of Radioactive Wastes, ARH-2059, Atlantic Richfield Hanford Company, Richland, Washington, April 15, 1971.

Burns, R. H. "Solidification of Low- and Intermediate-Level Wastes," Atomic Energy Review, Vol. 9, No. 3, pp. 547-599, 1971.

Cauwood, J. D., S. English and W. E. S. Turner. J. Soc. Glass Technol., Vol. 1, 153(1917), as described on p. 128, G. W. Morey, The Properties of Glass, 2nd Edition.

Clark, W. E. and H. W. Godbee. Laboratory Development of Progresses for Fixation of High-Level Radioactive Wastes in Glassy Solids: Wastes Containing (1) Aluminum Nitrates and (2) the Nitrates of the Constituents Of Stainless Steel, ORNL-3612, Oak Ridge National Laboratory, July 1964.

DeJonghe, P. "Extrapolation to a Disposal Program," pp. 104-105, in Treatment of Low- and Intermediate-Level Radioactive Waste Concentrates (Technical Reports Series No. 82) IAEA, Vienna, 1968.

Douglas, R. W. and T. M. El-Sharmy. "Reactions of Glasses with Aqueous Solutions," J. Amer. Ceramic Soc., pp. 1-8, Vol. 50, 1967.

Dukhovich, F. S. and V. V. Kulichenko. Radiation Stability of Vitrified Radioactive Preparations, Atomnaya Energiya, Vol. 18, No. 4, pp. 361-367, April 1965. Soviet Atomic Energy (English Translation) pp. 459-465.

Durham, R. W. "Fission Product Disposal in Glass," pp. 8-9, TID-7550, 1958.

Eliassen, R. and M. I. Goldman. "Fixation in Vitreous Matrices of High Activity Fission Product Wastes," pp. 576-589, TID-7613, 1961.

Elliot, N. N. and D. B. Auty. The Durability of "FINGAL" Glass, Part I. Discussion of Method and Effect of Leaching Conditions, AERE-R5151. Atomic Energy Research Establishment, Harwell, March 1967.

Fitzgerald, C. L., H. W. Godbee, R. E. Blanco and W. Davis, Jr. "The Feasibility of Incorporating Radioactive Wastes in Asphalt or Polyethylene," pp. 821-829, Nuclear Applications & Technology, Vol. 9, December 1970.

Godbee, H. W., W. E. Clark and C. L. Fitzgerald. "Physical Properties of Solids Incorporating Simulated Radioactive Wastes," pp. 566-584, Proceedings of the Symposium on the Solidification and Long-Term Storage of Highly Radioactive Wastes, February 14-18, Richland, Washington, CONF-660208, November 1966.

Godbee, H. W., C. L. Fitzgerald and J. O. Blomeke. "Diffusion of Radioisotopes Through Waste Solids," Trans. Am. Nucl. Soc., pp. 450-451, Vol. 12, 1969.

Golohanov, Y. N., N. E. Brezhneva, S. N. Ozivanov, A. A. Eremin and V. L. Zotov. The Chemical Stability and Crystallizability of a Glass Mass as Affected by Composition and Method of Preparation, Atomnaya Energiya, Vol. 15, No. 2, pp. 130-138, August 1963. Soviet Atomic Energy (English Translation) pp. 820-827.

Grover, J. R. and B. E. Chidley. Glasses Suitable for the Long Term Storage of Fission Products, AERE-R-3178. Atomic Energy Research Establishment, Harwell, July 1960.

Grover, J. R., M. N. Elliot and A. D. Jones. "Long Term Examination of Glasses for the FINGAL Process," Proceedings of the Symposium on the Solidification and Long Term Storage of Highly Radioactive Wastes, February 14-18, pp. 585-605, U. S. Atomic Energy Commission, November 1966.

Heimerl, W., H. Heine, L. Kahl, H. W. Levi, W. Latze, G. Malow, E. Schiener and P. Schubert. Research on Glasses for Fission Product Fixation, Summary Report. January 1968-June 1971, HMI-B109. Hahn-Meitner Institute, Berlin, September 1971.

Hemphill, L. "Evaluation of Experimental Clinkers," pp. 14-15, TID-7550, 1958.

Hespe, E. D. "Leach Testing of Immobilized Radioactive Waste Solids - A Proposal for a Standard Method," Atomic Energy Review, pp. 195-207, Vol. 9, 1971.

Jouan, A. and A. Magnier. "Leaching Installation for Radioactive Glass Blocks in the VULCAN Line," pp. 335-340 in Semi-Annual Report of the Department of Chemistry, June-November 1970, CEN-N-1479. Commissariat a l'Energie Atomique, Fontenay-aux-Roses. November 1971.

Kelly, G. D. "Ceramic Sponges for Radioactive Waste Disposal," pp. 157-179, TID-7613, 1960.

Kulichenko, V. V. Study of the Conditions for Burial of Radioactive Cements, Preliminary report of work done under IAEA Contract No. 329/RB. Report 66-4572 (English Translation) 1966.

Kulichenko, V. V., F. S. Dukhovich, N. V. Kryova and B. A. Salamatov. Study of the Physico-chemical Properties of Various Types of Solid Materials in Order to Determine the Possibility and Advisability of Using them for Fixing Radioisotopes in the Treatment of Radioactive Wastes, Preliminary Report of work performed under IAEA Contract 340/RB, period October 1965 to June 1966, at Physical Chemistry Institute, USSR Academy of Sciences, Moscow.

- Luce, R. W., R. W. Bartlett and G. A. Parks. "Dissolution Kinetics of Magnesium Silicate," Geochimica et Cosmochimica Acta, pp. 35-50, Vol. 36, 1972.
- McVay, T. N., R. L. Hamner and M. P. Haydon. "Fixation of Radioactive Wastes in Clay-Flux Mixes," pp. 335-338, TID-7517 (1956).
- Mendel, J. E. and J. L. McElroy. Waste Solidification Program, Volume 10, Evaluation of Solidified Waste Products, BNWL-1666, Battelle, Richland, Washington, July 1972.
- Merritt, W. F. "Permanent Disposal by Burial of Highly Radioactive Wastes Incorporated into Glass," pp. 403-408 in Disposal of Radioactive Wastes into the Ground, IAEA, Vienna, 1967.
- Morey, G. W. The Properties of Glass, 2nd Edition, Reinhold Publishing Corporation, New York, 1954.
- Nichols, J. P. "Safety Analysis," pp. 296-313 in Radioactive Waste Repository Project: Technical Status Report for Period Ending September 30, 1971, ORNL-4751, Oak Ridge National Laboratory, December 1971.
- Paige, B. E. Leachability of Glass Prepared from Highly Radioactive Calcined Alumina Waste, IDO-14672, Phillips Petroleum Company, Idaho Falls, Idaho, February 1966.
- Paige, B. E. Leachability of Alumina Calcine Produced in the Idaho Waste Calcining Facility, IN-1011, Phillips Petroleum Company, NRTS, Idaho Falls, Idaho, July 1966.
- Ralkova, J. and J. Saidl. "Incorporation of Radioisotopes into Melted Silicates," pp. 341-353, Treatment and Storage of High-Level Radioactive Wastes, Proceedings of the Symposium held by the International Atomic Energy Agency in Vienna, Austria, October 1962.
- Kalkova, J. and J. Saidl. "Solidification of High Level Wastes, Part 3. Diffusion and Elution Rates of Radionuclides Incorporated in Basalts," Kernenergie, Vol. 10, 1967, pp. 161-164, BNWL-TR-49, translated by E. Foskett.
- Rana, M. A. and R. W. Douglas. "The Reaction Between Glass and Water. Part I. Experimental Methods and Observations," pp. 179-195, Physics and Chemistry of Glasses, Vol. 2, No. 6, December 1961.
- Rastogi, R. C., J. D. Sehgal, K. Chandrin and K. T. Thomas, Investigation of Materials and Methods for Fixation of Low and Medium Level Radioactive Waste in Stable Solid Media, Final Report, BARC-400, Bhabha Atomic Research Centre, Bombay, India, 1969.

Sachse, G. and H. Rosenberger. "Solidification of Radioactive Concentrates in Glasses or Ceramics. Part 3. Studies of the Applicability of Volcanites from Native Deposits," pp. 15-23, Kernenergie, Vol. 11, January 1968.

Saidl, J. "Standard Leach Unit Proposal," Letter to Mr. L. H. Keher, Scientific Secretary, Division of Health, Safety, and Waste Disposal, International Atomic Energy Agency, Vienna, August 5, 1966.

Shreir, L. L. (Editor), Corrosion, Volume 2, George Newnes Limited, London, 1963.

Smith, A. J. Leaching of Radioactive Sludges Incorporated in Cement and Bitumen, AERE-M-2223 (1969).

Spector, M. L., E. Suriani, and G. L. Stukenbroeker, "Thermite Process for the Fixation of High Level Radioactive Wastes," pp. 117-122, I&EC Process Design and Development, Vol. 7, No. 1, January 1968.

Taylor, W. C. and R. D. Smith. "Solubility Characteristics of Glasses Basically Different in Composition," J. Am. Ceramic Soc., Vol. 19, pp. 331-335, 1936.

Thomas, H. C. "Tests of Stability," pp. 35-38, TID-7550 (1958).

Thomas, H. C. "Proposed Standard Leaching Test," pp. 87-89, TID-7550, March 1958.

Thomas, R. G., E. H. Rex and C. W. Christenson. "Studies on Fixation of Radioactivity in Fired Clays," pp. 352-367, TID-7517 (1956).

Thompson, R. J., J. E. Mendel and J. H. Kleinpeter. Waste Solidification Demonstration Program: Characterization of Nonradioactive Samples of Solidified High-Level Waste, BNWL-1393, Battelle-Northwest, Richland, Washington, June 1970.

TID-7517 (in 2 parts). Sanitary Engineering Aspects of the Atomic Energy Industry. A Seminar Sponsored by the AEC and the Public Health Service, Held at the Robert A. Taft Engineering Center, Cincinnati, Ohio, December 6-9, 1955. Issued October 1956.

TID-7550. Report of Working Meeting on Fixation of Radioactivity in Stable, Solid Media at the Johns Hopkins University, June 19-21, 1957, Issued March 1958.

TID-7613. Report of the Second Working Meeting on Fixation of Radioactivity in Stable, Solid Media at Idaho Falls, Idaho, September 27-29, 1960, Edited by J. M. Morgan, Jr., D. K. Jamison, J. D. Stevenson, February 1961.

Tuthill, E. J., G. G. Weth, L. C. Emma, G. Strickland and L. P. Hatch. Phosphate Glass Process for Disposal of High Level Radioactive Wastes, I&EC Process Design and Development pp. 314-321, Vol. 6, July 1967.

Upton, U. L. Fixation of High-Level Radioactive Wastes in Phosphate Glass - Hot Cell Glass Experiment, BNWL-200, Battelle-Northwest, Richland, Washington, January 1966.

Volf, M. B. Technical Glasses, Sir Issaac Pitman and Sons, Ltd., London, 1961, pp. 57-59.

Watson, L. C., H. K. Roe, R. W. Durham, E. J. Evans and D. H. Charlesworth. Methods of Storage of Solids Containing Fission Products, CRCE-736, Atomic Energy of Canada Limited, Chalk River, Ontario, June 1958.

Weyl, W. A. and E. C. Marboe. The Constitution of Glasses, A Dynamic Interpretation, Volume II: Part Two, Constitution and Properties of Some Representative Glasses, Interscience Publishers, New York, 1967.

Wilding, N. W. and J. W. Rhodes. Leachability of Zirconia Calcine Produced in the Idaho Waste Calcining Facility, IN-1298, Idaho Nuclear Corporation, Idaho Falls, Idaho, June 1969.

Wilding, N. W. and J. W. Rhodes. Characteristics of Radioactive Zirconia Calcine After Heating to High Temperature, IN-1486, Idaho Nuclear Corporation, Idaho Falls, Idaho, June 1971.

DISTRIBUTIONNo. of
CopiesUNITED STATES

- 1   AEC Chicago Patent Group
     A. A. Churm
- 2   AEC Division of Waste Management and Transportation
     R. W. Ramsey
     R. D. Walton
- 1   AEC Idaho Operations Office
     K. K. Kennedy
- 1   AEC Oak Ridge Operations Office
     E. H. Hardison
- 1   AEC Office of the Commissioners
     W. H. Beach, Technical Assistant
                 to commissioner C. E. Larson
- 186   AEC Technical Information Center
- 2   Allied Chemical Corporation
     Idaho Falls, Idaho 83401
     D. W. Rhodes
     C. M. Slansky
- 2   duPont Company, Aiken (AEC)
     A. S. Jennings
     R. W. Wallace
- 2   Union Carbide Corporation (ORNL)
     J. O. Blomeke
     H. W. Godbee

ONSITE

- 1   AEC/RL Patent Attorney
     R. W. Poteat

No. of
Copies

- 4   AEC Richland Operations Office
      B. J. Melton
      Production and Waste Management Programs Division
      O. J. Elgert
      R. B. Goranson
      N. T. Karagianes
- 2   AEC/RL Environmental, Safety and Technical Services Division
      P. G. Holsted (2)
- 8   Atlantic Richfield Hanford Company
      G. S. Barney
      L. E. Brownell
      M. H. Campbell
      R. E. Isaacson
      M. J. Kupfer
      W. W. Shultz
      A. E. Smith
      File Copy
- 29   Battelle-Northwest
      L. L. Ames
      J. L. Bates
      W. F. Bonner
      L. R. Bunnell
      T. D. Chikalla
      D. L. Condotta
      J. N. Hartley
      J. D. Kaser
      J. L. McElroy
      J. E. Mendel (10)
      A. M. Platt
      K. J. Schneider
      I. M. Warner
      W. K. Winegardner
      Technical Information Files (5)
      Technical Publications