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Strategy for Glass Waste Form Acceptance for Geologic Disposal

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Summary of Talking Points

1. What are the technical bases, including standards, test methods, and use of databases and models, for DOE's criteria for qualifying borosilicate glass waste forms as acceptable for disposal in a geologic repository?

2. What is DOE's technical basis for applying the results of short-term tests on reference glasses or glasses with simplified compositions to assessments of the long-term performance of more chemically complex HLW glasses in repository environments?

3. What is known about the influence of glass chemistry on crystallite precipitation during glass production and on glass corrosion, and how are crystallites taken into account in DOE's approach to designing glass for disposal in a repository?

4. Are data on natural and archeological glasses used to support DOE assessments of the long-term performance of HLW glass in a repository and, if so, how?

Development of Technical and Performance Standards for HLW Glass

- 1957 - National Academy of Sciences (NAS) recommended deep geologic disposal of HLW once made into a solid form (Disposal of Radioactive Waste on Land)

- Late 1970's DOE began evaluation of waste forms

- Dec 1982, Record of Decision (ROD) issued selecting borosilicate glass
  - Endorsed by EPA and several independent review groups
  - NRC had no objection

- 1982 Nuclear Waste Policy Act (NWPA) mandated that HLW be sent to a federal repository (based on 1957 recommendation)

- 1985 President ratified the DOE decision to send defense HLW to a civilian repository (Office of Civilian Radioactive Waste Management, OCRWM)

- Early 1990's Waste Acceptance System Requirements Document (WASRD) generated requiring DOE-EM to develop Waste Acceptance Product Specifications (WAPS)

THE DISPOSAL OF RADIOACTIVE WASTE ON LAND

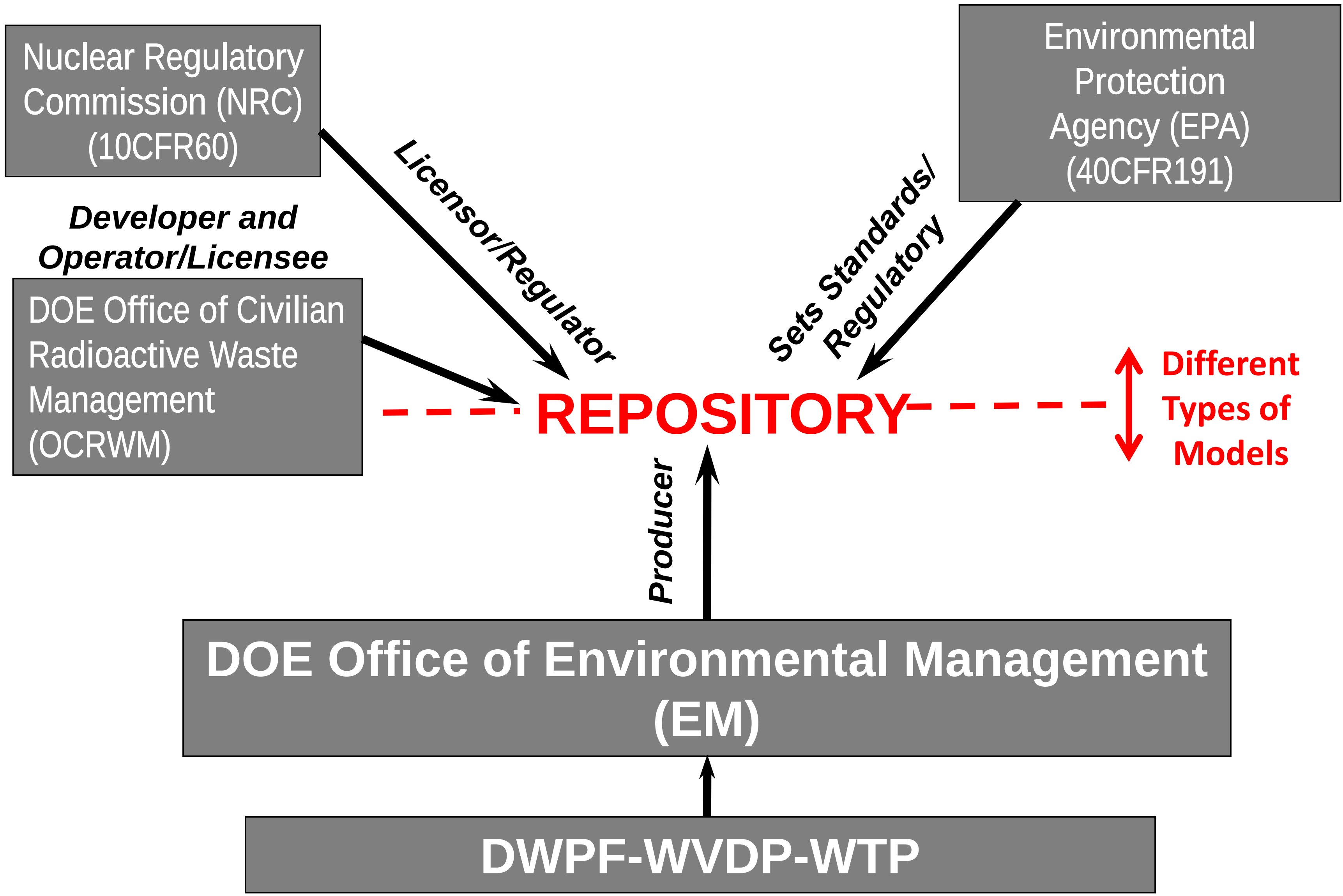
Report of the
Committee on Waste Disposal
of the
Division of Earth Sciences

Committee Members

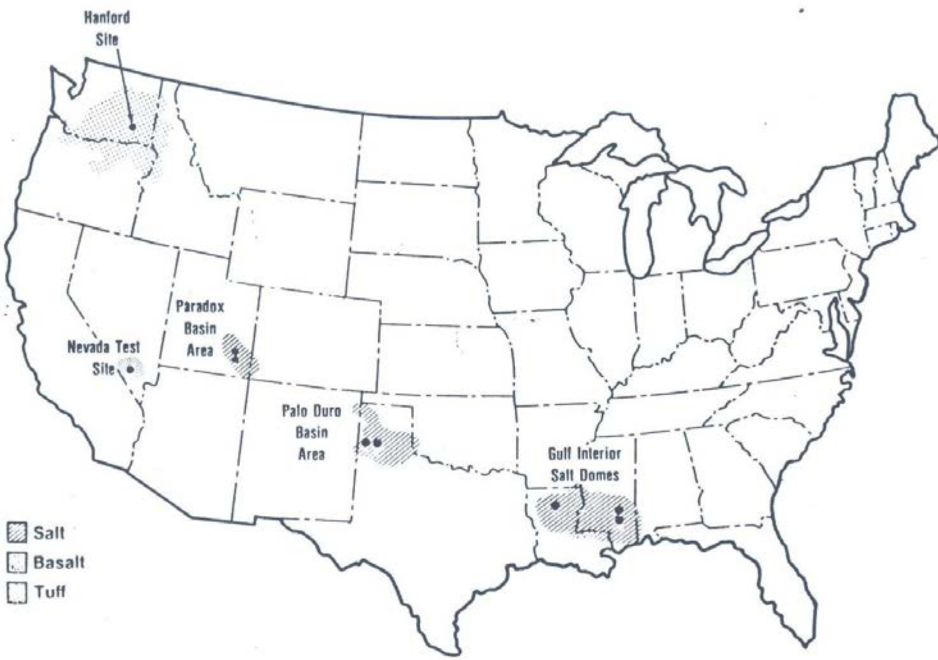
Harry H. Hess, Chairman
John N. Adkins — William B. Heroy
William E. Benson — M. King Hubbert
John C. Frye — Richard J. Russell
Charles V. Theis

Publication 519
Price $1.00
National Academy of Sciences - National Research Council
Washington, D. C.
September 1957

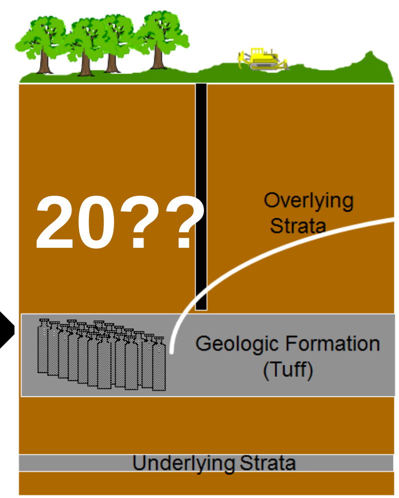
Waste Form Producer – Repository/Regulatory Interface



Nuclear Waste Policy Decisions 1982, 1987, 2009

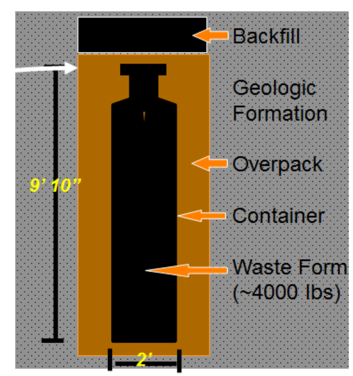


ISSUE: Waste forms being made now must be acceptable to a repository yet to be defined, sited, and/or built



Geologic System

Conceptual High Level Waste Repository

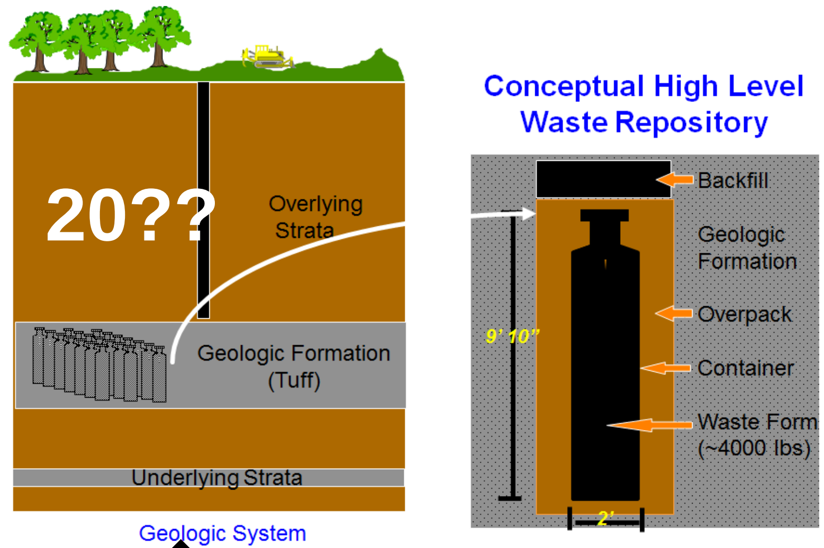


How Can The Waste Form Producers Comply?

OPTIONS

A. Relate glass dominated short term test results to a "repository relevant" test when a repository is chosen

B. Develop a glass durability standard that meets repository requirements defined by geochemical and HLW performance modeling: All production HLW glasses must be more durable than this standard glass



TECHNICAL JUSTIFICATION

- **Perform Long Term (LT) tests (HLW burial glasses and natural analogs)**
- **Perform repository relevant tests (rock cup tests in tuff, basalt, salt, granite with various groundwaters, at low Eh, etc.)**
- **Perform in situ tests in repositories (WIPP, STRIPA granite, Ballidon clay UK)**
- **Perform materials interactions tests (glass & metal, with/without rock present)**
- **Perform accelerated Short Term (ST) tests (without changing the durability mechanism) with HLW glass and analogs**
- **Relate LT and ST testing (radioactive and non-radioactive glass testing)**

Details of Waste Form Qualification Strategy Used in US (1982-Present)

Time Frame	Strategy
1982-1983	✓ develop "acceptable waste form durability" from geochemical modeling based on HLW performance modeling ✓ fractional dissolution rates between 10^{-4} to 10^{-6} parts per year (ppy) ✓ a glass waste form would take 10,000 to 1,000,000 years to totally dissolve ✓ early versions of 10 CFR Part 60.113 specified fractional release rates of 10^{-5} ppy ✓ which was in the middle of the range determined by HLW performance modeling ✓ 10^{-5} parts per year was adopted as the waste form specification (ONWI).
	✓ if the long-term fractional dissolution rate of a waste form was $\leq 10^{-5}$ parts per year for the most soluble and long-lived radionuclides then borosilicate glass would provide acceptable performance for any repository system
1987-Present	✓ develop tests (MCC and ASTM) that would provide an under-standing of the glass durability mechanisms from a combination of the test protocols ✓ ASTM C1220 (Leaching of Monolithic Waste Forms for Disposal of Radioactive Waste), ASTM C1285 Product Consistency Test (PCT), ASTM C1662 Single Pass Flow Through (SPFT) and ASTM C1663 Vapor Hydration Test (VHT), Pressurized Unsaturated Flow Test (PUF), ASTM C1308 (Accelerated Leach Test for Diffusive Releases from Solidified Waste)

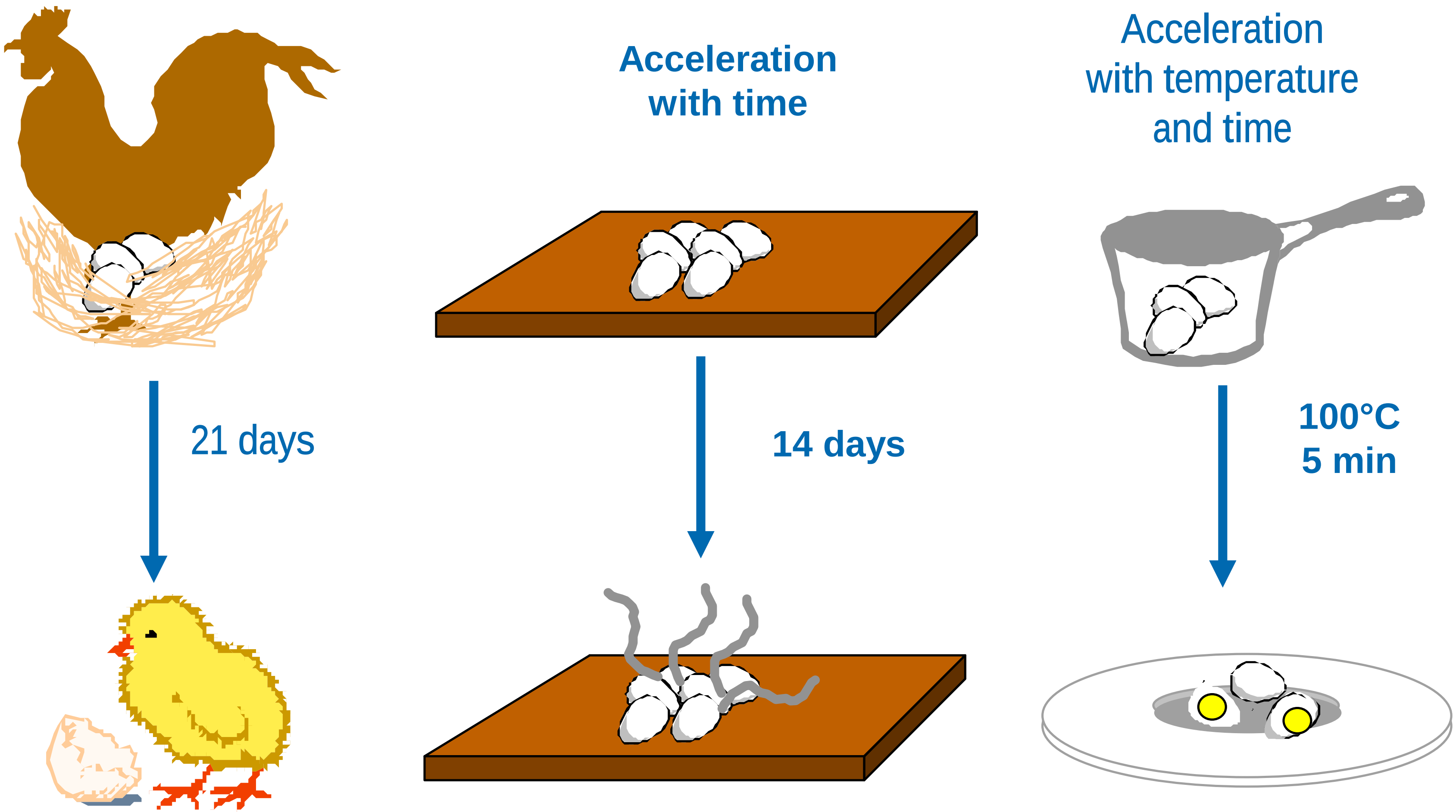
From L.P. Hatch, "Ultimate Disposal of Radioactive Wastes," Am. J. Science, 410-421 (1953) and Waste Forms Technology and Performance," Final Report, NAS Publication (2011).

Details of Waste Form Qualification Strategy Used in the US (Cont'd)

	Time Frame	Strategy
Testing	1987-present	✓ generate data for modeling the maximum radioactive release rate(s) in borosilicate glass by relating the release of soluble ^{99}Tc, ^{129}I, and ^{135}Cs to the release of soluble species such as Na, Li, and B soluble species leach at the same rate (congruently) ✓ this approach (with references) are part of the ASTM C1285 (PCT) protocol
	1987- present 1994-present	✓ develop a short term test and process control strategy for ensuring that every glass produced had a dissolution rate <EA glass at the L95% confidence level based on Na, Li, B ✓ this ensures acceptable performance control (part of the WAPS compliance strategy)
	1996-present	✓ Continue to test (qualify) the radionuclide response of production glasses to verify that radionuclide release consistent with the releases predicted by Na, Li, B
Standard	1992	✓ develop a borosilicate glass standard ✓ Environmental Assessment (EA) glass (1981 DWPF EA; Jantzen, et.al. 1992) bounded the upper release rate found to be acceptable from HLW performance modeling and 10 CFR Part 60.113

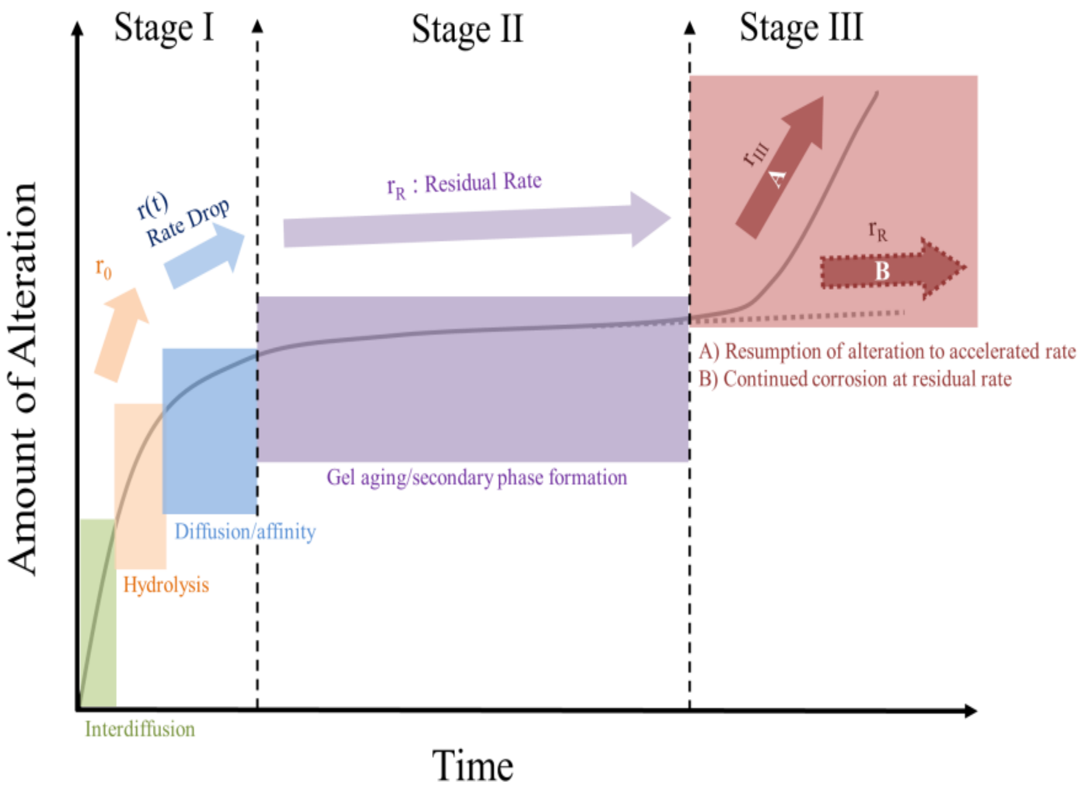
From L.P. Hatch, "Ultimate Disposal of Radioactive Wastes," Am. J. Science, 410-421 (1953) and Waste Forms Technology and Performance," Final Report, NAS Publication (2011).

Accelerated Testing: Must Simulate Correct Long Term Mechanism



* As defined at the August, 1998 NAS/NRC Workshop on Test Methods and Models to Simulate Accelerated Aging of Infrastructure Materials

Accelerated Testing: Must Simulate Correct Long Term Mechanism



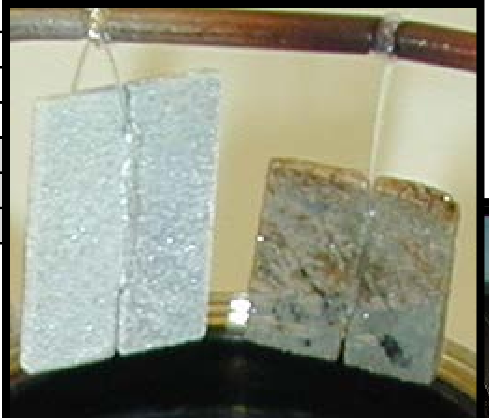
- Different times, temperatures, and pH regimes simulate different mechanisms
- Glass corrosion is complex and involves a variety of mechanisms
- Different tests are needed to study the different mechanistic regimes

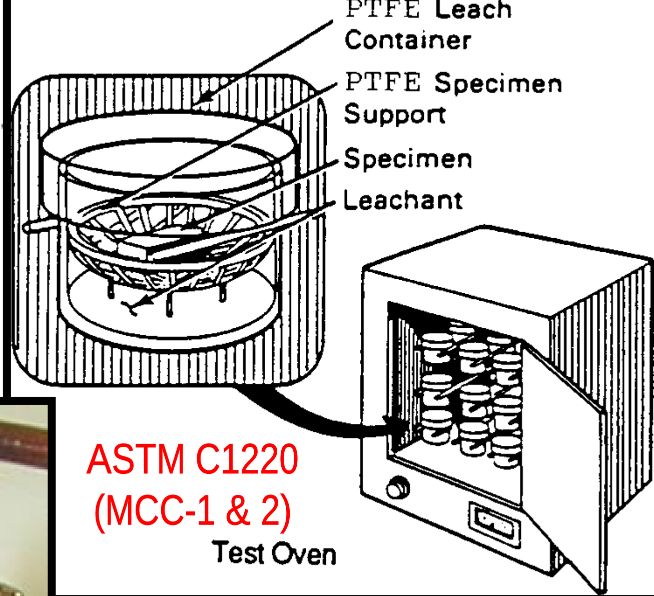
Tests Developed Since 1980

Waste Form (Since 1980-Present)	
Chemical Durability	MCC-1, 2, 3, 4, 5 (Soxhlet)
Aging Effects (thermal and radiation)	MCC-6, 7, 12, 13
Volatility	MCC-8,9,16
Physical Strength	MCC-10, 11, 15
Canister Container	
Corrosion Resistance	MCC-101, 102, 103, 104
Repository Interactions	
Canister/container corrosion	MCC-105[a]
Waste Form Durability	MCC-14[a]

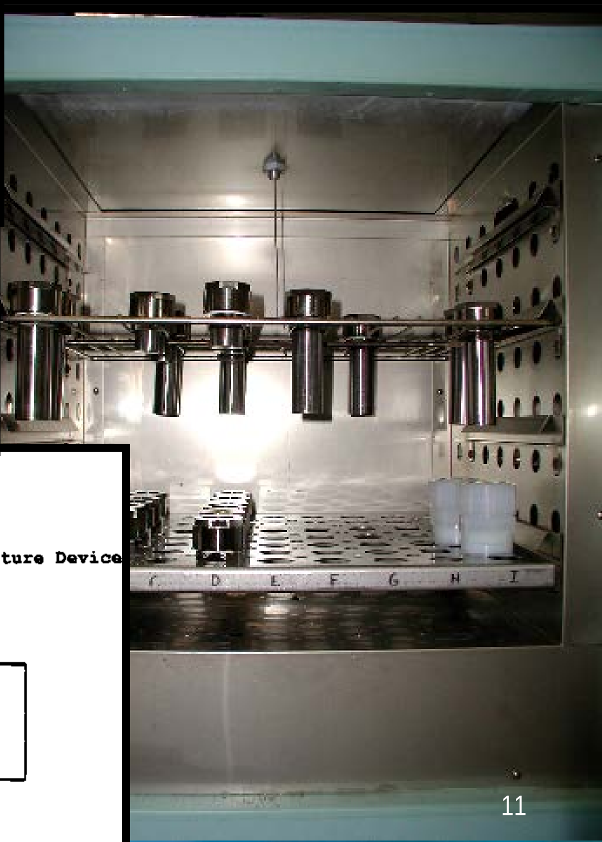
[a] The repository interactions tests are divided into site-specific subcategories, e.g., MCC-105.1 (basalt).

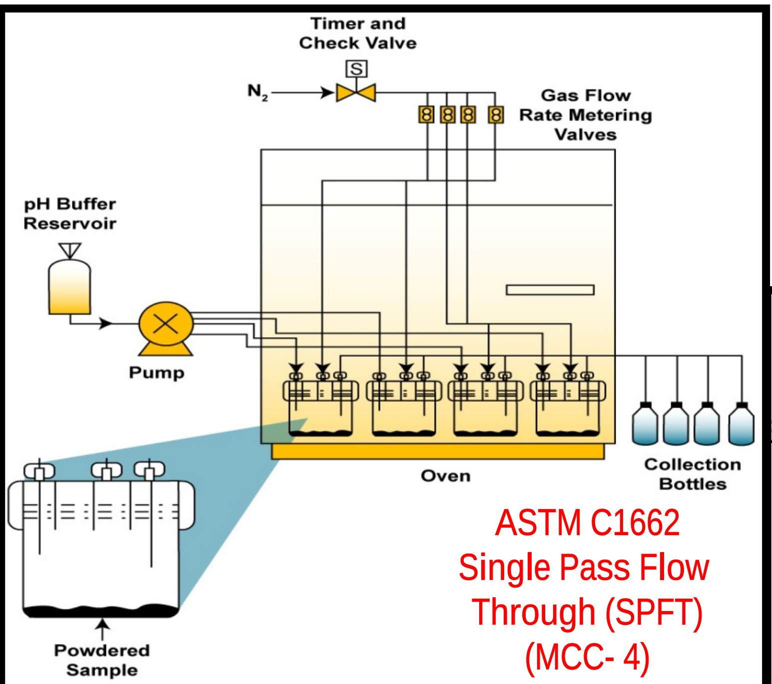
ASTM C1663 Vapor Hydration Test (VHT)





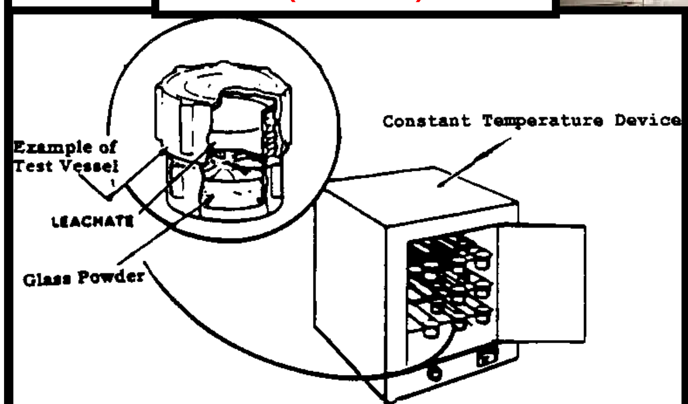
ASTM C1220 (MCC-1 & 2)





ASTM C1662 Single Pass Flow Through (SPFT) (MCC- 4)

ASTM C1285 Product Consistency Test (PCT A and B) (MCC- 3)



What Were the Criteria for PCT (ASTM C1285) Test Development in 1986?

– Waste Form Producers needed to ensure the acceptability of High Level Waste (HLW) glass made in DWPF to the geologic repository (testing and modeling)

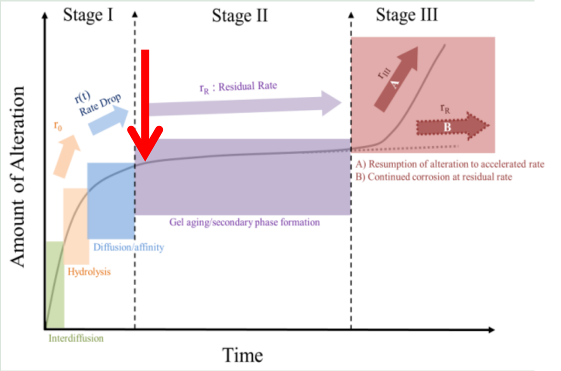
– Producers needed to define the durability of DWPF HLW glasses before and during production (Waste Acceptance Product Specifications 1.3 and 1.4)

  – WAPS 1.3 - DWPF must demonstrate (during production) control of the radionuclide release properties of the final waste form

  – WAPS 1.4 – determine the release properties of crystallized glass

– Producers needed a test sensitive to glass composition and glass homogeneity

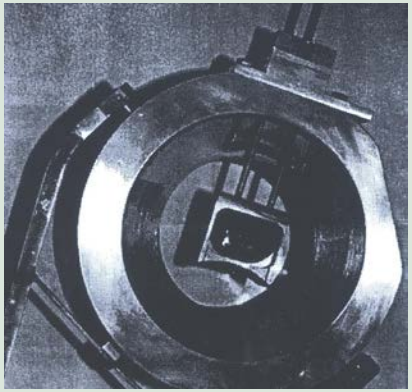
- Test factors rigorously controlled so that glass composition and homogeneity dominate the test response
  - –standardize particle surface area (S)
  - –standardize amount of glass to volume of solution
  - –standardize test duration
    - » short enough for use during production
    - » long enough to get good precision/reproducibility
  - –standardize test temperature



– Simple sample preparation/procedure for remote operation

– Acceptance by waste form developers and the repository

PCT Test Summary



PCT was shown to be more sensitive to glass composition and homogeneity than other glass durability tests

PCT can be routinely performed remotely on DWPF glass during production

Test durations of ≥7 days were shown to be adequate to distinguish between different glass durabilities

PCT test response has been related to other ASTM HLW glass test responses

ASTM Committee C26.13 (composed of waste form developers, repository representatives, and the Nuclear Regulatory Commission) peer reviewed the test from 1987 to present

Independent Confirmation of Test Discrimination Testing at PNNL (Shade & Piepels, 1990-1991; PNL-7530)

ASTM C1285 Significance & Use

These test methods (PCT-A and PCT-B) provide data useful for evaluating the chemical durability of glass waste forms as measured by elemental release. Accordingly, it may be applicable throughout manufacturing, research, and development.

Test Method A (short term) can specifically be used to obtain data to evaluate whether the chemical durability of glass waste forms have been consistently controlled during production

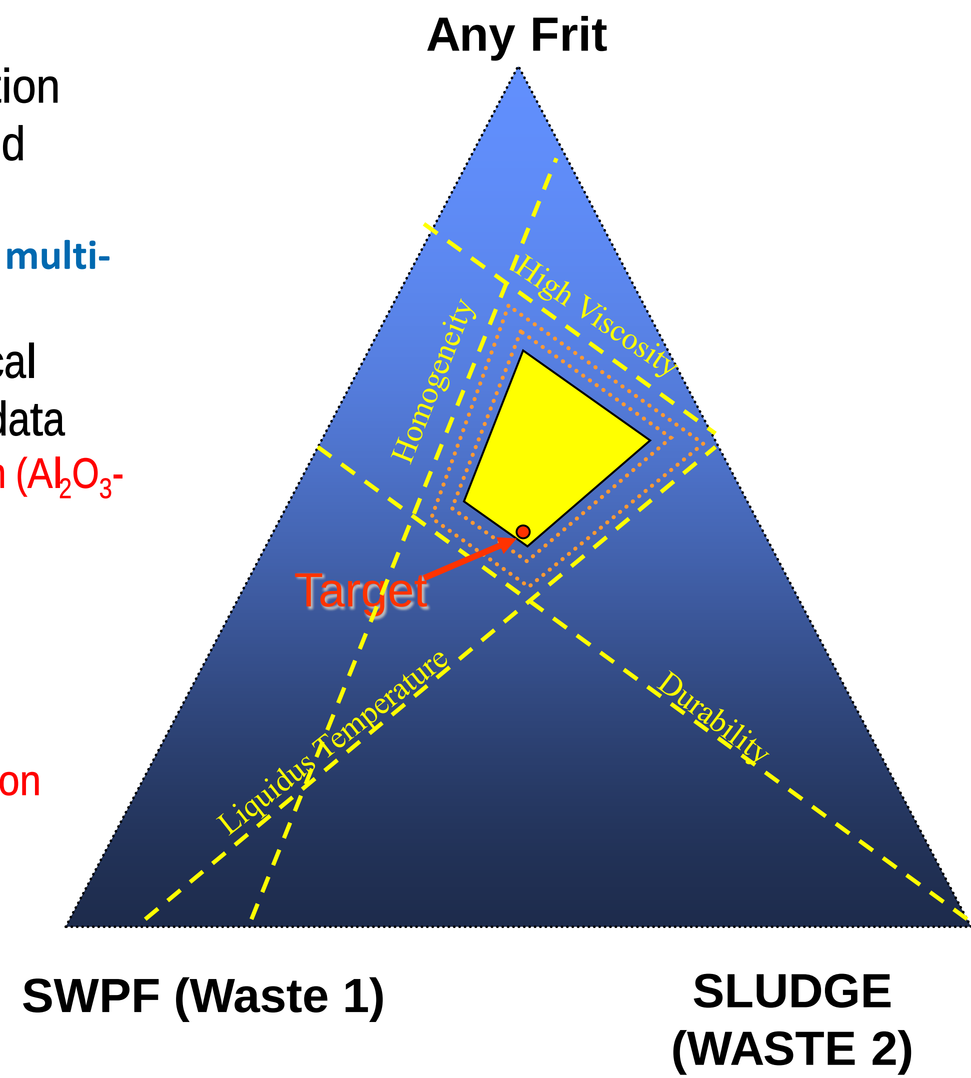
Test Method B (long term) can specifically be used to measure the chemical durability of glass waste forms under various test conditions, for example, varying test durations, test temperatures, ratio of sample-surface area (*S*) to leachant volume (*V*), and leachant types. Data from this test may form part of the larger body of data that are necessary in the logical approach to long-term prediction of waste form behavior (see Practice C1174).

Jantzen, C.M., Bibler, N.E., Beam, D.C., Ramsey, W.G., “Development of an ASTM Standard Glass Durability Test, the Product Consistency Test (PCT), for High Level Radioactive Waste Glass,” Spectrum 94, Am. Nuclear Soc., 164-169 (1994) and Jantzen, C.M. and Bibler, N.E., “The Product Consistency Test (PCT): How and Why it Was Developed,” Ceramic Transactions, V.207, 155-167 (2009).

PCT-A an Integral Part of DWPF Statistical Process Control (SPC)

CONTROL IS ON GLASS COMPOSITION DEFINED BY GLASS PROPERTIES

- To be a compliant glass, the glass composition must have properties that fall within the solid quadrilateral shown
  - Multivariate **theory used to control within multi-dimensional composition space**
- Each process model is based on geochemical principles and/or glass structure models + data
  - Homogeneity: Nolan's (1966) basalt system (Al_2O_3-[Fe_2O_3-FeO]-Na_2O-SiO_2)
  - Liquidus: Nolan, Bailey & Schairer (1966) crystallization in same basalt system and Burnham's quasicrystalline theory
  - Viscosity: glass polymerization
  - Durability: PCT-A versus glass polymerization (short range order) and thermodynamics
- SPC accounts for "model error", analytic error, tank transfer error, and heels
  - 95% confidence is obtained at max WL

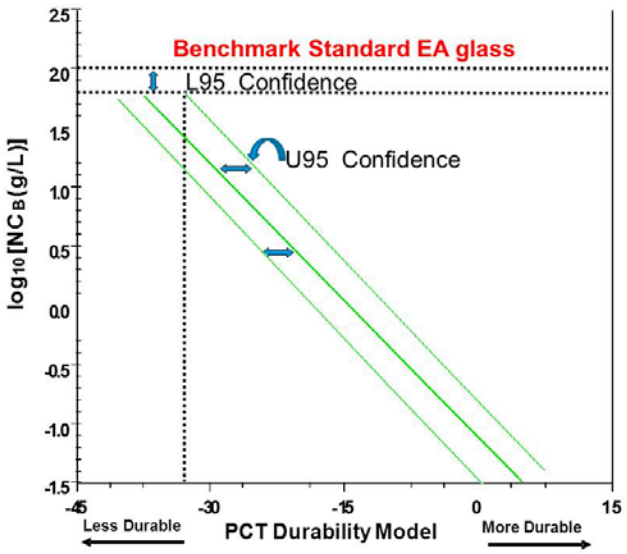


DWPF SPC Defines A Qualified Composition Range for Waste Acceptance

MINIMIZES CONFIRMATORY SAMPLES TO BE TAKEN DURING PRODUCTION

- Qualified glass range is processable
  - proven for the last 21 years of DWPF operation
- Process control is used to demonstrate acceptable performance by linking relationships

process control ↔ composition control ↔ dissolution rate control ↔ performance control ↔ acceptable performance



SPC Durability Model is known as THERMO™ (Thermodynamic Hydration Energy Reaction MOdel)

Compares the durability response of every melter feed batch (once made into glass) to the EA glass at 95% confidence; 2 standard deviations below EA glass

Production control using THERMO ™ is based on PCT-A testing

Repository modeling uses a variety of test methods, including PCT-A and PCT-B

Jantzen, C.M., Pickett, J.B., Brown, K.G., Edwards, T.B., and Beam, D.C., "Process/Product Models for the Defense Waste Processing Facility (DWPF)," US DOE Report WSRC-TR-93-0672, 464p. (September, 1995).

The Environmental Assessment (EA) HLW Borosilicate Glass Standard

DOE/EA-0179

Environmental Assessment

Waste Form Selection For SRP High-Level Waste

July 1982

U.S. Department of Energy

Assistant Secretary for Defense Programs

TABLE 1-1

Key Properties and Characteristics of Borosilicate Glass Waste Form

Property or Characteristic	Borosilicate Glass
Density, g/cm^3	2.75
Waste Loading, wt %	28
Toleration of Waste Variability	Acceptable
Long-Term Leachability,* $g/m^2 \cdot d$	10^{-3} to 10^{-4}
Fractional Release Rate from Full-Size Form,** yr^{-1}	10^{-5} to 10^{-6}
Radiation Stability	Very good
Impact Response,† wt % fines	0.14 to 0.18
Processability††	Relatively simple

* Based on plutonium leach rates in long-term tests at room temperature.

** Estimated from plutonium leaching data (conservatively assumes that release of radionuclides is not reduced by solubility limitations).

† Generation of particles less than 10 micrometers in size from single impact of 10 J/cm^3 energy density.

†† Relative ease of producing the waste form.

- EA glass is the glass that was qualified in the SRP HLW Environmental Assessment (DOE/EA-0179)
- Typical Properties and characteristics of borosilicate waste forms provided fractional releases based on Pu leaching of 10^{-5}-10^{-6} assuming no solubility limitations (MCC-1 testing)

HLW EA Glass Standard and Others Are Not Simplified Glasses

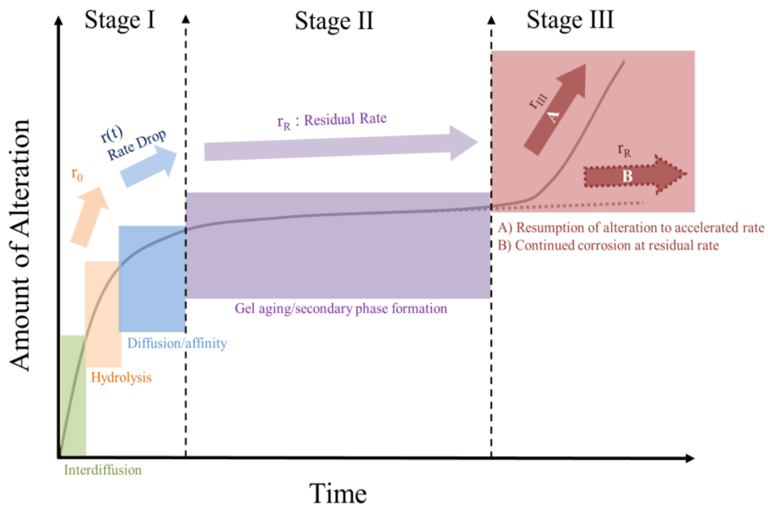
Typical Composition of SRP Waste Glass

Component	Concentration, wt % Waste Glass	Contribution From Waste
SiO_2	46.3	4.8
Fe_2O_3	5.9	5.9
Fe_3O_4	2.8	2.8
Na_2O	16.3	3.8
B_2O_3	10.9	-
Li_2O	4.2	-
MnO_2	1.6	1.6
Al_2O_3	3.2	3.2
NiO	0.6	0.6
MgO	1.6	0.2
U_3O_8	1.2	1.2
CaO	1.0	1.0
TiO_2	0.7	-
ZrO_2	0.4	-
La_2O_3	0.4	-
Other solids*	2.9	2.9
	100	28

* "Other solids" include zeolite, undissolved salts, and radionuclides. Chemically, radionuclides are less than 0.1% of the waste.

- For EA - Eliminated U_3O_8 and renormalized the composition
- For EA glass manufacture MnO_2 converted to MnO and part of the iron converted to FeO to give an $Fe^{2+}/\Sigma Fe$ ratio of 0.18 which corresponds to the REDuction/OXidation (REDOX) that the DWPF flowsheet targets for each waste glass batch
- 1000 pounds fabricated by Corning Glass Works for usage in DOE complex
- Other "standard" glasses can be used to ensure the PCT-A or PCT-B tests are in control include ARM-1 and ARG-1

Relation of PCT-A to PCT-B and Other HLW Glass Tests



Stage III behavior determined from long term PCT-B testing

- PCT-A has been compared to long-term PCT-B (Mueller et.al. 2004 & 2006)
- PCT-A has been related to long-term burial tests (Jantzen, et.al. 2008)
- PCT-B long term tests have been related to shorter term, higher temperature, Vapor Hydration Test (VHT) responses

  e.g. the HLW Environmental Assessment (EA) glass reaches the same stage of durability within 56 days at 20,000 m^{-1} or >313 days at 2000 m^{-1} when tested by PCT at 90°C or within 6 days when tested by VHT at 200°C (Bates, et. al. 1996)
- the rate of the short term crushed glass test (PCT-A) has been shown to be an upper bound for accelerated durability behavior (the resumption of dissolution or Stage III leaching behavior) (Ebert 2000)

Database: Accelerated Leach Testing of GLASS (ALTGLASS)

ALTGLASS constructed by SRNL (2013-Present) from Literature and SRNL Data

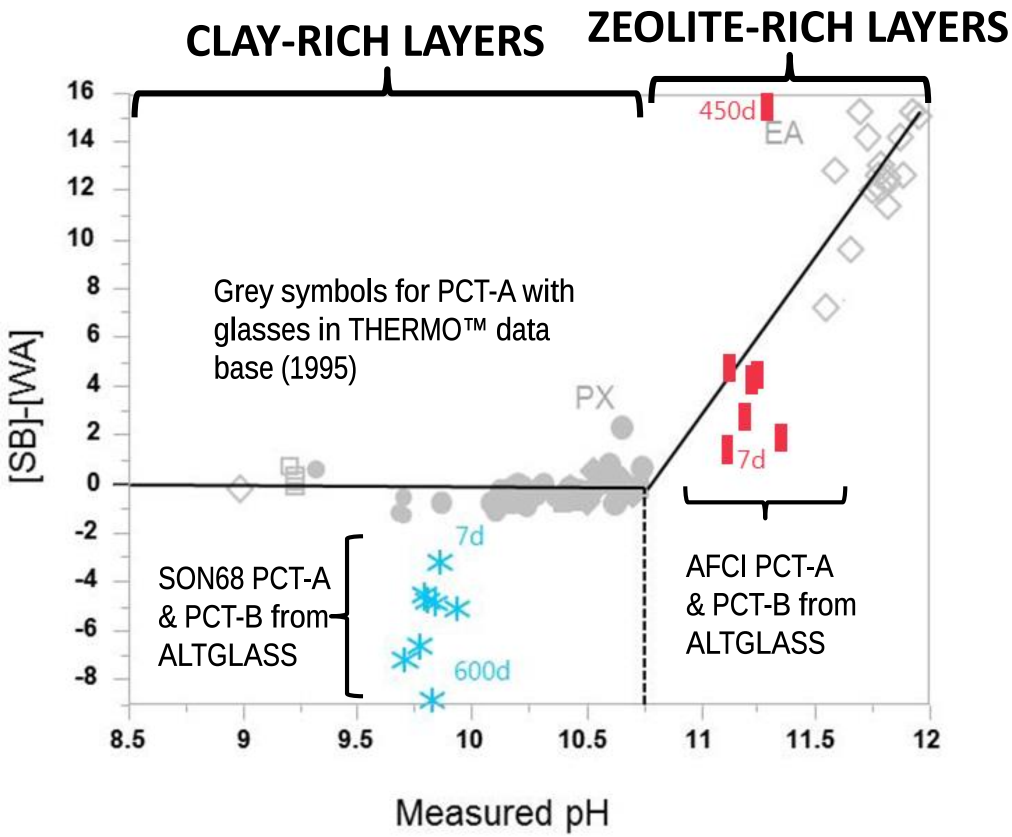
- Joint EM-NE-SC-International Technical Evaluation of Alteration Mechanism (I-TEAM) Glass Corrosion Program
- Contains ASTM C1285 ; PCT A (7day) and PCT B (up to 20 yrs)
- 490 Glasses (113 HLW and 377 LAW) – some exhibit Stage III and some do not

Database Is Being Used to Understand Many Aspects of the Relationship Between Glass Composition and Leaching Behavior (which impacts gel-leachate behavior)

- Dissolution processes of ion-exchange and diffusion form a “hydrogel layer”
- Hydrogels ripen into clays (causing no acceleration of long term durability) when there is little interaction with leachate species
  - *Solution is buffered*
- Hydrogels ripen/solution precipitates zeolites from strong interaction with leachate species (solution mediated), especially excess OH^- (Strong Base) (causing acceleration of long term durability)
  - *Solution not buffered, excess (Na,K,Li)OH in leachate interacts with alumino-silicate gel and Al in solution*
  - *Mimics industrial processing of zeolites from gel and NaOH*

Jantzen, C.M., Trivelpiece, C.L., Crawford, C.L, Pareizs, J.M, and Pickett, J.B., “Accelerated Leach Testing of GLASS: I. Waste Glass Hydrogel Compositions and the Resumption of Accelerated Dissolution, and): II. Mineralization of Hydrogels by Leachate Strong Bases.” International Journal of Applied Glass Science, 8[1] 69-83 and 84-96 (2017).

Relation of SRNL Strong Base-Weak Acid Model for PCT-A to PCT-B



The strong base-weak acid model represents effects of solution chemistry on glass dissolution behavior better than pH alone.

Dissolution of some glass compositions generate leachates enriched in strong base, high alkali containing glasses.

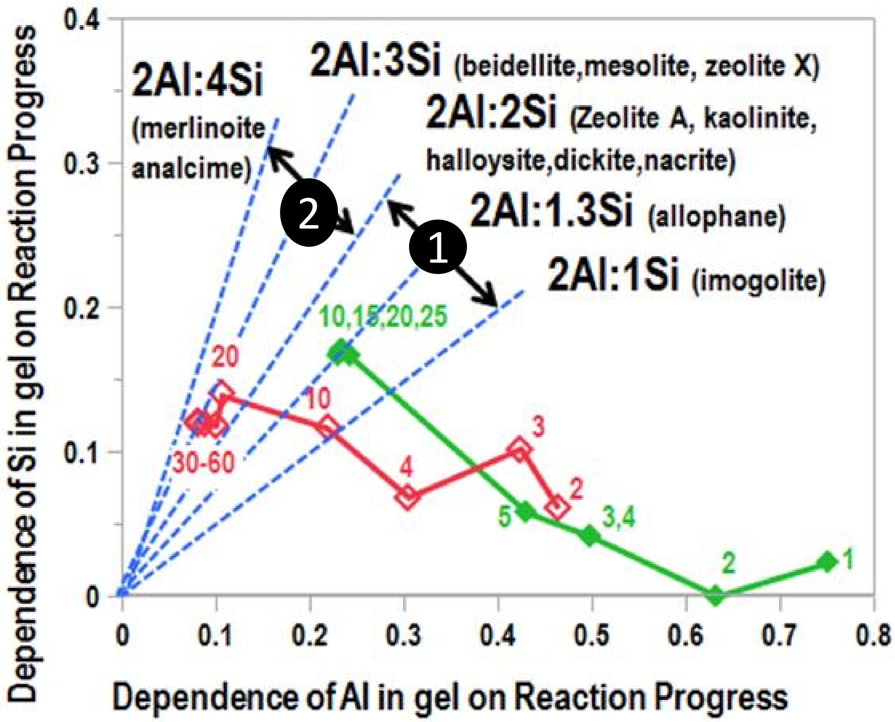
Dissolution of other glass compositions generate leachates enriched in weak acids.

$$Na_2O + SiO_2 + B_2O_3 + 6H_2O \rightarrow 2Na^+ + \underbrace{2OH^-}_{strong\,base} + \underbrace{H_4SiO_4}_{weak\,acid} + \underbrace{2H_3BO_3}_{weak\,acid}$$

$$\underbrace{[SB]-[WA]}_{leachate} \equiv \underbrace{[Na + Li + K + Cs]_{millimoles}}_{leachate} - \underbrace{[B + Si]_{millimoles}}_{leachate}$$

Jantzen, C.M., Pickett, J.B., Brown, K.G., Edwards, T.B., and Beam, D.C., "Process/ Product Models for the Defense Waste Processing Facility (DWPF): Part I. Predicting Glass Durability from Composition Using a Thermodynamic Hydration Energy Reaction MOdel (THERMO)," US DOE Report WSRC-TR-93-0672, 464p. (September, 1995).
Jantzen, C.M., et. al. International Journal of Applied Glass Science, 8[1] 69-83 and 84-96 (2017).

Calculated Evolution of Gel Composition from Database



❶ WA-enriched
❷ SB-enriched

Jantzen, C.M., et. al. International Journal of Applied Glass Science, 8[1] 69-83 and 84-96 (2017).

Hydrogel compositions were calculated from differences between measured solution concentrations and predicted based on dissolution congruent with boron

WA and SB enriched samples were sorted into resumption and non-resumption populations: gel composition trends with reaction time (progress) were investigated with stepwise regression

SB-enriched sample gels gave Al/Si ratios consistent with zeolite X (2 Al:3 Si) and analcime (2 Al:4 Si)

WA-enriched sample gels gave Al/Si ratios consistent with allophane (2 Al:1.3 Si) and Fe/Si ratios consistent with hisingerite /smectite/nontronite (2 Fe:2 Si)

Solution-Mediated Reactions That May Trigger Stage III Behavior

If leachate is not enriched in strong base:

$$\underbrace{3Na_2O \bullet 2Fe_2O_3 \bullet 2Al_2O_3 \bullet 12SiO_2}_{hydrogel} + 17H_2O \rightarrow$$

$$\underbrace{2(Fe^{+3})_2 Si_2O_5(OH)_4 \bullet 2H_2O}_{hisingerite} + \underbrace{2(Al_2O_3 \bullet SiO_2)}_{hydrogel} + \underbrace{6NaOH + 6H_2SiO_3}_{buffered\ leachate}$$

If leachate is enriched in strong base:

$$\underbrace{3Na_2O \bullet 2Fe_2O_3 \bullet 2Al_2O_3 \bullet 12SiO_2}_{hydrogel} + 2LiOH + 7H_2O \rightarrow$$

$$2\underbrace{NaLiAl_2Si_4O_{12} \bullet 2H_2O}_{analcime\ precursor\ gel} + \underbrace{2(Fe_2O_3 \bullet SiO_2)}_{hydrogel} + \underbrace{4NaOH + 2H_2SiO_3}_{SB\ dominated\ leachate}$$

The incorporation of these process dependencies and strong base-weak acid model into Stage 3 model as triggering mechanism is being evaluated.

Jantzen, C.M., et. al. International Journal of Applied Glass Science, 8[1] 69-83 and 84-96 (2017).

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Durability of Homogeneous vs. Inhomogeneous Glasses

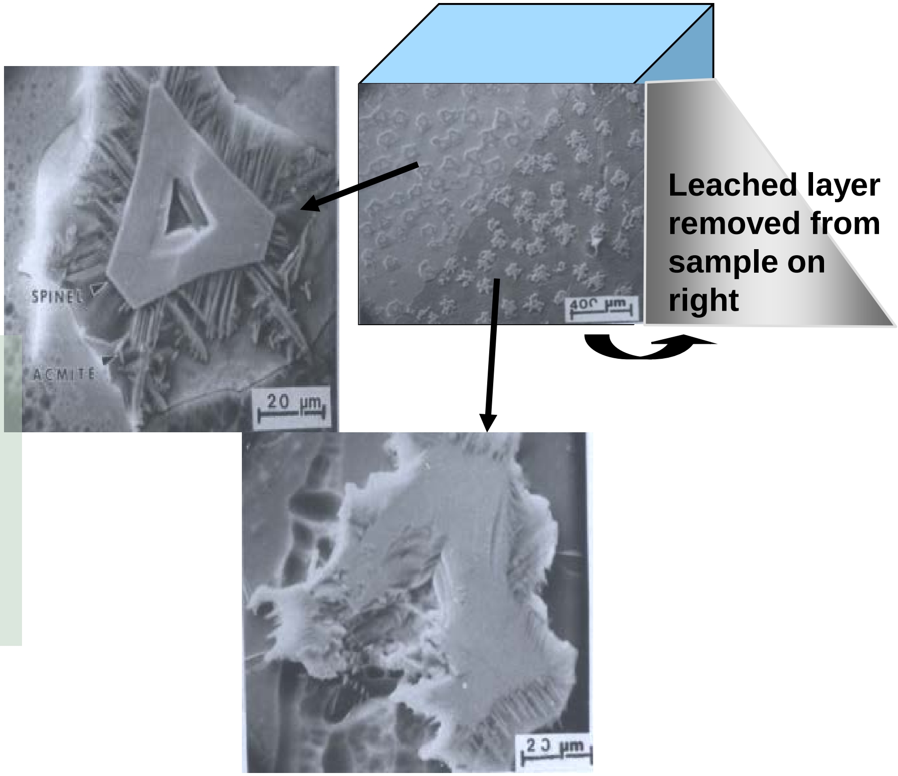
Homogeneous Glass

$$\sum Durability = \underbrace{durability_{(homogeneous)}}_{1st\ term} + \underbrace{durability_{\cancel{(amorphous\ phase\ separation)}}}_{\cancel{2nd\ term}} + \underbrace{durability_{\cancel{(crystallization)}}}_{\cancel{3rd\ term}}$$

$$+ \underbrace{durability_{\cancel{(accelerated\ grain\ boundary)}}}_{\cancel{4th\ term}}$$

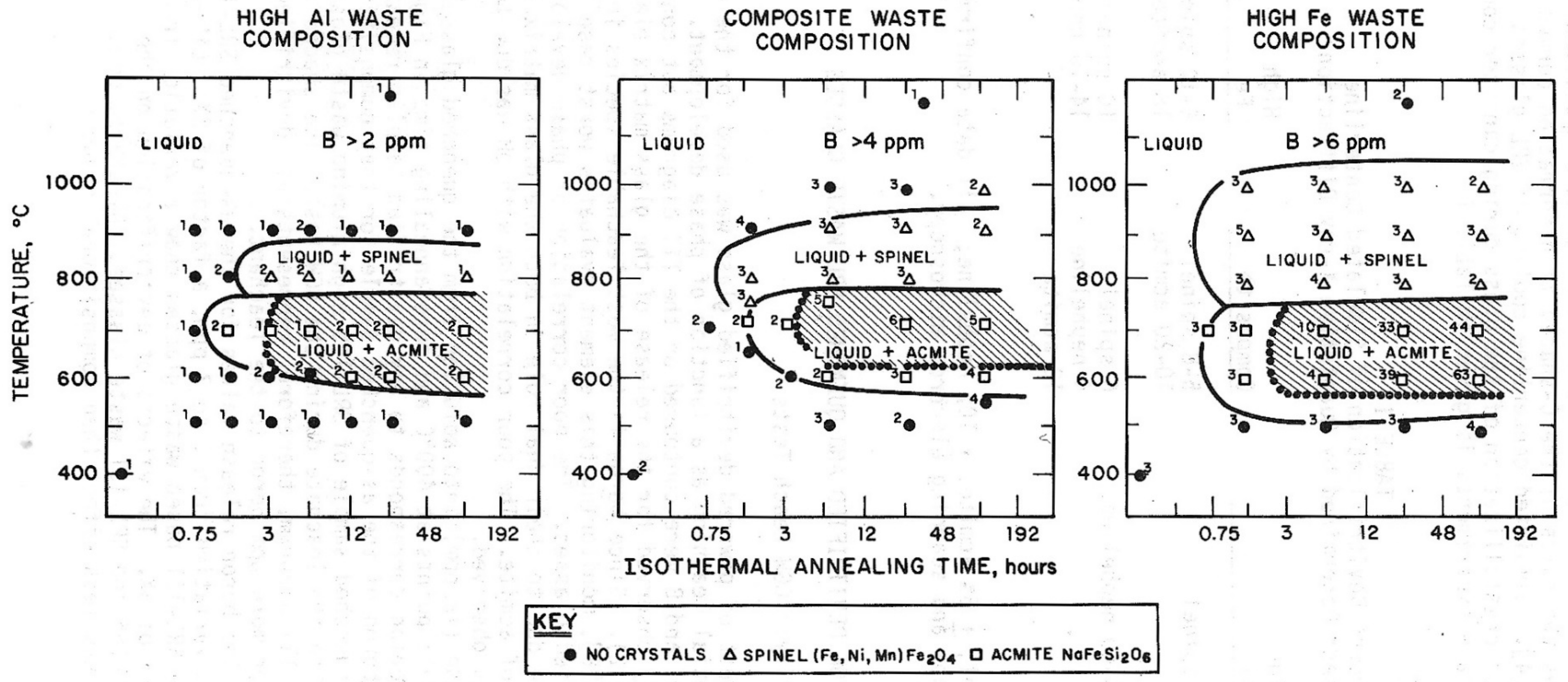
Phase Separated Glass and/or crystallized glass (requires that distribution of radionuclides amongst the phases be known)

Crystallization of spinel (isometric) produces no impact on glass durability, i.e. does not deplete surrounding glass of glass forming species that can degrade durability



Jantzen, C.M., Brown, K.G., and Pickett, J.B., International Journal of Applied Glass Science, 1 [1], 38-62 (2010).

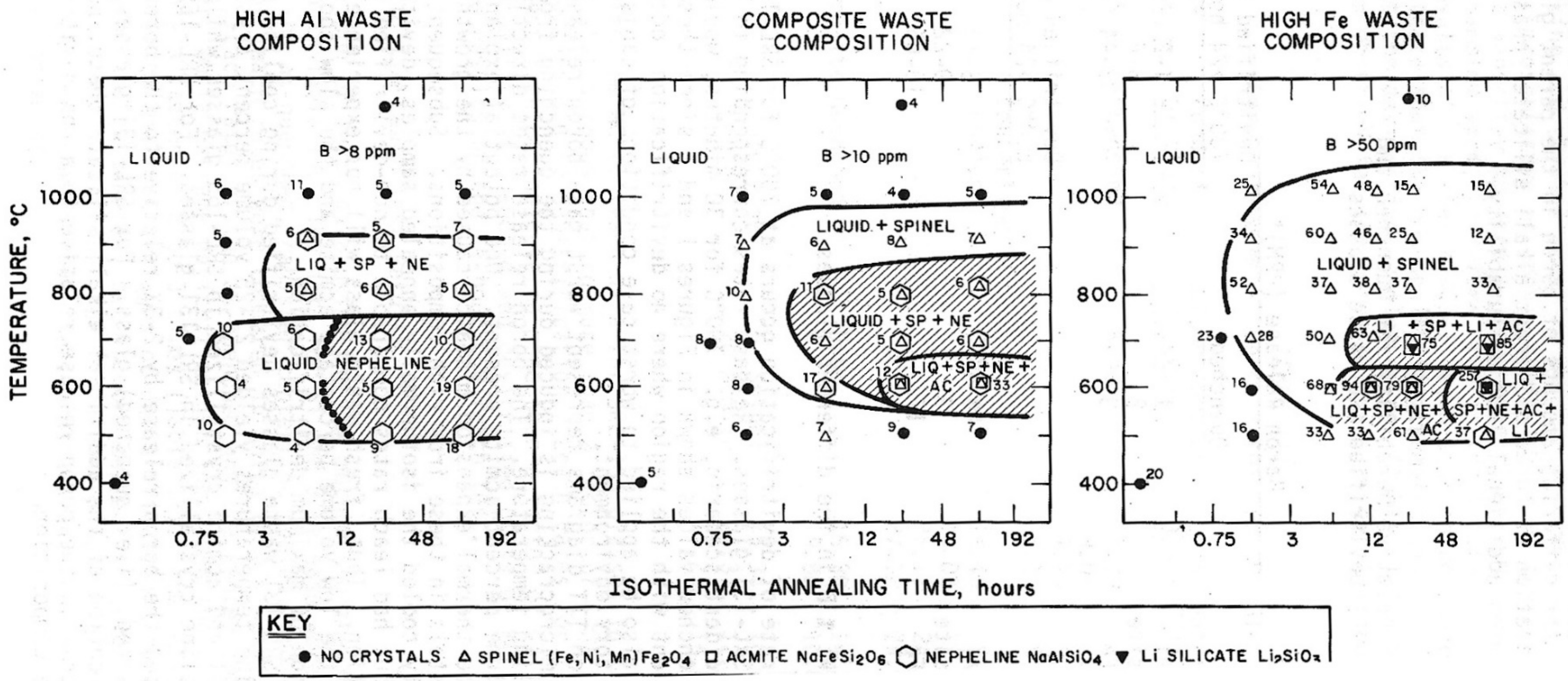
Effect of Crystallization on Durability - 165 Glass



- Also done for a variety of other glasses including the Waste Compliance Plan (WCP) glasses that span the range of compositions to be produced at SRS

Jantzen, C.M. and Bickford, D.F., "Leaching of Devitrified Glass Containing Simulated SRP Nuclear Waste," Sci. Basis for Nuclear Waste Management, VIII, C.M. Jantzen, J.A. Stone and R.C. Ewing (eds.), Materials Research Society, Pittsburgh, PA 135-146 (1985).

Effect of Crystallization on Durability -131 Glass



- Crystallization of nepheline ($NaAlSiO_4$; not isometric) does impact glass durability-depletes surrounding glass of glass formers (Si and Al)
- "nepheline discriminator" used as part of process control to avoid HLW glass compositions that could precipitate nepheline during canister cooling

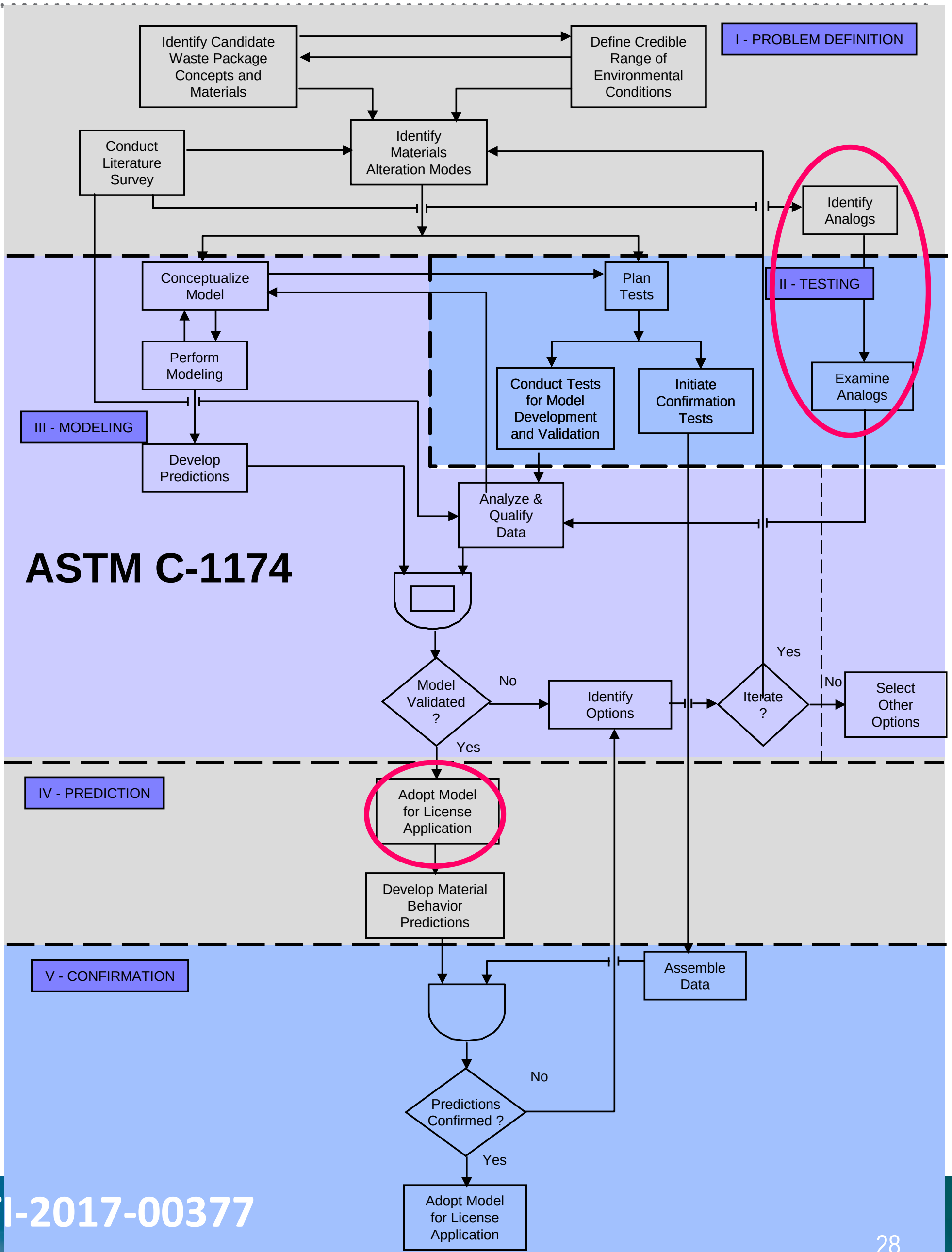
Jantzen, C.M. and Bickford, D.F., "Leaching of Devitrified Glass Containing Simulated SRP Nuclear Waste," Sci. Basis for Nuclear Waste Management, VIII, C.M. Jantzen, J.A. Stone and R.C. Ewing (eds.), Materials Research Society, Pittsburgh, PA 135-146 (1985).

ASTM C-1174 “The Roadmap to Predicting Long-term Behavior

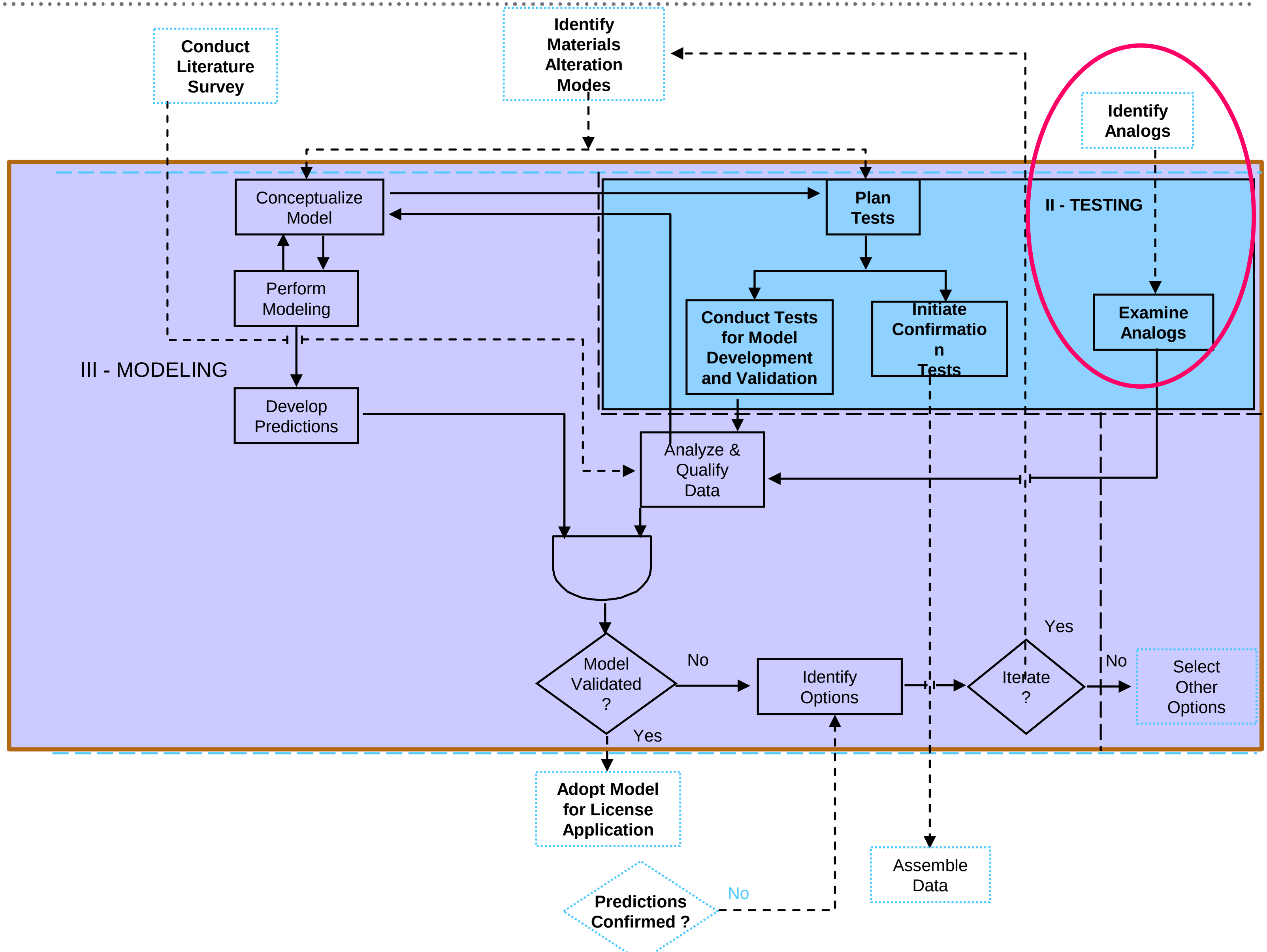
Title: “Standard Practice for Prediction of the Long-Term Behavior of Materials, Including Waste Forms, Used in Engineered Barrier Systems (EBS) for Geological Disposal of High-Level Radioactive Waste”

A roadmap for the steps involved in predicting long-term behavior:
(1) problem definition
(2) Testing (includes natural analogs and how to accelerate testing)
(3) modeling
(4) prediction
(5) model confirmation.

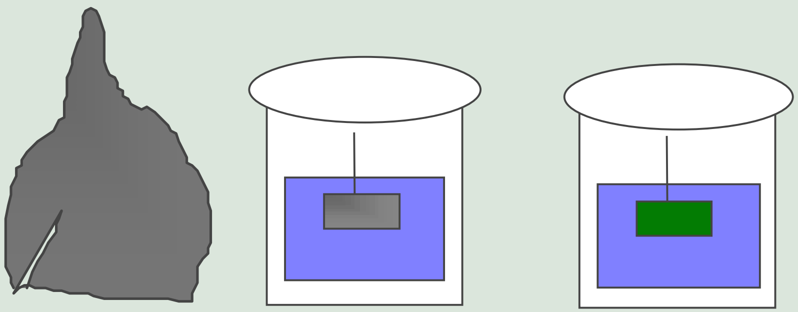
Many iterations between testing and modeling



ASTM C-1174 (Testing Module)



Use of Natural and Ancient Glass Analogs



Naturally Reacted Glass

Experiment On Natural Glass

Experiment On Waste Glass

- Natural Glasses
  - Obsidians
  - Basalt
  - Tektites
- Ancient Glasses
  - Libyan Desert Glass
  - Ancient and Medieval Glasses
- Other Glasses (Window, Pyrex)

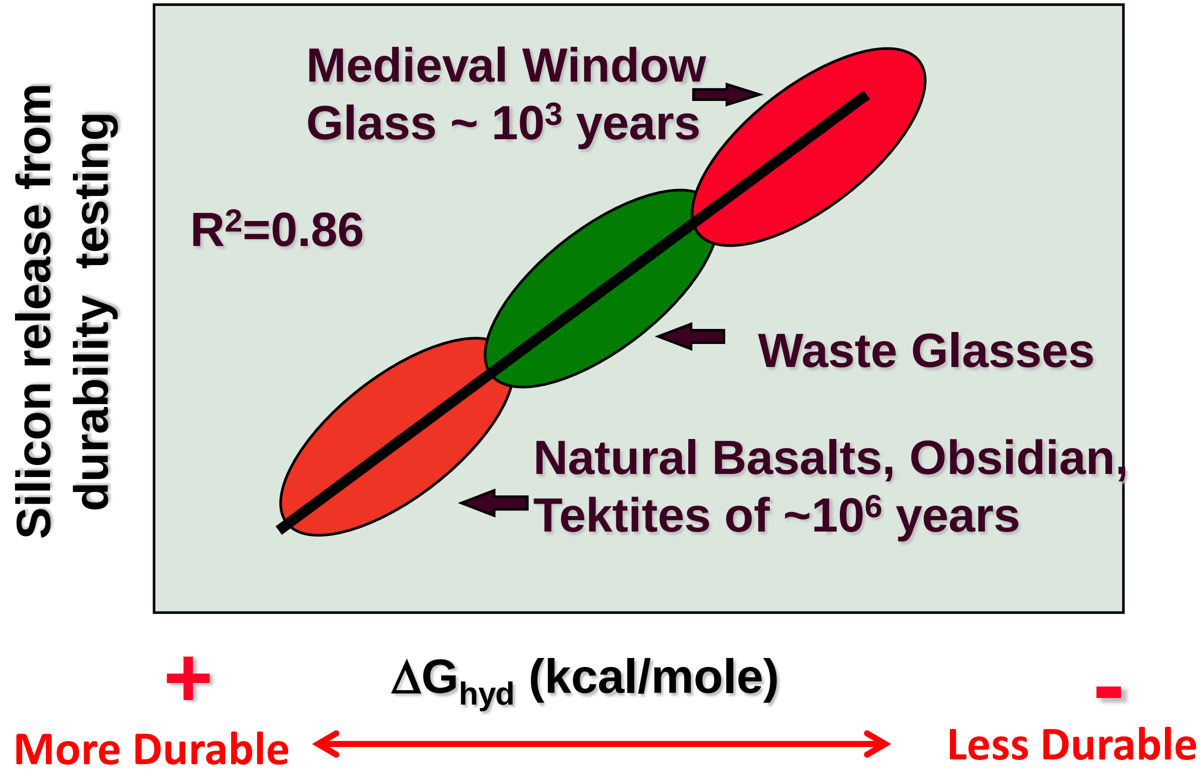
Helps determine leaching mechanisms, gel formation, gel conversion to clays and zeolites and relations to natural and ancient glasses

Jantzen, C.M. and Plodinec, M.J., "Thermodynamic Model of Natural, Medieval, and Nuclear Waste Glass Durability," J. Non-Cryst. Solids, 67, 207-233 (1984).

Historical Context for Waste Glass Durability

Glass Composition (mole fraction)

0.14 Na_2O
0.06 MgO
0.05 CaO
0.75 SiO_2



$0.14\ \Delta G_{hyd}\ \{Na_2O \cdot SiO_2 + 2H_2O \rightarrow 2Na^+ + 2\ OH^- + H_2SiO_3\} = 0.14\ (-28.815)$

$0.06\ \Delta G_{hyd}\ \{MgO \cdot SiO_2 + 2H_2O \rightarrow Mg^+ + 2\ OH^- + H_2SiO_3\} = 0.06\ (-13.888)$

$0.05\ \Delta G_{hyd}\ \{CaO \cdot SiO_2 + 2H_2O \rightarrow Ca^+ + 2\ OH^- + H_2SiO_3\} = 0.05\ (-16.116)$

$0.50\ \Delta G_{hyd}\ \{SiO_2 + H_2O \rightarrow H_2SiO_3\} = \underline{0.50\ (+5.59)}$

Sum ΔG° = -2.878 Kcal/mole

Jantzen, C.M. and Plodinec, M.J., "Thermodynamic Model of Natural, Medieval, and Nuclear Waste Glass Durability," J. Non-Cryst. Solids, 67, 207-233 (1984).

Responses to Talking Points

1. What are the technical bases, including standards, test methods, and use of databases and models, for DOE's criteria for qualifying borosilicate glass waste forms as acceptable for disposal in a geologic repository? *A variety of ASTM tests are used to determine glass durability mechanisms including PCT-B while PCT-A is used for production quality assurance. The main HLW glass durability standard is the EA glass, the main database of short term PCT-A and long term PCT-B is ALTGLASS, waste form producers use different models (i.e. THERMO™) from the repository modelers.*
2. What is DOE's technical basis for applying the results of short-term tests on reference glasses or glasses with simplified compositions to assessments of the long-term performance of more chemically complex HLW glasses in repository environments? *Glass standards are complex glasses usually without radionuclides. Glasses with radionuclides were tested and no differences in the leaching characteristics or mechanisms were noted with non-radioactive equivalents which gives confidence that it is acceptable to use these non-radioactive glasses.*
3. What is known about the influence of glass chemistry on crystallite precipitation during glass production and on glass corrosion, and how are crystallites taken into account in DOE's approach to designing glass for disposal in a repository? *Crystalline phases that are known to impact glass durability are avoided by careful glass formulation. Crystalline phases that do not have an impact on glass durability are allowed.*
4. Are data on natural and archeological glasses used to support DOE assessments of the long-term performance of HLW glass in a repository and, if so, how? Natural analog glasses have been a part of DOE assessments of long-term performance since the late 1970's. Natural analogs help determine leaching mechanisms, gel formation mechanisms, gel conversion to clays and/or zeolites, longevity of waste form (historical context).

Backup Slides

Waste Acceptance Product Specifications (EM-WAPS-Rev. 3)

WAPS 1.0 Waste Form Specifications (Glass or RW Equivalent and complies with NWPAA)

WAPS 1.1 Chemical Composition (projections and during production)

WAPS 1.2 Radionuclide Inventory (projections and during production)

WAPS 1.3 Product Consistency (more durable than the EA glass by two standard deviations; can project durability and/or meas. production glasses)

WAPS 1.4 Phase Stability

WAPS 1.5 Hazardous Waste

WAPS 1.6 IAEA Safeguards

WAPS 2.0 Canister Specifications

WAPS 2.1 Material

WAPS 2.2 Fabrication and Closure

WAPS 2.3 Identification and Labeling

WAPS 2.4 Canister Dimensions

WAPS 3.0 Canistered Waste Form

WAPS 3.1 Exclusion of free liquid/gases

WAPS 3.2 Tamper Indicating Seals

WAPS 3.3 Exclusion of explosive, pyrophoric and combustible materials

WAPS 3.4 Exclusion of organics

WAPS 3.5 Chemical Compatability

WAPS 3.6 Metric Tons of Heavy Metals

WAPS 3.7 Surface Decontamination

WAPS 3.8 Heat Generation

WAPS 3.9 Maximum Dose Rates

WAPS 3.10 Subcriticality

WAPS 3.11 Weight and Overall Dimensions

WAPS 3.12 Drop Test

WAPS 3.13 Handling Features

WAPS 3.14 Plutonium Concentration

WAPS 4.0 Quality Assurance

RW-0333P

HLW Vitrified in the US as of 2016

Vitrification Plant	Borosilicate Waste Glass Produced (metric tons)	Waste Loading Range (wt%)	Size of Canisters (meters)	Number of Canisters
DWPF Savannah River Site*	7200 (1996-2013)	28-40	0.61 x 3.05	4,242 made 3,928 proj.
West Valley Demonstration Project (WVDP)[t]	~573.8 (1996-2002)	~20.4-23.5	0.61 x 3.05	275 made 275 proj.
Hanford Waste Treatment Plant HLW**	31,968 (projected)	~35-40	0.61 x 4.57	0 10,586

*Chew and Hamm 2016
** Certa et. al. 2011
T Palmer , et. al 2004