Low Activity Waste Glass Waste Form Data Package for Hanford's Integrated Disposal Facility Performance Assessment - 16480

David J. Swanberg[1], Elvie E. Brown[1], Diana H. Bacon[2], Vicky L. Freedman[2], Joseph V Ryan[2], [1]Washington River Protection Solutions, [2]Pacific Northwest National Laboratory

ABSTRACT

A Direct Feed Low Activity Waste (DFLAW) flowsheet has been initiated to begin immobilization of Hanford tank wastes. In the DFLAW configuration, a pretreatment system will segregate the high level and the low activity wastes (LAW). The LAW will be immobilized in the Waste Treatment Plant (WTP) and disposed of in Hanford's Integrated Disposal Facility (IDF).

To obtain authorization to dispose of wastes in the IDF, a Performance Assessment (PA) must be completed in accordance with DOE Order 435.1. The PA shall include calculations of potential doses to representative future members of the public and potential releases from the facility to provide a reasonable expectation that performance objectives will not be exceeded as a result of operation and closure of the facility. To support the development of the PA, data from ILAW glass testing are needed to model the long term release of contaminants over the compliance period and verify that impacts to groundwater quality will be within performance objective limits.

Washington River Protection Solutions is conducting an ILAW glass testing program to produce the required data. This program is a combined effort involving National Laboratories with known expertise in glass waste form research. The work involves accelerated testing to develop parameters that can be used in computer simulations of glass corrosion and contaminant release in the IDF environment. The results of this work will be compiled in an ILAW glass waste form performance data package for the IDF PA.

INTRODUCTION

Radioactive byproduct wastes from nuclear weapon production are stored in underground tanks at the Hanford Site, located in the state of Washington. The waste tanks contain a complex and diverse mix of radioactive and chemical waste in the form of sludge, salts, and liquids, necessitating a variety of unique waste retrieval, treatment, and disposition methods. Generically, the tank waste can be characterized as the following:

Sludge – Insoluble materials largely consisting of metal hydroxides and oxides that precipitated when acidic wastes from spent nuclear fuel processing and other activities were neutralized and converted to high pH for storage in carbon steel tanks. The sludge fraction of the waste makes up the bulk of the material that will be processed via high-level waste (HLW) vitrification into a stable glass form.

Supernatant – Liquid waste with high sodium content and high pH.

Saltcake – a mixture of salts that precipitated from supernatant as the concentration was increased by evaporation to reduce tank storage space requirements. Saltcake must be re-dissolved and processed as supernatant waste. The supernatant and saltcake contain the majority of highly radioactive cesium which must be separated and processed with the sludge stream into HLW glass. The decontaminated supernatant will be processed via low-activity waste (LAW) vitrification into a stable glass form.

Potential contact-handled transuranic waste (CH-TRU) – a mixture of sludge and saltcake consisting of some 1.4 million gallons in 11 specific single-shell tanks (SSTs). The material in these tanks is being reviewed to determine the potential to transfer to the Waste Isolation Pilot Plant (WIPP) versus being processed on-site into HLW and LAW glass fractions.

In order to begin immobilization of tank waste as soon as practicable, a Direct Feed LAW (DFLAW) flowsheet has been initiated. In the DFLAW configuration, LAW feed will be provided to the LAW Pretreatment System (LAWPS). The LAWPS will separate the HLW and LAW fractions and provide qualified feed to the WTP-LAW Vitrification Facility. The HLW fraction will be returned to the double shell tank (DST) system.

Successful startup and operation of DFLAW requires the completion of engineering, design and construction of numerous facilities, flowsheet stewardship, program integration across facilities, generation of a series of permits, and development of the regulatory framework to dispose of the waste forms generated. This paper discusses the activities involved in the development and testing of the immobilized LAW (ILAW) glass waste form that will be produced in the LAW Vitrification facility.

The ILAW glass will be disposed in the Integrated Disposal Facility (IDF). However, a performance assessment (PA) must be performed to provide the regulatory basis for issuance of a Disposal Authorization Statement (DAS) addressing the radioactive constituents in the waste. The PA results may also be used to support a Resource Conservation and Recovery Act (RCRA) permit modification before the waste can be placed in the IDF. The PA utilizes computer models to project human health and environmental risks/impacts of IDF operation and closure using key data from expected waste forms and other IDF information. This paper delineates the development and testing of ILAW glasses and discusses the key data needed for the PA.

BACKGROUND

DOE Order 435.1 (*Radioactive Waste Management)* and its accompanying manuals delineates the prerequisites and processes of a low-level radioactive waste near-surface disposal PA. It states that,

“A site-specific radiological performance assessment shall be prepared and maintained for DOE low-level waste disposed of after September 26, 1988. The performance assessment shall include calculations for a 1,000-year period after closure of potential doses to representative future members of the public and potential releases from the facility to provide a reasonable expectation that the performance objectives identified are not exceeded as a result of operation and closure of the facility…"

PAs use integrated models to represent the engineered and natural systems in order to evaluate the long-term performance of a disposal facility. For ILAW glasses, it is postulated that when the glass corrodes, it will release components of the glass matrix along with the contaminants of concern (COCs) into the surrounding environment, which can then leach into the groundwater. ILAW glasses need to be tested to obtain parameters for the modeling of its long-term performance in the PA. The PA is required for the IDF to obtain the DAS issued by DOE/HQ and a RCRA permit needed for the operation of the IDF (see Figure 1).

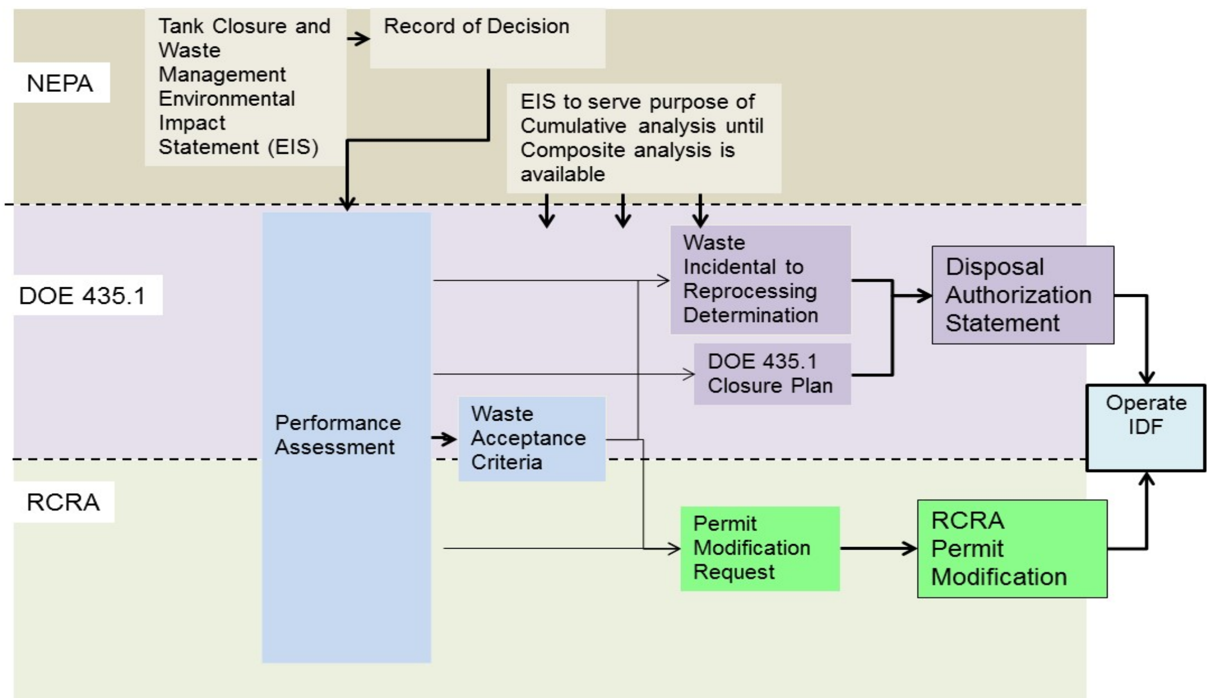

Figure 1. Regulatory framework for the authorization of IDF operation.

The IDF PA must comply with requirements delineated in the DOE Order and its accompanying manual. DOE M 435.1 prescribes numerous post-closure requirements that a low-level waste disposal facility must satisfy to obtain permission to operate. For some of these requirements (radiation dose limits to potential recipient), relevant exposure scenarios must be constructed and evaluated in a PA analysis to demonstrate compliance with the requirements.

The IDF PA must include a rigorous analysis using best available data and appropriate tools (computational models) to demonstrate that requirements specified in DOE O 435.1 (and its accompanying manuals) for the disposal facility will be met over the period of compliance, considering all agreed-upon exposure scenarios. The PA analysis will underpin the development of specific Waste Acceptance Criteria (WAC) for the IDF. The IDF WAC are currently being drafted but cannot be finalized until the PA analyses are complete, as shown in Figure 1. The WAC may be used to allow practical characteristics and limits to be established for the various waste types that are intended for IDF disposal.

The IDF PA will also serve a key role in supporting a Waste Incidental to Reprocessing (WIR) determination for the ILAW, as indicated in Figure 1. The PA will first undergo review for technical acceptability by the Low-Level Waste Federal Review Group (LFRG), and the WIR evaluation process will include consultation with the DOE Office of the Environmental Management (EM) and the US Nuclear Regulatory Commission (NRC).

The IDF is also a RCRA facility regulated by the Washington State Department of Ecology (Ecology) under *Washington Administrative Code* (WAC) 173-303, "Dangerous Waste Regulations." The RCRA regulations require risk assessments and environmental impact analyses to support the permitting process. The IDF PA will provide technical information needed by Ecology to support the required RCRA permit modification.

History of Hanford PA and ILAW Glass Program.

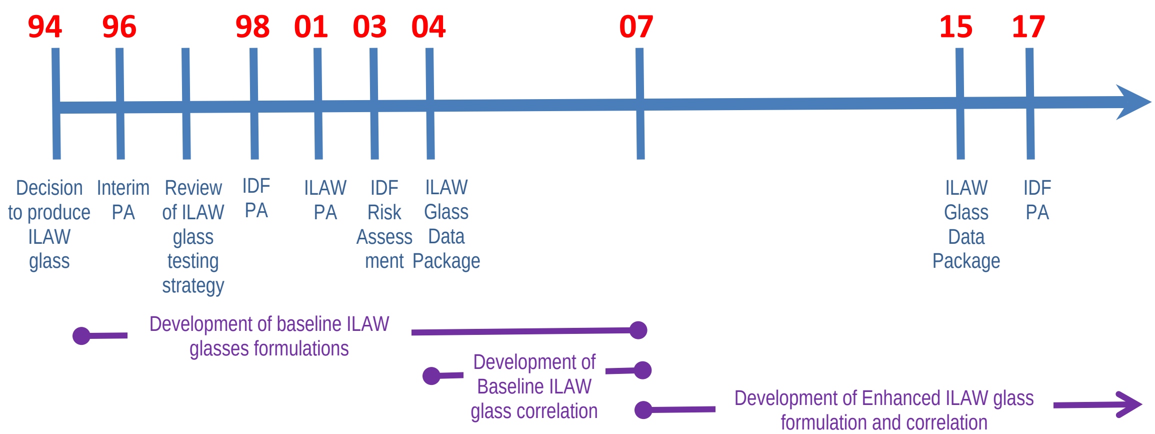

Figure 2. Timeline of ILAW glass program activities associated with the IDF PA.

The timeline of the IDF PA and supporting ILAW glass development program activities is shown in Figure 2. The first Hanford ILAW disposal PA occurred shortly after the decision to change the ILAW waste from grout to glass in the mid-1990s. An initial analysis was performed in 1994, which led to a data collection effort after realizing that certain parameters from actual glasses were needed to support the PA. Reference glasses were formulated, developed and tested and an interim PA was prepared in 1996 (Mann et al., 1996). The interim PA used a constant COC leach rate in the base case scenario as an input parameter to a simulation model to estimate the radiation dose from COCs in the groundwater below the disposal site.

The 1996 PA concluded that succeeding PAs would benefit from information on actual waste forms, disposal facility geologic features and other key data. Relative to ILAW glass, a testing strategy was developed to fill the information gaps (McGrail et al., 1998). A series of experimental techniques were outlined as part of the strategy that, when combined with scientific theories, would generate parameters needed to implement a transport model that could simulate long-term performance. Consequently, the 1998 PA was substantially more robust than the previous PAs because of increased understanding of waste form performance and disposal facility characteristics.

The ILAW PA was further refined in 2001 but was still based on a concept of disposing ILAW containers in large concrete vaults. Although the IDF was not yet designed and constructed, a 2003 risk assessment evaluated ILAW glass and other supplemental waste forms using a lined disposal trench concept that evolved to become the IDF (Mann, et al., 2003).

In addition to ILAW glass disposal, the mission of the IDF was expanded to include secondary wastes that would be generated during the Hanford Waste Treatment Plant (WTP) operations and other non-WTP waste forms. During this time frame, additional ILAW glass testing was conducted and a Data Package was prepared to support a PA for the IDF (Pierce, et. al., 2004). This coincided with construction of the IDF in the 200 East Area of the Hanford site in 2005. By this time the glass formulation approach had transitioned from reference glasses to formulations tailored to waste composition. Glasses were developed to address the ratio of sulfate to sodium in the waste since sulfate incorporation in the glass was recognized as a significant factor affecting waste loading or the amount of LAW that could be incorporated in each container of ILAW glass. This formulation approach was also used to establish a correlation between glass composition, processing characteristics, and waste form performance known as the Baseline ILAW glass correlation (Muller et al., 2004). The initial 2017 IDF PA analyses will use waste form performance data from baseline ILAW glass formulations (Freedman, et al., 2015).

The Baseline ILAW glass correlation was developed to provide practical, robust glass formulations for the initial operations of the WTP LAW Vitrification facility. However, increasing the throughput of WTP LAW melter could greatly reduce the cost per ton of LAW processed. Over the past several years, the ILAW glass formulation strategy has been focused on formulating glasses that can achieve significantly higher waste loadings than the Baseline glasses. This strategy will reduce the total amount of glass to be produced by the WTP and will reduce plant operating lifetime and cost. The development of this strategy started in 2007 and is an ongoing effort until the correlation is completed (Peeler et al., 2015). Testing for PA data is ongoing but will not be available in time for the initial release of the IDF PA, thus the information will be provided to support PA maintenance activities.

Under DOE O 435.1, it is stated that the PA shall be maintained to evaluate changes that could affect the performance, design, and operating bases for the facility. PA and composite analysis maintenance shall include the conduct of research, field studies, and monitoring needed to address uncertainties or gaps in existing data. Continuous testing shall be performed to provide data for PA maintenance.

METHODS

ILAW Glass Preparation

Glass formulation development typically starts with fabricating glass at the crucible scale using non-radioactive chemicals and glass former additives. This enables a large number of glass compositions to be tested in a relatively short period of time at modest expense. Glasses fabricated in this manner can quickly be characterized for processing properties such as melt viscosity and electrical conductivity as well as chemical durability in short term leach tests. Glasses that exhibit desirable properties at the crucible scale are then selected for testing in continuously–fed, scaled melter systems. In melter tests, a LAW simulant is combined with glass forming chemicals and fed to a melter operating at 1100 – 1150^{o}C. The glass is poured into containers and samples of the glass are periodically taken and subjected to the same durability tests as glasses made in crucible melts. To fully mimic the process that will be used in the plant, both crucible and melter glass samples may be re-melted and cooled slowly to represent the thermal conditions the glass would experience when poured into the full scale production container. Numerous studies have shown that glasses produced at the crucible scale have the same properties as glasses of the same composition produced in melters up to, and including pilot scale.

ILAW Glass Testing

It is postulated that corrosion/dissolution of the ILAW glass matrix is the fundamental mechanism that governs the rate at which the COCs leach into the surrounding environment and eventually reach the groundwater. It is imperative therefore to perform tests on representative glasses to ensure glasses with acceptable performance are disposed of in the IDF. To provide the key modeling input parameters for near-field reactive transport modeling, well-constrained and interpretable experiments have been developed to isolate and parameterize the key mechanisms of glass corrosion. As the glass contacts with water, as what is presumed in the IDF, there are three mechanisms that are important for characterizing glass corrosion, namely, ***matrix dissolution*** (e.g. *kinetic rate law*), alkali ***ion exchange***, and the effects of ***secondary phase formation***.

Matrix dissolution represented by kinetic rate law. Glass dissolution is a complex process that is considered to proceed through a number of reversible and irreversible reactions,

$$SiO_2\ (s) + 2H_2O \leftrightarrow SiO_2 \bullet 2H_2O \rightarrow H_4SiO_4\ (aq)$$

for which the rate-determining step is the decomposition of the activated complex ($SiO_2 \bullet 2H_2O$).

At present, the kinetic rate law based on Transition State Theory (TST) is considered to best describe the network hydrolysis and matrix dissolution of glass. This rate law was developed by Aagaard and Helgeson (1982) and applied to glass by Grambow (1985):

$$r = \vec{k} v_i a_{H^+}^{-\eta} exp\left(\frac{-E_a}{RT}\right)\left[1-\left(\frac{Q}{K_g}\right)^{\sigma}\right] \quad \text{Equation 1}$$

where,

r = dissolution rate, g m^{-2} d^{-1}

$\vec{k}$ = intrinsic rate constant, g m^{-2} d^{-1}

v_i = mass fraction of component *i*

a_{H+} = hydrogen ion activity

η = pH power law coefficient

E_a = activation energy, kJ/mol

R = gas constant, kJ/(mol·K)

T = temperature, K

Q = ion activity product of rate controlling reaction

K_g = pseudo-equilibrium constant of rate controlling reaction

σ = Temkin coefficient

The kinetic rate law parameters; $\vec{k}$, E_a, K_g, and η are determined from Single Pass Flow-Through (SPFT, ASTM C1662) laboratory experiments performed with ILAW glass samples. In SPFT experiments, the dissolution rate is measured by monitoring the effluent concentrations of the elements that comprise the glass matrix (e.g., boron, aluminum, silicon, or sodium). By manipulating the temperature, solution flow rate, pH, and concentrations of the glass components in the solution, data can be obtained to quantify the kinetic rate law parameters.

Ion Exchange. Ion exchange is the interdiffusion process of H^+, H_3O^+ and/or H_2O in the fluid phase being exchanged for network-modifying cations in the glass. In glasses with a high sodium content, the normalized release of sodium (Na) is expected to be higher than the release of other glass network components. The release of Na has been attributed to alkali ion exchange on the surface of the glass:

$$Glass \ldots Na + H^+ \xrightarrow{r_{IEX}} Glass \ldots H + Na^+, r_{IEX} = r_0 exp\left(\frac{E_{IEX}}{RT}\right) \quad \text{Equation 2}$$

where,
r_{IEX} = ion exchange rate, mol Na/m^2s
r_0 = intrinsic ion exchange rate constant, mol Na/m^2s
E_{IEX} = activation energy for ion exchange, J/mol

The rate of the ion exchange is denoted as r_{IEX}, which is a key input parameter in the PA, in addition to the rate law parameters. The ion exchange rate is an important consideration for waste glasses that have higher alkali proportions (sodium, lithium, and potassium) relative to aluminum and boron. In laboratory studies, ion exchange rate is quantified through analysis of the effluent from the SPFT experiment.

Secondary phase formation. When the components that dissolve out of the glass accumulate and reach their saturation point, secondary phases will form, especially phases involving silica. These secondary phase reactions can either slow down or accelerate glass corrosion. The information on the secondary phase reaction network from ILAW glass samples can be obtained from laboratory testing utilizing either the Long Term Product Consistency Test (PCT, ASTM C1285) or Pressurized Unsaturated Flow (PUF) test. The secondary phase reaction network can be obtained by identifying the secondary phases that form, determining the reactions involved and using geochemical modeling software to determine the parameters needed for the PA analyses.

Modeling of ILAW glass behavior in IDF PA. To simulate the dissolution behavior of glass as depicted by kinetic rate law, coupled with ion exchange and the secondary phase reactions, Equations 1 and 2 along with the reaction network can be directly input into a numerical modeling code capable of performing reactive transport calculations.

RESULTS

The compositions of selected ILAW glasses that have been tested and analyzed to obtain the parameters to be used for the IDF PA are listed in Table 1.

Table 1. Composition (mass%) of ILAW Glasses for IDF PA.

Oxide	LD6-5412[1]	LAWABP1[2]	LAWA44[3]	LAWB45[3]	LAWC22[3]
Al_2O_3	12	10	6.2	6.13	6.08
B_2O_3	5	9.25	8.9	12.34	10.06
CaO	4	NI	1.99	6.63	5.12
Fe_2O_3	NI	2.5	6.98	5.26	5.43
K_2O	1.46	2.2	0.5	0.26	0.1
La_2O_3	NI	2	NI	NI	NI
Li_2O	NI	NI	NI	4.62	2.51
MgO	NI	1	1.99	2.97	1.51
Na_2O	20	20	20	6.5	14.4
SiO_2	55.91	41.89	44.55	47.86	46.67
SO_3	0.21	0.1	0.1	0.84	0.34
TiO_2	NI	2.49	1.99	0.00	1.14
ZnO	NI	2.6	2.96	3.15	3.07
ZrO_2	NI	5.25	2.99	3.15	3.03
Others*	1.42	0.72	0.85	0.29	0.54

NI – Not Included
*Others include minor amounts of Ag_2O, BaO, CdO, Ce_2O_3, Cl, Cr_2O_3, Cs_2O, F, I, MnO, MoO_3, Nd_2O_3, NiO, P_2O_5, PbO_2, Pr_2O_3, Re_2O_7, SO_3, SeO_2, SrO, TeO_2, and Y_2O_3. Not all elements are present in every glass composition.
[1]McGrail, et al., 1997
[2]McGrail, et al., 2001
[3]Muller, et al., 2001

Glasses identified as LD6-5412 and LAWABP1 were developed and tested to support pervious ILAW PA analyses. LAWA44, B45, and C22 are part of the Baseline ILAW glass correlation and their compositions correspond to a range of expected waste stream compositions. The kinetic rate law and ion exchange parameters obtained from testing these glasses are listed in Table 2. These parameters may be used in performing the modeling analyses for the IDF PA.

Table 2. Summary of ILAW Glass Parameters for IDF PA

Para-meter	Meaning	LD6-5412[1]	LAWABP1[2]	LAWA44[3]	LAWB45[3]	LAWC22[3]
$\vec{k}$	forward rate constant (g/[m^2 d])	9.7×10^6	3.4×10^6	1.3×10^4	1.6×10^4	1.0×10^5
K_g	apparent equilibrium constant for glass based on activity product $a[SiO_2(aq)]$	1.14×10^{-4}	4.9×10^{-4}	*1.87×10^{-3}	*1.79×10^{-3}	*1.80×10^{-3}
η	pH power law coefficient	0.40 ±0.03	0.35 ±0.03	0.49 ±0.08	0.34 ±0.03	0.42 ±0.02
E_a	activation energy of glass dissolution reaction (kJ/mol)	74.8 ±1.0	68 ±3.0	60 ±7	53 ±3	64 ±2
σ	Temkin coefficient, assigned constant	1	1	1	1	1
r_{IEX}	Na ion-exchange rate (mol/[m^2 s])	1.74×10^{-11}	3.4×10^{-11}	5.3×10^{-11}	0 **	1.2×10^{-10}

*Values cited in the original publication (Pierce et al., 2004) were subject to a calculation error. Values in this table are corrected.
** No detectable ion exchange rate
[1]McGrail et al., 1997
[2]McGrail et al., 2001
[3]Pierce et al., 2004

In addition to the glasses listed in Table 1, additional glasses (100+ ILAW glasses) were analyzed for chemical alteration phases that formed as the glasses were exposed to long term accelerated leaching tests. The information obtained from those glasses was used to determine the representative secondary phase chemical reaction network for ILAW glass and parameters corresponding to those reactions. Phases that can form as part of the secondary phase reaction network are listed in Table 3. The reactions listed and the log K values used for the IDF PA modeling are important parameters as they describe the chemical feedback mechanisms that regulate the rate of glass corrosion/dissolution and thus the rate of contaminant release. The secondary phases typically slow the glass dissolution rate and provide a diffusion barrier on the surface of the glass. Certain phases however, can form rapidly and deplete concentrations of key elements in solution favoring further dissolution of the glass matrix.

Table 3. ILAW Glass Secondary-Phase Reaction Network		
Phase	**Reaction**	**Log K (90°C)***
Analcime ($Na_{0.96}Al_{0.96}Si_{2.04}O_6 \bullet H_2O$)	analcime + 3.84H^+ ↔ 0.96Al^{3+} + 0.96Na^+ + 2.04SiO_2(aq) + 2.92H_2O	3.40
Anatase (TiO_2)	TiO_2 + 2H_2O ↔ $Ti(OH)_4$(aq)	−6.56
Baddeleyite (ZrO_2)	ZrO_2 + 2H^+ ↔ $Zr(OH)_2^{2+}$	−5.20
Calcite ($CaCO_3$)	$CaCO_3$ + H^+ ↔ Ca^{2+} + HCO_3^-	0.91
Chalcedony (SiO_2)	SiO_2 ↔ SiO_2(aq)	−2.65
$Fe(OH)_3$(s)	$Fe(OH)_3$(am) + 3H^+ ↔ Fe^{3+} + 3H_2O	3.04
Gibbsite [$Al(OH)_3$]	$Al(OH)_3$ + 3H^+ ↔ Al^{3+} + 3H_2O	4.46
Sepiolite [$Mg_4Si_6O_{15}(OH)_2 \bullet 6H_2O$]	sepiolite + 8H^+ ↔ 4Mg^{2+} + 6SiO_2(aq) + 11H_2O	39.72
$Zn(OH)_2$-γ	$Zn(OH)_{2\text{-}\gamma}$ + 2H^+ ↔ Zn^{2+} + 2H^2O	11.88

The data presented here will likely be augmented or updated with new data prior to completion of the IDF PA which is currently expected to be issued in 2017. The new data will be aimed at expanding the target glass compositions to include advanced/enhanced waste loading ILAW glasses. In addition, ILAW glass testing is expected to continue through startup of DFLAW operations and beyond in order to make sure that waste treatment operations and glass production are being conducted in an efficient manner and delivering high quality products. Ongoing PA maintenance will be performed to continually evaluate changes that could affect the performance, design, and operating bases for the facility.

CONCLUSIONS

As part of the DFLAW, the ILAW glass to be produced in the WTP is planned to be disposed at the IDF. However, before the IDF can be used as a disposal site, DOE must approve a PA that provides the quantitative demonstration of IDF compliance with the performance objectives for the long-term protection of the public and the environment. One of the critical components of the IDF PA will be to provide a reasonable expectation that releases from the ILAW glass waste form do not result in performance objectives being exceeded. This paper provides a general discussion on the importance of ILAW glass development, testing and data package preparation. The ILAW glass data is relevant for PA analysis, without it, the IDF PA will not be able to produce estimates of the behavior of the glass waste form and take action, if needed, for protection of the public and the environment.

After the 2017 IDF PA is issued, PA support activities will transition to PA maintenance and will continue up to and beyond the startup of DFLAW operations. This will facilitate evaluation and testing of additional ILAW glasses to reduce uncertainty in the PA projections. It will also provide opportunities to evaluate flowsheet or process configuration changes that may improve efficiency while still producing glass that falls within the performance envelope established in the IDF PA.

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