Fluidized Bed Steam Reformed (FBSR) Mineral Waste Forms: Characterization and Durability Testing

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ABSTRACT

Fluidized Bed Steam Reforming (FBSR) is being considered as a potential technology for the immobilization of a wide variety of high sodium low activity wastes (LAW) such as those existing at the Hanford site, at the Idaho National Laboratory (INL), and the Savannah River Site (SRS). The addition of clay, charcoal, and a catalyst as co-reactants with the waste denitrates the aqueous wastes and forms a granular mineral waste form that can subsequently be made into a monolith for disposal if necessary. The waste form produced is a multiphase mineral assemblage of Na-Al-Si (NAS) feldspathoid minerals with cage and ring structures and iron bearing spinel minerals. The mineralization occurs at moderate temperatures between 650-750°C in the presence of superheated steam. The cage and ring structured feldspathoid minerals atomically bond radionuclides like Tc-99 and Cs-137 and anions such as SO_4, I, F, and Cl. The spinel minerals stabilize Resource Conservation and Recovery Act (RCRA) hazardous species such as Cr and Ni. Granular mineral waste forms were made from (1) a basic Hanford Envelope A low-activity waste (LAW) simulant and (2) an acidic INL simulant commonly referred to as sodium-bearing waste (SBW) in pilot scale facilities at the Science Applications International Corporation (SAIC) Science and Technology Applications Research (STAR) facility in Idaho Falls, ID. The FBSR waste forms were characterized and the durability tested via ASTM C1285 (Product Consistency Test), the Environmental Protection Agency (EPA) Toxic Characteristic Leaching Procedure (TCLP), and the Single Pass Flow Through (SPFT) test. The results of the SPFT testing and the activation energies for dissolution are discussed in this study.

INTRODUCTION

Fluidized Bed Steam Reforming (FBSR) is being considered as a potential technology for the immobilization of a wide variety of radioactive wastes. Studsvik built and tested a commercial Low-Level Radioactive Waste (LLRW) FBSR Processing Facility in Erwin, TN, which began commercial operations in July 1999 [1,2]. The Erwin facility employs the THermal Organic Reduction (THOR[sm]) process, developed by Studsvik, which utilizes pyrolysis[‡]/steam reforming technology. The reforming process has demonstrated effectiveness in pyrolyzing organics and separating sulfur and halogens from inorganic waste materials. Of special relevance is the capability of the THOR[sm] technology to convert nitrates to nitrogen and sodium salts to sodium compounds that are suitable for direct disposal and/or subsequent vitrification. If clay is added during processing a "mineralized" granular waste form is produced that is composed of various Na-Al-Si (NAS) feldspathoid minerals with cage and ring structures that stabilize alkali, alkaline earths, sulfate-sulfide species, halogens, and Tc-99.

Applications of the FBSR technology to produce a granular waste form from Hanford high sodium wastes commenced in 2001 when Studsvik demonstrated the technology at Hazen Research in Golden, Colorado [2,3]. Since the FBSR technology is applicable to aqueous high

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[‡] Pyrolysis chemically decomposes organic materials by heat in the absence of oxygen, e.g. C_xH_y + Heat $\rightarrow CH_4$ + C.

sodium organic containing wastes at Hanford, Idaho National Laboratory (INL), and the Savannah River Site (SRS), additional pilot scale tests of this technology were performed during 2003 and 2004 at the Science Applications International Corporation (SAIC) Science and Technology Applications Research (STAR) facility in Idaho Falls, Idaho. The pilot scale tests were performed on both Hanford and INL wastes at the STAR facility by a team of scientists from STAR, INL, and THOR[sm] Treatment Technologies (TTT). The characterization and durability of the mineral waste forms produced at the STAR facility during three pilot scale FBSR demonstrations was evaluated by the Savannah River National Laboratory (SRNL). Measurement of the STAR waste form product durability and a comparison to previous durability testing [4,5] of the Hazen waste form product is the focus of the present study.

The wastes processed at STAR and durability tested in this study were the acidic (pH~2) INL Sodium Bearing Waste (SBW)[6,7] and the Hanford basic (pH~14) Low Activity Waste (LAW) Envelope A [8]. The SBW wastes tested were representative of wastes stored in Tank WM-180 at the Idaho Nuclear Technology and Engineering Center (INTEC). They were ~2M Na^+, and ~1.9M Al^{+3} with a $NaReO_4$ spike (5.4 x 10^{-4}M) to simulate $NaTcO_4$, Cs, Cl and I. The LAW Envelope A waste had total organic carbon ~3.5 g/L, ~5M Na^+, a $NaReO_4$ spike (5.2 x 10^{-4}M) to simulate $NaTcO_4$, Cs, Cl, F, and I [9]. The waste forms tested included the granular mineral material produced in the fluidized bed and the finer mineral material from the filter, hereafter referred to as the filter fines (Table I). This study will summarize the Hanford LAW durability tests.

The durability of the bed and fines products from the STAR LAW Envelope A demonstration produced in this study were compared to the durability of the bed (SCT-02) and fines (PR-01) products from a Hanford Envelope C waste (AN-107) demonstration performed at Hazen Research in Golden, Colorado [4]. The Hanford Envelope C waste tested in 2001 had total organic carbon ~3.45 g/L, ~7.3M Na^+, a $NaReO_4$ spike (3.22 x 10^{-5}M) to simulate $NaTcO_4$, Cs, Cl, and F but no I.

All the pilot scale materials listed in Table I were tested with ASTM C1285 (Product Consistency Test) [4,10,11]. The Fluidized Bed Steam Reformer (FBSR) products (bed and fines) were determined to be ~2 orders of magnitude more durable than the LAW glass specification of NL_{Na}=2 g/m^2 when the PCT durability is expressed in the units given in the specification, e.g. when the BET surface area is used in the calculation instead of the geometric surface area as suggested by McGrail, et. al. [5]. When the geometric surface area is used and/or the leach rate is expressed in g/L (NC_i) which is not dependent on the surface area of the waste form being tested, then the leach rate of the FBSR products is comparable to LAW glass. The determination that FBSR mineral waste forms are ~2 orders of magnitude more durable than LAW glass is consistent with the findings of McGrail [5] during SPFT testing and preliminary Performance Assessment (PA) modeling [12] of these waste forms.

The PCT durability of the mineral waste forms listed in Table I were assessed against the mineral phases produced as identified by X-ray Diffraction (XRD). The PCT response for the SBW and LAW samples was determined to be a function of the aluminosilicate minerals formed and an aluminosilicate buffering mechanism that controls the leachate pH [10,11]:

- Cs and Na release are co-linear with Al release
- S, Re, and Si release are strong linear functions of the buffered leachate pH

Table I. Pilot Scale LAW Streams Processed by FBSR Between 2001-2004

Demonstration	Sample ID	Total Operating Time (TOT)	Bed Turnover (%)	Description
August 2004 LAW (Envelope A)[8]	Bed 1103	55 hrs and 30 min	97.4	Dynamic bed product
	Bed 1104	55 hrs and 30 min	99.7	Final bed product
	Fines 1125	55 hrs and 30 min	100	Final filter fines
December 2001 AN-107 LAW (Envelope C)[2,3]	SCT-02	4.8	Not reported	Dynamic bed product
	PR-01	23.3	Not reported	Dynamic filter fines

Environmental Protection Agency (EPA) Toxic Characteristic Leaching Procedure (TCLP) tests were also conducted on the LAW and SBW bed products given in Table I. Details are given elsewhere [10,11]. The TCLP testing indicated that the bed products and fines pass TCLP for chromium release at the Universal Treatment Standard (UTS) limits when the iron catalyst for nitrate destruction is used because the iron catalyst forms the magnetite spinel host phase that sequesters the chromium as Cr^{+3} into its insoluble structure.

The LAW Envelope A and 2004 SBW bed products were also tested with the Single Pass Flow Through (SPFT) test [13]. The SPFT durability of the LAW Envelope A bed product and fines from the STAR demonstration was compared to the SPFT durability of the bed product from the Hanford AN-107 (LAW Envelope C) demonstration in 2001 measured by McGrail [5]. Details of the SPFT testing and comparisons of the durability as measured at SRNL and Pacific Northwest National Laboratory (PNL) are the focus of this study.

EXPERIMENT

Excess carbon co-reactant was removed from the mineral product prior to chemical and durability analysis in order to express the composition and durability on a mineralized product basis, e.g. charcoal free. Elemental and anion compositions of the steam reforming materials were measured and reported elsewhere [10,11,13]. Removing the charcoal is considered conservative since the charcoal increases the measured BET surface area which would have decreased the apparent SPFT durability expressed in g/m^2•day[†].

The charcoal was removed by heating the samples to 525°C overnight as specified in a United States Geological Survey (USGS) procedure [14] for carbon removal in preparation for analysis. Samples were examined by XRD to verify that the phase assemblages had not changed after heating to remove the coal.

The FBSR bed products were sized using an electric grinder and a ceramic mortar and pestle, and sieved to a -100, +200 mesh size (75-150μm) which is the same size used by McGrail, et. al. [5]. The sized product was then washed for particle size control in accordance with procedure ASTM C1285. The sized product was washed six times with absolute ethanol in

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† $NR_i = \dfrac{c_i(sample) - c_i^0(blank)}{f_i \bullet (SA_{BET}/F)}$ where NR_i is the normalized release (g/m^2•day) and *SA/F* is the BET surface area of the final waste form in m^2 divided by the flow rate, F, in L/day

a sonication bath. Water was not used for washing so as to prevent the removal of any water-soluble phases prior to leach testing. After washing, the materials were placed in an oven at 90°C for drying overnight prior to being used for SPFT testing and BET surface area meansurement.

The SPFT testing was performed at 5 different pH values and 4 different temperatures for 3-14 day test durations [13]. Steady state conditions were achieved between 8-14 day test durations at a high flow rate (288 mL/day). For comparison McGrail et al. [5] tested various flow rates up to a maximum flow rate of 200 mL/day at the same 5 pH values but at only one temperature, 90°C, for up to 23 days. The 288 mL/day flow rate was chosen for this study because of the expected decline in release rate with decreasing flow rate and to ensure there would be no feedback from the leachant solutions. Sampling began on the third day of each test in order to allow the system to reach steady-state conditions, and then continued every other day until the test completion on the 14th day. Once collected, each sample was analyzed for the concentrations of Si, Al, Na, and S via ICP-ES, and for Re via ICP-MS. The fate of S dissolved from the waste form is important because S limits waste loading in glass waste forms but not in the FBSR mineral product. The fate of Re dissolved from the waste form is important as it is a surrogate for Tc-99.

DISCUSSION

When the release rates expressed as NR_i are plotted as a function of test duration given by Lorier, et. al. [13] it is observed that steady state conditions were achieved between 8-14 days at the high flow rates used. The steady state 8-14 day NR_i values for each element from replicate tests were averaged in this study and plotted against pH in Figure 1: each data point represents six replicate measurements used to determine the steady state release rates of Si, Al, Na, Re (simulant for Tc-99) and S in a manner analogous to that of Knauss et. al. [15] for borosilicate waste glasses.

Most mineral or ceramic waste forms leach incongruently because the multiple phases are "mixed oxides", e.g. zirconates, titanates, ferrates, aluminates, etc. [16].[‡] However, the FBSR mineral phases are all aluminosilicates of the feldspathoid family (nepheline, sodalite, and nosean) and they all have an $Na[AlSiO_4]$ configuration. The nosean and sodalite minerals have cage like structures that accommodate $1Na_2SO_4$ [17], $2NaCl$ [18], $2NaF$ [18], $2NaI$ [18], $1Na_2MoO_4$ [19], $1Na_2ReO_4$[20].

Figure 1 shows that during SPFT testing of the LAW bed product that Re, Al and S dissolution is parabolic with solution pH, while Na and Si release are almost linear. In general at temperatures >25°C, the release sequence is Re>S~Na>Si>Al. This data suggests that the aluminum, rhenium, and sulfur dissolution are related, perhaps from dissolution of the nosean/sodalite phase while the dissolution of silicon and sodium appear independent of which felspathoid $Na[AlSiO_4]$ configuration is dissolving so incongruent leaching effects may be minimized. Indeed, a plot of the Re release vs S release at all temperatures, e.g. 25°, 40°, 70°, and 90°C, indicate that these two species leach congruently and that the Al and S release is congruent indicating that the primary mineral phase contributing to the Re, S, and Al release may

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[‡] Incongruent dissolution of a waste form means that some of the dissolving species are released preferentially compared to others. Congruent dissolution of a waste form, like glass, is the dissolving of species in their stoichiomentric amounts. For congruent dissolution, the rate of release of a radionculide from the waste form is proportional to both the dissolution rate of the waste form and the relative abundance of the radionculide in the waste form. Thus for borosilicate glass ^{99}Tc is released at the same rate, congruently, as Na, Li and B.

be the nosean/sodalite phase. The Na and Si leach congruently at all temperatures but the Al and Si and the Al and Na leach incongruently.

The steady state release rates calculated in this study at flow rates of 288 mL/day are consistently lower than those reported by McGrail et.al. [5] for the 2002 LAW FBSR product at flow rates of 200 mL/day. McGrail et al. state that at flow rates ≥140 mL/day, that the release rates become independent of the flow rate and should represent a true forward rate of reaction which indicates that the differences in the release rates cannot be attributed to the differences in the flow rates. Other test parameters that varied between the McGrail et.al. [5] study and the test parameters in this study were (1) McGrail et.al. manually removed the carbon which Pariezs et al [10] determined leaves ~4 wt% carbon in the product which alters the measured BET surface area and (2) McGrail et.al. N_2 purged the leachant reservoirs which was not done in the current study. Although colloidal species are not anticipated at these high flow rates, the leachates in this study were filtered (0.45μm) while the leachates in the McGrail et al. [5] study were not.
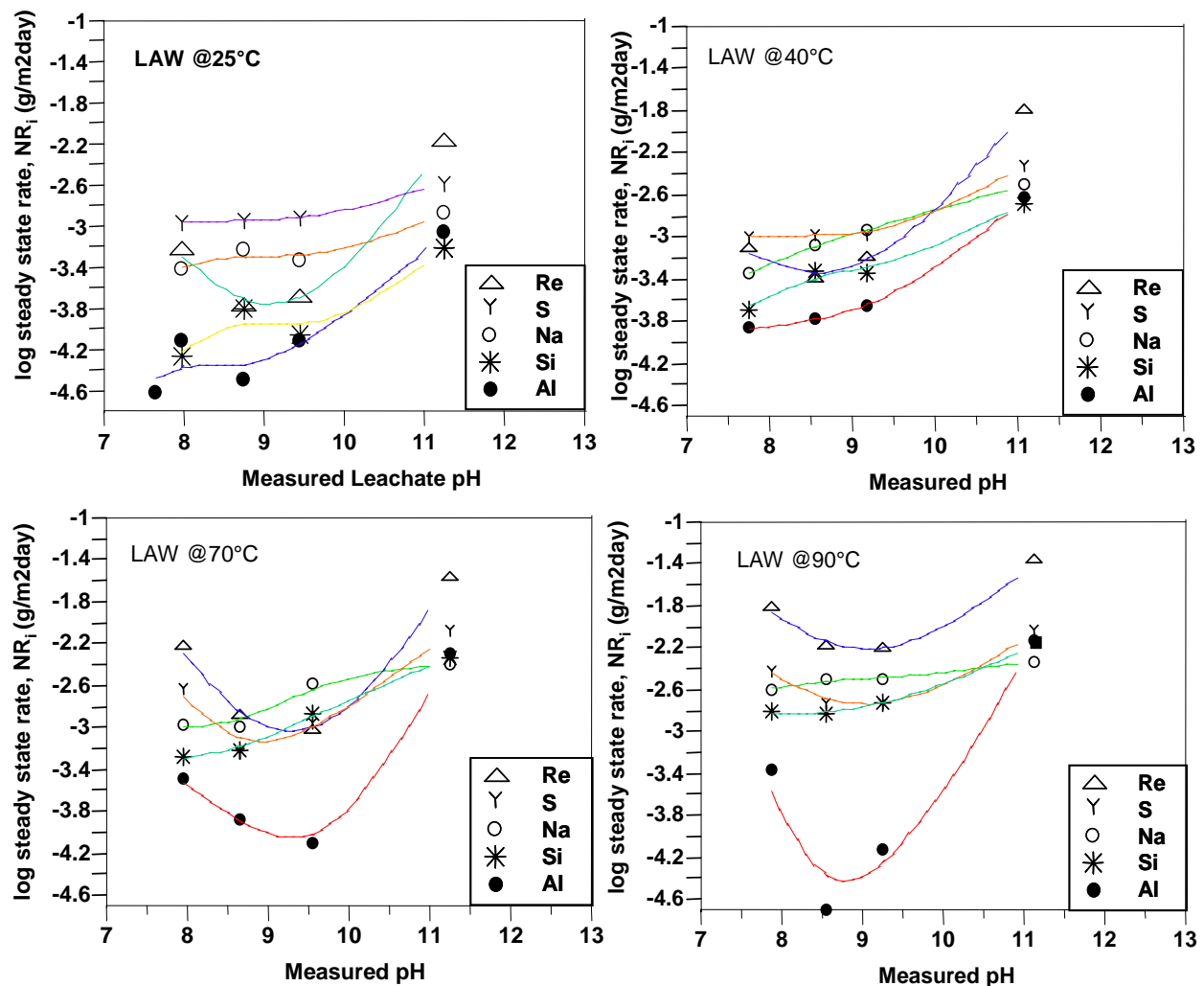

Figure 1. Dependence of steady state (8-14 day) normalized alkali (Na), Al, Re, S and Si on solution pH.

At the higher flow rates used in this study, Al dissolution is always lower than that of Si and Na and the lowest Al dissolution occurs at pH values between 8.5 and 9.5 in the 70-90°C temperature range (Figure 1). This pH is the pH of the aqueous equilibrium boundary between amorphous $Al(OH)_3$ gel (which may be colloidal) and the aqueous species $Al(OH)_4^-$ [19]. This may be explained by the fact that the FBSR products, those tested by McGrail et. al. [5] and in this study, have unreacted metakaolinite cores that are amorphous to X-ray Diffraction (XRD) and so, at a minimum, the leaching results represent a mixture of two different types of nepheline, nosean, and amorphous metakaolin (Table 2). Moreover, the alumina species in solution can participate in the early formation of amorphous colloids as noted by Tole [21]. Since the leachates in this study were filtered and the leachates in the McGrail et.al [5] study were not, this could be a major source of the differences observed in the leach rates and the data interpretation.

Table 2. Phases Identified by XRD and SEM

FBSR PRODUCTS	XRD	SEM
LAW Envelope C (AN-107) [3,4,5]	$NaAlSiO_4$ (nepheline) $Na_{1.53}Al_{0.92}Si_{0.92}O_4$ (Na-rich nepheline) $Na_6[Al_6Si_6O_{24}](Na_2SO_4)$ (nosean)	metakaolinite
LAW Envelope A [10,11]	$NaAlSiO_4$ (carnegieite) $Na_{0.89}Al_{0.9}Si_{1.1}O_4$ (Si-rich nepheline) $Na_6[Al_6Si_6O_{24}](Na_2SO_4)$ (nosean)	metakaolinite

A major assumption was made by McGrail et. al. during their SPFT data interpretation [5], e.g. S and Re dissolution represent the dissolution of the sodalite/nosean phases while the dissolution of Na, Si, and Al represent the dissolution of the nepheline component of the FBSR products. By subtracting the dissolution rate of the nosean (based on S or Re) from the dissolution rates of Na, Al, and Si and recalculating a residual dissolution rate for Na,Al, and Si dissolution at flow rates between 60 and 100 mL/day, McGrail, et. al. calculated a pH dependence of the reaction order (η) for nepheline dissolution of 0.25 at 90°C (Table 3). Using the same assumption in this study, Figure 2a for the dissolution of nosean and nepheline at 90°C can be generated and the nepheline dissolution rate calculated in this study was -4.66 with $\eta=0.22$ for Na, Al, and Si. Thus, based on this assumption, the reaction order in pH for the LAW Envelope A FBSR product ($\eta=0.22$) tested in this study is in agreement with the value of $\eta=0.25$ determined by McGrail et al. [5] using this methodology for the LAW Envelope C FBSR product (Table 3).

The assumption made by McGrail et. al. [5] assumes a linear response of the leach rate for all the elements with pH. The data generated in this study, at flow rates higher than those used by McGrail et. al [5], clearly demonstrates a parabolic dependence of the Re, Al and S releases with pH while Na and Si are approximately linear (Figure 2b). This data indicates that the leach rates of Re, S, and Al are similar (congruent with each other) while the leach rates of Na and Si congruent with each other but different but not congruent with Re, S, and Al. This indicates that the leaching of the $Na[AlSiO_4]$ framework structure of the nepheline and nosean structures may similar while the leach rate of alumina is highly variable.

Following the alternate assumption that the S and Re are in the nosean phase and represent the dissolution of this phase and that the nepheline like structure of the nosean phase

Na[AlSiO$_4$] leaches in the same manner as the Na[AlSiO$_4$] structural units in the nepheline and carnegieite phases, then a nosean dissolution rate of -3.20 g/m^{2}d^{-1} and a pH dependence of the rate (η) reaction order of 0.16 is calculated based on Re release and a rate of -3.45 and η=0.13 pH based on S release (Table 3) is calculated. The dissolution rates for nepheline/carnegieite Na[AlSiO$_4$] structures at various temperatures as measured in this study is given in Table 3.

When the reaction order for nepheline dissolution determined for the LAW nepheline components in this study and in the McGrail et al [5] study are compared to that measured by Tole [21] on single crystal natural nepheline (Table 3) it can be concluded that the nepheline reaction orders measured in this study for Na[AlSiO$_4$] structural units, irregardless of whether they are in nepheline, carnegieite, or nosean, are in agreement with the data of Tole (η=0.13) at comparable temperatures (80-90°C) than the data of McGrail et. al. [5]. In addition, the reaction order measured for nosean dissolution (based on S and Re) in this study are also comparable to the values measured by Tole for phase pure nepheline. This is not surprising as the nosean cage structures are formed from alternating silica and alumina tetrahedra of the Na[AlSiO$_4$] structural units. All the reaction orders measured on LAW nepheline FBSR product in this study and in the McGrail study (Table 3) are lower than the reaction order measured by Hamilton, et.al. [22] on nepheline glass. It should be noted that the reaction order given by Hamilton is based on Si release and is comparable to η=0.40 measured on a simple 5 component High Level Waste glass by Knauss, et. al. at 70°C [15].

Table 3. Reaction Order in pH for Nosean, Crystalline Nepheline, and Nepheline Glass

pH Range 7-11	Nosean/Sodalite	Nepheline			
	90°C	25°C	70°C	80°C	90°C
LAW Envelope A	0.13 (S) 0.16 (Re)	0.23*	0.17*	N.D.	0.13*
LAW Envelope C [5]	N.D.	N.D.	N.D.	N.D.	0.25
Natural Nepheline [21]	N.D.	0.2	N.D.	0.13	N.D.
Nepheline Glass [22]	N.D.	N.D.	N.D.	0.43	N.D.

* Based on Si release

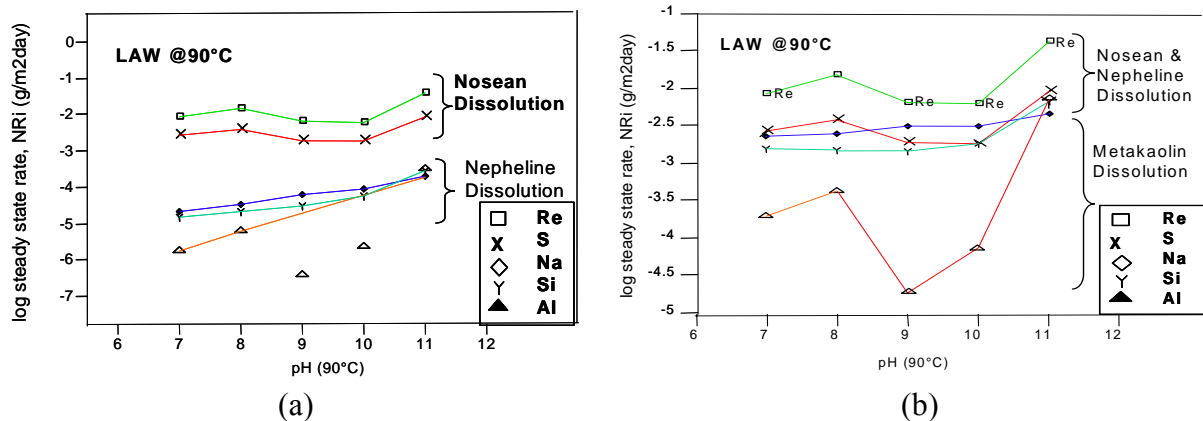(a)   (b)

Figure 2. Interpretation of dissolution in the Hanford LAW FBSR mineral waste forms determined from SPFT testing at high flow rates.

Table 4. Activation Energies for Nepheline and Nosean Dissolution (25-90°C)

pH Value	Si Dissolution Activation Energy (KJ/mole)	Re Dissolution Activation Energy (KJ/mole)	S Dissolution Activation Energy (KJ/mole)	Al Dissolution Activation Energy (KJ/mole)	Na Dissolution Activation Energy (KJ/mole)
7	-44.26	-35.05	-16.12	-30.18	-17.02
8	-43.45	-47.59	-18.83	-24.38	-26.02
9	-26.86	-48.06	-7.62	-23.75	-20.31
10	-40.58	-41.32	-7.21	N.D.	-26.07
11	-31.88	-23.83	-17.03	-28.91	-15.57

Because it is difficult to interpret SPFT test results on a multiphase mineral assemblage such as the FBSR product, and because data in this study was generated at four different leaching temperatures, activation energies for individual elements at varying SPFT buffer solution pH values were tabulated (Table 4). This could not be done in the McGrail et. al. study because only one temperature, 90°C, was studied. The activation energies were calculated from the raw data after conversion to release rates in moles/m^{2}sec. Activation energies were only reported if the fit of the 1/T regressions was in the range of 0.85-0.99.

CONCLUSIONS

Single Pass Flow Through Testing (SPFT) of a granular nepheline-nosean bearing feldspathoid waste forms produced from Hanford Envelope LAW wastes have been completed. Reaction orders and activation energies of dissolution have been defined that will be useful during Performance Assessment (PA) evaluation of this new waste form.

ACKNOWLEDGMENTS

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