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CHARACTERIZATION AND PERFORMANCE OF FLUIDIZED BED STEAM REFORMING (FBSR) PRODUCT AS A FINAL WASTE FORM (U)

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CHARACTERIZATION AND PERFORMANCE OF FLUIDIZED BED STEAM REFORMING (FBSR) PRODUCT AS A FINAL WASTE FORM

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ABSTRACT

A demonstration of Fluidized Bed Steam Reforming (FBSR) was recently completed on a Hanford Low Activity Waste (LAW) simulant. This technology produced stable mineral phases (feldspathoids) when co-fired with clay. The mineral phases are cage structured and were determined to retain anions such as $SO_4^{=}$ as well as cations such as Re (simulant for Tc) in the mineral cages. The mineral phases are produced at moderate steam reformer operating temperatures between 650-750°C. The FBSR mineral waste form exhibited incongruent leaching characteristics during Product Consistency Testing (PCT or ASTM C1285). The radionuclides (Cs and Re as simulants for Cs^{137} and Tc^{99}) are released in significantly lower concentrations than Na. In addition, the Na release is less than the 2 g/m^2 Hanford contract requirement for vitrified LAW. FBSR mineral waste forms are EPA regulatory compliant at the Universal Treatment Standard (UTS) making delisting an attractive option for this waste form.

INTRODUCTION

The Hanford LAW is a basic high Na^+ molarity (~8.1M) sodium nitrate – sodium hydroxide solution that also contains significant amounts of sulfate, chloride, fluoride and organic compounds as well as certain heavy metals and radionuclides. LAW is the low activity salt supernate fraction of Hanford High Level Liquid Waste (HLLW). In December 2001, Fluiduized Bed Steam Reforming (FBSR), was investigated for stabilization of LAW waste by THOR Treatment Technologies (TTT) using the patented THermal Organic Reduction ($THOR^{sm}$) process. This process, developed by Studsvik, utilizes pyrolysis[*]/steam reforming to destroy both organics and nitrates/nitrites in the waste. The FBSR demonstrations were performed on ~150 gallons of a Hanford AN-107 LAW simulant simultaneously being used for a vitrification demonstration. The radionuclide Tc^{99} was simulated with Re while Cs^{137} was simulated with stable cesium (Cs^{133}).

The non-radioactive AN-107 simulant of 8.1M Na^+ was successfully tested in a 6-inch TTT pilot scale facility.[†] Other demonstrations performed by TTT showed that LAW waste could be transformed into Na_2CO_3, $NaAlO_2$, or Na_2SiO_3 feed material for the LAW Hanford melter (Table I). Addition of no solid co-reactant yielded a sodium carbonate product. Sodium will combine with carbon dioxide in the reformer gases to provide a sodium carbonate product. The generation of sodium carbonate in this type of application has been known since the 1950s in fluid bed denitration systems [1]. Addition of an $Al(OH)_3$ co-reactant will provide an $NaAlO_2$ product, addition of SiO_2 will provide an Na_2SiO_3 product, while addition of clay will provide a final mineral waste form product (Table I). The latter has been shown to perform well as a final waste form [2,3]. Testing on Hanford LAW surrogates has shown that over 95% of the sulfur compounds, fluorides and chlorides in the waste feed react in the steam reformer with the clay co-reactant and become an integral part of the final mineral waste product structure [2].

Table I. THOR^{sm} Pilot Scale Demonstrations with Simulated LAW Wastes

# Pilot Runs	Solid Additive	Mineral Product	Purpose
5	Clay	Feldspathoid minerals (nepheline and sodalite) that stabilize problematic anions such as Cl, F, and SO_4	Stabilization of LAW or salt supernates as a final mineral waste form
3	Sand	Sodium silicate	Pretreatment of LAW for vitrification and/or recycle of melter off-gas blowdown.
2	$Al(OH)_3$	Sodium aluminate	Pretreatment of LAW for vitrification and/or recycle of melter off-gas blowdown.
3	None	Sodium carbonate	Pretreatment of LAW for vitrification and/or recycle of melter off-gas blowdown.

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[*] Pyrolysis chemically decomposes organic materials by heat in the absence of oxygen

[†] The solution was diluted to 5.2M Na^+ to homogenize the feed before processing due to the observation of precipitated solids in the feed tank

EXPERIMENTAL

Scoping FBSR tests were performed by TTT between December 6, 2001 and December 20, 2001 using the Studsvik THOR^{sm} process. The FBSR waste forms were made Hanford AN-107 LAW simulant. The results of the final waste form testing performed by TTT under reducing FBSR conditions will be summarized in this report (Scoping Tests 1 and 2 and Production Run 2). Additional testing performed under oxidizing FBSR conditions will be discussed comparatively since there was only one set of tests run under these conditions.

Sample SCT02-098-FM was dissolved using a lithium borate fusion and a second dissolution performed via an Na_2O_2 fusion (ASTM C1463). Each dissolution was performed twice on two different days. Each dissolution was analyzed twice, once with no dilution and once with a 10X dilution. All four replicate analyses were averaged. A glass standard, Batch 1 glass, was analyzed simultaneously for quality assurance. The sample was analyzed for anions (phosphorous and sulfur) and cations by both Inductively Coupled Plasma Emission Spectroscopy (ICP-ES) and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS) (ASTM C1463). In addition, the sample was dissolved in a Na_2O_2 dissolution with a water uptake and analyzed for additional anions by Ion Chromotography (IC) (ASTM D4327). The PCT leachates were analyzed for cations by both Inductively Coupled Plasma Emission Spectroscopy (ICP-ES) (ASTM C1109) and Inductively Coupled Plasma Mass Spectroscopy (ICP-MS).

Dissolution of sample SCT02-098-FM was also performed by H_2SO_4/HF in the presence of NH_4VO_3 followed by colorimetric determination of Fe^{2+} and total iron ($\sum$Fe) in order to determine the <u>RED</u>uction/<u>OX</u>idation (REDOX) equilibria of the sample in terms of the $Fe^{2+}/\sum Fe$ ratio [4]. A standard glass, the EA glass [5], with a known and reproducible REDOX, was used during the analysis for quality assurance.

X-Ray Diffraction (XRD) analyses were performed by TTT and confirmed at SRTC for sample SCT02-098-FM. XRD was performed at SRTC both before and after durability testing.

The waste form durability and chemical analyses specific to Scoping Test 02 when the FBSR was operated under reducing conditions will be discussed in detail. Coal was used to assist in denitration of the waste at temperatures between 715-735°C. Clay, small amounts of excess SiO_2, and iron oxide were the only waste form additives. The waste loading achieved with Scoping Test 02 was ~27 wt% since 73 wt% additives were used. This corresponds to a Na_2O loading of ~20 wt% in the mineral waste form.

Durability testing of the FBSR sample SCT02-098-FM from Scoping Test 02 was performed at SRTC. Durability testing was performed using ASTM C1285-97 (PCT-A test protocol). The PCT-A test was run for 7 days at 90°C in stainless steel vessels. Triplicate samples were tested along with two standard glasses; the ARM-1 standard and the Environmental Assessment (EA) glass [5] standard used to assess the durability of HLW vitrified waste forms. Testing of the FBSR final waste form using the EPA TCLP protocol was performed by Evergreen

Analytical, an EPA certified laboratory, under subcontract to TTT. The results of the testing of samples from the FBSR process fabricated under both reducing and oxidizing conditions are summarized in this report.

RESULTS AND DISCUSSION

Chemical Analysis of the FBSR Mineral Product

A complete chemical analysis of FBSR Scoping Test 02 Sample SCT02-098-FM was performed. The average of the replicate analyses is given in Table II. The replicate REDOX analyses are given in Table III. The average REDOX of the two replicate analyses in Table III were used to calculate the relative proportions of FeO and Fe_2O_3 in the FBSR mineral waste form as given in Table II.

The REDOX measurements in Table III indicate that the mineral phases are not overly reduced at a $Fe^{+2}/\Sigma Fe$ of 0.15 even in the presence of coal added for denitration of the feed. REDOX ratios of 0.15 are too oxidizing for any metallic iron to be present in the FBSR product.

The data in Table II indicate that Cs, Re, SO_4, Cr, and Pb are retained in the FBSR mineral phases and do not volatilize during processing. This was confirmed by the TTT analyses of the off-gas during processing

Based on the analysis provided in Table II, a waste loading of 27 wt% was calculated by assuming that all of the SiO_2, all of the Al_2O_3 and all of the Fe_2O_3 and FeO are waste form additives.

Table II. Chemical Analysis of FBSR Sample SCT02-098-FM

Analytic Method	Oxide	Wt% 5/31/02	Wt% 8/15/02	Analytic Method	Oxide	Wt% 5/31/02	Wt% 8/15/02
ICP-ES	Al_2O_3	31.7436	38.924	ICP-ES	P_2O_5	0.2176	0.2474
ICP-ES	B_2O_3	0.2576	BDL	ICP-ES	PbO	0.0248	0.0199
ICP-ES	CaO	0.7332	0.5681	ICP-MS	PbO	0.0175	NA
ICP-ES	Cr_2O_3	0.0716	0.0278	ICP-MS	ReO_2	0.0005	0.0006
ICP-MS	Cs_2O	0.0029	0.005	ICP-ES	SiO_2	34.8706	30.0572
ICP-ES	Fe_2O_3	5.4471	6.23	ICP-ES	SO_4	1.1175	NA
ICP-ES	FeO	0.8749	1.001	IC	SO_4	NA	2.6335
ICP-ES	K_2O	0.6975	0.6794	IC	Cl	NA	0.318
ICP-ES	La_2O_3	0.0117	NA	IC	NO_2	<0.0005	NA
ICP-ES	Na_2O	19.8156	16.7826	IC	NO_3	<0.0005	NA
ICP-ES	NiO	0.0814	0.0350		SUM	95.9681	97.525

BDL = Below Detection Limit, NA = Not Analyzed

Table III. Replicate Redox Analyses of FBSR Sample SCT02-098-FM

Analysis	EA Standard	AN107-A	AN107-B
Fe^{+2}	0.088	0.058	0.056
$\Sigma Fe_{(total)}$	0.458	0.377	0.376
$Fe^{+2}/\Sigma Fe$	0.192	0.154	0.149

X-Ray Diffraction Analysis of the FBSR Mineral Product

The phases identified by TTT in the FBSR Sample from Scoping Test 02 (Sample SCT02-098-FM) are given in Table IV. The phases identified for the same sample at SRTC are given for comparison in Table IV. Analysis at SRTC indicated the presence of a minor second iron oxide phase, magnetite (Fe_3O_4) in addition to the hematite (Fe_2O_3). Nepheline (the hexagonal type) is the major component with subordinate amounts of nosean and corundum. A cubic structured nepheline was not observed in this sample but was observed in the production run of a similar material. The relative amounts of the two types of nepheline and sodalite (nosean) will vary with optimization of waste additives, e.g. types of clay or other aluminosilicates, and processing parameters.

Table IV Phases Identified in FBSR Sample SCT02-098-FM

Mineral Phases Identified by TTT	Mineral Phases Identified by SRTC before PCT-A Testing	Mineral Phases Identified by SRTC after PCT-A Testing
$Na_8Al_6Si_6O_{24}(SO_4)$ (Nosean)	$Na_8Al_6Si_6O_{24}(SO_4)$ (Nosean)	$Na_8Al_6Si_6O_{24}(SO_4)$ (Nosean)
$NaAlSiO_4$ (Nepheline)	$NaAlSiO_4$ (Nepheline)	$NaAlSiO_4$ (Nepheline)
Al_2O_3 (Corundum)	Al_2O_3 (Corundum)	Al_2O_3 (Corundum)
Fe_2O_3 (Hematite)	Fe_2O_3 (Hematite)	Fe_2O_3 (Hematite)
	Fe_3O_4 (Magnetite)	Fe_3O_4 (Magnetite)

The sodium aluminosilicate (NAS) mineral phase assemblage(s) given in Table IV are anhydrous feldspathoid phases such as sodalite. The sodalite family of minerals (including nosean) are unique because they have cage-like structures formed of aluminosilicate tetrahedra. The remaining feldspathoid minerals, such as nepheline, have a silica "stuffed derivative" ring type structure. The cage structures are typical of sodalite and/or nosean phases where the cavities in the cage structure retain anions and/or radionuclides which are ionically bonded to the aluminosilicate tetrahdra and to sodium. The cage structured feldspathoid system of minerals has the basic structural framework formula $Na_6[Al_6Si_6O_{24}]$. The square brackets in the formula are used to delineate the alumina:silica ratio of the aluminosilicate cage structure which is 1:1.

The feldspathoid mineral, sodalite has the formula $Na_8[Al_6Si_6O_{24}](Cl_2)$. The cage is occupied by two sodium and two chlorine ions in natural sodalites [6]. The formula can also be written as $Na_6[Al_6Si_6O_{24}]\bullet(2NaCl)$ to indicate that two NaCl are ionically bonded in the cavities of the cage structure while the remaining Na:Si:Al have a 1:1:1 stoichiometry [6]. When the 2NaCl are replaced by Na_2SO_4, the mineral phase is known as nosean , $(Na_6[Al_6Si_6O_{24}](Na_2SO_4))$ which is one of the feldspathoid cage structured minerals found in the FBSR waste form. Since the Cl^-, $SO_4^=$, and/or S_2 are atomically bonded inside the sodalite cage structure, these species do not readily leach out of the respective FBSR waste form mineral phases.

Other minerals in the sodalite family, namely hauyne and lazurite which are also cage structured minerals, can accommodate either SO_4 or S_2 depending on the REDOX of the sulfur during the steam reforming process. Regardless of the FBSR REDOX the feldspathoid minerals can accommodate sulfur as either sulfate or sulfide. Sodalite minerals are known to accommodate Be in place of Al and S_2 in the cage structure along with Fe, Mn, and Zn, e.g. helvite $(Mn_4[Be_3Si_3O_{12}]S)$, danalite $(Fe_4[Be_3Si_3O_{12}]S)$, and genthelvite $(Zn_4[Be_3Si_3O_{12}]S)$ [6]. These cage-structured sodalites were minor phases in High Level Waste (HLW) supercalcine waste forms[*] and were found to retain Cs, Sr, and Mo into the cage-like structure, e.g. Mo as $(Na_6Al_6Si_6O_{24})$ $(NaMoO_4)_2$ [7]. In addition, sodalite structures are known to retain B [8], Ge[9], I[9,6], and Br[9,6] in the cage like structures. Indeed, waste stabilization at Argonne National Laboratory-West (ANL-W) currently uses a glass-bonded sodalite ceramic waste form (CWF) for disposal of electrorefiner wastes for sodium-bonded metallic spent nuclear fuel from the EBR II fast breeder reactor [10,11].

A second feldspathoid mineral found in the FBSR waste form is nepheline $(NaAlSiO_4)$ [12]. Nepheline is a hexagonal structured feldspathoid mineral. The ring structured aluminosilicate framework of nepheline forms cavities within the framework. There are eight large (nine-fold oxygen) coordination sites and six smaller (8-fold oxygen) coordination sites [6]. The larger sites nine-fold sites can hold large cations such as Cs, K, and Ca while the smaller sites accommodate the Na. The K analogue is known as leucite $(KAlSi_2O_6)$. In nature, the nepheline structure is known to accommodate Fe, Ti and Mg as well.

The remaining aluminosilicate mineral found in the FBSR waste form is a sodium rich cubic structured nepheline derivative $(Na_2O)_{0.33}Na[AlSiO_4]$ (PDF#39-0101). This nepheline derivative structure has large (twelve-fold oxygen) cage like voids in the structure [13]. This cage structured nepheline is not known to occur in nature but the large cage like voids should be capable of retaining large radionuclides, especially monovalent radionuclides such as Cs.

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[*] Supercalcines were the high temperature silicate based “natural mineral” assemblages proposed for HLW waste stabilization in the United States (1973-1985).

Durability Testing of the FBSR Mineral Product

The PCT-A was performed in triplicate on sample SCT02-098-FM in conjunction with glass durability standards, e.g. the ARM-1 and EA glasses. Stainless steel vessels (304L) were used as specified in the PCT-A leaching protocol for the first set of tests. These tests were repeated in Teflon® vessels to demonstrate that Re release was independent of vessel type. The logarithm of the NL_i was taken for each replicate and then averaged per ASTM C1285-02.

The PCT responses measured in this study for Sample SCT02-098-FM and the standard glasses tested are summarized in Table V. It is evident that the leach testing was in control as the response for the EA glass is within the allowable standard deviations of the reference response shown in Table V.

The leaching of Sample SCTO2-098-FM demonstrates that the normalized Na release is 1.74 g/m^2 within the 2 g/m^2 Hanford specification. In addition, it is obvious that the AN-107 FBSR waste form leaches incongruently instead of congruently* as most vitrified waste forms. Incongruent dissolution of a waste form means that some of the dissolving species are released preferentially to others. Incongruent dissolution is diffusion-controlled and can be either surface reaction-limited under conditions of near saturation or mass transport-controlled. Preferential phase dissolution, ion-exchange reactions, grain-boundary dissolution, and dissolution-reaction product formation (surface crystallization and recrystallization) are among the more likely mechanism of incongruent dissolution, which will prevail, in a complex polyphase ceramic waste form [14].

Incongruent dissolution is only detrimental to a waste form if a radionculide species is released preferentially to a matrix element. In the FBSR final waste form the radionuclide release (Cs and Re) is retarded preferentially to the matrix element, Na, release (Table V) or conversely, Na is released from one of the phases preferentially compared to the nosean phase which retains the Re. This finding is noteworthy because the Hanford specification for Na release for vitrified waste forms is an indicator for the congruent release of Tc^{99} since Na and B and Tc^{99} are all released at similar stoichiometric rates (congruently) from vitrified waste forms [10, 11,15,16,17,18,19,20,21,22,23,24].

The incongruent release of Cs and Re is not attributed to the use of stainless steel vessels as numerous studies have shown that neither Cs nor Tc^{99} have an affinity for stainless steel vessels [19,15]. In addition, the PCT triplicate analyses were rerun in Teflon® vessels and the Re release indicated in Table V is comparable to the Re release measured in this study in the stainless steel vessels. The solids remaining after PCT testing were analyzed by X-ray Diffraction and all of the same phases were present as before PCT testing (see Table IV).

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* Congruent dissolution of a waste form 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 Tc^{99} is released at the same rate, congruently, as Na^+, Li^+ and B.

Table V PCT Performance of Sample SCT02-098-FM and the Glass Standards Tested Compared to the Durability Response of Known Standards

Sample	pH	$NL_{(B)}$ g/m^2	$NL_{(Na)}$ g/m^2	$NL_{(Cs)}$ g/m^2	$NL_{(Re)}$ g/m^2	$NL_{(Si)}$ g/m^2
EA	11.64	7.76	6.05			2.21
ARM-1	10.47	0.29	0.27			0.15
AN-107	11.95	1.27	1.74	0.16	0.29 (0.22)*	0.35
AN-107*	11.98	BDL	*f*	*f*	0.22	0.48
EA REF	11.85	8.37	6.67			1.96
LAW REF	10.90	0.55	0.54			0.16
AN-102 RAD	10.60	0.29	0.35			0.12

* = rerun in Teflon vessels, *f* = analysis indicated that vessel blanks were contaminated with Na and Cs

Regulatory Testing of the FBSR Mineral Waste Form

The mineral phases formed during the FBSR process were subject to the EPA TCLP leaching protocol. All of the mineral phases, regardless of particle size, appear to have met the LDR Universal Treatment Standards (UTS) as shown in Table VI.

Delisting the final LAW waste form may also be accomplished by delisting the final waste form at the point of generation so that the UTS are not applicable. The EPA calculates delisting levels and risk levels for a given waste form using their DRAS code (EPACMTP model) for calculation of major pathways for human exposure to a given waste. If the allowable concentrations in the TCLP leachate of the waste, as calculated by DRAS, are higher than the Toxcitiy Characteristic (TC) level for the TC constituents, then the delisting level for the TC constituents can be capped at the TC regulatory limits. The UTS levels may or may not apply to a delisted waste. This is still highly debated even within the EPA. However, a waste form that meets the EPA UTS treatment standard limits should be easily delisted.

The results of the TCLP testing is provided in Table VI for FBSR products produced under reducing (Scoping Test 01 and Production Test 02) and oxidizing FBSR conditions (Production Test 01) since no TCLP testing had been performed on sample SCT02-098-FM from Scoping Test 02.

Table VI. TCLP Testing of FBSR Final Mineral Waste Form Products

Element of Concern[†]	**TCLP Releases for FBSR Under Reducing Conditions (ppm)**	**TCLP Releases for FBSR Under Reducing Conditions (ppm)**	**UTS Limits Federal Register, V.63, No. 100 p.28748-9 May 28, 1998 (ppm)**
Cr	0.015-0.060	0.001-0.018	0.60
Pb	0.005-0.023	0.002-0.067	0.75
Ni	0.001-3.11	0.66-2.80	11

* 1.0 or still hazardous

† As, Ba, Cd, Hg, Se, Ag, Zn, Sb, Be, Tl and V not in simulant

CONCLUSIONS

The following can be concluded about the use of Fluidized Bed Steam Reforming (FBSR) as a final waste form for Hanford's LAW waste:

- FBSR is a robust technology capable of accommodating wide ranges of feeds and additives including high concentrations of sulfate
- FBSR's ability to retain sulfate can lead to increased waste loadings and accelerated stabilization of Hanford's LAW vs. LAW vitrification
- FBSR's mineral waste species exhibit a superior durability to LAW glass in both ASTM C1285 and EPA TCLP testing
- FBSR's durability is incongruent and the radionuclides (Cs and Re as simulants for Cs^{137} and Tc^{99}) are released at a rate lower than that of Na (Na release is <2 g/m^2)
- FBSR is a medium temperature process: temperatures are low enough not to vaporize radionuclides but high enough to destroy volatile organic compounds (VOC's) in the presence of a reductant and a catalyst
- FBSR waste form mineral phases are cage like structures that trap radionuclides and anions
- FBSR waste form mineral phases alter to zeolites that have the same cage-like structures and will likely still retain the radionuclides and anions
- FBSR waste forms are regulatory compliant at the Universal Treatment Standard (UTS) making delisting of the final waste form more assured (at an estimated cost savings of $5952/ft^3$ for disposal since disposal of listed mixed waste is $6000/ft^3$ while disposal as radioactive (low level) waste once the waste is delisted costs only $42/ft^3$) [25]

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