MOLTEN SALT BREEDER REACTOR WASTE MANAGEMENT

DARRYL D. SIEMER*†

12N 3167E, Idaho Falls, Idaho 83402

Received December 7, 2012 Accepted for Publication June 13, 2013 doi:10.13182/NT12-164

The fuel reprocessing (recycling) system invoked by the developers of Oak Ridge National Laboratory's molten salt–based breeder (of ²³³U from ²³²Th) reactor (MSBR) would generate high-level reprocessing waste consisting of ~3 mol % fission product fluoride salts in a matrix consisting primarily of sodium and potassium fluoride salts. This technical note discusses a management scenario for such waste that invokes the following steps: (a) mixing of the waste salt with dilute nitric acid with a pug mill; (b) volatilization/separation of the bulk of the fluoride as hydrofluoric acid (HF) with a wiped film evaporator; (c) vitrification of the thus "converted" (to nitrate) salt waste to an iron phosphate glass waste form with a stirred melter; (d) reduction of the nitric acid/NO_x in the combined off-gas to elemental nitrogen with hot charcoal; (e) condensation of

REPROCESSING

TECHNICAL NOTE

KEYWORDS: molten salt breeder reactor waste management, liquid fluoride thorium reactor waste management, integral fast reactor waste management

the water and HF in the reduced off-gas; (f) neutralization of that solution with an alkali (sodium and/or lithium and/or potassium) hydroxide; (g) drying of that solution to produce the fluoride salts utilized by the process; and finally, (h) offgas disposal after treatment implemented with a condenser, wet electrostatic precipitator, catalytic converter, and highefficiency particulate air filters. This scenario's advantages relative to those that invoke the preparation of a synthetic fluoride mineral (cation-substituted fluorapatite) waste form include much higher effective waste loading, lower cost, and a product (glass) more consistent with stakeholder expectations.

Note: Some figures in this technical note may be in color only in the electronic version.

I. INTRODUCTION

The U.S. government's approach to reprocessing waste management continues to provide utility company chief executive officers, lawmakers, and antinuclear activists with readily documentable evidence of why the implementation of any sustainable (i.e., breeder reactor–based) nuclear renaissance in this country would be prohibitively expensive.^{1–3} This writer's most recent attempt to address this situation involved the development of a process that would reduce the cost of preparing the high-level reprocessing waste (HLRW) salts generated by the Integral Fast (Breeder) Reactor (IFR) for disposal; a description of that work is published in a previous issue of this journal.⁴ This follow-up technical note outlines how the same approach [separating/recycling the halide and

converting the rest to an iron phosphate (Fe-P) glass] could be applied to the HLRW salt wastes envisioned for molten salt breeder reactors (MSBRs).

II. MOLTEN SALT BREEDER REACTORS

The key distinguishing characteristic of any molten salt reactor (MSR) is that its fuel consists of a fissile cation dissolved in a low-viscosity, extremely high boiling, ionic solvent that serves double duty as its primary heat transfer medium. The fact that the fuel is a liquid renders its continuous "reprocessing" [i.e., removal of fission products (FPs), some of which are extremely valuable,^a also known as fuel salt cleanup] relatively

^{*}Retired consulting scientist for Idaho National Laboratory †E-mail: d.siemer@hotmail.com

^aFor instance, the "moly cows" containing the \sim 3 g (total) of ⁹⁹Mo sold to/utilized by U.S. hospitals every year currently cost U.S. citizens \sim \$360 million. One reason for the excessive cost

simple, which in turn means that it should be possible to operate these reactors continuously (no shutdowns for refueling) with relatively small fissile inventories. Only two of the many possible ways of implementing an MSR are genuinely sustainable: (a) the deliberately moderated (generally by graphite) molten fluoride salt breeder reactor of ²³²Th to ²³³U [or MSBR; also known as a liquid fluoride thorium reactor⁵ (LFTR)], which was primarily developed by Oak Ridge National Laboratory (ORNL) personnel,^{6b} and (b) the "faster" (unmoderated), molten chloride salt–based breeder of ²³⁸U to ²³⁹Pu and/ or ²³²Th to ²³³U investigated elsewhere. Since the latter's waste could be treated in the same fashion as that generated by the IFR, this techical note addresses only fluoride salt wastes.

The product of any competent radioactive waste treatment process should be highly resistant to corrosion (leaching) by water and not readily dispersible (i.e., monolithic, not "dusty"). Because durable radioactive waste-type glasses-all of which are oxide basedcannot accommodate much of any of the halides, most of the management scenarios envisioned for MSBR/LFTR radioactive wastes have assumed the same paradigm embraced by the IFR's developers; i.e., the goal is to produce a synthetic analog of a reasonably leach-resistant natural crystalline halide mineral. In practice, this boils down to making a cation-substituted sodalite [also known as a ceramic waste form (CWF)] of chloride salt-based wastes^{4,7} and a cation-substituted fluorapatite [Ca₅(PO₄)₃F] of fluoride-based wastes.^{8,9} My recent study⁴ of the IFR's waste management scenario identifies several reasons why it would make more sense to recycle the halide and vitrify only the cations:

1. Doing so would permit much higher waste loadings [less waste form/GW(electric)·yr would have to be made, temporarily stored, transported, and eventually buried] because glasses can accommodate much more of these wastes' primary cationic components (up to ~8 mM alkali/g) than can such crystalline minerals as the halides that accompany them: i.e., sodalite (Na₄Al₃Si₃O₁₂Cl) is only 7.3 wt% (or 2.0 mM/g) chlorine, and fluorapatite [Ca₅(PO₄)₃F] is only 3.8 wt% (2.0 mM/g) fluorine. 2. Greater acceptability: the United States has officially considered vitrification to be the "best demonstrated available technology" for treating its reprocessing waste for several decades,^{10,11} and maximizing recycle in waste treatment processes is both politically and technically correct.

3. Vitrification is intrinsically simpler and therefore cheaper to implement than is hot isostatic pressing (Ref. 11, p. 143 and p. 153), the technology usually invoked to generate high-quality crystalline mineral waste forms.

4. Unlike most of the cation separation schemes championed by the U. S. government's radioactive waste management experts, 11,12 halide separation is intrinsically easy (the addition of aqueous mineral acid followed by a quick boil-off generally suffices¹³) and is therefore economically feasible.^c

Since technical papers that do not assign even semiquantitative numbers to the concepts that they address are of limited value, this technical note assumes the waste streams generated by the 1.0 GW(electric) MSBR/LFTR described in Ref. 6^b (Table I)—the most detailed such report I have seen.

Table I illustrates that an MSBR/LFTR's "real" HLRW would be similar to that generated by the IFR (Ref. 4) in that it consists primarily of nonradioactive alkali halide salts (in this case, \sim 97 mol % of the total), not FPs and minor actinides (MAs). This is important because the characteristics of both the waste form and the process that makes it will be dominated by its major components.

III. EXPERIMENTAL

Since my previous paper⁴ details how the iron phosphate glass (Fe-P) specimens are both made and characterized, this section will be relatively brief. The direct approach utilized for making them from alkali fluoride salt(s) is the same as that shown⁴ to be suitable for chloride salts. It involves the gradual (to prevent boil over) heating of a mix of glass-forming additives (concentrated phosphoric acid and ferric oxide) with a salt waste surrogate consisting of the raw fluoride salt(s) [mostly alkali fluoride(s) along with a few mole percent MgF₂ and/or CaF₂, and/or ZrF₄], in an alumina crucible situated on an electric hotplate followed by placing that crucible into a preheated ~1100°C glass kiln for ~30 min. The "indirect" approach is identical except

is that the United States no longer possesses the ability to manufacture ⁹⁹Mo, which means that it must be imported. Since ⁹⁹Mo is a high-yield ($\sim 6\%$) FP that readily forms a volatile fluoride, continuously recovering it from an MSBR fuel salt slip stream would be simple.

^bThe best source of MSR/MSBR-related technical documents (e.g., papers/reports about chloride salt–based fast MSRs) is K. Sorensen's Energy from Thorium blog/website (click on its ORNL pdf document repository link). This repository includes a textbook, MSR-related technical reports produced both within the DOE complex and elsewhere, and papers published in technical journals.

^cFor instance, the decision/promise to separate the immense quantities of mostly alkali metal-based salt wastes in Hanford's tank farm into two fractions—one of which must contain 99.9% of the ¹³⁷Cs before vitrifying both—has generated most of the technical issues that have rendered that "cleanup" project prohibitively expensive.

What?	Source	gram mole/yr
LiF NaF MgF ₂ KF FP	Discarded fuel salt (still bottoms) Adsorbant Adsorbant Derived from the KOH mostly used for off-gas scrubbing Miscellaneous fission products in the above wastes	$5.6E + 04^{a} 7.1E + 04 3.1E + 03 1.7E + 05 7.4E + 03 $

TABLE I

LFTR Salt Wastes*

*Per GW(electric)·yr; from Table 7 in Ref. 6. aRead as 5.6×10^4 .



Fig. 1. Aluminum electrode voltammeter.

that the fluoride waste surrogate is converted to its nitrate analog by dissolving FPit in dilute nitric and boiling to dryness before the glass formers are added. In any case, the atom-wise proportions of P:Fe: Salkalies in the feed mixture should be in the range of 1: (0.4-0.8): $(0.9-1.5_{(max)})$, which values depend upon the waste surrogate's chemical makeup and how important "optimization" is deemed to be.⁴ The resulting products are (a) weighed to provide an estimate of fluoride retention, (b) visually examined to determine how "glassy" they are, and (c) subjected to a streamlined version of the product consistency test (PCT). The latter compares the degree of dissolution^d of 75- to 150-µm sample particles with that of same-sized particles of the U.S. Department of Energy (DOE) Environmental Assessment (EA) high-level waste (HLW) borosilicate glass reference material in 90°C water.14,15

form characteristic (the degree of gross matrix dissolution in hot water) that is relevant for glass-type waste forms; (c) it is reasonably reproducible; and most importantly, (d) a scientifically valid version of it can be performed with equipment that any reasonably competent experimentalist can cobble together with readily available materials.⁴ Because fluoride salts exhibited different behavior than did the chloride salts discussed in the previous paper,⁴ the formal PCT protocol was modified (again) so that it could indicate whether the waste form specimen actually behaves like glass, i.e., determine if its leach rate is governed by saturation or gross matrix dissolution.^e This modification simply involves replacing the leachate with fresh deionized water when successive conductivity measurements indicate that little further dissolution is taking place. If doing this quickly generates another leachate exhibiting conductivity similar to that of its predecessor, the sample is behaving like a moderately water-soluble mineral (e.g., BaF2)-not like a goodquality glass.^e Since the fate of fluoride is the key to understanding this process, an aluminum electrode voltammeter (Fig. 1)

The PCT test is used because (a) it has become

DOE's defacto standard leach test; (b) it measures a waste

was fabricated to perform the necessary analyses. It consists of three operational amplifier circuits: two homemade electrodes and a plastic test tube electrolysis cell. The first operational amplifier (left side) biases a tiny aluminum metal working electrode 0.5 V positive with respect to the much larger stainless steel auxiliary electrode,^f both of which are immersed in the sample solution containing

^dThese determinations are based upon the conductivities (glass dissolution releases inorganic ions that render leach water proportionately conductive) of leachates, not chemical analyses. The actual measurement is made by temporarily removing the leach vessel from the PCT oven, taking up a few drops of its contents (leachate) with a plastic eyedropper, squirting it into a tared plastic test tube situated on a sensitive digital balance, diluting it with an appropriate amount of distilled water, and then sucking that solution up into a tiny conductivity cell with a syringe.

^eSlavish adherence to the formal PCT-A protocol is apt to generate grossly misleading (overly optimistic) results if it is (mis)applied to materials that do not behave like glasses. This can have serious consequences to U.S. taxpayers because it may lead to inappropriate reprocessing waste treatment technology choices.

^fThe stainless steel auxiliary electrode also serves as the reference electrode because its absolute potential is determined (fixed) by the reduction potential of dilute nitric acid.

fluoride in $\sim 0.1 M$ nitric acid. The solution is stirred by air bubbles introduced with an aquarium pump through the tip of a fine-bore plastic tube situated under the electrodes. Under such conditions, the rate at which the aluminum dissolves (oxidizes) is directly proportional to the sample solution's free hydrofluoric acid (HF) concentration.¹⁶ The second operational amplifier's feedback loop forces electrons generated by the aluminum's oxidation through a ground-referenced 10⁵-ohm load resistor and thereby generates an output voltage directly proportional to the sample solution's fluoride concentration. The third amplifter comprises a low-pass (RC = 1.35 s) filter that smooths the second amplifier's noisy output^g so that a conventional digital voltmeter can accurately measure it. To minimize other errors, all determinations are done via the method of standard additions.

IV. RESULTS AND DISCUSSION

Extension of the principles/process previously demonstrated for IFR-type wastes⁴ to MSBR/LFTR wastes proved to be less straightforward than hoped. While most of the direct vitrification (direct vit) fluoride salt waste products *appeared* to be more durable than DOE's reference glass (see Fig. 2) because a smaller fraction of their alkali metals ended up in solution under PCT conditions, both their appearances (see Fig. 3) and physical characteristics (often "crumbly") suggested that they were glass composite materials^h rather than the true glasses produced from their chloride analogs.

There were several other indications that fluoride did/ does not behave like chloride:

1. The mass of the glasses made with the chloride salt waste surrogates invariably matched (within experimental error, approximately $\pm 1\%$) what they would/should if 100% of the chlorine (equivalent weight = 35.46) in the feed mixture had been replaced by oxygen (equivalent weight = 8). The mass of specimens made with the fluoride (equivalent weight = 19) salt waste surrogates did not (they were usually significantly heavier).

2. Finished chloride salt Fe-P glasses did not lose a significant fraction of their mass when reheated for extended periods at 1100°C—their fluoride analogs did.

3. The primary anion in PCT leachates generated from direct vit fluoride salt specimens was invariably fluoride, not the $HPO_4^{=}/H_2PO_4^{-}$ ions that predominate in water leachates generated from good-quality Fe-P glasses.

4. Water rinsates generated by rinsing off the dust adhering to ground/sieved particles of direct vit fluoride Fe-P specimens prior to PCT testing contained significantly more total salt than did rinsates generated from their chloride-based cousins.

5. And, most importantly, my latest modification of the PCT protocol indicated that their dissolution is solubility limited and therefore that they would be less durable in some "repository failure" scenarios than Fig. 2's leach curves would suggest.ⁱ

The solution to this conundrum is both obvious and easy to implement—remove most of the fluoride before vitrification.

Since the phosphoric acid utilized in my Fe-P glass recipes^j obviously could not displace sufficient fluoride,^k another acid had to be used to affect that separation. Nitric acid is the most suitable candidate¹ because it readily displaces HF from fluoride salt solutions, and both it plus the NO_x subsequently generated during vitrification are readily converted to innocuous gases (water and elemental nitrogen) that are easy to dispose of.

Figure 4 depicts the results of nitric acid boil-off experiments performed to determine how fluoride removal might best be accomplished. In one test series (dotted line), successively increasing (but all small) volumes of 3 M nitric acid were added to ~20-mg samples of dry sodium fluoride before the combination was heated to dryness in a Teflon beaker. In the other series (solid line), the same amounts of 3 M HNO₃ were added to aliquots of a NaF solution (0.5 molal) containing the same 20 mg of the salt accompanied by much more water. In all cases, after the boil-off, the residue was dissolved in ~0.2 M nitric acid, and that solution's fluoride content was determined with the aluminum electrode voltammeter.

^g"Noisy" due to the chaotic nature of bubble-induced sample solution stirring.

^hIn this case, glass composite material means particles of a moderately soluble fluoride-containing mineral phase dispersed in glass (which gives it a dull, not glassy, appearance). Mineral dissolution rate is generally limited by its solubility product ("ksp") and the amount of liquid water available to saturate, not by the rate at which anything within each sample particle can diffuse through an already depleted and strongly adherent surface layer—the mechanism obtaining with a good-quality "pure" glass.

ⁱAlmost all credible "repository failure" scenarios invoke flooding of the burial zone/waste form with liquid water. If such groundwater were *moving* (not absolutely stagnant), it would not achieve saturation and therefore quickly dissolve/ disperse anything with solubility-limited leach behavior.

^JPrevious studies of Fe-P–type glasses generally added the phosphorous in forms (e.g., P_2O_5 , AIPO₃, CaHPO₄, etc.) less likely than H_3PO_4 to promote HF (and chloride) volatilization. ^kOne reason for this is that phosphoric acid is not "strong" enough ($K_{a1} = 7.25 \times 10^{-3}$) to quantitatively convert fluoride ion to HF. Another is that the molar ratio of fluoride to phosphoric acid in my glass formulations is typically above unity in order to achieve good (high) waste loadings.

¹Sulfuric acid's drawbacks are that SO_x cannot readily be rendered innocuous and sulfate tends to form troublesome salt phases in glass melters.



Fig. 2. Fraction of total alkali leached from several direct process fluoride salt–based Fe-P glasses and EA borosilicate glass under PCT conditions.



Fig. 3. Direct vit products (everything is mixed together and melted in the crucible) Fe-P–sodium fluoride plus H_3PO_4/Fe_2O_3 (a glass composite material); CWF alternative⁴ = equivalent amount of NaCl plus H_3PO_4/Fe_2O_3 (a genuine glass); bsg EA = remelt of DOE's benchmark borosilicate glass.

These experiments revealed that (a) excess water (steam?) facilitates the removal of fluoride from its salts with nitric acid and (b) under optimum conditions, fluoride removal/volatilization is near stoichiometric; i.e., each mole of nitric acid volatilizes 1 mol of HF.^m Subsequent experiments revealed that the HF should be boiled off before the ferric oxide and/or phosphoric acid is/are added.

^mSimilar experiments performed with fluoride salts of cations that form strong fluoride complexes (e.g., ZrF_4 —a minor component of the combined waste) revealed a much lower degree of fluoride removal. To a chemist this is not surprising and is probably why subsequent experiments indicated that the fluoride should be removed *before* the ferric oxide is added (ferric iron also forms strong fluoride complexes).



Fluoride boil-off curves

Fig. 4. Fluoride displacement/volatilization with nitric acid.



Fig. 5. PCT leach curves of Fe-P specimens made with sodium fluoride both directly (solid line) and after most of its fluoride had been removed via boil-off with nitric acid.

For "large scale" experiments involving the preparation of sufficient glass for leach testing, that separation was performed by dissolving the waste salt surrogate in a slight stoichiometric excess of 2 *M* HNO₃ (nitrate:fluoride mole ratio ≈ 1.15) in a Teflon-coated pan and then baking it to dryness on an electric hotplate. Figure 5 compares the leach characteristics of products made from NaF both with and without this separation. The converted specimen appeared like and exhibited the leach characterics⁴ of those made with an equivalent amount of NaNO₃ or NaCl while the latter looked like Fig. 3's leftmost specimen and exhibited crystalline mineral-like (saturation-limited) leach behavior (note the large "steps" induced in its PCT leach curve by the water changes). Figure 6 depicts Fe-P

NUCLEAR TECHNOLOGY VOL. 185 JAN. 2014

specimens made with a 1:1 mole-wise mix of converted NaF plus KF and equivalent amounts of technical-grade NaNO₃ and KNO_{3-t}; they are equally "glassy."

V. OFF-GAS TREATMENT AND CHEMICAL RECYCLE

Figure 7 depicts a hypothetical flow sheet for the overall waste treatment process based upon HSC 5 thermodynamic modeling.¹⁷

The MSBR's combined waste streams (again, mostly consisting of alkali metal fluoride salts) are pug mill– mixed with water and nitric acid, and the resulting solution is fed to a steam-heated, wiped film evaporator



Fig. 6. Glasses made with converted (defluoriated) NaF plus KF (left) and NaNO3 plus KNO3 (5.8 mM/g total alkali in each).



Fig. 7. Process flow sheet.

that boils off most of its water and HF. The "converted" (to nitrate) dry salt mixture is then run into a stirred glass melter along with sufficient ferric oxide and phosphoric acid to produce a good-quality Fe-P glass. The H_3PO_4 is introduced through a packed, countercurrent scrub column in order to return the particulate matter and fumes in the combined mixer/evaporator/melter off-gas streams to the melter.

A fluidized bed reactor contacts the scrubbed off-gas with \sim 500°C charcoal granules that serve to chemically

reduce all NO_x species (HNO₃, NO₂, NO, etc.) to N₂ and the majority of any concomitant gaseous or semivolatile FPs (possibilities include ¹²⁹IF₅, ¹²⁵SbF₅, ⁹⁹TcF₅, ⁹³MoF₆, H⁹⁹TcO₄, etc.) to less volatile (mostly elemental) species apt to be readily adsorbed by charcoal. When the initially much larger charcoal granules are ground down/oxidized to tiny (<15-µm) particles, they plus their adsorbed FPs are blown out of the reactor, cooled to ~125°C with sprayed water, and captured by a blowback filterequipped cyclone. The filtered "de-NO_xed" off-gas passes through a packed bed condenser that converts its gaseous water and HF to a dilute acid solution that is neutralized with NaOH (and/or ⁷Li OH and/or KOH) and then dried to generate useful chemicals (i.e., fluorine is recycled). The so-regenerated water vapor is recondensed and the liquid recycled to the pug mill mixer. Noncondensible off-gases (at this point primarily N₂ and CO₂) pass through an automotive-type catalytic converter to destroy trace concomitants (e.g., CH₄, H₂, CO, NH₃, etc.), then through a wet electrostatic precipitator (WESP), and are finally high-efficiency particulate air (HEPA) filtered before being discharged to the atmosphere.

Of the many possible treatment options for the semivolatile FP-laden carbon dust captured by the blowback filters, the most attractive is to simply recycle it to the melter. Recent studies suggest that an efficient off-gas scrubbing/recycling system would eventually put most of the semivolatile FPs into the glass¹⁸ and that implementing their recycle in this fashion is unlikely to put sufficient elemental carbon into the melter to upset its redox state.ⁿ

VI. CONCLUSIONS

Vitrifying a LFTR's reprocessing wastes would be intrinsically more difficult than that of a "fast" breeder reactor's chloride-based salt wastes because of the additional equipment (e.g., pug mill mixer and wiped film evaporator) required to affect the necessary degree of halide removal. However, there is nothing exotic about either such equipment or the way that it would be employed.

The properties of the product glasses will be much like those prepared from either the chloride-based salt waste surrogates used in Ref. 4 or DOE's high-sodium tank wastes^{20–22} because they are all basically the same thing—durable glasses composed primarily of alkali metals, phosphorous, ferric iron,^o and oxygen that serve

to immobilize relatively small amounts of concomitant radioactive/toxic materials.

The amount of waste form made will be determined by the radioactive wastes' alkali metal content, not its FPs or MAs. The waste quantities described in Table I combined with a waste loading of 8 mol alkali/kg translates to ~37 tonnes (~12.8 m³)^p of Fe-P glass waste form per GW(electric)·yr. Approximately 160 tonnes (50 m³) of theoretically dense (3.2 g/cm³) fluorapatite would be needed to immobilize the same waste.

The production of 37 tonnes of glass per year would require relatively small/inexpensive equipment [e.g., a 0.3-m (1-ft)–diam glass melter] provided that it was closecoupled to a continuous fuel salt reprocessing/recycling system.

If future environmental management decision makers continue to insist upon a borosilicate glass, in principle it should be possible to produce one by substituting powdered silica and boric acid (or a glass "frit" containing them) for the phosphoric acid and ferric oxide depicted in Fig. 7. However, the production of a borosilicate glass is likely to be more expensive/difficult because both off-gas treatment and fluoride/water recycle are apt to be complicated by volatile silicon (SiF₄) and boron (BF₃) species.

While Fig. 7's flow sheet includes several as-yet unproven features and will therefore probably need some revision/optimization, I feel that it is reasonable to expect that the flow sheet could be made to achieve its goals.

Finally, I encourage any individual or group possessing both the will and wherewithal (e.g., access to a "real" laboratory) to do so, to correct, verify, clarify, and/or improve upon anything described in either this or my previous paper.

NOMENCLATURE

- CWF Ceramic waste form: the sintered-together mix of powdered glass and synthetic sodalite that represents ANL's solution to the IFR salt waste immobilization problem [also called glass bonded zeolite (GBZ) or glass bonded sodalite (GBS)].
- Fe-P Iron phosphate glass: the alternative to borosilicate glass developed by D. Day and his students at the University of Missouri-Rolla. Its advantages vis-à-vis borosilicate glasses include higher waste loadings, easier fabrication, and superior leach resistance.

IFR Integral fast reactor.

ⁿThis assertion, "... it is highly unlikely that...," is based upon (a) the stoichiometry of NO_x reduction and (b) the fraction of coarse granular carbon ("coke") fed to a fluidized bed off-gas treatment pilot plant (carbon reduction reformer) subsequently blown out of it as dust (4.8 wt%). The latter datum was obtained from a proprietary report¹⁹ describing Hazen Research's last attempt to demonstrate the \$571 million steam reforming process that is eventually supposed to convert Idaho National Laboratory's remaining liquid reprocessing waste to a water-soluble and readily dispersible waste form.

^oPrevious studies^{20–22} of Fe-P's potential for mitigating DOE's vitrification cost issues have capitalized upon the fact that aluminum can be substituted for ferric iron. Still-preliminary work performed subsequent to the acceptance of this technical note suggests that total substitution of aluminum for iron may render the conversion of fluoride-based wastes to their nitrate analogs unnecessary.

^pFe-P volume/year = 2.97×10^5 mol alkali/yr (Table I) / 8 mol alkali/kg (alkali glass loading) / 2.9 kg/ ℓ (glass density) = 12 800 ℓ (12.8 m³)/yr (calculation is similar for fluorapatite).

- LFTR Liquid fluoride thorium reactor: a less evocative term for a thorium-fueled MSBR capable of break-even fissile regeneration.
- MSBR Molten salt breeder reactor.
- PCT Product consistency test: the relatively rapid (nominally 7-day) leach test originally developed to monitor the durability of the glass produced by the Savannah River Site's HLW glass melter.

ACKNOWLEDGMENTS

I thank K. Sorensen for creating the "Energy from Thorium" web/blog site, which, first, reminded me why I decided to become a scientist and, second, provided a wealth of technical information. I also thank J. Christian and D. Day, who donated supplies without which this project could not have been accomplished. Finally, I would like to thank the legions of anonymous "techies" whose volunteer labor has rendered Wikipedia, Google, and the Engineer's Tool Box invaluable to any technical hobbyist.

REFERENCES

1. J. MAHAFFEY, *Atomic Awakening*, pp. 304–305, Pegasus Books (2009).

2. B. COHEN, *The Nuclear Energy Option*, Chap. 12, Plenum Press (1990); http://www.phyast.pitt.edu/~blc/book/chapter12 .html (current as of Dec. 7, 2012).

3. Barriers to Science: Technical Management of the Department of Energy Environmental Remediation Program, National Academies Press (1996); http://www.nap.edu/catalog .php?record_id=10229 (current as of Dec. 7, 2012).

4. D. D. SIEMER, "Improving the Integral Fast Reactor's Proposed Salt Waste Management System," *Nucl. Technol.*, **178**, 341 (2012).

5. R. MOIR and R. HARGRAVES, "Liquid Fluoride Thorium Reactors: An Old Idea in Nuclear Power Gets Reexamined," *Am. Sci.*, **98**, 304 (2010).

6. C. D. SCOTT and W. L. CARTER, "Preliminary Design Study of a Continuous Fluorination Vacuum Distillation System for Regenerating Fuel and Fertile Streams in a Molten Salt Breeder Reactor," ORNL 3791, Oak Ridge National Laboratory (Jan. 1966).

7. C. E. TILL and Y. I. CHANG, "Plentiful Energy: The Story of the Integral Fast Reactor," *CreateSpace*, p. 182 (Dec. 2011), doi:10.1007/s11661-999-0202-x.

8. D. LEXA, "Preparation and Physical Characteristics of a Lithium-Beryllium-Substituted Fluorapatite," *Metall. Mater. Trans. A*, **30**, *1*, 147 (Jan. 1999).

9. R. W. MOIR and E. TELLER, "Thorium-Fueled Underground Power Plant Based on Molten Salt Technology," *Nucl. Technol.*, **151**, 334 (2005).

10. "Glass as a Waste Form and Vitrification Technology: Summary of an International Workshop," National Academies Press (1996); http://www.nap.edu/openbook.php?record_ id=5488 (current as of Dec. 7, 2012).

11. Alternative High-Level Waste Treatments at the Idaho National Engineering and Environmental Laboratory, pp. 66–68, 143, 153, National Academies Press (1999); http://www.nap .edu/catalog.php?record_id=9743 (current as of Dec. 7, 2012).

12. "External Technical Review of the Flowsheet for the Hanford Waste Treatment Plant (WTP)," ERT-1, U.S. Department of Energy, Office of Environmental Management (Mar. 2006); http://energy.gov/sites/prod/files/em/Summary-FlowsheetfortheHanfordWasteTreatmentPlant-March2006.pdf (current as of Dec. 7, 2012).

13. J. BRADY and G. HUMISTON, *General Chemistry— Principles and Structure*, 3rd ed., p. 639, John Wiley and Sons (1982).

14. "Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses and Multiphase Glass Ceramics: The Product Consistency Test (PCT)," ASTM C1285-02(2008), ASTM International (2008).

15. C. M. JANTZEN, N. E. BIBLER, and C. E. BEAM, "Characterization of the Defense Waste Processing Facility (DWPF) Environmental Assessment (EA) Glass Standard Reference Material," WSRC-TR-92-346, OSTI ID 6876372, Westinghouse Savannah River Company (1992).

16. C. R. CASTOR and J. H. SAYLOR, "Amperometric Determination of Fluoride," *Anal. Chem.*, **24**, *8*, 1369 (1952), doi:10.1021/ac60068a034.

17. "HSC Chemistry—Chemical Reaction and Equilibrium Thermodynamics," Version 5, Chempute Software (2002).

18. I. L. PEGG, K. S. MATLACK, and I. S. MULLER, "Technetium Retention During LAW Vitrification," presented at Environmental Management Waste Processing Technical Exchange 2010, Atlanta, Georgia, November 16–18, 2010; http://www.srs.gov/general/srnl/techex_2010/pdfs/S02-03.pdf (current as of Dec. 7, 2012).

19. "Pilot Plant Report for Treating Sodium-Bearing Waste Surrogates Carbonate Flowsheet," RT-ESTD-PMR-001, Rev. 0, Idaho Cleanup Project, THOR Treatment Technologies and Washington Group International (Oct. 2006).

20. G. J. SEVIGNY et al., "Iron Phosphate Glass–Containing Hanford Waste Simulant," PNNL 20670, Pacific Northwest National Laboratory (Aug. 2011).

21. R. LEERSON, "Fe Phosphate Glass for the Vitrification of INEEL SBW and Hanford LAW," MS Thesis, University of Missouri–Rolla (2002).

22. D. S. KIM et al., "Iron Phosphate Glass as an Alternative Waste-Form for Hanford LAW," PNNL 14251, Pacific Northwest National Laboratory (2003).