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TO: SRS Citizens Advisory Board (CAB), and
SRS Department of Energy (DOE)

Public Comment
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From: Dr. John B. Pickett

I am a retired WSRC/Dupont (Principal Technical Advisor) engineer. I had 25 years experience at the SRS, primarily in SRNL and M-Area. I retired in 2002.

I recently noticed an article which stated that the contractor for the Defense Waste Processing Facility (DWPF), Savannah River Remediation (SRR) LLC, is planing to install "bubblers" into the DWPF melter. I have some serious concerns with this concept.

I believe I may be uniquely qualified to comment on these concerns. I am quite familiar with the bubbler system being proposed by SRR and the Vitrious State Lab.(VSL) I was an author of a set of contract specifications for a private vendor to treat the low level/ mixed waste in M-Area. The contract was awarded to G.T. S. Duratek, and they constructed and operated a glass melter in M-Area for 6-7 years. The melter utilized the VSL's bubbler technology. I acted as the STR (Subcontract Technical Representative) for the 1st 3 years of the contract. I had a number trips to VSL to work with their researchers there, and I maintained close oversight of the M-Area operations.

W/r to the DWPF operation, I am a co-author on 2 of the main process control models for the DWPF (the glass durability model "THERMO"), and the Reduction/Oxidation model "REDOX".

Based on this experience I have a number of concerns w/r to the use of bubblers in the DWPF melter.

- 1) Based on the proximity of the bubbler to the melter wall, and the rate of gas flow, the bubbler WILL erode the K-3 refractory. It will also cause increased corrosion/erosion at the melt pool line. It may be possible to estimate the time it will take to erode the K-3, if VSL has conducted simulant studies at the proposed configurations and flow rates. I know that the bubblers in both of the M-Area melters caused significant erosion. I believe that the 2nd M-Area melter had a 12 inch layer of K-3 refractory (similar to the DWPF), so it finished the waste treatment before breaching. I would estimate no longer than 1-2 years before the K-3 in the DWPF melter is breached.

This is the 2nd DWPF melter - the first was replaced after 7 years due to lid heater problems. The 2nd melter is now about 7 years old and may have 4-5 years left. Removing it 2-3 years earlier than necessary would not be good, but not terrible.

- 2) I am more concerned about the effects of the bubbler system on the Redox control, the DWPF off-gas system, and the downstream waste effects on the tank farm evaporators and the Saltstone Performance Assessment. The DWPF currently uses a system of formic and nitric acids, added to the DWPF feed tanks to control the Redox

in the melter. This control is crucial to insure that noble metals are not reduced to molten metals or sulfides, which would short out the DWPF electrodes. The redox is also critical to insure that the many of the fission product components (such as Tc-99 and Ru-104) in the sludge are incorporated into the glass matrix.

The formic/nitric acid feed works in conjunction with a "Cold Cap" on the surface of the glass in the melter. The sludge feed forms a layer 2-4 inches thick, where the oxidation/reduction reactions actually occur. This cold cap is critical to this step. However, with a bubbler system, there will be no cold cap at all, a very thin cold cap, and/or thin patches of cold cap. These types of cold caps will NOT provide redox control.

How will the redox be controlled in the DWPF with a bubbler system in use? Would various types of gases be used? How frequently will the final glass product be measured to insure that the radioactive constituents are sequestered in the glass and that the durability limit is met?

In addition to redox control, the cold cap prevents significant solid carryover to the melter off gas system, and keeps the plenum temperature above the melt pool 300-400 degrees (C) cooler. The combination of a bubbler, no melt pool cold cap, and a hotter plenum temperature will cause molten glass strings to freeze and plug the off gas discharge at the top of the melter. I know this will happen in the DWPF, because it happened with the M-Area melter. That melter had 2 foot diameter off-gas discharge port, and it plugged with glass fibers and sludge every few days. An operator would climb up on top of the melter, and rotate a beater bar to break the glass and let it fall back into the melt pool.

I understand that the DWPF has a device in the off-gas discharge port (off-gas film cooler brush) to break up and remove plug-age, but I don't know if it will be rugged enough for 24 hour service and solid glass shards. Can the expected lifetime of the DWPF off-gas film cooler brush be estimated, and how long (how much downtime) would be required to replace it?

The cold cap also plays a key role in minimizing carry over of volatile constituents in the DWPF sludge, such as Ruthenium(Ru), Technicium(Tc), chlorides(Cl), fluorides(F), Cesium (Cs), Strontium (Sr), and sulfides. A bubbler system will cause MUCH greater loss of these constituents than the current DWPF process. For example, Tc is currently retained at >99% in the glass, due to the cold cap and Redox control. I would expect no better than 30-50% retention of Tc with a bubbler system. The Tc would then be returned to the tank farm, and eventually end up the Saltstone grout. This is a concern due to fact that Tc is very mobile and the Saltstone facility has limits on how much Tc it can accept. Has the level of Tc retention been determined in simulant runs? What will be the effect of additional Tc on the Saltstone feed, the Saltstone waste water treatment permit, and Saltstone Performance Assessment?

The additional carry over of Cl, F, and S acidic gases would be a serious concern if the DWPF off-gas system were constructed with stainless steel. However, the DWPF off-gas is constructed of Hastalloy, which is more resistant to acidic gases. However, even Hastalloy is not impervious, and it will eventually be corroded and/or dissolved. If the

increased rate of Cl, F, and sulfur off-gas releases were determined in simulant studies, perhaps the lifetime of the DWPF off-gas system could be estimated.

Without a cold cap, additional Cs and Sr will also be returned to the tank farm. This will have to be re-treated, and then recycled back to the melter. The additional treatment time (for Cs and Sr removal) and cost should be calculated based on known Cs and Sr carryover to the off-gas condensate systems (with and without the bubbler system).

The amount of silicon (from the glass forming frit feed) will probably also increase in the DWPF off-gas condensate (w/o a cold cap). This will adversely affect the tank farm evaporator, as the concentration of Si in the evaporator feed is critical to keep the evaporator from plugging. This possibility should be explored in more detail.

QUESTIONS:

- Based on the "post-mortem" inspection carried out by G.T.S. Duratek on the 2nd M-Area melter, how long will the K-3 refractory in the DWPF melter last?
- How will the redox be controlled in the DWPF with a bubbler system?
- Would various types of gases be used?
- How frequently will the final glass product be measured to insure that the radioactive constituents are sequestered in the glass and that the durability limit is met?
- Can the expected lifetime of the DWPF off-gas film cooler brush be estimated, and how long (how much downtime) would be required to replace it?
- Has the level of Tc retention been determined in simulant runs?
- What would be the effect of additional Tc on the Saltstone feed, the Saltstone Waste Water treatment permit, and Saltstone Performance Assessment?
- What is the rate of additional acidic gas carryover to the off-gas system, and can the effect on the Hastalloy be estimated?
- How long (how many months, or years) would be required to replace the DWPF off-gas system, if and when the Hastalloy is corroded?
- How much additional Cs and Sr would be carried back to the tank farm?, and how much would it cost to retreat the Cs/Sr to return it to the DWPF ? (note, at least 4-5 iterations should be used)
- Based on simulant studies, how much additional silicon and/or borates would be carried over to the tank farm evaporator?, and
- Would this affect the evaporator plug-age rate?

REFERENCES

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

C.M. Jantzen, D.C. Koopman, C.C. Herman, J.B. Pickett, and J.R. Zamecnik, "Electron Equivalents REDOX Model for High Level Waste Vitrification", **Environmental Issues and Waste Management Technologies IX. Ceramic Transactions V. 155**, J.D. Vienna and D. R. Spearing, (Eds), American Ceramic Society, Westerville, OH, 79-91, (2004).

J.B. Pickett, C.M. Jantzen, and L.C. Martin, "First Delisting Petition Approval by the U.S. EPA For a Vitrified Mixed Waste", Environmental Issues and Waste Mgt. Technologies VIII. Ceramic Transactions, V. 143, S.K. Sundaram, J.D. Vienna, and D.R. Spearing, (Eds.), American Ceramic Society, Westerville, OH, 83-94 (2003).

J.B. Pickett and C.M. Jantzen, "TCLP Leaching Prediction from the THERMO® Model for Borosilicate Glasses", Environmental Issues and Waste Management Technologies, VII, G.L. Smith, L.K. Sundaram, and D.R. Spearing (Eds.), Am. Ceram. Soc., Westerville, OH, p. 323-333 (2002).

J. B. Pickett, S. W. Norford, J. C. Musall, and D. J. Bills ((2001). "Vitrification and Privatization Success", Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries VI. Ceramic Transactions, V. 102, from the 102nd Annual Mtg. of the Am. Ceramic Society, St. Louis, MO, April 29-May 3, 2000, D. R. Spearing, G. L. Smith, and R. L. Putnam (Eds.), American Ceramic Society, Westerville, OH.

J. B. Pickett, S. W. Norford, and G. A. Diener (1999). " First Commercial U. S. Mixed Waste Vitrification Facility: Permits, Readiness Reviews, and Delisting of Final Wasteform", Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries IV. Ceramic Transactions, V. 93, from the 100th Annual Mtg. of the Am. Ceramic Society, Cincinnati, OH, May 3-6, 1998, J. C. Marra and G. T. Chandler (Eds.), American Ceramic Society, Westerville, OH.

C. M. Jantzen, K. G. Brown, K. J. Imrich, and J. B. Pickett, (1999). "High Cr₂O₃ Refractory Corrosion in Oxidizing Melter Feeds: Relevance to Nuclear and Hazardous Waste Vitrification," Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries IV. Ceramic Transactions, V. 93, from the 100th Annual Mtg. of the Am. Ceramic Society, Cincinnati, OH, May 3-6, 1998, J. C. Marra and G. T. Chandler (Eds.), American Ceramic Society, Westerville, OH.

C. M. Jantzen, J. B. Pickett and K. G. Brown, (1998). "COMP-U-GLASS: Glass Property Optimization from Glass Composition", Proceedings of XVIII International Congress on Glass, July 5-10, 1998, San Francisco, CA, M. J. Choudhary, N. T. Huff, and C. H. Drummond, (Eds.), American Ceramic Society, Westerville, OH (1998).

K.G. Brown, C.M. Jantzen, and J. B. Pickett, "The Effects of Formate and Nitrate on REDuction/OXidation (REDOX) for the Defense Waste Processing Facility", U. S. DOE Report, WSRC-RP-97-34, Rev.1 (June 30, 1998).

C. M. Jantzen, K. G. Brown, J. B. Pickett, and T. B. Edwards, "Statistical Process Control for Vitrification of Waste Glasses Using Compositional - Property Relationships", Invited Presentation at American Nuclear Society Special Session, 6/1-5/97, Orlando, FL.

- C. M. Jantzen, J. C. Whitehouse, M. E. Smith, J. B. Pickett, and D. K. Peeler (1998). "Waste Vitrification Projects Throughout the U.S. Initiated at the Savannah River Site," Environmental Issues and Waste Management Technologies in the Ceramic and Nuclear Industries III. Ceramic Transactions. V. 87. from the 99th Annual Mtg. of the Am. Ceramic Society. Cincinnati, OH. May 3-7, 1997. J. C. Marra and D. K. Peeler (Eds.), American Ceramic Society, Westerville, OH
- S. W. Norford and J. B. Pickett, "M-Area Vendor Treatment Facility", Presentation at the AIChE Spring National Meeting, 3/9-13/97, Houston, TX.
- R. E. Hottel, J. B. Pickett, J. R. Kosko, and L. D. MacLean, "Privatization of Waste Management Programs at the Savannah River Site", Presentation at Waste Management '96 Conference. Tucson AZ. 2/25-2/29/96. University of Arizona, Tucson, AZ.
- C. M. Jantzen, J. B. Pickett, K. G. Brown, T. B. Edwards, and D. C. Beam, "Process/Product Models for the Defense Waste Processing Facility (DWPF): Part I. Predicting Glass Durability from Composition Using a Thermodynamic Hydration Energy Model (THERMO) ," U. S. DOE Report WSRC -TR-93-0672, 464 p., (September, 1995).
- C. M. Jantzen, J. B. Pickett, W. G. Ramsey and D. C. Beam, 1994. "Treatability Studies on Mixed (Radioactive and Hazardous) M-Area F006 Sludge; Vitrification via the Reactive Additive Stabilization Process (RASP)", Proceedings of International Topical Meeting on Nuclear and Hazardous Waste Management SPECTRUM '94. 8/14-18/94. American Nuclear Society, La Grange Park, IL 60525.
- J. B. Pickett, J. C. Musall, A. F. Hayes, and E. A. Campbell, "Novel Procurement Concepts Utilized to Award Subcontract for Vitrification of an F006 Mixed Waste Sludge", 1994. Proceedings of International Topical Meeting on Nuclear and Hazardous Waste Management SPECTRUM '94. 8/14-18/94. American Nuclear Society, La Grange Park, IL 60525.

PATENTS

#5,846,278; December 9, 1998, "Method of Determining Glass Durability, "THERMO".