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Vacuum Assisted Filtered Salt Sampling Progress

September 2022

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ABSTRACT

Several different design iterations for a filtered salt sampling technique have been tested in non-radiological molten salts. A piston vacuum assembly was designed and reliably and repeatably used to take salt samples through multiple different porous quartz frit sizes (as small as 5 – 10 μm). This design has been modified slightly and sent into the HFEF hot cell for upcoming testing with used fuel electrorefiner salt.

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ACRONYMS

FSS – Filtered Salt Sampler

$GdCl_3$ – Gadolinium chloride

HFEF – Hot Fuels Examination Facility

INL – Idaho National Laboratory

LiCl-KCl – Lithium chloride-potassium chloride eutectic

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Filtered Salt Sampling Progress

1. Introduction

Recent experience with sampling salt for analytical analysis in electrorefining salt (LiCl-KCl-UCl_3) at INL has, at times, revealed high variability of uranium concentration in the salt among duplicate samples. During the most recent work performed at HFEF standard deviations amongst what should be duplicate samples have ranged from as low as 0.6% up to as high as 76%. A possible explanation for this variability has been proposed involving entrainment of solid particles in the molten salt bath. To avoid variability due to entrained particles, a method for taking filtered salt samples is being developed for use in systems within both remotely operated hot cells and gloveboxes. This method had been initially tested by submerging a quartz tube with a quartz filter in the end in the salt and relying solely on the hydrostatic head to push salt through the frit into the tube. Results from this testing is covered in the previous report entitled "Initial Design, Research, and Experimentation for Filtered Salt Sampling," (INL/EXT-21-64908). Following that work the design has been updated to include a method of pulling a small amount of vacuum on the quartz tube to pull the salt sample in through the frit. Two methods were tried: (1) a rubber pipette bulb and (2) a small piston pump. Testing of these new filtered salt sampler (FSS) designs will be described and discussed in this report.

2. Background

Previous tests were performed relying on the hydrostatic head of the molten salt to flow salt through the frit and into the quartz sample tube. Varying success was experienced while testing this method using a surrogate molten salt in an argon atmosphere glovebox depending on the porosity of the frit in the sampler. With the finer frits (16 – 40 micron) salt was not able to flow through into the sampler. With the coarser frits (160 - 250 micron), salt was able to flow through into the sampler, however it would also quickly flow back out upon removal from the bulk salt prior to freezing. Based on these results it was decided that further testing would involve applying a small amount of vacuum to the samplers to pull molten salt up into the finer frit samplers.

3. Sampler Design

Two methods were used to generate the desired small pressure differential across the quartz frit for salt sampling. First, a rubber bulb was fitted to the top of a quartz sampler tube and was squeezed and released to generate the pressure difference. This approach was a direct precursor to the second method, which was to design a better-sealing assembly out of off-the-shelf components such as Swagelok fittings, a pressure gauge, and a double-acting piston. In effect, these designs are the same in function but different in quality of hardware and results.

3.1 Rubber Bulb Design

The first design change came in the form of attaching a standard laboratory rubber pipette bulb to the top of a roughly 21" long quartz tube with a frit in the end of various porosities. The same rubber bulb could be attached to several fritted quartz tubes repeatedly; however, it was difficult to reliably form a seal between the rubber bulb and the quartz. The unreliability of this design is discussed further in Section

4. With the primary issue of this design being the reliability of the quartz to bulb seal, the next design was focused on creating a repeatable and effective seal to the quartz sampler tube.

3.2 Piston Vacuum Design

The improved sampler design focused on creating a better seal to the quartz sampler tub and confirming that a pressure differential can repeatably force salt through the quartz frit. The new sampler design is referred to as the piston vacuum assembly utilizes off-the-shelf pipe fittings capable of making better seals than the rubber bulbs from the previous design.

The assembly can be divided into two parts: the reusable piston vacuum hardware and the single use fritted quartz tube. The sampler is assembled by inserting a long quartz tube into the appropriate fitting on the piston vacuum hardware, then the quartz tube is lowered into the molten salt. The piston on the piston vacuum hardware is then extended, lowering the internal pressure of the assembly relative to ambient. This encourages the molten salt to flow through the frit and into the sampler due to the pressure differential and eventually return or nearly return the internal pressure of the assembly to the surrounding ambient pressure. This results in a filtered salt volume of no more than the piston's displaced volume, which is easily controllable per sample if desired. The sampler is then removed from the furnace and allowed to cool, followed by breaking the quartz tube to retrieve the filtered salt samples. The quartz tube is no longer usable, but the piston vacuum assembly continues to be usable.

3.2.1 Piston Vacuum Assembly

The piston vacuum assembly is a simple assembly constructed of various Swagelok fittings, pipe adapters and tees, a piston, pressure gauge, and a grip intended for use by remote manipulators within HFEF. Fabricating this assembly is trivial and only requires that the fittings be purchased, treated with a thread sealant such as an adhesive or Teflon tape, and then tightened together for final assembly. A picture of the final assembly can be seen in Figure 1.

Figure 1. Completed piston vacuum assembly.

The purposes of key components of this part of the assembly are detailed here. The piston (top right item in Figure 1) is a double-action piston that displaces the same volume each time it is actuated. For the purposes of this project, this piston is an input that can be used to change the internal volume of the sampler. Because the sampler is completely sealed except for the end of the quartz tube, increasing the volume of the sampler by actuating the piston results in several psi of pressure change relative to ambient. This can be read on the gauge at the top of the assembly; however, it is not necessary for the experiment and was added due to its low cost and effort required. The final key component of the piston vacuum assembly is the Swagelok Ultra-Torr fitting which is located on the bottom of the assembly. This is the point where the quartz tube is attached to the sampler. The rubber O-ring inside of the fitting is compressed by the tightening of the fitting, resulting in the O-ring compressing against the outer surface of the quartz tube and forming a reliable seal. The reliability of this connection between the Swagelok Ultra-Torr and the quartz sampler tube is much better than the rubber bulb from the previous design, as discussed in Section 4.

3.2.2 Quartz Tubing

The quartz tubing design has been changed to consist of a standard quartz tube with a single quartz frit fused inside of the end of the sampler tube. In previous experiments, concerns were raised over the possibility that the quartz frits had been directly exposed to the flame of the torch used to fuse the frits in place. Special instructions for fabricating these samplers were written in order to completely remove this possibility. These instructions were essentially to fuse the quartz frit inside of the quartz tube approximately 1/3 of the distance through the quartz tube and then to cut the quartz tube approximately 0.25” below the frit. Though not investigated in previous tests, this fabrication change may have improved results going forward.

Additionally, some fritted quartz sampler tubes were fabricated with three frits in one end of the tube. These frits ranged from coarsest to finest, with the coarsest located nearest the end of the quartz tube. In theory this sampler would promote salt flow through the frits due to the coarsest frit posing the least significant barrier to salt flow because of its possibly lower surface tension. As the salt flows thought the coarser frit, it may better overcome the surface tension at the surface of the finer frit, resulting in easier fluid flow. A pressure differential may make this design choice redundant or show it to be completely ineffective, however it was a low effort and low-cost addition to this sampling work and deemed worth investigating.

The quartz frits selected for this work were unchanged from the original set of experiments. Quartz frits ranging from 160-250 micron to 5-10 micron porosity were fused inside of quartz sampler tubes and used in several experiments. Upon close investigation after observing the fused frits with a microscope, the surface finish of each frit seemed to be less uniform than expected. On a cursory inspection of each new sampler via a microscope, it appeared that samplers of the same frit size fell into one of two categories: one where the surface of the frit was relatively flat with small, deep cracks between flat portions and another where the surface is uniform in roughness. The different groups were marked appropriately and may be evaluated whenever possible for differences in sampling performance.

Retrieving salt samples from within the quartz sampler tubes will be done using a small fixture equipped with a glass tubing cutter and a trough. The glass tubing cutter consists of an extremely hard cutting wheel which scores the exterior of whatever tube is rotated along it, allowing for the tube to be broken along the scored perimeter. The trough is at the height of the glass tubing cutter, allowing for the long quartz sampler tubes to rest in the trough and also line up accurately for the glass tubing cutter to be effective. This combination of tools has been mounted to rigid base and sent into HFEF's main hot cell for future use with filtered salt sampling work. It is expected that salt samples will release from the quartz tube easily due to contracting during the cooling process after being cut as well, which is a result of the thermal expansion of the salt being much higher than the thermal expansion of the quartz sampler tube.

4. Experiments and Observations

Two different methods of pulling salt up through the frits into the sample tube were tested. In the first set of tests two simple methods in addition to simply dipping the sampler into the salt were tested. The first method involved lowering the sampler into the molten salt, allowing some time for it to come to temperature, capping the top of the tube with the glovebox glove and removing the sampler. The next method used a simple rubber pipette bulb attached with a hose clamp to the quartz sample tube. The second set tested the use of a small piston assembly (discussed in Section 3) to provide the vacuum. All tests were performed in the Molten Salt Furnace-IV in an argon atmosphere glovebox in the Engineering Development Laboratory using LiCl-KCl with ~ 5 wt% $GdCl_3$ salt at approximately 500 °C. Four different arrangements of quartz frits were used in the tests. There were three different frit porosities used: 160 - 250 µm (0-1 & 0-2), 16 - 40 µm (3-1 & 3-2), and 5 - 10 µm (VF-1 & VF-2), as well as all three stacked with the coarsest frit at the bottom, closest to the opening of the tube (ST-1 & ST-2).

4.1 Filtered Salt Sampler: Capped and Pipette Bulb

The first series of tests involved three different methods. The first method was what had been done in previous work, lowering the sampler down into the salt bath, allowing hydrostatic head to push salt through the frit(s) and pulling the sampler up out of the bath after some time for temperature to equilibrate. The second method involved lower the sampler into the salt, allowing hydrostatic head to push salt through the frit(s) for some equilibration time and then manually capping the top of the tube with the glovebox glove before pulling the sampler from the bath. In the third method, a rubber pipette

bulb was placed over the top of the sampler and squeezed prior to lowering into the salt (Figure 2). After some time to equilibrate, the bulb was allowed to reinflate, creating a vacuum in the sampler, and pulling salt up through the frit(s). A summary of the tests performed along with qualitative results is shown in Table 1. An example of a salt sample taken during this series of tests is shown in Figure 3.

Figure 2. Rubber pipette bulb attached to quartz tube sampler.

Table 1. Summary of filtered salt sampler tests performed with and without cap and pipette bulb.

Test #	**Sampler # / Frit Size**	**Cap/Bulb**	**Time Held**	**Results**
1	3-1 / 16 - 40 µm	None	1 min	No salt in tube
2	3-1 / 16 - 40 µm	None	4 min	Salt in tube (17 mm) No drip upon removal 0.919 g sample size
3	0-1 / 160 - 250 µm	None	4 min	Salt in tube Salt dripped out on removal
4	0-1 / 160 - 250 µm	Cap	4 min	Salt in tube (50 mm) No drip on removal w/cap 3.083 g sample
5	VF-1 / 5 - 10 µm	None	4 min	No salt in tube
6	VF-1 / 5 - 10 µm	Bulb	4 min	Salt in tube

7	VF-1 / 5 - 10 µm	Bulb	10 min	Salt in tube (20 mm) No drip on removal 1.079 g sample
8	VF-2 / 5 - 10 µm	Bulb	10 min	No salt in tube
9	ST-1 / 5 – 10, 16 – 40, and 160 – 250 µm	None	4 min	Salt in tube (3 mm) 0.442 g sample
10	ST-1 / 5 – 10, 16 – 40, and 160 – 250 µm	None	5 min	Salt in tube (10 mm) 0.849 g sample
11	ST-1 / 5 – 10, 16 – 40, and 160 – 250 µm	Bulb	5 min	No salt in tube

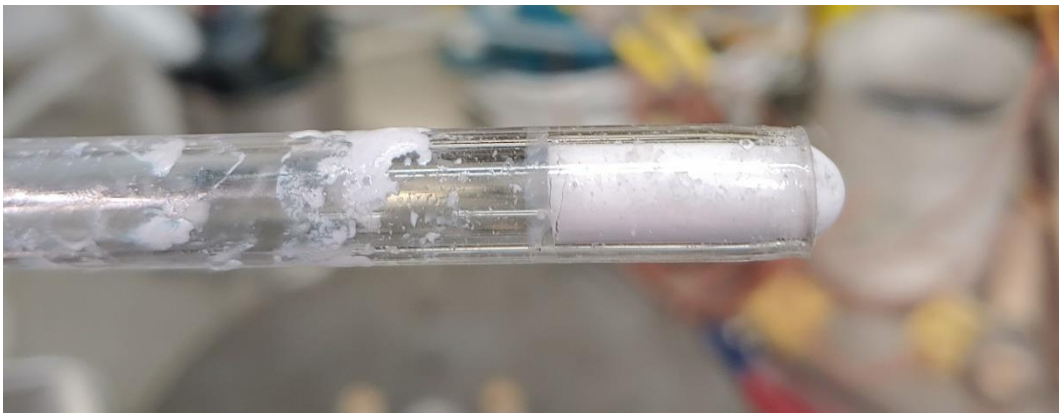

Figure 3. Salt sample taken with the filtered salt sampler and rubber pipette bulb.

As can be seen in the summary Table 1, there was a large amount of variability in the tests performed with the pipette bulb. At times, a salt sample was successfully taken, while at other times with similar conditions, a sample was unable to be procured. It was determined that the seal between the quartz tube sampler and the pipette bulb could not be reliably and repeatably formed. There was, at times, leakage causing no salt to be pulled into the sampler. Because of this a more robust method for pulling a vacuum on the sampler tubes was developed as discussed in Section 3.

4.2 Filtered Salt Sampler: Piston Vacuum Assembly

The second series of tests was performed to test the piston vacuum assembly presented in Section 3. In all of these tests, the sampler was lowered into the molten salt bath and allowed some time to equilibrate prior to attaching the piston assembly and applying vacuum to pull salt through the frit(s) into the sampler. In all but the final test approximately half of the piston volume was used for sampling. The sampler with piston vacuum assembly is shown inserted into the furnace in Figure 4. A summary of the

tests, along with qualitative results, is shown in Table 2. A photo of a sample taken with the piston vacuum assembly prior to the salt freezing is shown in Figure 5.

Figure 4. Filtered salt sampler with piston vacuum assembly in Molten Salt Furnace.

Table 2. Summary of filtered salt sampler tests performed with and without piston vacuum assembly.

Test #	**Sampler # / Frit Size**	**Method**	**Time Held**	**Results**
1	0-1 / 160 - 250 µm	Cap	10 min	Salt inside Majority of salt rapidly flowed back out after cap was removed
2	3-1 / 16 - 40 µm	Cap	10 min	Salt inside Salt remained after removing cap (freezing?)

3	VF-1 / 5 - 10 µm	Piston	13 min	Very small amount of salt inside
4	VF-1 / 5 - 10 µm	Piston	10 min / 5 min after piston activation	Salt inside (10 mm)
5	VF-1 / 5 - 10 µm	Piston	10 min / 5 min after piston activation	Salt inside (25 mm)
6	VF-2 / 5 - 10 µm	Piston	10 min / 5 min after piston activation	Salt inside (6 mm)
7	VF-2 / 5 - 10 µm	Piston	10 min / 5 min after piston activation	Salt inside (10 mm)
8	VF-2 / 5 - 10 µm	Cap	10 min	Salt inside (5 mm)
9	ST-2 / 5 – 10, 16 – 40, and 160 – 250 µm	Cap	10 min	No salt in tube
10	ST-2 / 5 – 10, 16 – 40, and 160 – 250 µm	Piston	10 min / 5 min after piston activation	Salt in tube (13 mm)
11	0-2 / 160 - 250 µm	Piston (full volume)	10 min	Salt in tube

Figure 5. Molten salt sample taken with piston vacuum assembly.

As seen in the results in Table 2, a successful method using the piston vacuum assembly was developed. The piston assembly was able to reliably and repeatably pull salt through even the finest (5 - 10 µm) and stacked frits (160 – 250, 16 – 40, and 5 - 10 µm). Based on the success of these tests, the assembly was modified slightly for ease of use with manipulators and sent into the HFEF hot cell for testing in a kilogram-scale used fuel electrorefiner (Figure 6).

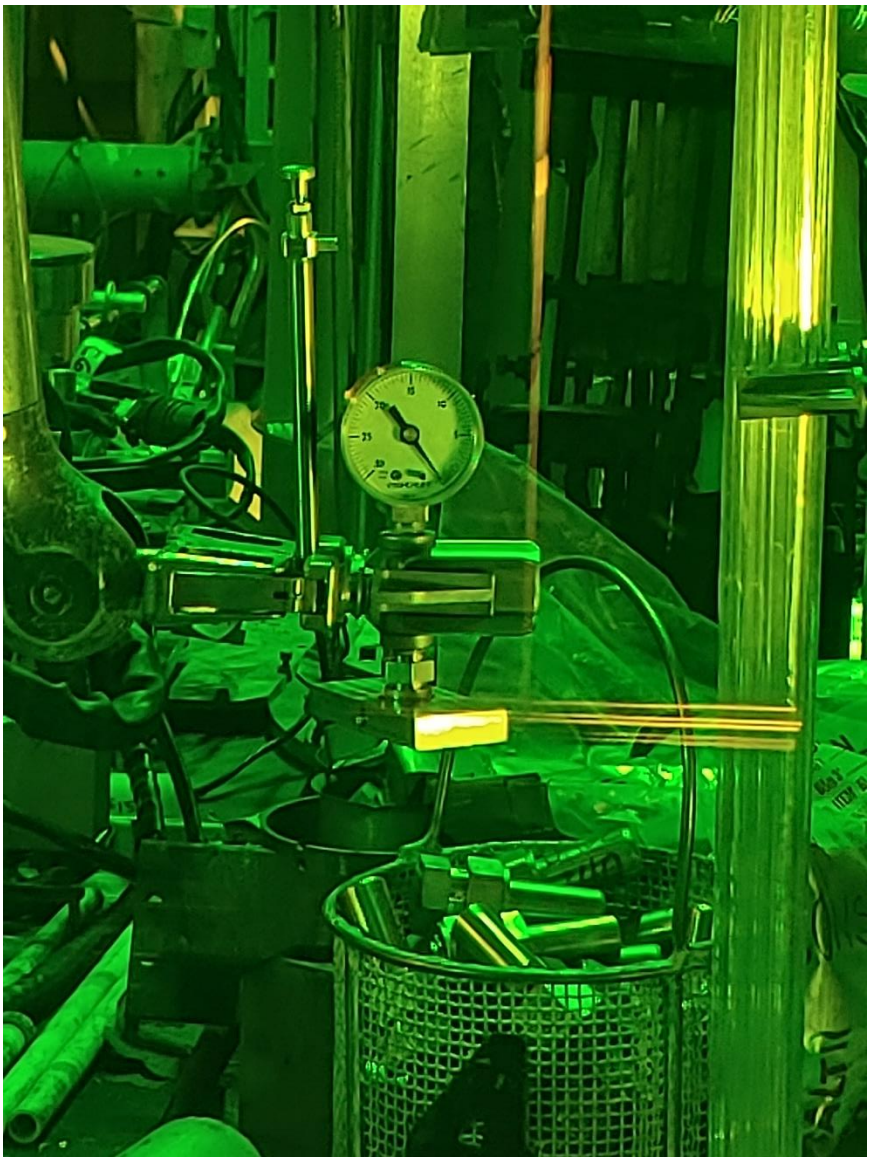

Figure 6. Piston vacuum filtered salt sampler in HFEF hot cell for testing with used electrorefiner salt.

5. Summary

Several different methods of applying vacuum as part of a filtered salt sampling technique were tested using a LiCl-KCl-$GdCl_3$ molten salt at 500 °C. A piston vacuum assembly was designed and reliably and repeatably used to take salt samples through multiple different porous quartz frit sizes (as small as 5 – 10 µm). This design has been modified slightly and sent into the HFEF hot cell for upcoming testing with used fuel electrorefiner salt.

6. Future Work

Filtered salt samplers have been sent into the HFEF hot cell for upcoming testing with spent fuel electrorefiner salt. These samplers will be tested alongside the baseline sampling technique (cold finger

dip), analyzed for elemental and isotopic contents at the Analytical Research Laboratory at INL, and compared. Additional work remains on how best to collect the salt sample from the quartz tube, minimizing contamination of salt from the exterior of the sample tubes. Additionally, future work should involve testing with surrogate insoluble impurities of known particle sizes in non-radiological salt to determine the samplers' effectiveness at separating salt and salt soluble constituents from insoluble solid particles in the salt bath.