Contract No:

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Extraction of Mercury Utilizing 3D Printed Media – Mercury Extraction Coalescer (MEC)

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Overview & Relevance

This project was undertaken to address the concern of mercury species present in chemical reprocessing facilities and environmental remediation sites. Typical elemental mercury removal is performed by steam stripping. As efficient as this process is, the vaporized elemental mercury is often condensed along with acidic condensate. The acidic conditions re-dissolve the elemental mercury, transferring it back into the aqueous stream, and undoing the energy-intensive steam stripping process.

This project looked to design and produce a 3D printed component to act as a mercury vapor/liquid coalescer utilizing structural and chemical features to segregate elemental mercury from an aqueous solution or process offgas and improving elemental mercury retention and removal from the recirculated condensate stream.

The aim of this project is to take inspiration from nature, for example the lotus leaf, and the advantage of advanced manufacturing techniques, in this case 3D printing, to produce a functional solution to a serious environmental issue, the accumulation of elemental mercury in aqueous environments.

Project Timeframe

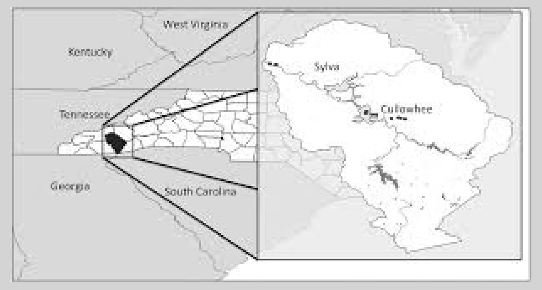
- Project start date: Oct 2018
- Project end date: Sept 2019
- Percent Complete: ~75%

Budget (as of 9/13/2019)

- Total project funding: $190k
- Funding received in FY18: $21k
- Funding received in FY19: $144k
- Encumbered Funds: $30k
- Total project spending: $175k (92%)

Partners

- Dr. Carmen Huffman with Western Carolina University in Cullowhee, North Carolina



Overall Objectives:

- Objective 1
  - Develop a 3D printed component with a tuned surface to act as a segregating coalescer for elemental mercury
- Objective 2
  - Integrate a 3D printed component into current chemical reprocessing and environmental remediation testing equipment with minimal impact to current testing procedures

Barriers Addressed:

- Printing Media
  - To achieve sufficiently fine detail along with flexibility of composition, the correct 3D printing technique and material had to be selected. After examining available printing technology and example components, a printer utilizing UV-curable resin that prints using stereolithography was selected (FormLabs Form2).

- Integrable Design
  - Current testing apparatus were utilized as design parameters so that 3D printed components could be designed is such as way as to be integrated into testing processes with minimal redesign of equipment. Large scale facilities often do not have the ability to redesign entire process lines; therefore, designs that are easily integrated are integral to the adoption of enhancements.
- Testing Protocols
  - The steam stripping process of elemental mercury from an aqueous solution is very simple, theoretically. In reality, significant work must be done to ensure that the mercury is sufficiently dispersed as small particles near the surface of the liquid. With this in mind, initial simple testing had to be abandoned and replaced with more advanced testing protocols. To maximize efforts, testing for mercury stripping was combined with testing of new antifoams given the facts that the equipment and chemicals required were the same and the two tests could be performed concurrently without interference.

Approach

Advanced Structures

Various physical geometries were designed proceeding from the simplistic to more and more advanced.

Initial designs aimed at taking advantage of the simple differences in physical properties of elemental mercury and aqueous solutions such as density and surface tension. Structures were designed to physically segregate the liquids as they coalesced from the vapor stream.

Design 1 (Multi-turn Insert) utilized an overflow pathway that would allow the heavier mercury to settle-out in the component and the lighter aqueous solution to flow over the barriers and proceed onward toward condensate collection.

Design 2 (Centrifugal Insert) utilized a spiral path with channels built into the walls that would segregate the heavier mercury as it moved to the outer portion of the curved path.

Figure 1: Design 1, the Multi-Turn Insert

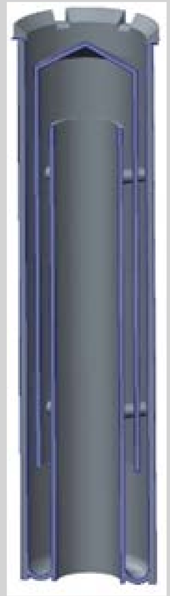


Figure 2: Design 2, the Centrifugal Insert

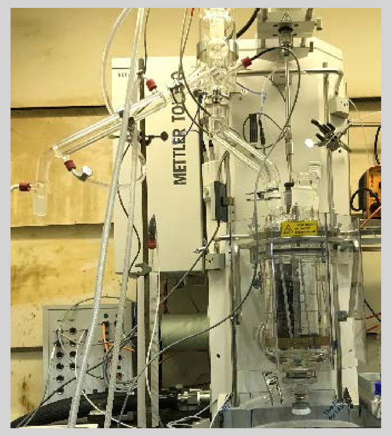


Figure 3: the chemical processing equipment to be integrated with

Testing Integration

The original designs were developed as cylindrical inserts. This shape is the most integrable due to the abundance of process pipes that exist as connectors between processing units.

For chemical reprocessing integration, the components were designed to be inserted between the offgas stream condenser and the offgas condensate collection vessel (known as the Mercury Water Wash Tank, or MWWT). The insert would act as a segregation point to prevent elemental mercury from sitting in contact with acidic condensate.

For environmental testing, the components were sectioned and processed alongside current aqueous remediation media. The materials would be equilibrated in a “contaminated” solution and then evaluated for uptake of the target species.

Technical Progress

Successful Printed Coalescers in Multiple Materials

A number of increasingly complex designs were successfully printed.

The first designs (Multi-Turn Insert and Centrifugal Insert, Figure 4) were printed in both 3in. and 1in. diameter configurations for integration into different equipment adapters (Figure 5). Successive designs (also shown in Figure 4) began to stretch toward the bounds of achievable geometries and scales.

Coalescers were printed in two different materials via the same printing process, showcasing the flexibility of the manufacturing technique to produce a variety of components with minimal adjustments.

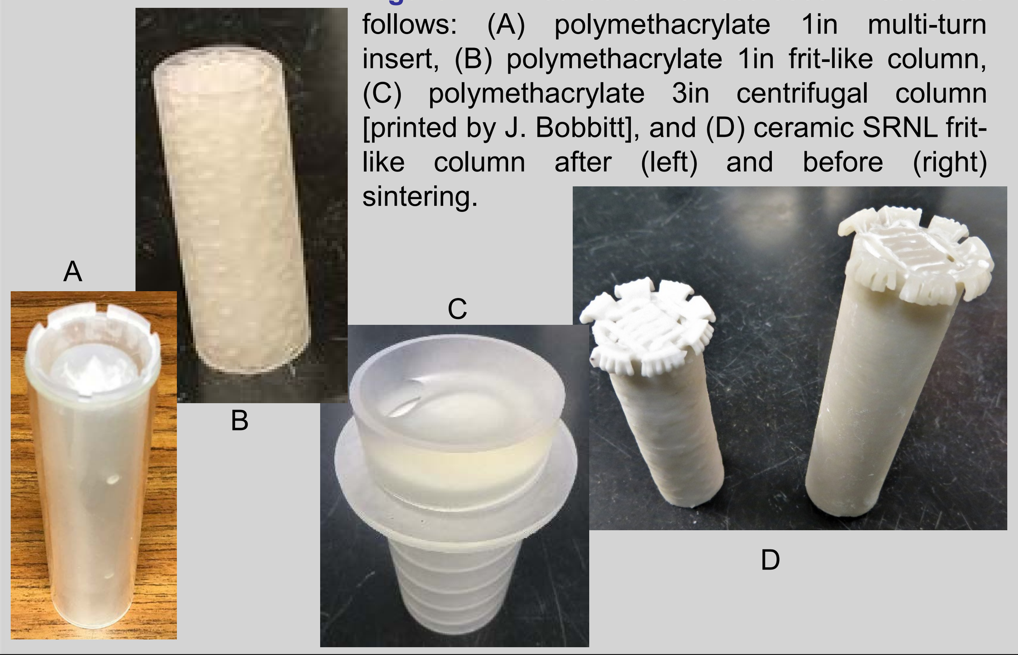


Figure 4: A collection of coalescer inserts as follows: (A) polymethacrylate 1in multi-turn insert, (B) polymethacrylate 1in frit-like column, (C) polymethacrylate 3in centrifugal column [printed by J. Bobbitt], and (D) ceramic SRNL frit-like column after (left) and before (right) sintering.

Equipment Integration with Minimal Testing Interruption

Figure 5: The incorporation of both a 3in. (left) and 1in. (right) insert between condenser connections (tops) and MWWTs (bottoms) through Teflon bushings.

We were able to perform a simple redesign of the connections between two pieces of testing glassware from the chemical reprocessing rig (Figure 5) to generate a space capable of housing the coalescer. The redesign of the glassware setup more closely mimics the process piping that would be located in the large scale facility, further supporting the ease of facility integration.

Initial testing with other environmental remediation techniques was performed by Dr. Carmen Huffman and her research group at Western Carolina University (shown in Figure 6).

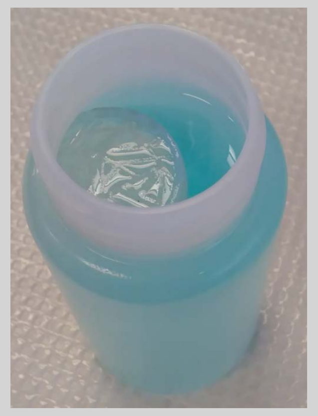


Figure 6. Sectioned piece of SRNL Frit-like column placed in copper (II) chloride pH 4 buffered solution.

Proof-of-Concept Testing

Testing with a 1in Multi-Turn insert was performed utilizing the newly acquired Mettler Toledo RC1 reaction calorimeter (Figure 3). Chemical reprocessing was performed to compare the system performance to the standard custom glassware testing rig historically utilized.

The testing was performed during day-only operations with abbreviated processing steps. Shortening the process resulted in an overall reduction in the amount of mercury steam stripped from the reaction vessel (due solely to time at boiling), but did not affect the chemistry or materials generated to the offgas stream.

The impact of the coalescer on the process was negligible except in the final location of elemental mercury in the equipment. Whereas mercury will typically be located in the MWWT, the condensate collection vessel, or remaining in the reaction vessel; in this case, a significant fraction of the mercury was extracted into the coalescer as well. When testing concluded, the coalescer was removed from the insert and the adsorbed/collected mercury was dissolved and analyzed via inductively coupled plasma atomic emission spectroscopy (ICP-AES); results in Table 1 and images of the coalescer post-dissolution in Figure 7.

Table 1. Concentration of Mercury Present Adsorbed/Collected by MEC

Multi-Turn Insert MEC	
Mass of Hg (g) collected in MEC	1.55 g
Fraction of total Hg from test (%)	23.8 wt.%

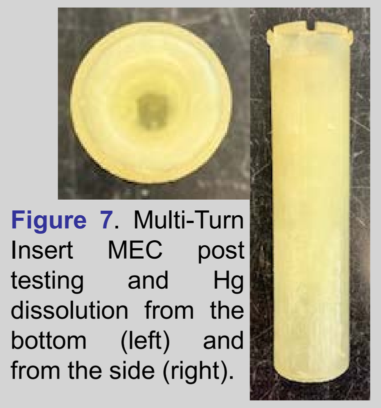


Figure 7. Multi-Turn Insert MEC post testing and Hg dissolution from the bottom (left) and from the side (right).

Collaborations

Savannah River National Laboratory

- Environmental Stewardship
- Mechanical Systems and Custom Equipment Development

Western Carolina University

- Subcontract to SRNL
- Dr. Carmen Huffman
- Utilization of 3D printed components in non-radioactive environmental remediation studies

Remaining Challenges and Barriers

- **Challenge 1: More complex microstructures**

To more closely mimic natural structures (i.e. lotus leaf), we will have to push the bounds of how the 3D printers are designed to perform. Advances are being made quickly to improve printing technologies and media and we will have to take advantage.

- **Challenge 2: Testing scheduling**

Given the required components to perform testing with mercury, scheduling testing that is both efficient and cost-effective demands collaboration with other projects performing co-located experiments. The tests can be performed in parallel with no interference, but must be coordinated into the larger overall schedule of testing.

Proposed Future Work

- **Future Task 1: Reactive printed components**

As evidenced by the ceramic resin, materials may be incorporated into the printing media and, therefore, the final printed component. We will develop reactive materials to incorporate into the resin to improve the selective chelation of mercury from the aqueous reaction solutions.

- **Future Task 2: Advanced environmental remediation testing**

The long-term goal of this project is to develop a methodology for the removal of mercury and other heavy metals, not just from chemical reprocessing facilities like the Savannah River Site, but to be integrable to general environmental remediation sites. Continued work with collaborators on incorporation into that testing would be ideal.

Project Summary

- **3D printed components were designed and developed targeting the segregation of elemental mercury from aqueous media.**
- **Increasingly complex designs were successfully printed from multiple materials.**
- **Integration of 3D printed components was accomplished with minimal impact to current testing methodologies and equipment.**
- **The 3D printed component was shown to significantly separate elemental mercury from the targeted offgas/condensate stream.**

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