Recommendations on the Ultrasonic Spectroscopy System for Deployment at Hanford's AY-102 Tank

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Executive Summary

As the DOE's Hanford site begins preparations for the transfer of high-level radioactive waste (HLW) from the double-shell tanks (DST) to the Waste Treatment and Immobilization Plant (WTP), the influence of waste feed consistency on the waste stabilization process – and final stabilized waste form - is currently under analysis. In order to characterize feed consistency prior to transfer, a suite of instrumentation will be required to monitor the waste preparation and mixing process in real time. FIU has focused its instrumentation efforts on identifying improvements to the in-situ, near-real time monitoring of the mixing process. Specifically, this project has identified innovative technologies applicable for in-tank monitoring during the mixing process.

After the current technology baseline plan and previous research efforts were reviewed, FIU performed an extensive literature and technology search for applicable systems that could provide waste parameters within the HLW tank environment. The literature search focused on available methodologies for in-situ analysis of slurries, emulsions and suspensions applied in all industries. In particular, the monitoring of bulk density and/or particle concentration/characteristics measurements was the focus of the search. For the technology search, vendors of applicable techniques were identified and contacted. The searches resulted in several academic, commercial and governmental reports/articles applicable to the monitoring needs of the HLW tanks. Instrumentation specifications were collected and reviewed to identify the technology capabilities and limitations. These capabilities were also used for a comparative analysis between technologies. Based on the information, all the technologies were down selected to five applicable systems/methods that could provide useful information if deployed in a HLW tank. The five applicable methods applicable to the in-situ monitoring of the waste feed consistency were focused beam reflectance measurement (FBRM), optical back-reflectance measurement (ORM), ultrasonic spectroscopy (USS), Lamb/Stoneley wave viscosity measurement, and vibration-based densitometers. Based on literature results and commercial options, the ultrasonic and vibration-based techniques showed the most promise for developing a technology that could be used for in-situ measurements within the aggressive environment of a HLW tank. Specifically, the vibration-based densitometers and USS systems could provide information on the density and concentrations of the mixed slurry. These techniques had the potential to be engineered for monitoring at various depths within the tank.

In order to evaluate a technology, the USS was tested on an array of slurries, and the density compared to a commercially available density meter that utilizes an accurate and repeatable technique for density measurement. A setup was conceived that allowed the testing of the USS and a commercially-available Coriolis mass flow meter side-by-side. The setup used a 10-gallon tank for agitation of the simulated slurry mixtures, with both systems sampling at the same location; the USS system probe was lowered into the tank, while a pick-up tube was used to transfer mixture to the Coriolis meter. In addition, the USS was subjected to several tests using a benchtop setup for more controlled evaluations of its concentration tracking capabilities. Both these systems were subjected to solutions and suspensions that would simulate some of the physical and rheological properties of the HLW slurry found in AY-102. The simulated slurries
consisted of one to three distinct solids suspended in a water or NaNO₃ solution as the supernatant.

FIU tested the USS between May and October of the previous year with delays associated with hardware and software issues that plagued the device under evaluation. In August, the system had to be returned to the vendor as a result of a hardware fault. A new system was sent to FIU in October, and was used to complete testing. The results indicated that the technology could provide a measurement of density, but the frequency at which it occurs varies with slurry characteristics. If one frequency is selected for analysis, the density values vary more than 10% when compared to the reference value. One major issue that occurred during testing was that the spectral response profile of the USS system changed between hardware changes, and this was something that the vendor attributed to an issue with the reference file used for the tests.

In order to address issues with system inconsistency, additional bench-scale tests were performed at ITS facilities during the first quarter of this performance period. The testing utilized similar materials, concentration profiles for NaNO₃ and solids loading profiles in an attempt to determine the root cause for the inconsistency of results, as well as to compare the results to the FIU data. The USS utilized consisted of a through-transmission configuration, which had a slightly larger transducer than those tested at FIU. The ITS test results indicated good agreement between the measured and calculated densities for solutions, and certain material and water/NaNO₃ suspension. The technology showed significant variance between the measured and calculated densities for complex mixtures (multiple solids). This variance was again attributed to the significant attenuation of the acoustic pulse after traversing the suspension. This attenuation caused issues in the detection of the correct return echo from the suspensions. With incorrect detection, the measured density value showed significant errors (> 20%).

Based on the results of the testing at ITS facilities, it is evident that the technology still requires the following: applied research in the design and selection of the necessary transducers to provide the needed sensitivity; additional engineering analysis to improve signal acquisition and detection at low signal levels; and basic research into the influence of complex mixtures that contain matter that displays an inverse relationship between speed of sound and concentration. With these limitations, the technology is not at the readiness level required for deployment at the WTP waste feed mixing tank AY-102. The USS has several technical limitations that must be addressed before system would be at a TRL readiness level commensurate with a potential deployment in 2-3 years.

**Background**

The US Department of Energy's (DOE) Hanford site is completing development of the Waste Treatment and Immobilization Plant (WTP), and preparing for the processing of over 54 million gallons of high-level radioactive waste (HLW). This waste shall be mixed within storage tanks, transferred to staging tanks, and processed by the WTP for final disposal. As the WTP development is completed, the window to integrate additional instrumented systems into the process loop will close. One such area that could benefit from additional instrumentation is the
HLW tanks where this waste is currently stored. These +1M gallons of waste must be mixed into slurry that can be retrieved via pipelines to other tanks, or to WTP. The ability to characterize slurry in-tank would greatly optimize the mixing and retrieval processes. After several years of experience in the development of remote monitors for HLW tanks, FIU has undertaken a task to evaluate additional characterization and monitoring technologies that could be scaled-up for deployment inside the HLW tanks at Hanford.

During the 2010 performance period, FIU conducted a literature search for promising technologies that could be utilized in-situ and provide process engineers with near real-time information on slurry parameters during the mixing process. In particular, technologies that could monitor the slurry bulk density were highlighted and reviewed via addition literature search and screening. The search identified two technologies/methodologies that could be deployed in a HLW tank, and could provide useful bulk density information at various depths in the tank. These included a Coriolis-principle based densitometer, and an Ultrasonic Spectroscopy-based technology (USS). The Coriolis meter utilized was a commercially available Promass 63F by Endress-Hauser, and the USS was a commercially available system from Industrial Tomography Systems plc (ITS). FIU evaluated both these technologies in a laboratory setting with slurry simulants. The technology results were inconclusive; the USS system(s) showed varying results for the same slurry characteristics. These variations were attributed to a system performance issue. In order to address these variations, an additional round of testing was performed at ITS on a bench-scale setup with more controlled suspensions. This report summarizes the tests performed as well as addresses a timeline for data analysis.

**Experimental Approach**

In order to mitigate system issues from affecting the test success, it was determined that the tests should be performed at ITS facilities. The vendor would be capable of addressing any system issues during testing, and would follow the test requirements and goals defined by FIU. In order to collect sufficient information to ensure that the technology could proceed into a prototype development stage, the tests were divided into two phases: a (1) NaNO₃ concentration ladder phase, and a (2) solids loading ladders tests with different materials. The first phase involved concentration ladder of aqueous NaNO₃ solutions. A mixture was prepared and static (no mixing) measurements were taken in the cell for the following concentrations: 0.1%, 0.5%, 1%, 2%, 3%, 5%, 7%, 10%, 15%, 20%, 25%, 30%, and 37.5%. For the second phase, distilled water was used as the continuous phase in aluminum hydroxide, zirconium oxide and stainless steel mixtures and further on the ultrasonic measurements of the mixtures. Measurements were taken for the following concentrations (by volume): 0.1%, 0.5%, 1%, 2%, 3%, 5%, 7%, 10%, 15%, 20%, and 25%.

In this test program, the USS was configured in a through-transmission mode. This would address the expected large attenuation caused by the solids utilized. It is expected that a field deployable system will have to utilize a through-transmission system in order to address the
large attenuation expected in Hanford’s HLW tanks\(^1\). All measurements were carried out using a USS in-line measurement cell in the static and dynamic conditions. The experiments consisted of 10 repeated measurements for each sample. The sample was then removed from the cell for disposal and new material was added. In the case of sodium nitrate concentration ladder, the spacing between transducers was set to 22-mm. That spacing was reduced to 10-mm for suspensions phase. The system was calibrated using deionized water as a reference media. Additional parameters of the experiment: room temperature, the volume of the sample 350 ml.

**Results**

The test report provided by ITS is presented in Appendix A. Analysis of the NaNO\(_3\) solutions show good agreement with the expected wave speed, and agrees with the data collected during FIU testing. The attenuation data shows that these solutions experience less attenuation than a water mixture. Analysis of the density measurement for the Aluminum Hydroxide solid loads indicates good agreement with the calculated value, even into the 30% concentration. The measurements collected from the suspensions of Zirconium oxide in water showed the inversely proportional velocity to the concentration of the sample. The complex mixture data showed significant variations in the density as compared to the calculated value, which indicates significant attenuation of the ultrasonic pulse.

**Conclusion & Path Forward**

The results from this final phase of testing indicates that several areas of the technology still require substantive applied research in order to overcome current limitations. The technology holds potential as a simplified bulk density and concentration method, but it requires additional work in order to overcome current commercial design limitations. There are three areas that require more applied research to overcome the limitation. First, the design and selection of the necessary transducers to provide the needed sensitivity must be reviewed; the benefits of advanced piezo-composite transducers can improve signal sensitivity and fidelity. Second, additional engineering analysis must be performed to improve signal acquisition and detection at low signal levels. The utilization of higher sensitivity acquisition system, as well as better control of thermal voltage levels can improve minimum detectable signal level. Finally, basic information on the complex mixtures, especially those that contain matter that displays an inverse relationship between speed of sound and concentration, must be investigated. The attenuation characteristics at varying spectra of the expected waste stream must be properly considered when evaluating the response to a broadband ultrasonic pulse.

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Appendix A

ITS Final Test Report submitted to FIU