

**Contract No:**

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy.

**Disclaimer:**

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied: 1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or 2. representation that such use or results of such use would not infringe privately owned rights; or 3. endorsement or recommendation of any specifically identified commercial product, process, or service. Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

# Effects of Oxygen and Air Mixing on Void Fractions in a Large Scale System

*Robert A. Leishear, Savannah River National Laboratory, Aiken, SC*

*Michael L. Restivo, Savannah River National Laboratory, Aiken, SC*

*Hector N. Guerrero, Savannah River National Laboratory, Aiken, SC*

*David J. Sherwood, Hanford Waste Treatment & Immobilization Plant Project,  
Richland, Wa.*

## Abstract

Oxygen and air mixing with spargers was performed in a 30 foot tall by 30 inch diameter column, to investigate mass transfer as air sparged up through the column and removed saturated oxygen from solution. The mixing techniques required to support this research are the focus of this paper. The fluids tested included water, water with an anti-foam agent (AFA), and a high, solids content, Bingham plastic, nuclear waste simulant with AFA, referred to as AZ01 simulant, which is non-radioactive. Mixing of fluids in the column was performed using a recirculation system and an air sparger. The re-circulation system consisted of the column, a re-circulating pump, and associated piping. The air sparger was fabricated from a two inch diameter pipe concentrically installed in the column and open near the bottom of the column. The column contents were slowly re-circulated while fluids were mixed with the air sparger. Samples were rheologically tested to ensure effective mixing, as required. Once the fluids were adequately mixed, oxygen was homogeneously added through the re-circulation loop using a sintered metal oxygen sparger followed by a static mixer. Then the air sparger was re-actuated to remove oxygen from solution as air bubbled up through solution. To monitor mixing effectiveness several variables were monitored, which included flow rates, oxygen concentration, differential pressures along the column height, fluid levels, and void fractions, which are defined as the percent of dissolved gas divided by the total volume of gas and liquid. Research showed that mixing was uniform for water and water with AFA, but mixing for the AZ101 fluid was far more complex. Although mixing of AZ101 was uniform throughout most of the column, gas entrapment and settling of solids significantly affected test results. The detailed test results presented here provide some insight into the complexities of mixing and void fractions for different fluids and how the mixing process itself affects void fractions in Bingham plastic fluids, which have a measurable yield stress.

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy. This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied: 1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or 2. representation that such use or results of such use would not infringe privately owned rights; or 3. endorsement or recommendation of any specifically identified commercial product, process, or service. Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

## Introduction

This paper focuses on void fractions and mixing of fluids in an 8.4 meter tall by 0.76 meter diameter bubble column. This paper is the last of three papers which describe mass transfer tests performed in this system, using various fluids<sup>1,2,3</sup>. Void fraction data was obtained for 24 different test conditions, at different fill levels in the bubble column. Superficial sparging velocities of 2, 5, and 10 mm/second were applied through the central sparger to each of the simulants at three different column fill levels, where the superficial velocity is defined as the average volumetric flow rate divided by the liquid surface area in the column. The primary fluid of concern was a Bingham plastic fluid, but mixing effectiveness was first demonstrated using Newtonian fluids, which were water and water with AFA added. Void fractions were one of the primary interests of this research, where void fractions are defined as the volume of gas divided by the total solution volume while bubbles rise in solution. This paper provides experimental void fraction data, and compares experimental results to theory. To do so, a system description is provided, followed by a discussion of mixing techniques, and a discussion of void fractions.

## System Description

The primary mixing system consists of a bubble column, a centrifugal pump, and associated piping, supported by auxiliary components to fill the column and aid in mixing. A simplified schematic of the system is shown in Figure 1, a complete system is shown in Figure 2, and the installed bubble column is shown in Figure 3. Detailed component descriptions are provided as required throughout the paper, material properties of the tested fluids are summarized in Table 1. The fluids included process water, process water mixed with anti-foam agent which was cloudy white in color (Dow Corning, Q2-3183, antifoam agent), and AZ101 simulant which was reddish brown in color and is a mixture of chemicals used to simulate nuclear waste, which also included anti-foam agent.

**Table 1: Fluid Properties**

| Fluid          | Yield Stress,<br>Pascals | Consistency /<br>Viscosity,<br>Centipoise |
|----------------|--------------------------|---|
| Water          | 0                        | 1   |
| Water with AFA | 0                        | 1   |
| AZ101 with AFA | 13                       | 11.5                                      |
| AZ101 with AFA | 30                       | 27.7                                      |

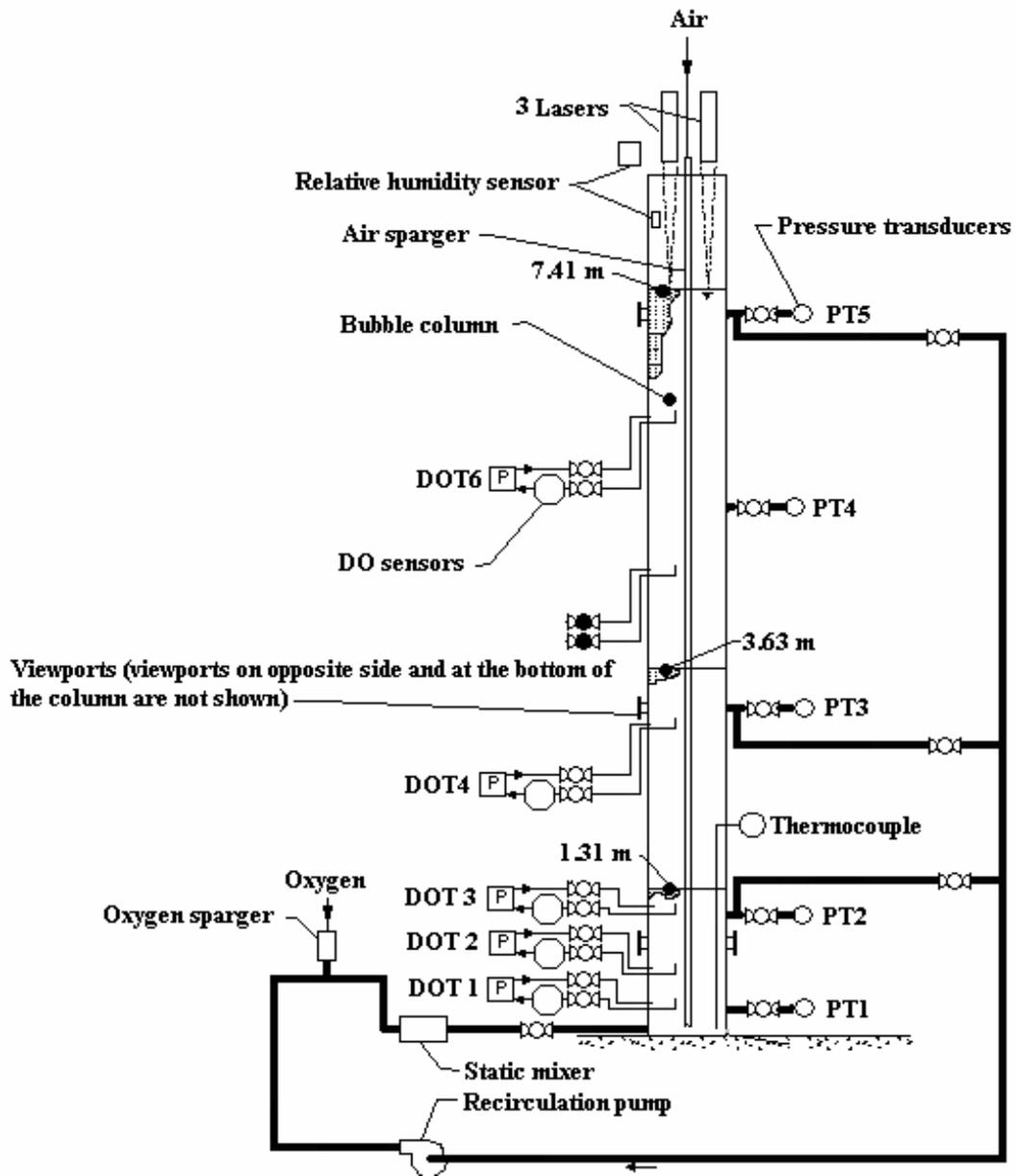
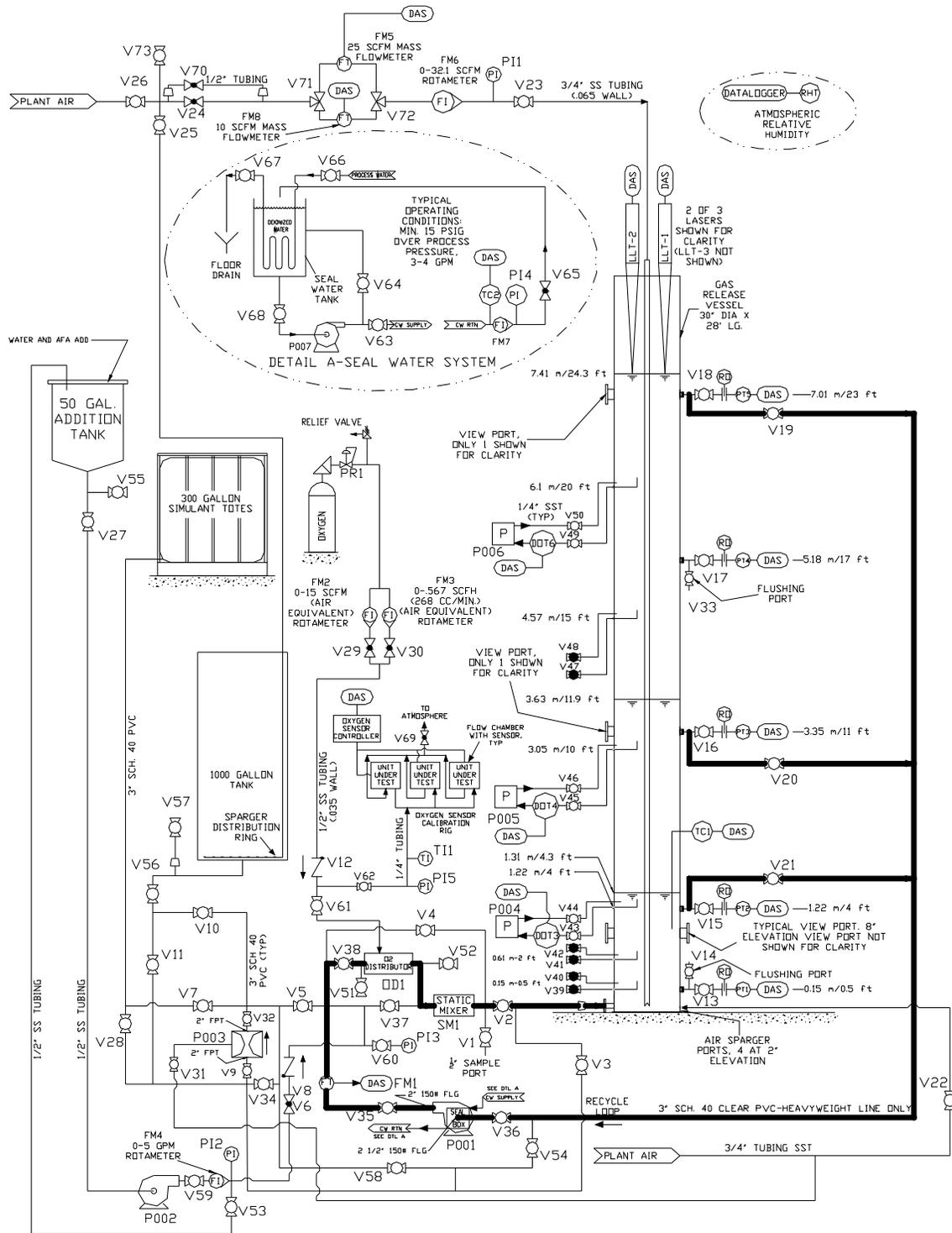


Figure 1: Simplified System Schematic<sup>1</sup>



ELECTRONIC P&ID, REVISED 8/15/2007

Figure 2: System Schematic<sup>3</sup>



**Figure 3: Bubble Column Design and Installation<sup>1</sup>**

## Mixing

There were four separate mixing processes required for these tests. One process added AFA to either water or AZ101 simulant as required. A second process thoroughly mixed the test fluids prior to testing. The third process saturated the test fluid with oxygen, and the fourth process removed oxygen from solution by sparging with air. Utilizing these processes, tests were performed at three different liquid levels in the bubble column, as shown in Figure 1 (1.31 meters, 3.63 meters, and 7.41 meters).

### Adding AFA

To effectively mix AFA into solution, a common practice of simply mixing the contents for a total of four volume changes was employed. Referring to the left hand side of Figure 1, the re-circulating pump, P002, was used to re-circulate the contents of the 50 gallon addition tank. Anti-foam agent (1.2 kg) was added to 27 gallons of water and re-circulated at 5 gpm to ensure that four volume changes occurred. Once the solution was well mixed in the addition tank, the AFA was added to the primary mixing system, which is shown in Figure 2 as the line shown as a heavy line. The re-circulating pump for this system operated at 100 gpm while the AFA was added at a uniform rate to the fully mixed test fluid, and the bubble column contents were then re-circulated for four volume turnovers after the AFA was added. When AFA was added to water, mixing effectiveness was easily verified. A transparent pipe is vertically attached to the side of the bubble column, and the column contents were mixed near the top of this column through this pipe just below the 7.41 meter column level, as shown in Figure 1. Visual inspection up the length of the transparent, PVC pipe verified uniform mixing, as shown in Figure 4. Once the AFA was mixed with water, the solution remained well mixed. Similarly, AFA was added to the opaque AZ101 solution, which is shown in Figure 4.



**Figure 4: Bubble Column and Transparent External Piping**

### ***Initial Mixing of AZ101 Simulant***

To ensure a homogeneous mixture before testing of the AZ101 simulant, effective mixing was required. To do so, the tank contents were re-circulated at 100 gpm for four volume turnovers. While the contents were re-circulating, the air sparger in the center of the column injected 20 scfm (10.46 scfm = 10 mm/second superficial velocity) of air and a circular sparging ring at the bottom the column (not shown) injected another 20 scfm of air. Effective mixing was verified by taking samples from the column at equal time increments, and then measuring the yield stress and consistency to ensure that properties were consistent between samples. Once an adequate mixing time was established, all further testing was preceded by mixing at the established time.

### ***Mixing of Oxygen into Solution***

Oxygen mixing was verified by measuring the dissolved oxygen concentration in solution, but mixing of oxygen in water helped verify the mixing process, since it was the only transparent fluid used in the tests. Oxygen was added to the water as it re-circulated at 100 gpm through the re-circulating pump and the column. Oxygen mixing was accomplished by using both a sintered metal filter which can create air bubbles down to 1 - 2 micron diameters and a helical static mixer which can create bubbles down to 20 – 30 microns. To ensure that air bubbles were as small as possible and that the bubbles were uniformly

distributed, these two components were used in series, as shown in Figure 1. Oxygen was added at 3 scfm, and mixing of oxygen in the three inch diameter pipe is shown in Figures 3 – 5. Once the oxygen entered the column from the three inch pipe, bubbles almost immediately coalesced and formed larger bubbles of ¼ to ½ inch in diameter near the bottom of the column, as shown in Figure 7. The bubbles then rose to the top of the column in approximately 30 seconds and became a more uniform size of 1/8 to ¼ inch in diameter as they rose.

The 3 scfm oxygen flow rate was optimized based on measurements of dissolved oxygen (DO) concentrations in the bubble column. Dissolved oxygen sensors were installed at various elevations, as shown in Figure 1, but only three sensors were available and they were moved to different locations for different tests. To validate mixing, the dissolved oxygen concentration was monitored at three different column levels until the DO concentration reached equilibrium at each of those levels. Typical test results are shown in Figure 8.

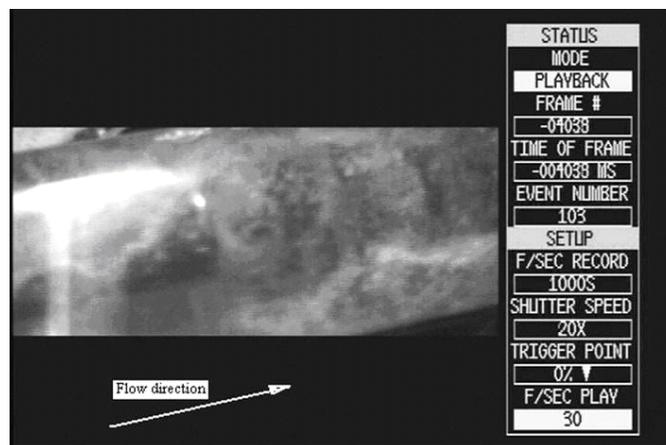


Figure 5: Bubbles Prior to the Static Mixer<sup>1</sup>

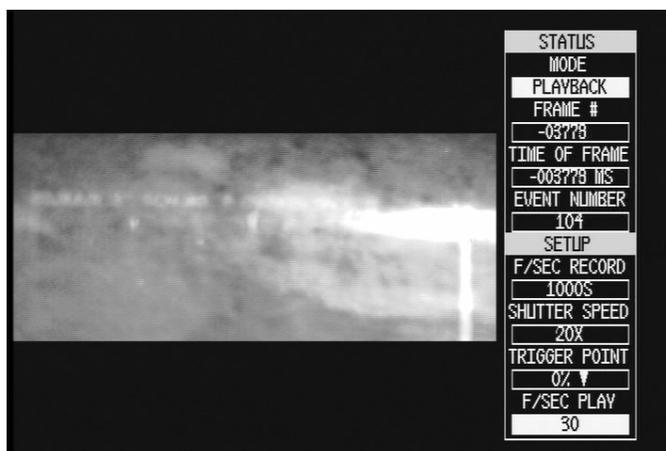
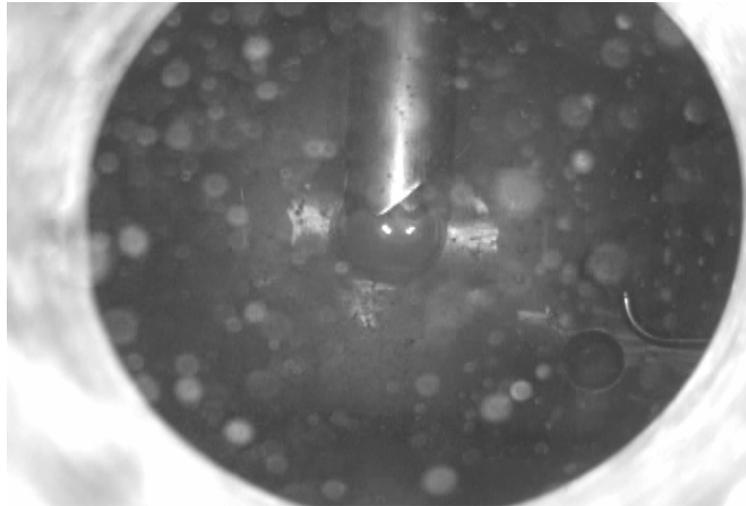
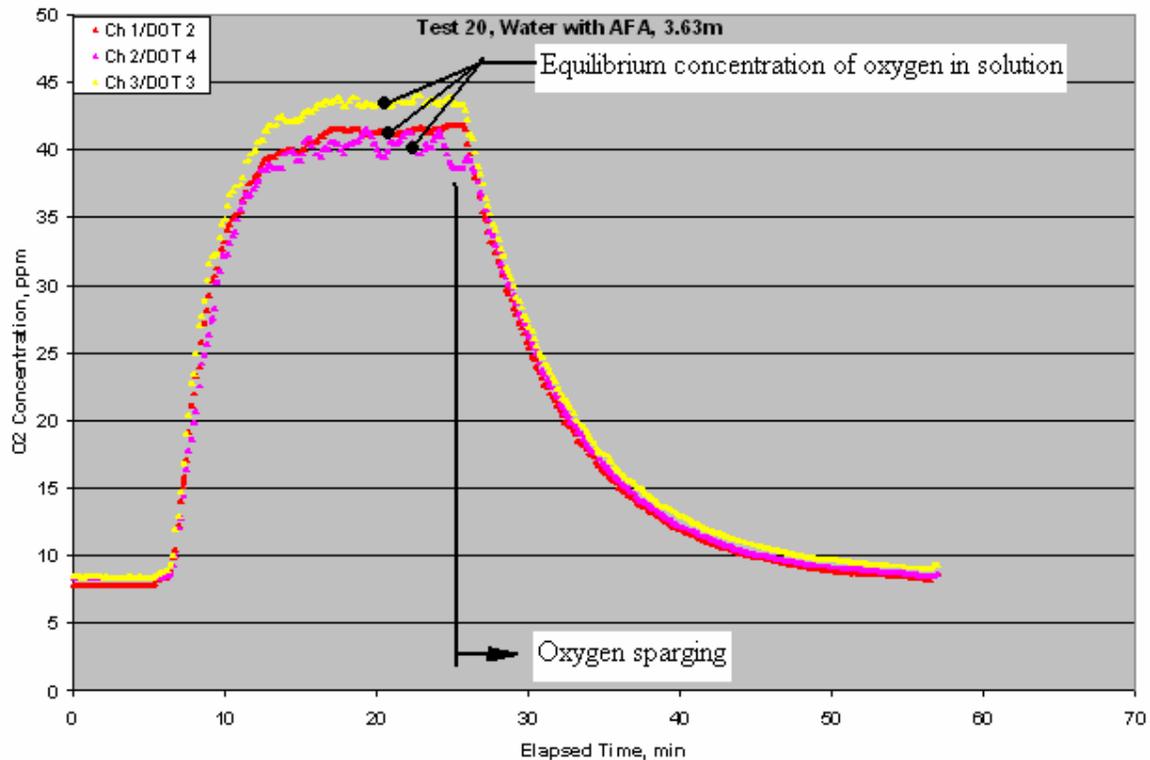


Figure 6: Uniform Distribution of Bubbles Leaving the Static Mixer<sup>1</sup>



**Figure 7: Bubbles Exiting the Mixing pipe Near the Column Bottom**

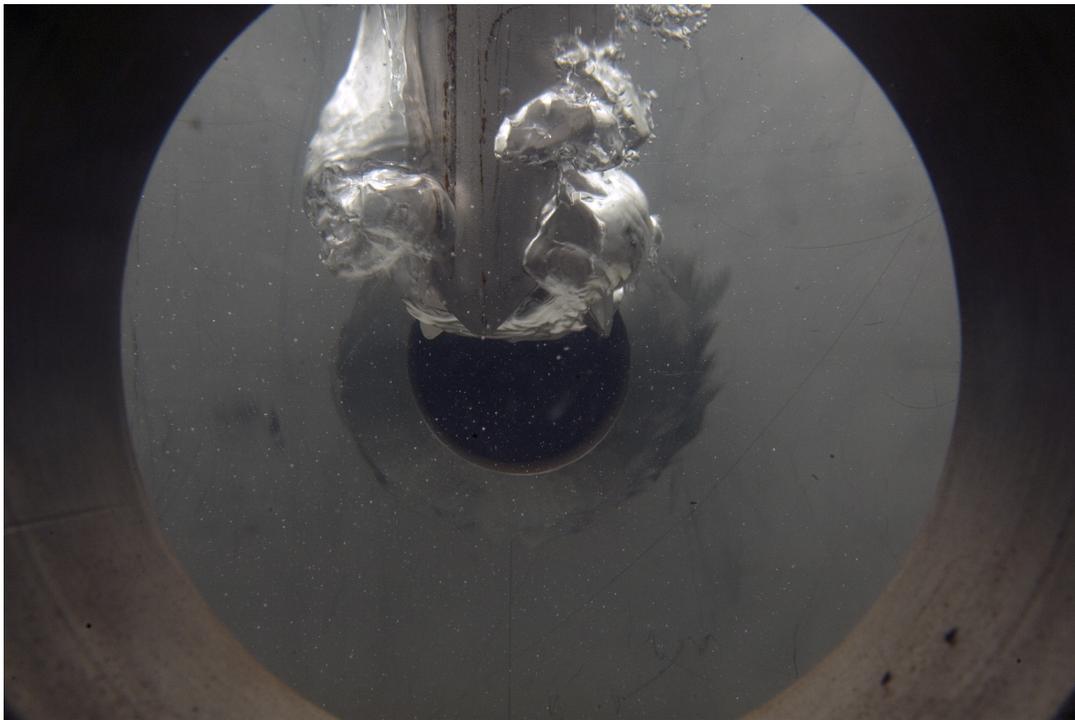


**Figure 8: Typical Mass Transfer Test Results**

### ***Mixing of the Column Using the Air Sparger***

To mix the column at three required superficial velocities, only the central sparger was used, and again DO concentrations were used to monitor mixing as shown in Fig. 8. For the water and water with AFA, mixing was consistent since the fluids were homogeneous. However, the AZ101 properties changed during testing due to fast settling solids in solution. Overnight, the simulant typically settled, and an inch of water separated from solution on the

liquid surface in the column. During tests, it was obvious from the DO readings that the material settled during tests and affected the DO sensor readings. As the material settled, oxygen was trapped near the bottom of the tank, and bubbled out sporadically. Even though the material was thoroughly mixed at the beginning of the tests, this mixing problem persisted. The sparger was inadequate to mix the simulant near the column bottom, since the bubbles exiting the sparger formed a rising cone of bubbles. Mixing was uniform at higher elevations in the column since the mixing bubbles formed a uniform field of bubbles as they rose to the top of the column. To demonstrate this statement, typical photos of bubble formation in the column are shown in Figures 9 and 10 for water. Similar results were obtained for the AZ101 simulant, but photos were only available of the fluid surface since it was opaque, as shown in Figure 11. Even though the bubble sizes were markedly different, the DO concentrations proved that mixing was effective throughout most of the column length, except at the bottom.



**Figure 9: Bubbles Near the Column Bottom Viewed through a 4 Inch Nozzle**

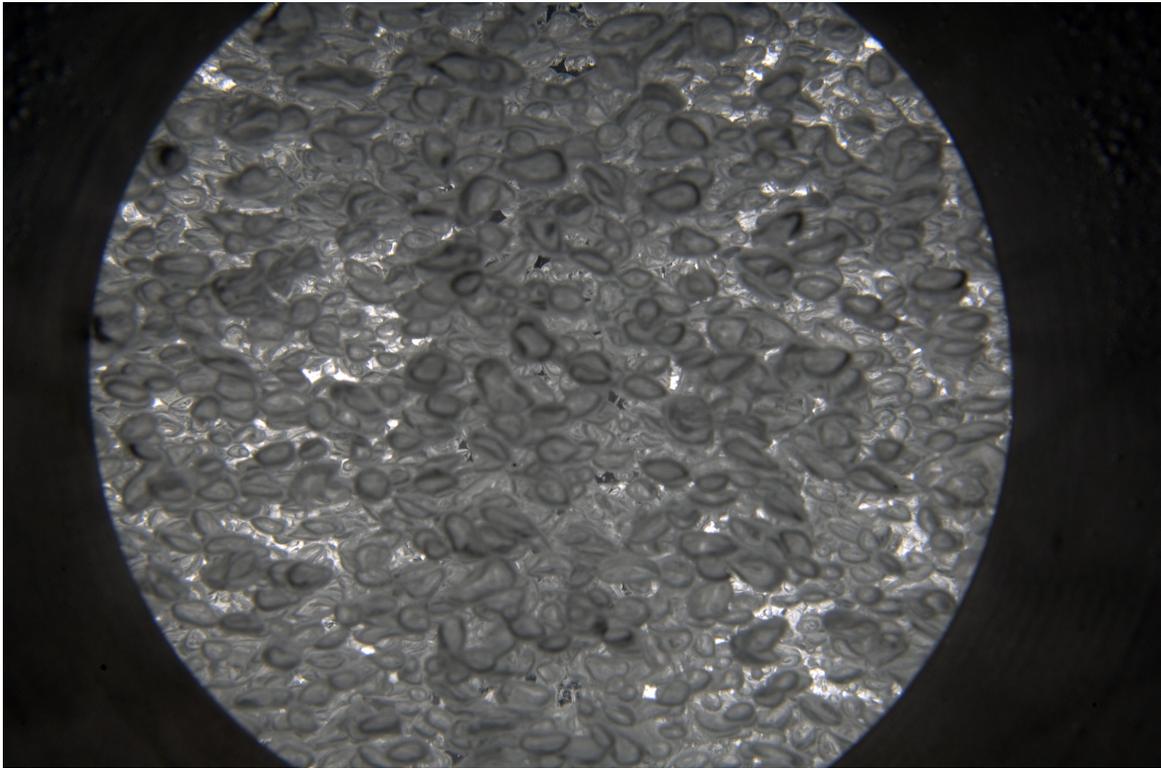


Figure 10: Bubbles Near the Column Top Viewed through a 4 Inch Nozzle

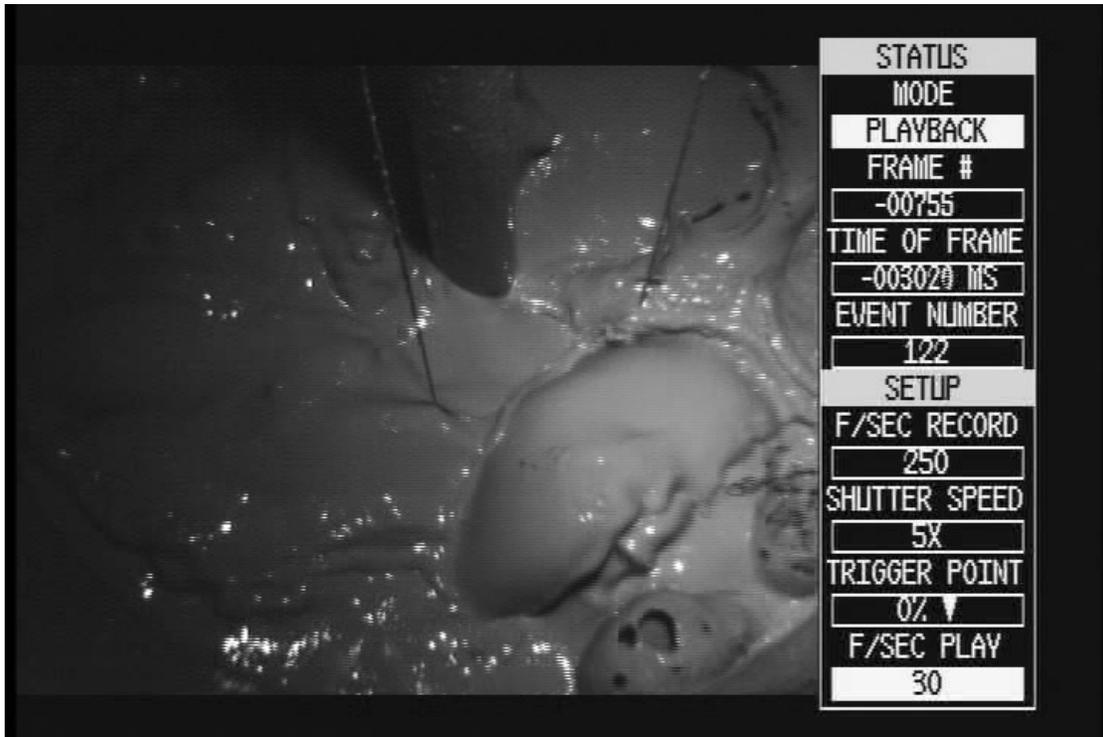


Figure 11: Bubbles on the Surface of the AZ101 Simulant

## Void Fractions

Void fractions were measured with lasers (Figure 12) and pressure transducers as shown in figure 1. Typical laser test results are shown in Figure 13. Calculations were performed to determine the void fractions,  $\phi$ , for each of the tests, using the gas volume,  $V_g$ , and the stimulant volume,  $V_s$ , such that

$$\phi = \frac{V_g}{V_g + V_s}$$

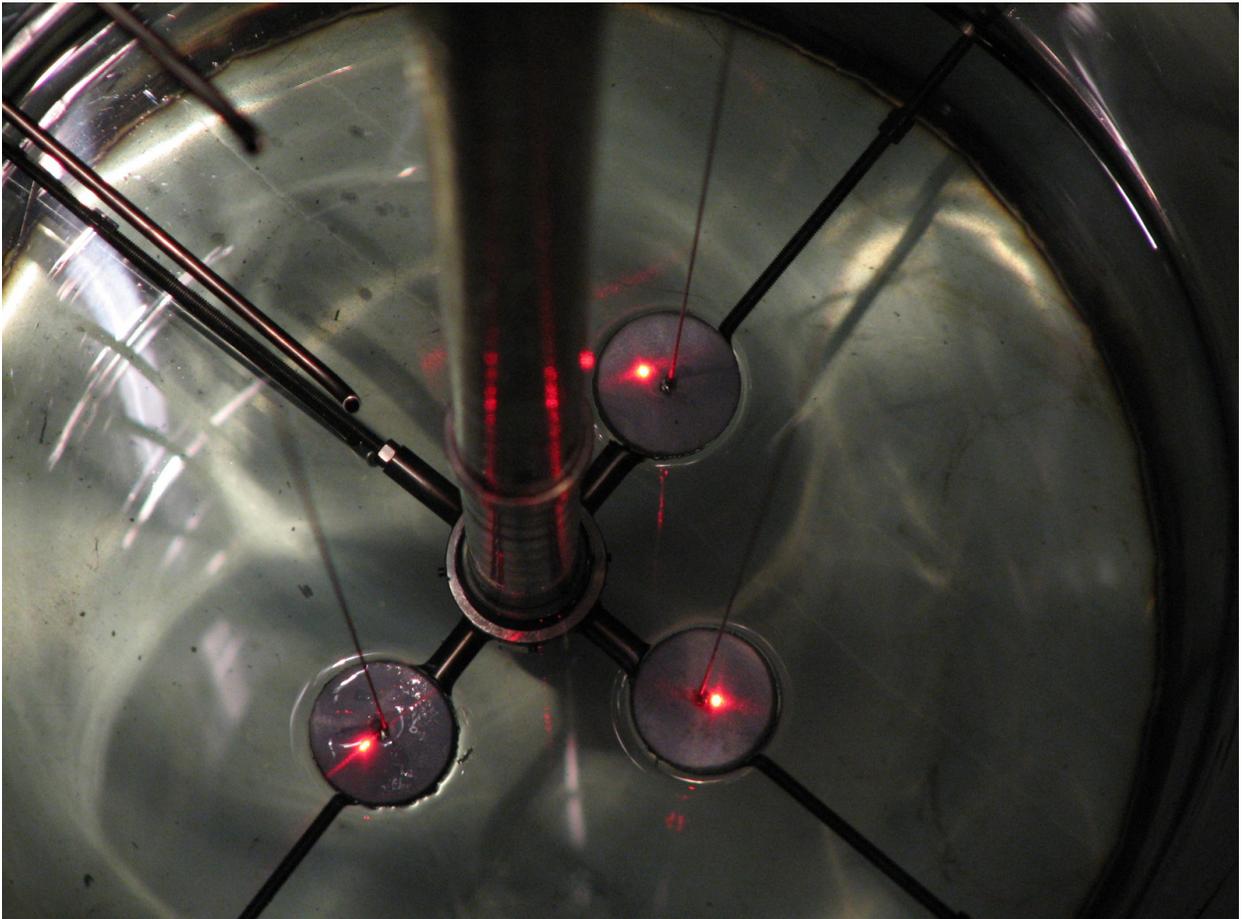
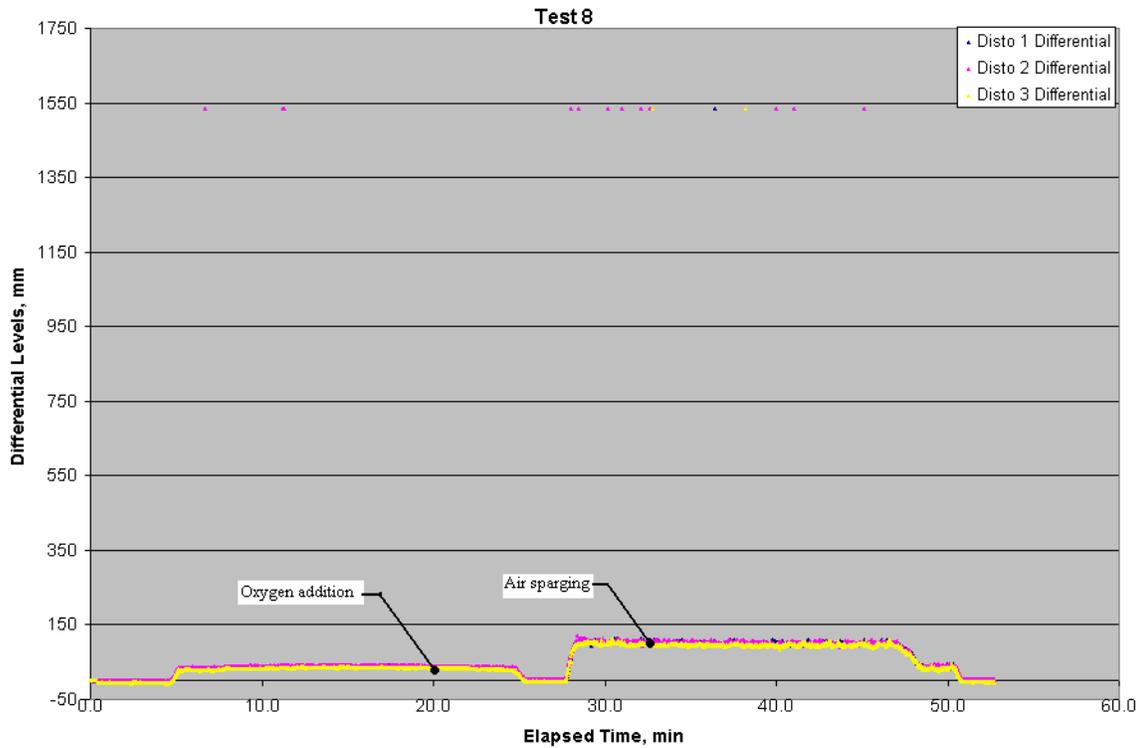


Figure 12: Laser Measurements for Water Tests



**Figure 13: Laser Measurement Data**

## Test Results

All of the test results are displayed in Figures 14 – 21. Testing with water and water with AFA provided reasonable results, but the AZ101 testing raised concerns. In all testing, mixing at the lower levels of the column affected test results, due to bubble distribution and flow turbulence. For AZ101 tests, the void fraction uncertainty is affected by the way that the tests are performed. The calculated uncertainty showed that the maximum instrument uncertainty with respect to the void fraction is 0.5% for the water tests and 0.8% for the AZ101 tests. The values for water were reasonable, but the values for AZ101 were frequently outside of the measurement uncertainty. Following initial uncertainty calculations, a sample of the AZ101 was retested after mechanical agitation to determine if trapped air was present in the fluid. Testing showed that approximately 1.7 % of the volume was trapped air. The measured void fractions are actually changes in void fractions, rather than overall void fractions. This phenomenon affects any void fraction measurement in laboratory testing or elsewhere. Once air is sparged into solution, there will always be trapped air, unless mechanical agitation is introduced. The quantity of trapped air is variable. Bubbles are released from AZ101 solution after sparging has stopped, which further complicates the situation. In addition, as soon as sparging stops, the simulant begins to settle rapidly. For the tests performed here, the simulant was mixed to obtain homogeneity immediately prior to test, but DO concentrations near the tank bottom are inconsistent with typical results, when Figure 8 is compared Figure 22. Actual field results may vary for cases where settling has occurred prior to sparging. For example, in 60 days the yield stress of the material tripled during storage, which implies that additional gas holdup may initially exist in radioactive

environments prior to sparging, since a higher yield stress material will hold up more gas. In short, trapped air affects void fraction results.

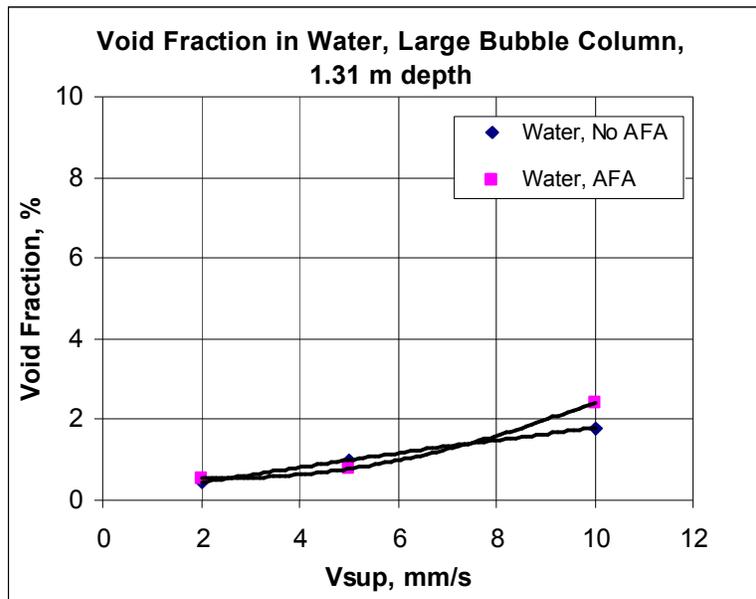


Figure 14: Average Void Fractions for Water without AFA Tests at 1.31 Meters

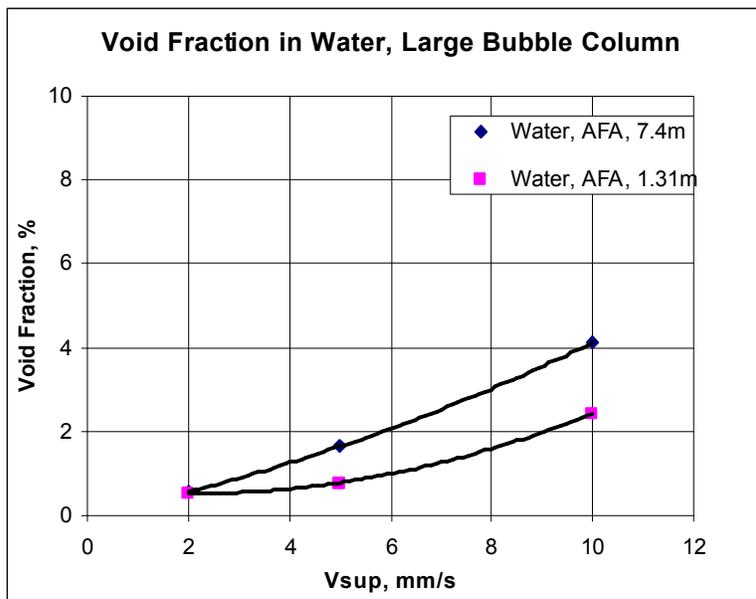
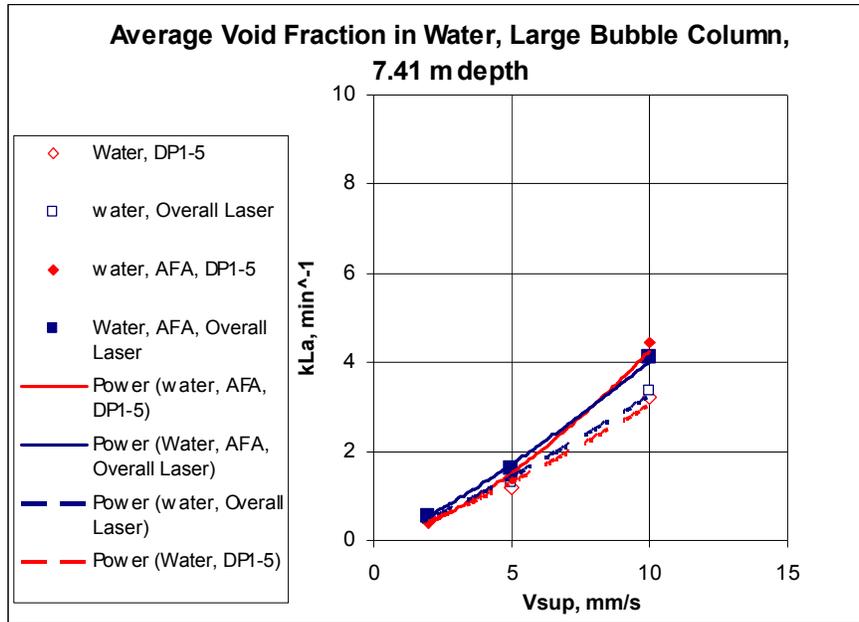
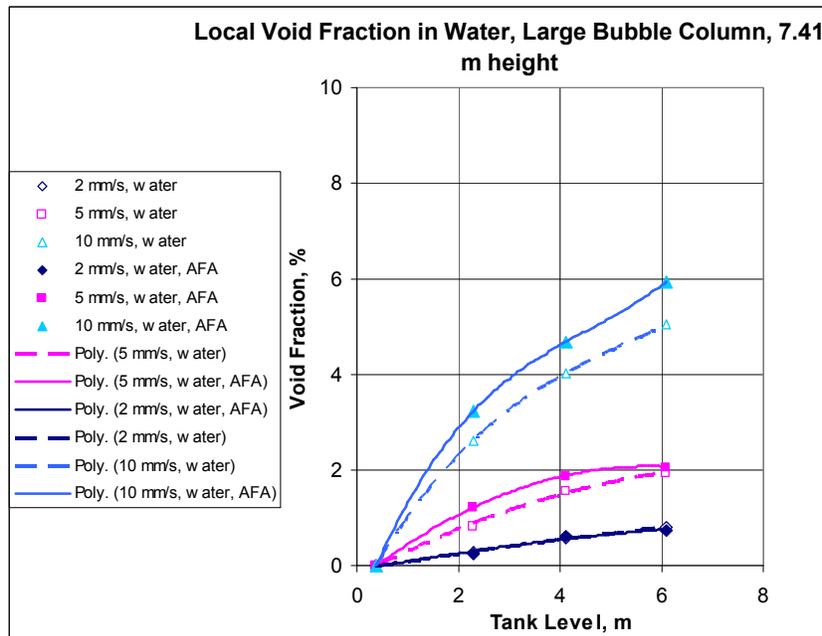


Figure 15: Comparison of Void Fractions for Water with AFA Tests



**Figure 16: Average Void Fraction for Tests at 7.41 Meters**



**Figure 17: Local Void Fraction for Tests at 7.41 Meters**

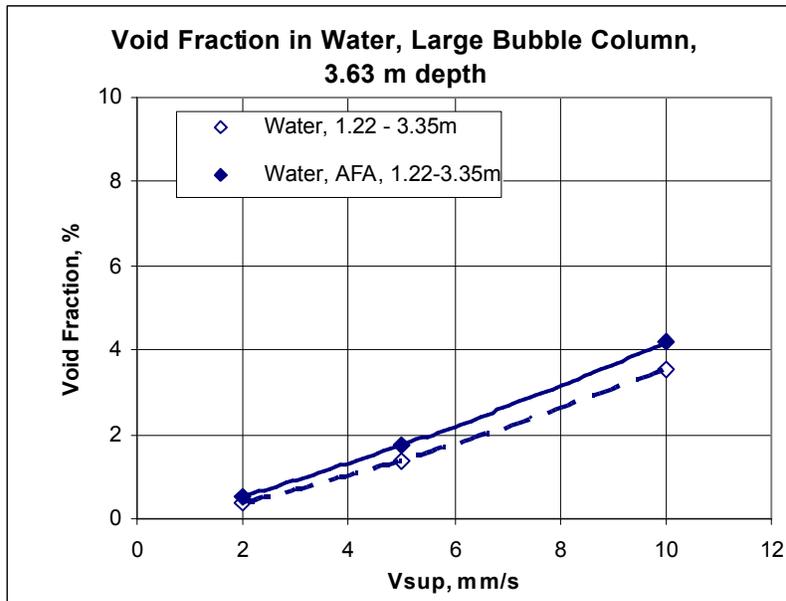


Figure 18: Local Void Fractions for Tests at 3.63 Meters

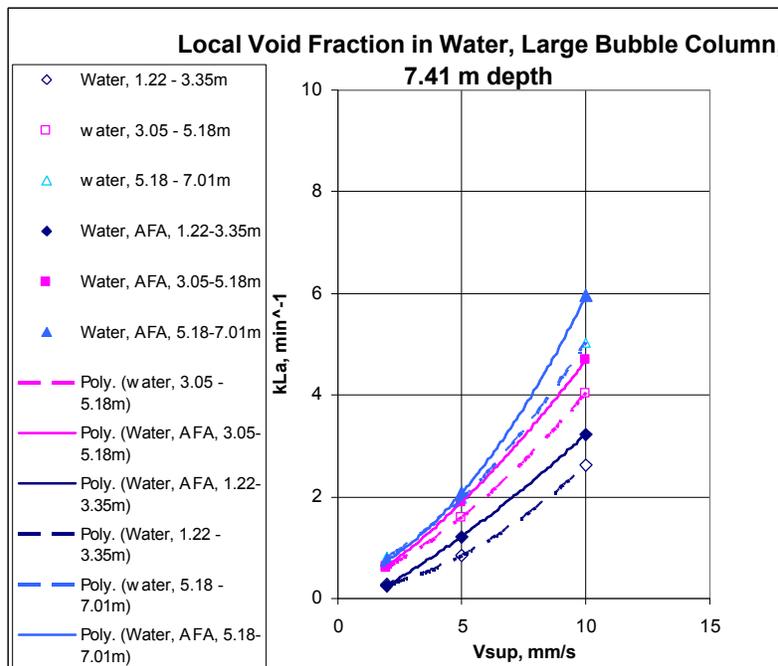
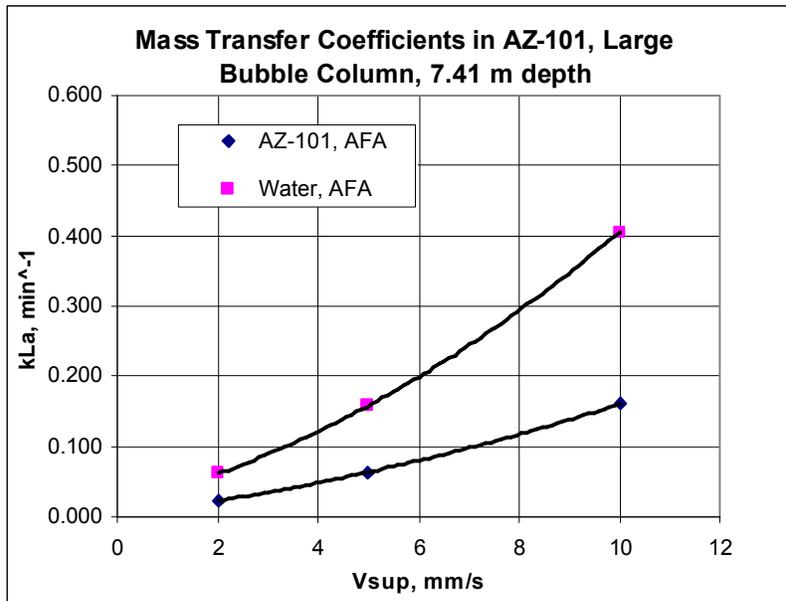
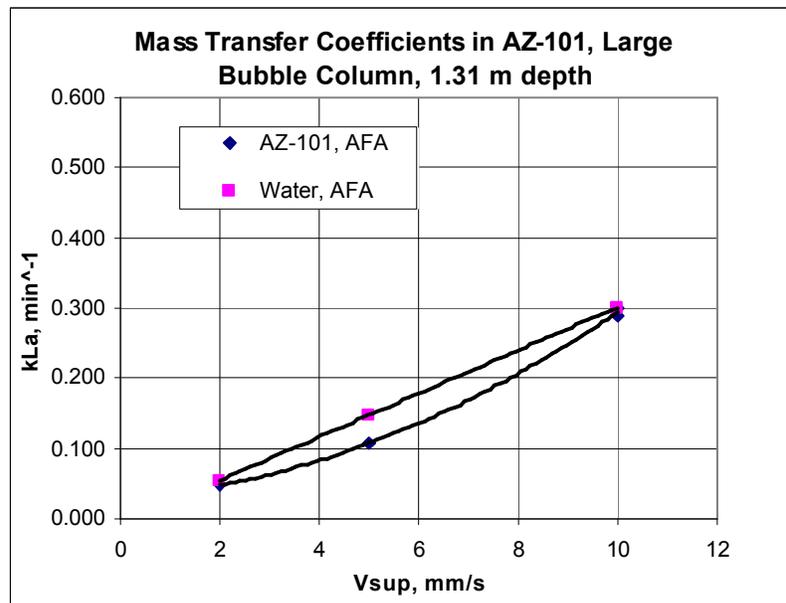


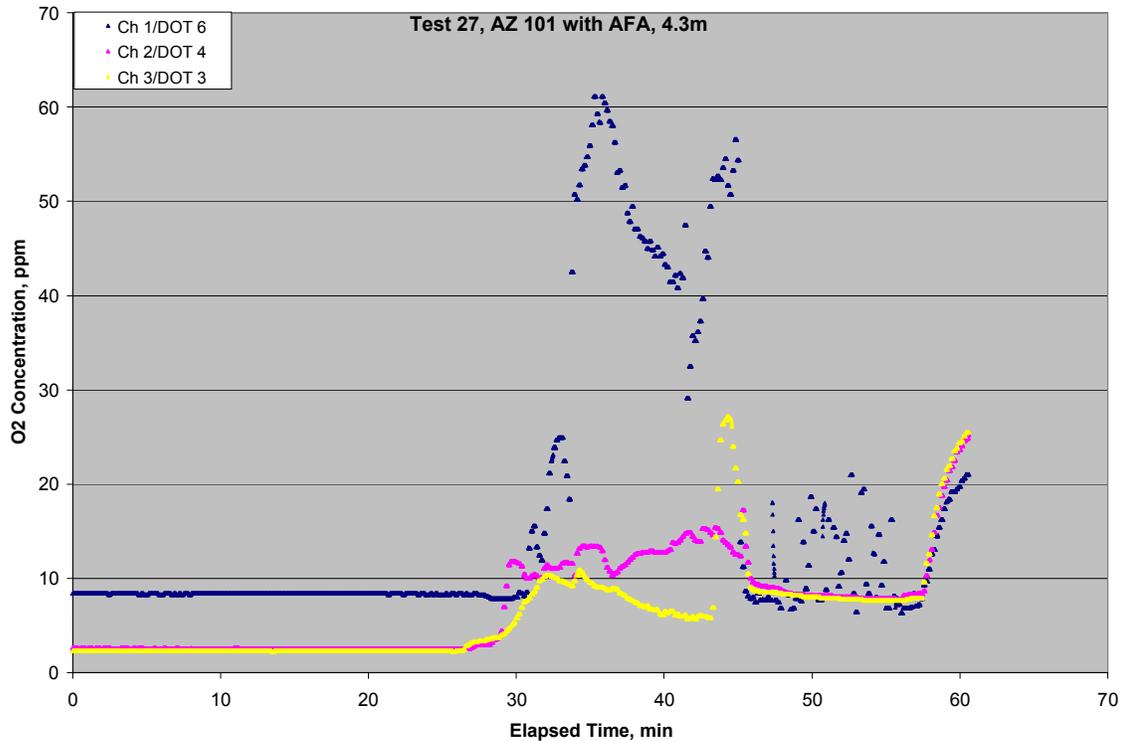
Figure 19: Local Void Fractions for Tests at 7.41 Meters



**Figure 20: Local Void Fractions for AZ101 Tests at 7.41 Meters**



**Figure 21: Local Void Fractions for AZ101 Tests at 1.31 Meters**



**Figure 22: DO Concentrations Near the Column Bottom**

## Conclusions

This paper focused on mixing and void fraction results for large scale mass transfer tests, which were performed on several simulants including process water, process water with anti-foam agent (AFA), and AZ101 simulant with AFA. To do so, a 28 foot tall by 30 inch diameter, stainless steel column was constructed, along with supporting equipment to perform the required tests. The tests required simulant testing at various levels in the column, and consisted of saturating the simulants with oxygen. Oxygen was injected into the system, rather than generated in situ using chemical reactions to create distributed oxygen throughout the solution. Once saturation was obtained, a vertical, centrally located sparge tube injected air into solution at the bottom of the column to obtain specified superficial velocities, which are the average velocity across the simulant surface in the column. The injected air displaced the oxygen from solution, and the effects of this process were measured. In particular, the effects of mixing on the determination of void fractions were considered, and void fractions were shown to be markedly affected if air was used to mix a Bingham fluid prior to test, since trapped air remained in solution between tests. Also, poor mixing near the bottom of the column was evidenced by poor mixing from the air sparger, fast settling of solids in solution, and trapped air bubbles in solution. This paper concludes a series of three papers, which considered mixing, bubble formation, and mass transfer in a large scale system.

## **References**

1. Leishear, R. A., Restivo, M., Guerrero, H., Sherwood, D., "Mass Transfer Coefficients for a Bingham Fluid and Water With and Without Anti-foam Agents", 2008 Pressure Vessel and Piping Conference, ASME, N. Y.
2. Leishear, R. A., Restivo, M., Sherwood, D. "Bubble Formation in a Large Scale System", FEDSM2008-55013, 2008 Fluids Conference, ASME, New York
3. H. N. Guerrero, C. L. Crawford, M. D. Fowley, R. A. Leishear, M. L. Restivo, "Effects of Alternate Antifoam Agents, Noble Metals, Mixing Systems, and Mass Transfer on Gas Holdup and Release from Non-Newtonian Slurries", WSRC-TR-2007-00537, Savannah River National Laboratory, S. C.