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Accurate determination of density, surface tension, and vessel depth using a triple bubbler system☆

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ABSTRACT

A triple bubbler sensor was developed for use in high temperature molten salt systems used in pyroprocessing to determine the density, surface tension, and depth of the molten salt. Preliminary bubbler testing was performed in aqueous solutions to calibrate and validate the bubbler sensor. Independently determined fluid property values and depths were compared to the calculated values and the percent differences were less than 0.16%, 3.38%, and 0.31% for density, surface tension, and depth, respectively. In addition, uncertainties were within 0.04%, 1.25%, and 0.15% for density, surface tension, and depth, respectively.

Introduction

An important aspect of safeguards, material accountancy, and process monitoring of nuclear facilities involving liquids is vessel/tank level measurements to accurately determine the fluid volume [1,2]. With knowledge of the fluid density and constituent concentration, the volume can then be related back to the total mass of special nuclear materials (SNM) contained in the vessel which is critical information for material tracking and safeguards. Numerous level measurement technologies exist, each with advantages and limitations, many of which are application specific due to liquid characteristics, environmental conditions, and the desired accuracy [3,4]. In aqueous reprocessing of spent nuclear fuel (SNF), the standard level measurement approach has been a bubbler system [5,6]. In this approach, two bubbler tubes are immersed in the fluid—one at a fixed position (reference) while the other is dynamic in depth. From the pressure measurements from the two tubes, an accurate depth can be obtained.

In electrochemical reprocessing (pyroprocessing) of SNF, the fluid media is a highly hygroscopic molten salt at high temperature. In this process, uranium from the SNF is electrochemically dissolved at an anode and transported through the molten salt where it deposits on a cathode [7]. As part of the process thermodynamics, fission products, rare earth elements, and actinides accumulate in the salt—thus changing the physical properties (density, surface tension, melting point, etc.) over time. Due to radiation, high temperature, and accessibility constraints of the material and equipment, most density and level measurements approaches are not feasible. Consequently, methods to model the density based on the elemental composition of the salt have been developed [8]. To measure the salt level, a simple dipstick method is often used. In the dipstick approach, a cold rod is lowered to the bottom of the salt and then removed and the wetted length of the rod is then measured. This approach has poor precision and accuracy for vessel level measurements. The motivation for this work is to develop instrumentation to accurately determine (via in situ measurements) depth and density in molten salts in a timely manner to enhance material accountancy, safeguards, and process monitoring of pyroprocessing molten salt.

Research has been done to explore bubbler methods for molten salt applications [9–11]. The Idaho National Laboratory (INL) developed a fixed double bubbler approach to determine the density and level of molten salts [10]. In this approach, two bubbler tubes were submerged in the fluid at different depths with a fixed distance between the tube tips. From the differential pressure between the tubes, the density could be calculated in situ. Results from the double bubbler in an aqueous media showed that the

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density of the fluid could be determined within 1% error and the depth within 3% error. The largest contribution to the depth error was the estimation of the surface tension needed in the depth calculation. A standard bubbler approach to measure surface tension uses two bubbler tubes at the same depth with different diameters [12,13]. The differential pressure between the tubes can be used to calculate the surface tension. Being that the largest error associated with the INL double bubbler [10] stemmed from the estimation of the surface tension, there may be significant improvements by adding an additional tube—thus enabling the in situ determination of density, surface tension, and depth in near real-time.

Kim et al. [11] developed a dynamic bubbler approach to measure the tank level of molten salts. With this method, a bubbler tube with flowing gas was slowly lowered into the molten salt. At the liquid surface, a distinct pressure signal was observed. With this approach, the level in molten KNO_3 was determined to within 1.1% at 500 °C. Whereas the dynamic bubbler system [11] combined with the density estimations [8] may provide sufficiently accurate results (assuming the salt analytical results have low uncertainties) for material accountability purposes, it is advantageous to develop a sensor capable of accurately measuring or determining both the density and depth in situ.

The addition of a third bubbler tube to the INL double bubbler has the potential to significantly enhance the accuracy of the determined density and depth as well as to provide determinations of the surface tension. Galbreth et al. [14] performed some preliminary experiments using this concept and demonstrated the feasibility of such an approach. The goal of this work is to further develop and test a novel triple bubbler sensor (two at the same depth and one offset) and to assess the sensor accuracy for density, surface tension, and depth in different fluids. The approach is to test the triple bubbler sensor in several room temperature aqueous solutions at varying depths to calibrate the sensor and to evaluate the overall accuracy prior to testing in a more complex molten salt. The success of this work can significantly enhance the nuclear material accountancy and safeguards of SNM in fluids—not only for molten salt applications but potentially for aqueous applications as well.

Bubbler theory

Techniques for determining density and depth in a fluid using the maximum bubble pressure are well established in ISO 18213 standards 1–6 [6]. In addition, the technique for determining surface tension is well established in ASTM D 3825-90 [13] using a maximum bubble pressure method. A schematic of the triple bubbler system is shown in Fig. 1. Tubes 1 (t_1) and 2 (t_2) have approximately the same radius (r) and are used to measure the density and depth. Tube 3 (t_3) has a smaller radius and is used in conjunction with t_2 to measure the surface tension. Tube 4 (t_4) is positioned within the gas space of the vessel and is used to determine the differential pressures between t_1, t_2, and t_3 and the liquid surface.

The maximum pressure (P) of a bubble at the bottom of a tube is the combination of the static pressure (P_s) and the overpressure or bubble pressure (P_B) as shown below:

$$P = P_s + P_B \quad (1)$$

The pressure of the bubble fluctuates as a function of its development as shown in Fig. 2. As the bubble starts to form, the bubble radius is much larger than the tube radius and the pressure is at a minimum (represented by the trough). Note that the minimum pressure is not quite the static pressure. As the bubble continues to grow, the pressure increases to a maximum when the bubble radius is equal to the tube radius, this is the maximum pressure used in the calculations. As the bubble continues to grow (radius is again larger than the tube radius), the pressure decreases until the bubble separates and floats to the surface and the processes starts again. The surface age is the time it takes for the bubble pressure waveform to go from trough to peak. According to ASTM D 3825-90 [13], the static surface tension is determined ideally with a surface age of two seconds. At these bubbling rates, the gas flow rate is low and line losses are negligible. However, with these flow rates, convection or turbulence in the fluid can disrupt the bubbles.

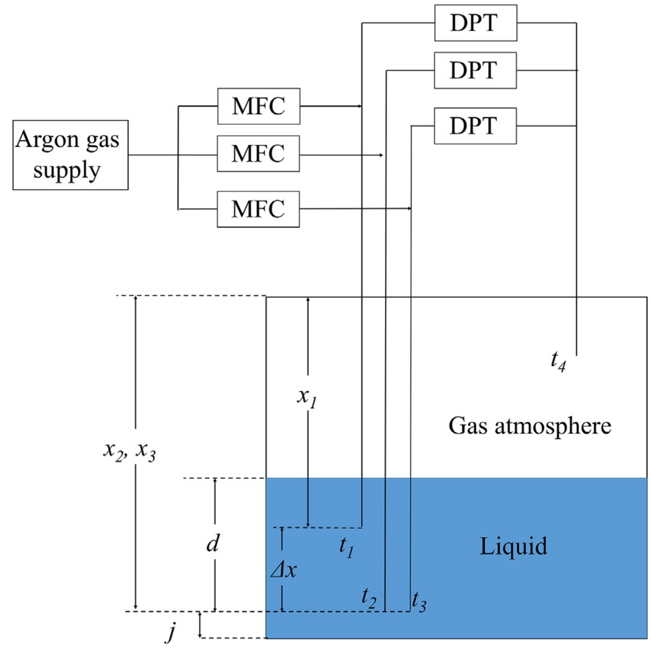

Fig. 1. Schematic of the triple bubbler system with the mass flow controller (MFC) and differential pressure transducers (DPT).

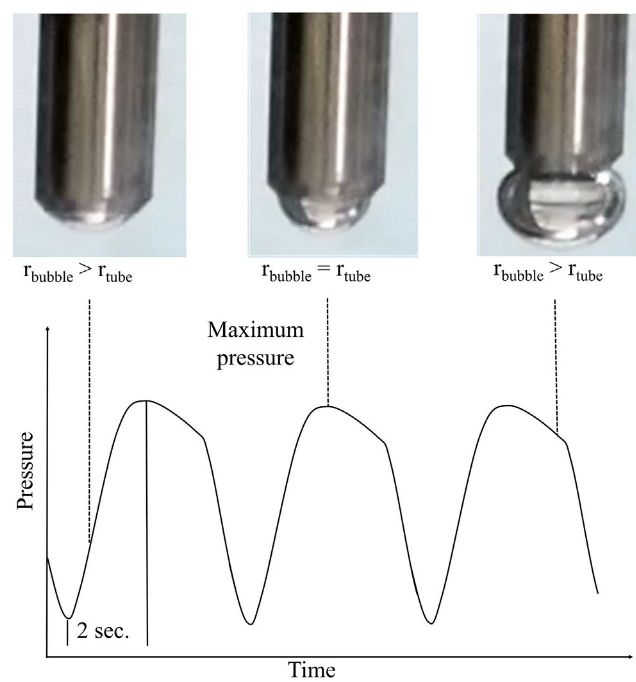

Fig. 2. Pressure to bubble radius relationship (photos are of t_1 in DI water).

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