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Note

Observation of Interface Deformation in Sodium Polytungstate Solution–Silicone Oil System due to Single Rising Bubble

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The interfacial behavior between sodium polytungstate solution (SPTS) and silicone oil (SO) due to a single rising bubble was directly observed to investigate the influence of the Eötvös number on the flow characteristics. We found that the transient behavior of the jet under the bubble strongly depended on the SPTS density in the range of 1 000–3 000 kg/m^3. Although the SPTS film generated in the SO influenced the detention time of the jet under the bubble, the lifetime of the film did not depend on the SPTS density.

KEY WORDS: molten metal–slag interface; bubbly flow; film; wake flow pattern; interfacial tension.

1. Introduction

Physical properties such as density, viscosity, and interfacial energy play a decisive role in the kinetics of reactions and transport phenomena in liquid metal–slag systems at high temperatures. To understand and model these phenomena, it is necessary to obtain experimental data pertaining to these properties. One of the most important factors that influence the rate of the chemical reaction at a metal–slag interface is the interfacial energy. When gas bubbles ascend up through the interface, mass transport gains high speed but also becomes complicated. Bubble-bursting phenomena in a gas–slag–iron system have been investigated by in-situ observation using X-ray transmission microscopy.[1–5] These studies have found two mechanisms of droplet formation: jet entrainment and film entrainment. Nevertheless, the analysis of the dynamic behavior of a molten metal–slag interface by in-situ X-ray observation has also proven difficult owing to an extremely limited observation domain. Therefore, computer simulations have been widely adopted to reproduce and predict the behavior of molten iron–slag systems. Highly sophisticated computational fluid dynamics models have been developed and applied to study the interfacial flow of high-temperature melts.[6–12] Nevertheless, even when high-performance computers are used, the analysis scale is not sufficient to clarify the interfacial phenomena completely.

Most of the available information on bubbles passing through an interface is based on investigations at low temperatures. Room-temperature media such as water, oil, and mercury have been widely used to perform non-dimensional analysis.[13–16] The system consisting of the two immiscible melts—a low-melting-point metal and a molten salt—is particularly interesting. The entrainment mechanisms in Pb–salt systems have been investigated in earlier studies,[17,18] and recently, an additional rupture phenomenon wherein an elongated metal column is formed by two or three bubbles passing through the interface of a Sn, Al–Cu alloy–salt system has been proposed.[19,20] Researchers have investigated the temperature dependence of the detention time of a single bubble passing through the interface between molten Sn and LiCl–KCl eutectic melt.[21] Although these studies have indicated that a slight change in the Eötvös number ($=\Delta\rho \mathbf{g} D^2/\sigma$) influences the interfacial shape considerably even in similar material systems, it is still not well understood as to which properties play a dominant role in determining the column shape and film detention time because so far, it has not been possible to consider the influence of a single physical property. In this study, we employed the sodium polytungstate solution (SPTS)–silicone oil (SO) system to clarify the influence of a linear change in density on the interface deformation in a two-immiscible-liquid system.

2. Experimental

An acrylic tank (inner dimensions: 0.040 × 0.090 × 0.295 m) was first filled with two different liquid phases (height of 0.090 m each): SPTS ($Na_6[H_2W_{12}O_{40}]$ in H_2O, Measure Works Co., Ltd.) as the lower liquid phase, and SO (KF-96-10cs, Shin-Etsu Chemical Co., Ltd.) as the upper liquid phase. Then, a constant volume of air was injected through a glass tube using a syringe pump. The air was trapped in a hemisphere-shaped plastic cup (inner diameter: 0.026 m); to this cup, a nichrome wire that was rotated by 180° was attached to allow a single bubble to rise. The bubble volume was controlled to be in the range of (0.5–5.0) × 10^{-6} m^3 using the syringe pump. It is an early stage of arising bubble, and it does not reach the terminal velocity.[22] However, this domain corresponds with calculation condition in previous report.[9] Interface changes were recorded using a high-speed video camera (FASTCAM, Photoron Co., Ltd.) at a rate of 2 000–10 000 frames per second and a resolution of 1 024 × 1 024 pixels. For each image capture, the position of the interface of each phase was analyzed using image processing software. Measurements were repeated at least five times in each condition. This procedure is similar to that used in our previous study.[23] Using this procedure, we observed that the rising bubble began to modify the SPTS–SO interface by pulling the SPTS phase upward through their interface.

We used SPTS in this study because this compound has been successfully used in the manufacture of heavy liquids. It has major advantages when compared to zinc chloride solutions or the highly toxic halogenated carbons, *i.e.*, low viscosity, neutral pH, and easily adjustable density. The

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heavy liquid solution is prepared by dissolving SPT in distilled water. The solution density can be continuously changed by controlling the SPTS concentration. The maximum attainable density of the solution in water at room temperature (293 K) is 3 100 kg/m^3. The chemical properties of the prepared solution are listed in **Table 1**. The density and viscosity are derived as a function of the SPTS density based on previous reports.[24–26] However, to the best of our knowledge, the interfacial energy of SPTS in each fluid phase is still unknown. Thus, the pendant drop method was performed to measure the interfacial energy using an optical contact angle meter (DM-501, Kyowa Interface Science Co., Ltd.). The interfacial shapes from the obtained images are used to determine the interfacial tension σ by the Young–Laplace method; *i.e.*, the following equation is used wherein $\Delta\rho$ and **g** are considered to be known parameters.[27,28]

$$\beta = \frac{\Delta\rho \mathbf{g} b^2}{\sigma} \quad \text{..... (1)}$$

where β is the shape factor and b is the principal curvature of the drop contour curve. **Figure 1** shows the interfacial tension of SPTS as a function of density. The surface tension of SPTS is in the range of 72–80 mN/m, and the interfacial tension between SPTS and SO is in the range of 44–52 mN/m. It can be considered that these ranges are sufficiently small relative to the change in density. We also confirmed that the solubility of SPT powder in SO is significantly small by performing a dissolution test for 1 h.

Table 1. Physical properties of solution components at room temperature (293 K).

	distilled water	SO	SPTS1.5	SPTS2.0	SPTS2.5	STPS3.0
density ρ, kg/m^3	998	935	1 500	2 000	2 500	3 000
viscosity μ, mPa·s	1.00	9.35	1.4	3.1	10.2	38.1
surface tension σ, mN·m^{-1}	72.8	20.1	unknown			

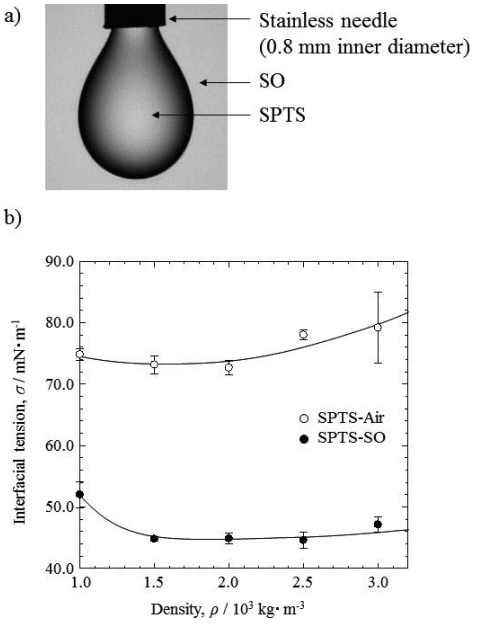


Fig. 1. Measurement results of surface tension and interfacial tension by pendant drop method: (a) snapshot of SPTS droplet in SO and (b) relationship between interfacial tension and density.

3. Results and Discussion

Figure 2 shows the obtained snapshots of a constant volume bubble rising through a water–SO interface. Rising velocity is approximately 0.3 m/s when the bubble passes through the interface. When the bubble passes through the water–SO interface, a thin film of SPTS covers the floating bubble and a wake structure is formed below the bubble. Reiter and Schwerdtfeger called it a jet.[13] **Figure 3** shows the behavior of the bubble passing through various SPTS–SO interfaces. The column immediately below the bubble becomes narrower as the SPTS density increases. The density of SPTS continuously varies without any significant change in its viscosity and interfacial tension. A thicker column is generated with decreasing the SPT density at t = 0.2 s, whereas it is broken easily with increasing the SPT density. Thus, the column shape is dominated by gravity force due to the density difference between the two liquid phases. On the one hand, the SPTS–SO interface becomes unstable when the density ratio is small. On the other hand,

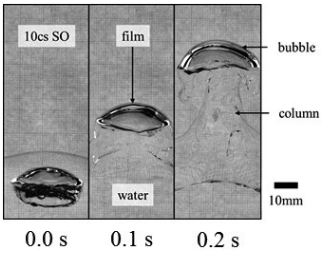


Fig. 2. Snapshots of single air bubble with constant volume (5 × 10^{-6} m^3) rising through distilled water–SO interface.

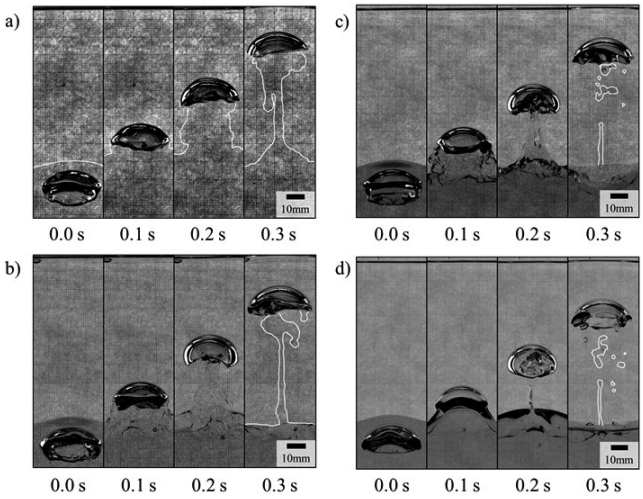


Fig. 3. Snapshots of single air bubble with constant volume (5 × 10^{-6} m^3) rising through SPTS–SO interface for (a) SPTS1.5, (b) SPTS2.0, (c) SPTS2.5, and (d) SPTS3.0. The refractive indices of SPTS and SO are approximately equal; therefore, auxiliary lines are provided to help readers identify the SPTS–SO interface.

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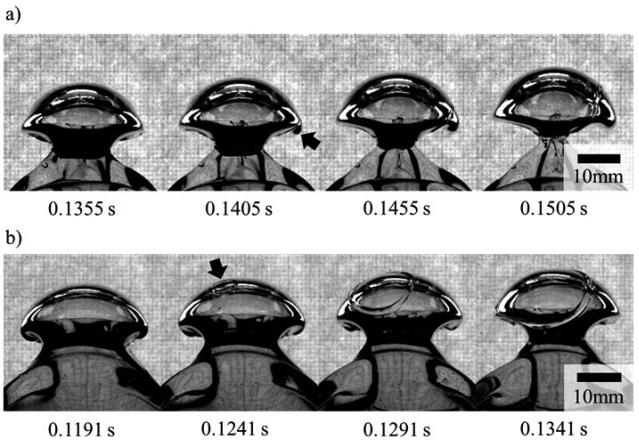


Fig. 4. Representative snapshots of the interface of the SPTS film for SPTS3.0. Rupture sites are shown in the black vectors.

a thin cylindrical column is formed as the density difference increases. If the length of the column is greater than $2\pi r$, stable droplets form along most of its length and narrow regions or "necks" form between bulges on the liquid thread (Plateau–Rayleigh instability). The thickness of the column decreases continuously until it breaks, sometimes leaving behind a number of additional "satellite" droplets.[29] Reiter and Schwerdtfeger[30] indicated that the size and volume of the produced droplets are uniquely determined by the density ratio and interfacial tension ratio of the two liquids using dimensionless correlation. Because their methodologies are based on the predicted geometry of the column, it is possible to obtain a continuous correlation. However, significant differences in the duration time of the film are not observed in this study. The films are preserved until $t = 0.1$ s under all conditions. **Figure 4** shows the snapshots of the film rupture behavior. After film rupture, the column quickly settles down, and it is clear that there is a correlation between the duration time and column geometry. An interesting point is that further rupture sites are not always located in the bubble top but can occur in the bubble bottom as well. Thus, the density only has a minor influence on the duration of the film. In a previous paper,[21] the authors mentioned that the film duration time is determined by the surface fluctuation due to interfacial tension and the reduction in the film thickness due to drainage. In the case of SPTS3.0, in spite of the fact that the viscosity becomes relatively large, changes in the film duration time is small. Therefore, in this system, the interfacial tension must be the dominant factor governing the duration of the film. Because the column shape varies sharply according to the density, the diameter of the generated SPTS droplets in SO also varies greatly. Such critical changes are difficult to predict using the correlation equation of Reiter and Schwerdtfeger.[30] The resulting droplet diameter cannot ignore the effects of changes in the film duration of the three-dimensional wake structure described above. It has been reported that stress fields produced by surfactants or nanoscale bubbles influence the rupture site of a film.[31,32] To study the mechanism of randomness in the sizes of formed droplets, changes in the physical properties of the bubble surface should be considered.

4. Conclusions

By using the SPTS–SO two-liquid-phase system, changes in the interfacial shape when a single bubble passes through the interface are investigated. By controlling the concentration of SPTS, it is possible to continuously vary the density difference without significantly changing the viscosity and interfacial tension. The column shape can vary greatly depending on the density of the lower liquid phase. It is experimentally demonstrated that the duration of the thin film does not depend on the density of the lower liquid phase.

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