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

Shungo NATSUI,* Ryota NASHIMOTO, Daiki NAKAJIMA, Takehiko KUMAGAI, Tatsuya KIKUCHI and Ryosuke O. SUZUKI

Division of Materials Science and Engineering, Faculty of Engineering, Hokkaido University, Kita 13 Nishi 8, Kita-ku, Sapporo, 060–8628 Japan.

(Received on August 30, 2016; accepted on October 18, 2016; J-STAGE Advance published date: December 15, 2016)

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/m³. 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 (\(= \Delta \rho g D^2/\sigma\)) 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 (Na₅[H₂W₁₂O₄₀] in H₂O, Measure Works Co., Ltd.) as the upper liquid phase, and SO (KF-96-10cs, Shin-Etsu Chemical Co., Ltd.) as the lower 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 m³ 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
A 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/m³. 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. 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 by the Young–Laplace method; i.e., the following equation is used wherein Δρ and g are considered to be known parameters.

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

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.

### 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. 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,

| Physical properties of solution components at room temperature (293 K). |
|----------------------|-----------------|-----------------|-----------------|------------------|
|                     | distilled water | SPTS1.5 | SPTS2.0 | SPTS2.5 | SPTS3.0 |
| density ρ, kg/m³     | 998             | 935     | 1 500  | 2 000  | 2 500  | 3 000  |
| viscosity μ, mPars    | 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.

Fig. 2. Snapshots of single air bubble with constant volume (5 × 10⁻⁶ m³) rising through distilled water–SO interface.

Fig. 3. Snapshots of single air bubble with constant volume (5 × 10⁻⁶ m³) 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.
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.

Acknowledgment

This work was financially supported by the Japan Society for the Promotion of Science (JSPS) “KAKENHI” and research grant of the Japan society for the promotion of mining.

REFERENCES

1) B. S. Terry and P. Grieveson: ISIJ Int., 33 (1993), 166.
2) Z. Han and L. Holappa: ISIJ Int., 43 (2003), 292.
3) Z. Han and L. Holappa: ISIJ Int., 43 (2003), 1698.
4) Z. Han and L. Holappa: Metall. Mater. Trans. B, 34 (2003), 525.
5) L. Holappa, L. Forsbacka and Z. Han: ISIJ Int., 46 (2006), 394.
6) T. Oda, N. Satofuka and H. Nishida: Computational Fluid Dynamics 2002, Springer, Berlin, Heidelberg, (2003), 529.
7) M. Nakano and K. Ito: ISIJ Int., 56 (2016), 1297.
8) N. Kochi, Y. Ueda, T. Uemura, T. Ishii and M. Iguchi: ISIJ Int., 51 (2011), 1011.
9) S. Natsui, R. Washimoto, H. Takai, T. Kagamai, T. Kikuchi and R. O. Suzuki: Chem. Eng. Sci., 141 (2016), 342.
10) T. Tagawa, Y. Anzai and T. Kagamai: Tetsu-to-Hagané, 101 (2015), 101.
11) T. Himeno and T. Kagamai: Tetsu-to-Hagané, 101 (2015), 109.
12) O. Nakamura, T. Kagamai and K. Takatani: Tetsu-to-Hagané, 101 (2015), 117.
13) G. Reiter and K. Schwerdtfeger: ISIJ Int., 32 (1992), 50.
14) M. Iguchi, T. Chihara, N. Takanashi, Y. Ogawa, N. Tokunitsu and Z. Morita: ISIJ Int., 35 (1995), 1354.
15) L. T. Khajavi and M. Barati: ISIJ Int., 50 (2010), 654.
16) T. Kagamai, Y. Kishimoto, O. Nakamura, T. Tagawa, T. Himeno and S. Shimazaki: Tetsu-to-Hagané, 101 (2015), 93.
17) D. Poggi, R. Minto and G. Davenport: J. Met., 21 (1969), 40.
18) R. Minto and W. G. Davenport: Can. Min. Metall. Bull., 65 (1972), 70.
19) D. Y. Song, N. Maruoka, G. S. Gupta, H. Shibata, S. Y. Kikamai and S. Kambe: Metall. Mater. Trans. B, 43 (2012), 973.
20) H. Yoshida, J. Liu, S. J. Kim, X. Gao, S. Ueda, N. Maruoka, S. Ono and S. Kitanuma: Metall. Mater. Trans. B, 47 (2016), 2498.
21) S. Natsui, R. Washimoto, H. Takai, T. Kagamai, T. Kikuchi and R. O. Suzuki: Metall. Mater. Trans. B, 47 (2016), 1532.
22) R. Cliff, J. R. Grace and M. E. Weber: Bubbles, Drops, and Particles, Chapter 7, Academic Press, New York, (1978), 169.
23) S. Natsui, H. Takai, T. Kagamai, T. Kikuchi and R. Suzuki: Mater. Trans., 55 (2014), 1707.
24) M. R. Gregory, R. Murray and K. A. Johnston: N. Z. J. Geol. Geophys., 30 (1987), 317.
25) M. E. Torresan: The Use of Sodium Polytungstate in Heavy Mineral Separations, US Geological Survey, Reston, Virginia, (1987), 87.
26) M. D. Hoover, G. L. Finch and B. T. Castorina: J. Aerosol Sci., 22 (1991), 215.
27) J. M. Andrews, E. A. Hauser and W. B. Tucker: J. Phys. Chem., 42 (1938), 1001.
28) Y. Rotenberg, L. Boruvka and A. W. Neumann: J. Colloid Interf. Sci., 93 (1983), 169.
29) G. Tryggvason, R. Scardovelli and S. Zaleski: Direct Numerical Simulations of Gas–liquid Multiphase Flows, Cambridge University Press, (2011), 204.
30) G. Reiter and K. Schwerdtfeger: ISIJ Int., 32 (1992), 57.
31) S. Ishihara, H. Tsukiyama, Y. Tagawa and M. Kameda: Proc. Konsouryu Symp. 2016, The Japanese Society of Multiphase Flow, Kyoto, (2016), C133.
32) K. Tomaru and M. Shirazaki: Proc. Konsouryu Symp. 2016, The Japanese Society of Multiphase Flow, Kyoto, (2016), F214.