# Rheological Characteristics of Molten Metal for Casting Design

Soo-Han Park, Hwan-Goo Seong, Young-Hwan Song and Bo-Young Hur\*

i-Cube Center, Human Resource Training Project for Regional Innovation, ERI

School of Nano and Advanced Materials Engineering, Gyeongsang National University, Jinju, 660-701, Korea

Fax: 81-55-753-2270, e-mail: suhan2121@nate.com; uw98hgs@gnu.ac.kr;

yhsong4u@nate.com \*hurby@gnu.ac.kr

The surface tension and viscosity are defined as the attraction between the molecules of the melt and the friction of fluids in motion, respectively. Both terms are considered to be dominant factors in various metallurgical phenomena, including embracing gas absorption, nucleation of gas, nucleation and growth of non-metallic inclusions, and surface oxide/melt reactions. In this study, therefore, the experimental measurements of melt properties for several metals, including pure Al, Al alloys and Mg-Al alloys (AZ91 & AM60), were assessed with variation of temperature and alloying element concentration. The surface tension and the viscosity of individual melts have been measured, using the ring method and the rotation method, under either pure Ar or SF<sub>6</sub>+CO<sub>2</sub> atmosphere. The primary results through present experiment showed that both properties of these metals decreased with increasing temperature. The addition of elements to both pure Al and Mg also decreased the surface tension and increased the viscosity. The optimum viscosity was evaluated for different alloy melts, including Al-Si, Al-Mg, Al-Cu. In detail, greater than 1.5 wt% for Al-2wt%Mg melt, 2~2.5 wt% for Al-1wt%Si melt, and 1.5 wt% for Al-4wt%Cu melt. The surface tension values of Mg alloys were less than 500~600 (mNm<sup>-1</sup>). However, the optimum viscosity values for both alloy melts were obtained with 1.5wt% Ca addition and more than stirring time of 10 min. Key words: surface tension, viscosity, Mg alloys, metallic foam.

## 1. INTRODUCTION

Surface tension and viscosity are crucial thermo-physical properties of molten metal essential for standardization of conventional and emerging casting technology which leads to an improvement in product quality and reduction in production cost. These factors in molten metals, particularly, influence the quality control in manufacturing metallic foams<sup>1)</sup> which would bring synergetic effects on advancement transportation and machinery parts<sup>2-5)</sup>. However, little experimental works associated with the surface tension and viscosity has been performed yet because of complexity of molten metal. Although some researchers<sup>6)</sup> have given considerable efforts on elucidation of aforementioned factors in molten metals quantitative data are still scarce as ever.

In the present study, therefore, the influence of various alloying elements on the surface tension and viscosity of molten metal with low melting points were investigated in a quantitative manner by using modified apparatus. The objective of the present study was to establish the appropriate melt condition suitable for manufacturing metallic foams and further provide basic data suitable for fabrication of metallic foams employing a melt-foaming process.

#### 2. EXPERIMENTAL PROCEDURES

Surface tension was measured by the modified drop weight method, which applies the capillary phenomenon to measuring the maximum force and contact angle when the ring is pulled out from the melt surface<sup>6)</sup>. The viscosity was estimated experimentally by measuring

changes in torque that is a resistance force of the melt against the rotating rotor  $^{6)}$ .

In this experiment, the high purity argon or  $SF_6+CO_2$ gas sealing was used to prevent surface oxidation. The flowing rate was set to  $25(\ell/\min)$ . The measured samples were Al and Mg alloys with and without adding Calcium, Silicon, Copper, and Magnesium. The temperature range of measurement was set from 660°C to 950 °C(from 600°C to 850 °C for Mg alloys).

The temperature was measured by Pt-Rh thermocouples, which were put into the bottom and the side of the crucible.

The maximum force  $(F_{max})$  that is measured by the ring method can be recalculated to a surface tension  $(\sigma_{st})$  by equation (1).

$$\sigma_{st} = \frac{F_{\max}}{4\pi R \cdot \cos\theta} \cdot f\left(\frac{R^3}{V}, \frac{R}{r}\right) \tag{1}$$

Where  $4\pi R$  is wetted length,  $F_{max}$  is the total maximum force, f is the Harkins Jordan factor <sup>7</sup> and  $\theta$  is the contact angle ( $\theta = 90^{\circ}$ ).

Viscosity  $(\eta_{visco})$  can be calculated from the measured torque (T) on the rotation rotor.

$$\eta_{visco} = \frac{15T}{\pi^2 r_1^3 N(\frac{r_1}{ab} + \frac{h}{a})}$$
(2)

Where T is the measured torque and N is the revolutions per min of the rotor.  $r_1$  is the radius of rotor : 26mm, a is side gap : 4mm, b is bottom gap : 5mm and h is wetted height : 100mm.

In addition, to compare the foamability among pure Al, recycled Al alloy and AM60, foaming tests were carried out. The testing conditions were taken as table 1.

Thickening	Agent	Ca (1.5wt% add.)
	Stirring	15minutes
Blowing	Agent	$\begin{array}{c} \text{TiH}_2 \text{ (3wt\% add.)} \\ \text{CaCO}_3 \text{ (2wt\% add.)} \end{array}$
	Stirring	30 seconds
Temperature	Thickening & blowing	700°C for Mg alloys
	Curing	600°C for Mg alloys

Table 1. The condition of foaming test.

#### 3. RESULTS and DISCUSSION

3.1 Surface Tension and Viscosity of Aluminum Alloys

The variation in surface tension with melt temperature was measured experimentally prior to fabrication of metallic foams. Figure 1 shows the changes in surface tension as a function of the melt temperature, showing decreasing surface tension with increasing temperature. A linear regression equation fitting to the present experimental measurements were expressed as

$$\sigma(T) = 842 - 0.024 (T - T_m) [mNm^{-1}]$$
[1]

According to Eotovo's  $law^{6)}$ , the temperature dependence of the surface tension were correlated as follows.

$$\frac{dT}{d\sigma} = \frac{k_{\sigma}}{V^{2/3}} \left[ \frac{2(T_c - T)}{3\sigma} \frac{d\sigma}{dT} - 1 \right]$$
 [2]

Where  $k_{\sigma}$  is constant and ~6.4×10<sup>-8</sup> (JK<sup>-1</sup>mol<sup>-2/3</sup>), density of aluminum is about 2.38×103 (kgm<sup>-3</sup>), and  $(d\rho/dT) = -3.5\times10^{-1}$  (kgm<sup>-3</sup>K<sup>-1</sup>). Using equation [2], the calculated surface tension of aluminum at its melting temperature were approximately 914 (mNm<sup>-1</sup>). It was decreased with increasing the melt temperature, following a  $(d\rho/dT) = -0.35$  relationship.



Fig.1. Temperature dependences of surface tension of pure Al and Ref. 7



Fig.2. Effect of added Ca and Mg on the viscosity of pure Al ant 700~710°C.



Fig.3. Effect of added Ca and Mg on the viscosity of pure Al at 670-680°C.

As can be illustrated in Figure 3, the linear regression equation established by using maximum bubble pressure (MBP) Hanging drop (HD) methods were given in following form.

$$\sigma(T) = 868 - 0.152 (T - T_m) [mNm^{-1}]$$
[3]

From these, it was shown that experimental measurements obtained in the present study were slightly lower than ones reported previously.

As shown in Figure 1, however, the slopes in linearly decreasing fitting curves for both cases were quite similar.

The influence of alloy composition on the surface tension was also examined experimentally at constant melt temperature of 700~710 °C. Figure 2 displays the changes in surface tension with various alloying elements and their contents. In this case, the data reported in open literature<sup>8)</sup> and collected in commercial simulation package (Anycating<sup>TM</sup>) were cited to compare the present experimental measurements. With additions of Si and Cu elements, the measured surface tensions of dilute aluminum was decreased (to about 40 (mNm<sup>-1</sup>) at 0.5 wt.%Si) and beyond changed constant irrespective of increases in its content. This was in good agreement with previously reported results.

With additions of Mg and Ca elements, however, the surface tensions of dilute aluminum after a rapid drop was continuously decreased with increasing its content. Nevertheless, the experimental measurements of surface tension for the Ca diluted melt were much lower than the calculated ones as shown in Figure 2. This noticeable difference in surface tension values were likely to be due to influx of the surface oxide layer of the diluted melt and/or preformed CaO caused by a mechanical stirring employed for adding alloying elements.

On the other hand, the influences of Ca and Mg elements on the viscosity of aluminum melt are shown in Figure 3. In this case, the present measurements were performed after the mechanical stirring of 400 rpm followed by dilution of aluminum melt with Ca or Mg elements. For both cases, the viscosities of diluted melts were increased with their contents. According to previous study performed by present authors<sup>9</sup>, the optimum content of Ca element in the aluminum melt might be about 1.5 wt% in fabricating high quality metallic foams using a melt forming technique. In that case, the surface tension and viscosity were approximately  $550~600 \text{ (mNm}^{-1})$  and 10~14 (mPa.sec), respectively. As further study, similarly, the optimum stirring time and Ca content were here assessed by

adding Ca element to three alloy melts: Al-Mg, Al-Si, and Al-Cu.

Figure 4 and Figure 5 show changes in viscosity of various diluted melts with stirring time after addition of different contents of Ca element. In these cases, the stirring speed and melt temperature were 400 rpm and 700 C, respectively. For all cases investigated, the melt viscosities measured were slightly increased with stirring time and Ca contents. In one case of Mg and Ca addition shown in Figure 4, its measured viscosity values were larger for the diluted melt with higher contents compared to those for the melt diluted with low contents of elements. The role of Ca element increasing the melt viscosity to viscosity of 10~14 (mPa.sec) about was clearly shown for the diluted melts with Si and Cu elements (Figure 5 and Figure 6). These results indicated that the addition of Ca element for appropriate melt viscosity should be appropriately adjusted with the types of melts of interest. In detail, greater than 1.5 wt% for Al-2wt%Mg melt, 2~2.5 wt% for Al-1wt%Si melt, and 1.5 wt% for Al-4wt%Cu melt.



Fig. 4. Viscosity vs. stirring time diagram with and without Ca addition.



Fig. 5. Viscosity vs. stirring time diagram with Ca addition.



Fig. 6. Viscosity vs. stirring time diagram with Ca addition.

3.2 Surface Tension and Viscosity of Magnesium Alloys

In previously report<sup>10</sup>, The surface tensions measured at near the melting temperature were about 473 (mNm<sup>-1</sup>) for AZ91 alloy and 557 (mNm<sup>-1</sup>) for AM60.

Individual ones were decreased in a similar slope with increasing the melt temperature. Yet, the estimated linear relation was  $473 - 0.545 (T - T_m)$  for AZ91 alloy melt and  $557 - 0.468 (T - T_m)$  for AM60 alloy melt.

The viscosities of both alloy melts were also decreased with increasing the melt temperature. The viscosity values obtained for AZ91 alloy melt were higher than those for AM60 alloy melt, which might be related to adsorption behavior of solute (aluminum and zinc) in liquid AZ91.

The Ca element and stirring time affecting the surface tension and viscosity of both alloy melts are displayed in Figure 7. For both alloy melts, the measured surface tension values were decreased with increasing Ca contents as shown in Figure 7(a). In contrast, their viscosity values at 1.5 wt% Ca contents were increasing stirring time. In this case, the AZ91 alloy melt was found to have higher viscosity than that of AM60 alloy melt over the stirring time of 20 sec. It was shown from these results that for both alloy melts the optimum viscosities were reached after addition of 1.5 wt% Ca followed by the 10 min stirring.



Fig. 7. Effect of added Ca on the (a) surface tension and (b) viscosity of magnesium alloy at  $600 \sim 610$  °C.

3.3 Mg alloy foams manufacturing

There have been reported that  $TiH_2$  can be used as blowing agents to fabricate Al alloys(Al-Si and Al-Cu etc.) foam successfully<sup>11,12</sup>.

The melting points of Al alloy and Mg alloy are quite similar, so these two agents (TiH2 and CaCO<sub>3</sub>) are possible used for making Mg alloy foam.

But Figures 8(a) and (b) show the Mg alloy foams prepared by using TiH<sub>2</sub> have very coarse pore structures while Figures 8(c) and (d) show the Mg alloy foams prepared by using CaCO3 as blowing agent have homogeneous pore structure. Although the difference of pore stabilization mechanism by using these two blowing agents is needed more investigations, there are two possible reasons to explain the difference. One is that, for AZ91 case, after adding 1.5wt% Ca, although the viscosity of AZ91 melt is at same level as that of thickened Al melt, which is about 10~14(mPa.s), the surface tension of AZ91 melt, which is about  $450(mN^{-1})$ , is so low compared with thickened Al melt, which is about 550~600(mN<sup>-1</sup>) that bubbles likely can not stay stable in the Mg alloy melt if TiH<sub>2</sub> is used as blowing agent; On the other hand, the blowing gas released by  $CaCO_3$  decomposition in the melt cam react with Mg alloy melt to form a large number of solid particles<sup>6,13)</sup>. It is these solid particles that can slow down the cell wall rupture process and hence inhibit cell coarsening while H<sub>2</sub> released by decomposition of TiH<sub>2</sub> does not have these effects.



Fig.8. Mg alloy foams prepared by different blowing agents.

## 4. SUMMARY

The surface tension and viscosity of the aluminum and magnesium melts were investigated, in terms of the melt temperature, alloy composition, and the stirring time, to establish the optimum melt properties for sound quality of metallic foams. The following conclusions can be drawn.

- 1. The surface tension obtained at near the melting temperature for pure aluminum melt was in the range of 842~914 (mNm<sup>-1</sup>). Its temperature dependence was fitted to a linear equation,  $\sigma(T) = 842 0.024 (T T_m)$ . The addition of Ca and Mg element to the pure aluminum melt decreased the surface tension of the corresponding melts; decrease to 550 (mNm<sup>-1</sup>) for 2 wt% Ca but to 625 (mNm<sup>-1</sup>). It was due possibly to the surface-activity effect.
- The viscosity of aluminum melt, after 2 wt% Ca addition and 20 min stirring, was increased up to about 14 (mPa·sec). This was related to in-situ

formation of high melting point CaO and/or Al<sub>2</sub>O<sub>3</sub>.

- The optimum viscosity values were evaluated for different alloy melts, including Al-Si, Al-Mg, Al-Cu. In detail, greater than 1.5 wt% for Al-2wt%Mg melt, 2~2.5 wt% for Al-1wt%Si melt, and 1.5 wt% for Al-4wt%Cu melt.
- 4. The surface tension values obtained for both alloy melts, AZ91 and AM60, were less than 500~600 (mNm<sup>-1</sup>). However, the optimum viscosity values for both alloy melts were obtained with 1.5wt% Ca addition and more than 10 min.

#### Acknowledgment

This research was financially supported by the Ministry of Commerce, Industry and Energy (MOCIE) and Korea Industry Technology Foundation (KOTEF) through the Human Resource Training Project for Regional Innovation.

### References

[1] J. Banhart, M. Ashby and N. Fleck, "Cellular metals and Metal Foaming Technology", Verlag MIT Publishing (2001) pp.63-68

[2] B.Y. Hur, H.J. Ahn, D.C. Choi, S.H. Cho, K.D. Park, Y.J. Kim and S.H. Jun, Proceedings of the Symposium on Solidification Process of Metals, 87 (2000)

[3] B.Y. Hur, Y.C. Cho, J.G. Lee, D.S. Lee and S.H. Cho, Patent 378395 (1995)

[4] L.M. Niebylski and C.P. Jarema, Ethyl Corporation Richmond Va, Patents No. 3,790,365.

[5] R.C. Gradinger, R. Kretz, H.P. Degiscer and F.G. Ramerstorfer, Junior-Euromat, Lausanne, Switzerland (1996)

[6] T. Iida and P.I.L. Guthrie, "The Physical Properties of Liquid Metals", Clarendon Press, Oxford (1998) pp.109-198

[7] W.D. Harkins and H. F. Jordan, J. Am. Chem. Soc, .52, 1751 (1930).

[8] Siber Hegner Company, Korea,

"www.siberhegner.com".

[9] S.Y. Kim, B.Y. Hur, C.K. Kwon, D.K. An and S.H. Park, Proceedings of the 65<sup>th</sup> World Foundry Congress, 499 (2002).

[10] S.H. Park, K.H. Song, Y.S. Um and B.Y. Hur, Materials Science Forum, **510-511**, 742-745 (2006)

[11] S.H. Park and B.Y. Hur, Materials Science Forum, **486-487**, 464-467 (2005)

[12] S.H. Park, U.S. Um and B.Y. Hur, Solid State Phenomena, **116-117**, 656-660 (2006)

[13] J.W. Gibbs, The Collected Works of J.W. Gibbs, Vol.1, "Thermodynamics", Yale Univer. Press, New Haven, CT, (1948), pp.300

[15] D. Skupien and D.R. Gaskell, Met. & Mater., Trans.B, **31**, 921 (2000)

[16] A. Pamies, C.G. Cordovilla and E. Louis, Scripta Materialia, **18**, 869 (1984)

[17] J.P. Anson, R.A.L. Drew and J.E. Gruzleski, Met. & Mater., Trans. B, **30**, 1999 (1998),.

[18] I. Egry, J. Mater. Sci., 26, 2997 (1991)

(Received December 28, 2006;Accepted May 15, 2007)