Letter to the Editor

An improved method for measuring surface tension of liquid metals and alloys

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Abstract

According to the maximum bubble pressure method, a method for fast measuring of surface tension of liquid metals and alloys has been improved. By experiments and analysis, a relationship between surface tension and such parameters as the pressure difference in bubbles, the number of bubbles in a fixed time, and the capillary diameter has been established, according to which surface tension of liquid metals and alloys can be obtained directly. Experimental results show that there are small deviations between the measured values and the reference values of the surface tension.

Key words: surface tension, improved method, liquid metals and alloys, fast measurement

1. Introduction

Surface tension is one of important physicochemical properties of high temperature liquid metals and alloys [1, 2]. It is the foundation of studying the interface dynamics of liquids [3, 4], it also plays a very important role in optimizing and simulating crystal growth parameters [5], generating electro meniscus phenomenon [6], evaluating mould filling capacity, graphite shape and surface roughness [7, 8]. Therefore, metallurgists and physicists have been seeking an effective method for fast measuring surface tension of liquid metals and alloys for a long time. Up to now, although many methods have been invented [9–11], the most widely used one in foundry is the maximum bubble pressure method. However, to precisely measure the surface tension, the dipping depth of the capillary must be controlled strictly. Besides, the time of generating a spherical bubble is much longer than two minutes, and more important is that it is difficult to generate the spherical bubble successfully [12]. So, it is not adequate for fast measurement. On the contrary, to reduce the influence of external factors, surface tension of liquid metals and alloys must be measured quickly. Furthermore, shorter measurement period can increase the number of measurements in a finite time, and thus statistically improve the measuring precision. Therefore, there is a need to develop an effective method.

2. Methodology

Already 150 years ago, the maximum bubble pressure method was proposed by Simon to measure the surface tension. In this method, pressurized gas is blown slowly in the liquid metals and alloys through a capillary and the pressure in the bubble is measured. Surface tension σ can be calculated [13] by the maximum pressure $P_{\rm m}$, the capillary radius r, the dipping depth h of the capillary and the density ρ of liquid metals and alloys.

However, the dipping depth of the capillary must be fixed strictly and measured exactly, and the speed of blowing bubbles must be very slow. To avoid these two disadvantages, an improved method for measuring surface tension of liquid metals and alloys has been developed. In this improved method, the blow speed in the liquid metals and alloys through the capillary is relatively fast compared with that in the maximum

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Fig. 1. Relationship between the pressure in the bubble and the time of blowing gas.

bubble pressure method, and the dipping depth of the capillary can be altered in a fixed range. In addition, the measuring period is shorter and the operation becomes easier.

Its basic principle is as follows. Pressurized inert gas is blown in the liquid metals and alloys through a capillary tube at a relative fast speed that the bubble can form and vanish continuously, and the pressure in the bubble is monitored by computer. The relationship between the pressure in the bubble and the time of blowing gas is shown in Fig. 1. The top shape of the pressure curve is like the sinusoid when the speed of generating bubbles is stable. The numbers 1, 2, 3 and 4 denote four states of the bubble, respectively, namely, not blowing gas, not generating the bubble, the bubble with maximal curvature radius and the bubble escaping from the capillary. The direct factors of deciding the horizontal position of the sinusoid are the dipping depth h of the capillary and the density ρ of liquid metals and alloys. When the capillary is dipped deeper and the density of liquid metals and alloys is bigger, the horizontal position of the sinusoid will become higher because of the increment of the static pressure on the end of the capillary. During the course of generating bubbles, according to the Laplace's formula [14], the additional pressure caused by surface tension can be expressed as:

$$\Delta P = \sigma \left(\frac{1}{R_1} + \frac{1}{R_2} \right),\tag{1}$$

where ΔP is the additional pressure, or the amplitude of the sinusoid, R_1 and R_2 are two principal curvature radiuses of bubbles, and they have relation to the capillary diameter. Hence, besides surface tension σ of liquid metals and alloys, the additional pressure ΔP is affected by the capillary diameter ϕ_x . That is to say, the shape of the sinusoid is mainly influenced by the surface tension and the capillary diameter. When the blowing velocity is fixed, the greater the surface tension of liquid metals and alloys is, the bigger the amplitude is and the longer the period is. Because the amplitude and the period of the sinusoid reflect the additional pressure caused by surface tension and the speed of generating bubbles, respectively, they are taken as basic parameters calculating surface tension. When surface tension retains the same, the bigger the capillary diameter is, the bigger the amplitude is and the longer the period is. Therefore, the capillary diameter is also considered as a parameter. To simplify calculation, the period Δt of the sinusoid is replaced by the number N of bubbles in a fixed time.

According to the above three parameters, through experiments on many kinds of liquid metals and alloys, the basic model of calculating surface tension has been got, which is shown in the following equation:

$$\sigma = a \cdot \Delta P + b \cdot N + c \cdot \Delta \phi_x + d, \tag{2}$$

where $\Delta \phi_x$ is the difference between actual value ϕ_x and standard value ϕ_0 of the capillary diameter (defined as 2.00 mm, and it is replaced by the conversion result, 600, of the 10bit A/D converter when the data are acquired by computer), and a, b, c, d are coefficients determined by many experiments on many kinds of liquid metals and alloys.

When surface tension is measured, the capillary must be dipped in liquid metals and alloys, so it is disposable and it is indispensable that the capillary diameter be measured quickly every time. In this method, the pressure in the capillary before being dipped in liquid metals and alloys is used to represent the actual value of the capillary diameter, and the real-time pressure can be monitored by computer automatically. At the same time, during the blowing gas, the pressure curve can be recorded by computer, and thus the amplitude of the sinusoid and the number of bubbles in a fixed time can be got automatically. Accordingly, if these coefficients and constants of Eq. (2) had been determined, surface tension of liquid metals and alloys would be calculated instantaneously.

Besides surface tension of liquid metals and alloys and the capillary diameter, the factors that determine the amplitude and the period of the sinusoid are the dipping depth of the capillary and the blowing velocity. When surface tension of liquid metals and alloys and the capillary diameter retain the same, the bigger the dipping depth of the capillary is or the smaller the blowing velocity is, the greater the amplitude is and the less the number of bubbles in a fixed time is. Consequently, the surface tension calculated by Eq. (2) may be affected by the dipping depth of the capillary and the blowing velocity. On the condition of different dipping depth and blowing velocity, we have experimented on Hg at room temperature by using the capillary with standard value and the results are shown in Table 1. Experimental results, however, have

Table 1. Effect of the dipping depth and the blow velocity on the calculated surface tension (the capillary with standard value is used and the time of fast blow is 5 seconds)

		37	ϕ_x	$\sigma \ ({ m mN}$		
(mm)	ΔP	IN		(Calculated value)	(Reference value)	(%)
$ \begin{array}{r} 6\\ 9\\ 12\\ 15\\ 18\\ 21\\ 24\\ 27\\ \end{array} $	$ 103 \\ 106 \\ 119 \\ 125 \\ 139 \\ 161 \\ 190 \\ 208 $	$18.4 \\ 15.2 \\ 14.6 \\ 13.2 \\ 11.4 \\ 9.7 \\ 7.8 \\ 6.9$	600	503 473 483 475 474 483 498 511	480	$\begin{array}{c} 4.8 \\ -1.5 \\ 0.6 \\ -1.0 \\ -1.3 \\ 0.6 \\ 3.8 \\ 6.5 \end{array}$
15	194 192 158 137 126 120 93 86	$7.5 \\ 8.6 \\ 10.2 \\ 11.6 \\ 13.2 \\ 14.8 \\ 15.7 \\ 17.2$	600	$500 \\ 509 \\ 484 \\ 473 \\ 477 \\ 486 \\ 462 \\ 470$	480	$\begin{array}{c} -9.6 \\ 6.0 \\ 0.8 \\ -1.5 \\ -0.6 \\ 1.3 \\ -3.8 \\ -2.1 \end{array}$

shown the deviation of surface tension between the calculated values and the reference values [15] will be below 1.5 % by using appropriate coefficients a and b (a = 1.2294 and b = 10.5368) when the dipping depth is over the range of 10–20 mm and the blowing velocity is 2–3 bubbles per second. So, the dipping depth of the capillary and the blowing velocity can be eliminated from Eq. (2) and their effect on calculating surface tension can be neglected.

3. Results and discussion

To obtain appropriate coefficients a, b, c, d of Eq. (2), we experimented on many kinds of liquid metals and alloys, i.e., Al at 933 K, Fe at 1803 K, Hg at room temperature, aluminium alloy over the 973–1013 K temperature range, graphite cast iron over the 1603–1683 K temperature range, magnesium alloy over the 1013–1053 K temperature range, etc. Because surface tension of different liquid metals and allovs will differ, actual surface tension is measured by RTW-08 Melt Physical Property Analyzer (the surface tension is measured according to the ring detachment method, the measurement range is at 200- $2000 \,\mathrm{mN} \,\mathrm{m}^{-1}$ and the precision is ± 1.5 %). Also, three parameters calculating surface tension are fast measured automatically. Through regressive calculation and statistical analysis, the appropriate coefficients have been obtained, and the surface tension of liquid metals and alloys can be calculated by the following equation:

$\sigma = 1.2294 \cdot \Delta P + 10.5368 \cdot N + 0.2031 \cdot \Delta \phi_x +$ + 182.6371.(3)

We know from experimental results, that surface tension of the measured liquid metals and alloys is from $276 \,\mathrm{mN}\,\mathrm{m}^{-1}$ (the value of the modified aluminium magnesium alloys at 1013 K to 1862 mN m^{-1} (the value of pure iron at 1803 K). Therefore, Eq. (3) can be applicable for the liquid metals and alloys with surface tension over the $280-1860 \,\mathrm{mN} \,\mathrm{m}^{-1}$ range. A device has been constructed according to this improved method and Eq. (3). With this device the surface tension of liquid metals and alloys can be measured in five seconds. The comparison of surface tension measured by this device and its reference values, which are measured by RTW-08 Melt Physical Property Analyzer, is shown in Table 2. The results of both Hg and graphite cast iron show small deviations from the reference values.

4. Conclusions

In this work, we have proposed an improved method for measuring the surface tension of liquid metals and alloys. By this method, the surface tension can be calculated by the sum of different contributions from the pressure difference in bubbles, the number of bubbles in five seconds and the capillary diameter, and the appropriate coefficients have been obtained by experiments. Comparative experiments show that there are small deviations between the measured values and the reference values of surface tension.

Olisata	<i>T</i> (K)	ΔP	Ν	ϕ_x	$\sigma~({\rm mN~m^{-1}})$		D : /:
Objects					(Calculated value)	(Reference value)	(%)
Hg	293 296 298 298	$117 \\ 139 \\ 147 \\ 122$	$13.5 \\ 12.9 \\ 12.6 \\ 13.4$	$638 \\ 561 \\ 544 \\ 623$	476 482 485 479	482 474 480 486	-1.2 1.5 1.0 -1.4
Graphite cast iron	$1664 \\ 1672 \\ 1675 \\ 1678$	744 696 803 875	$11.5 \\ 12.4 \\ 10.8 \\ 10.6$	$712 \\ 541 \\ 607 \\ 589$	$1241 \\ 1157 \\ 1285 \\ 1368$	$1228 \\ 1176 \\ 1264 \\ 1357$	$1.1 \\ -1.6 \\ 1.7 \\ 0.8$

T a b l e 2. Comparison of the measured surface tension and reference values

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References

- [1] BOCHNAK, R.: Kovove Mater., 27, 1989, p. 281.
- [2] PARADIS, P. F.—ISHIKAWA, T.—YODA, S.: J. Appl. Phys., 97, 2005, p. 53506.
- [3] BAIDAKOV, V. G.—CHERNYKH, G. G.—PRO-TSENKO, S. P.: Chem. Phys. Lett., 321, 2000, p. 315.
- [4] KRUSINA, J.—MYSLIVEC, T.: Kovove Mater., 16, 1978, p. 510.
- [5] YILDIZ, M.—DOST, S.: Cryst. Res. Technol., 42, 2007, p. 914.
- [6] MATSUURA, H.—TANIKAWA, T.—FURUKAWA, H.: Jpn. J. Appl. Phys., 43, 2004, p. L602.

- [7] SHI, D.—LI, D.—GAO, G.—WANG, L.: Kovove Mater., 46, 2008, p. 59.
- [8] SAKURAGI, T.: Int. J. Cast Met. Res., 18, 2005, p. 202.
- [9] MATSUMOTO, T.—NAKANO, T.—FUJII, H.—KA-MAI, M.—NOGI, K.: Phys. Rev. E, 65, 2002, p. 31201.
- [10] ROACH, S. J.—HENEIN, H.: Metall. Mater. Trans. B, 36, 2005, p. 667.
- [11] ISHIKAWA, T.—PARADIS, P. F.—YODA, S.: Appl. Phys. Lett., 85, 2004, p. 5866.
- [12] CHRISTOV, N. C.—DANOV, K. D.—ANANTHA-PADMANABHAN, K. P.—KRALCHEVSKY, P. A. —LIPS, A.: Langmuir, 22, 2006, p. 7528.
- [13] MOODY, M. P.—ATTARD, P.: Phys. Rev. Lett., 91, 2003, p. 56104.
- [14] ONISCHUK, A. A.—PURTOV, P. A.—BAKLANOV, A. M.—KARASEV, V. V.—VOSEL, S. V.: J. Chem. Phys., 124, 2006, p. 14506.
- [15] LANG, G.: J. Inst. Met., 101, 1973, p. 300.