International Journal of Heat and Fluid Flow 000 (2016) 1–6

Contents lists available at ScienceDirect

International Journal of Heat and Fluid Flow

journal homepage: www.elsevier.com/locate/ijhff

Measurement of dynamic surface tension for liquid metal by capillary jet method

Tatsuro Wakimoto[a,*], Kenji Katoh[a], Yoshiaki Ueda[b], Manabu Iguchi[c]

[a] Department of mechanical and physical engineering, Graduate school of engineering, Osaka City University, 3-3-138 Sugimoto, Sumiyoshi-ku, Osaka 558-8585, Japan
[b] Department of mechanical engineering, Setsunan University, Neyagawa 572-8508, Japan
[c] Department of mechanical engineering, Osaka Electro-Communication University, Neyagawa 572-8530, Japan

ARTICLE INFO

Article history:
Available online xxx

Keywords:
Surface tension
Jet
Capillary
Liquid metal
Oxidization
Dynamic surface tension

ABSTRACT

Most liquid metals are easily oxidized, causing their surface tensions to vary over time. In this study, a novel method for measuring the dynamic surface tension of a liquid metal during oxidization is proposed. This method is based on the fact that the descent trajectory of a capillary jet ejected horizontally from a small orifice at slow speed depends on the surface tension as well as inertial and gravitational forces. We derive a theoretical model to predict the jet trajectory and determine the dynamic surface tension of the liquid metal by matching the predicted theoretical trajectory to the experimentally measured one. Actual measurements for Wood's alloy at various oxygen concentrations demonstrate that the surface tension decreases to an equilibrium value on a time scale of 10–11 ms at oxygen densities of less than 1.5%, whereas it increases on a time scale of 10–17 ms at greater oxygen densities. This method has the ability to measure dynamic surface tension on a time scale of 1.5–55 ms at an accuracy of ±1.5 ms, making it suitable for measuring the dynamic surface tension of liquid metal in an oxidation process.

1. Introduction

The surface tensions of liquid metals decrease rapidly upon exposure to environments such as oxygen because oxygen molecules are adsorbed to the liquid metal's surface (Egry et al., 2010; Ricci and Passerone, 1993). It takes a certain amount of time before the surface tension reaches an equilibrium value. The temporary change in surface tension, which is referred to as the dynamic surface tension, has a great influence upon various techniques related to liquid metals such as metallurgy, solder-joint formation, and metal–particle production processes (Hayes et al., 1993; Las and Chen, 2005). Several studies have investigated the equilibrium surface tensions of liquid metals at various oxygen concentrations (Egry et al., 2010). However, very little work is currently available in the published literature on the dynamic surface tension of liquid metals during oxidation.

Some methods for measuring dynamic surface tension have been developed, particularly for surfactant solutions that also have time-dependent surface tensions for adsorption of surfactant molecules to the liquid surface. One commonly used is "maximum bubble pressure method", which is based on the relationship between surface tension and bubble pressure. When gas is injected into a liquid bath through a capillary tube, a bubble is formed repeatedly at the tube's end. The dynamic surface tension is then obtained from the maximum value of oscillating gas pressure and the repetition frequency of bubble generation. Another representative is "oscillating jet method" . If liquid is ejected downward from an elliptical nozzle exit, the long and short axes of the cross-section of the elliptical jet alternate in the downward direction because of surface tension. Since a fresh liquid surface is created at the nozzle exit and advected downstream, the dynamic surface tension is obtained from the wavelength of the oscillating jet and the downward length from the nozzle exit. However, a highly-responsive precise pressure sensor is required in the former method, and accurate determination of the wavelength is difficult in the latter method. These disadvantages are more serious for high-temperature molten metals.

Howell et al. (2004) measured the dynamic surface tension of molten Sn/Pb solder in nitrogen atmosphere containing oxygen at 1% concentration using the oscillating jet method and revealed that the surface tension varies within several milliseconds. However, in their measurement, the measurable time scale was limited within 1–2.5 ms and the achievement of equilibrium surface tension was unconfirmed as observing the decaying oscillating jet with small

* Corresponding author.
E-mail address: wakimoto@mech.eng.osaka-cu.ac.jp (T. Wakimoto).

http://dx.doi.org/10.1016/j.ijheatfluidflow.2016.12.007
0142-727X/© 2017 Published by Elsevier Inc.

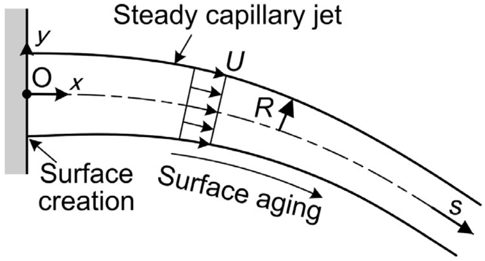

Fig. 1. Schematic of the steady capillary jet.

amplitude was difficult. A simple and novel measurement method for dynamic surface tension with a longer measurable time scale is needed.

Therefore, we propose a method using a capillary jet, as schematically illustrated in Fig. 1. Assuming the jet ejects horizontally at a velocity $u < 1$ m/s from a circular orifice with radius $R_0 \approx 1$ mm, typical physical properties of most liquid metals (e.g., density $\rho \approx 10^4$ kg/m^3, surface tension $\sigma \approx 500$ mN/m) leads to unity order for the Weber number ($\mathrm{We} = \rho u^2 R_0/\sigma$) and Bond number ($\mathrm{Bo} = \rho g {R_0}^2/\sigma$). Here, g is the gravitational acceleration. This implies that the descent trajectory of the jet is affected by not only inertia and gravity but also surface tension.

In addition, since the liquid surface of the jet is freshly created at the orifice exit and ages as it travels downstream, the trajectory depends on the dynamic surface tension. To predict the trajectory of the jet, we developed a theoretical model based on a force balance among inertia, pressure, gravity, and surface tension. Dynamic surface tension is determined based on the condition in which the theoretically predicted trajectory agrees with an experimentally measured one.

In a previous report, we applied this method to surfactant solution and verified that the dynamic surface tension measured by this method is coincident with those measured by the abovementioned conventional methods (Katoh et al., 2009; Wakimoto et al., 2010). In this study, we demonstrate that this method is also applicable to liquid metals using actual measurements of the dynamic surface tension of Wood's alloy, which has a low-melting point.

2. Theoretical analyses for predicting the jet trajectory

In this section, we describe the theoretical analyses used to predict jet trajectory. We consider a capillary jet with local velocity U and radius R that is ejected horizontally with a uniform velocity profile from a circular orifice, as illustrated in Fig. 1. The coordinate origin O is defined as the intersection point of the central axis of the jet and the orifice exit. The horizontal and vertical coordinates are represented by x and y, respectively, and a coordinate along the central axis of the jet is expressed by s. Since the liquid surface is created freshly at the orifice exit and then advected downstream, the surface tension at an arbitrary point with coordinate s depends on the travel time from the orifice exit.

Assuming a capillary jet, the capillary number $\mathrm{Ca} = \rho \nu U/\sigma$, which represents the ratio of viscous force to surface tension, becomes much lower than unity for the typical physical properties of liquid metal (e.g., kinematic viscosity $\nu \approx 10^{-6}$ m^2/s, $\rho \approx 10^4$ kg/m^3, and $\sigma \approx 500$ mN/m) and for the jet velocity ($U < 1$ m/s) in the experiment described below. This quite small capillary number indicates negligible viscous force.

Fig. 2 shows a small control volume of a jet with length ds. The geometric relation between the jet radius R and the its divergence angle α yields the following formula:

$$\frac{dR}{ds} = \tan\alpha. \tag{1}$$

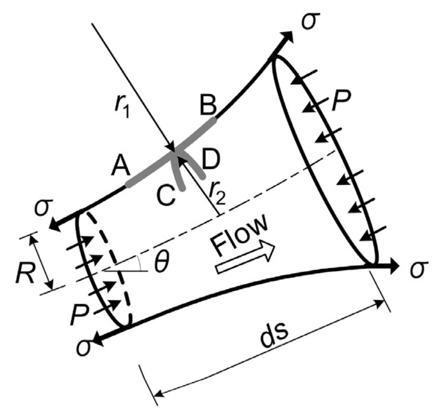

Fig. 2. Element of the capillary jet.

We consider the variation in the momentum of the jet and the forces acting on the control volume. Assuming that the cross-section of the jet is circular, the momentum flux M through the control volume is estimated as

$$M = \pi R^2 \rho U^2 = \frac{\rho Q^2}{\pi R^2}, \tag{2}$$

where Q is the volumetric flow rate.

The pressure P acting on the cross-section of the control volume is derived from the following estimation. As illustrated in Fig. 2, by defining r_1 as the curvature radius of the outline A–B of an upper liquid surface along the flow direction and r_2 as the other curvature radius of the outline C–D appearing in the plane perpendicular to outline A–B, the gauge pressure P_U on the upper liquid surface can be obtained from Laplace's equation,

$$P_U = \sigma\left(\frac{1}{r_1} + \frac{1}{r_2}\right) = \sigma\left\{-\cos\alpha\frac{d(\theta+\alpha)}{ds} + \frac{\cos\alpha}{R}\right\}, \tag{3}$$

where θ is the oblique angle of the jet. In a similar manner, the gauge pressure P_L on the lower surface is given by

$$P_L = \sigma\left\{\cos\alpha\frac{d(\theta-\alpha)}{ds} + \frac{\cos\alpha}{R}\right\}. \tag{4}$$

P_U and P_L are nearly identical since r_1 (≈10 mm) becomes larger than r_2 (≈1 mm) under the experimental conditions of this study. By averaging Eqs. (3) and (4), the gauge pressure P in the jet can be estimated as

$$P = \sigma\left(-\cos\alpha\frac{d\alpha}{ds} + \frac{\cos\alpha}{R}\right). \tag{5}$$

We can obtain the following equations to describe the balance between the momentum variation and the forces owing to pressure, surface tension, and gravity acting on the control volume in the directions along s and perpendicular to s:

$$\frac{dM}{ds} = -\frac{d}{ds}\left(\pi R^2 P + 2\pi R\sigma\cos\alpha\right) - \pi R^2 \rho g \sin\theta \tag{6}$$

and

$$M\frac{d\theta}{ds} = -\pi R^2 P\frac{d\theta}{ds} + 2\pi R\sigma\cos\alpha\frac{d\theta}{ds} - \pi R^2\rho g\cos\theta. \tag{7}$$

To solve Eqs. (6) and (7), the elapsed time t_s from the initial surface formation and the dynamic surface tension σ, which depends on t_s, are required at coordinate s. The surface age t_s is related to the jet velocity U as follows:

$$\frac{dt_s}{ds} = \frac{1}{U} = \frac{\pi R^2}{Q}. \tag{8}$$