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(Review)

Interfacial Phenomena and Thermophysical Properties of Molten Steel and Oxides
- Fundamental Research of Steel Processing using Electrostatic Levitation Furnace (ELF)-

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Abstract

In the present-day steel processing, interfacial phenomena between molten steel and oxides, which are called slag or mold flux, play important roles in materials design. Therefore, interfacial tension must be known for process controlling. From this requirement, we proposed the interfacial tension measurement technique between molten steel and oxides using the modified oscillating drop method with levitation techniques. The interfacial tension data using traditional technique based on the sessile drop method have been obtained only at melting temperature of iron due to dissolution of containers and the substrate into molten steel and oxides in higher temperature regions. The technique of our proposal to technique to obtain temperature dependence of interfacial tension between molten iron and oxides uses a core-shell form droplet including interface between two liquids using the electrostatic levitation, which can achieve the containerless conditions. The measurements are performed in International Space Station using the electrostatic levitation furnace (ELF) in KIBO.

Keyword(s): Thermophysical Property, Interfacial Tension, Electrostatic Levitation, Liquid Droplet.
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1. Introduction

Immiscible two liquids make a core-shell droplet under the microgravity condition. The core-shell formation is dominated by the surface and interfacial free energies. This means that we can know the interfacial tension, which is the interfacial free energy per unit area, using the core-shell droplet. We can measure the surface tension from the surface oscillation frequencies using the levitated liquid droplets, which is called surface oscillation technique. Based on the ideas, we have tried to measure the interfacial tension of immiscible high-temperature liquids using core-shell droplet by the surface oscillation technique. The interfacial tension measurement is required from the industrial applications, especially in the steel industries, such as the smelting, the continuous casting and the welding. In the continuous casting process, mold flux made of molten oxides covers steel melts in the casting pool. In the casting pool, the interfacial tension prevents pulling down the oxide into the steel melts. In the welding process, the welding flux made of oxide encloses steel melts in order to prevent oxidation of steel melts and in order to control welding part shape. For welding part shape controlling the interfacial tension between welding flux and steel melts plays important rules. In these processes, the interfacial tension is empirically changed by the oxide composition change. Therefore, in order to control systematically the interfacial tension between molten oxide and steel melts, the interfacial tension values between many kinds of molten oxides and steel melts must be obtained[1)]. However, oxide compositions were limited due to temperature in the previous measurements using a sessile drop with x-ray radiograph method[2)], since it is difficult to raise temperature over the melting point of iron with the conventional method using container.

Therefore, we planned the measurements of the interfacial tension between molten oxides and steel melts using core-shell droplet with oscillating drop technique under microgravity conditions[3)]. For this plan of the interfacial tension measurement, we must use the containerless and noncontact method. We must select the electrostatic levitation method to achieve both requirements for the measurement of interfacial tension between molten oxide and steel melts formed core-shell

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droplet by the oscillating drop technique. The electrostatic levitation furnace (ELF) [4], will be installed in the International Space Station (ISS), KIBO during 2015, we can use this facility for our measurement plan. In this paper, we review our research project using ELF in ISS, KIBO for the interfacial tension between molten oxides and steel melts.

2. Basis of Oscillating Drop Technique using Core-Shell Droplet

2.1 Normal Mode Analysis of Core-Shell Droplet

Under the microgravity conditions, immiscible liquids form core-shell droplet. The surface oscillation of core-shell droplet is analyzed from the equation of motions[5]. Under the conditions of core-shell droplets dealing with incompressible and non-viscous fluids, the equation of motion of core-shell droplet describes following equations using velocity potential in spherical coordinates $\psi(r,\theta,\phi,t)$;

$$\rho\frac{\partial^2\psi}{\partial t^2}=\frac{\sigma}{R}\left(2\frac{\partial\psi}{\partial r}+\frac{\partial}{\partial r}\hat{L}\psi\right) \tag{1}$$

$$\hat{L}=\frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right)+\frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\phi^2} \tag{2}$$

For core and shell liquids velocity potentials ψ_1,ψ_2 and outer atmosphere velocity potential ψ_3 are given by following using boundary condition of continuous at two interfaces;

$$\begin{aligned}\psi_1(r,t)&=\left[A(t)r^l+B(t)r^{-(l+1)}\right]\mathrm{Y}_l^m(\theta,\phi)\\ \psi_2(r,t)&=A_2(t)r^l\mathrm{Y}_l^m(\theta,\phi)\\ \psi_3(r,t)&=B_3(t)r^{-(l+1)}\mathrm{Y}_l^m(\theta,\phi)\end{aligned} \tag{3}$$

$$A_2(t)=A(t)-\frac{l+1}{l}B(t)R_1^{-(2l+1)},B_3(t)=B(t)-\frac{l}{l+1}R_2^{(2l+1)} \tag{4}$$

Here, $\mathrm{Y}_l^m(\theta,\phi)$ is spherical harmonic function, R_1,R_2 are core and shell radius respectively, (l,m) is mode of oscillation. From the equation of motion and velocity potentials, we obtained the conditions of oscillation frequencies,

$$\omega_\pm^2=K_\pm\left(W/J\right) \tag{5}$$

$$K_\pm=\frac{1}{2}\left(\frac{\sigma m_1}{\tau^3}+\frac{m_2\tau^3}{\sigma}\right)\pm\sqrt{\frac{1}{4}\left(\frac{\sigma m_1}{\tau^3}-\frac{m_2\tau^3}{\sigma}\right)^2+1} \tag{6}$$

$$\frac{W}{J}=\frac{\omega_o^2\tau^8}{\sigma}\frac{1}{(1+\Delta\rho)\tau^{10}+(2/3)\Delta\rho} \tag{7}$$

In eqs. (6) - (7), parameters are follows,

$$\tau=\sqrt{\frac{R_2}{R_1}},\quad \sigma=\sqrt{\frac{\sigma_2}{\sigma_{12}}},\quad \Delta\rho=\frac{3}{5}\frac{\rho_1-\rho_2}{\rho_2} \tag{8}$$

$$m_1=\left(1+\Delta\rho\right)\tau^5-\Delta\rho\tau^{-5},\quad m_2=\frac{3}{5}\tau^5+\frac{2}{5}\tau^{-5} \tag{9}$$

Here, ρ_1,ρ_2 are density of core and shell liquids respectively, σ_2 is surface tension of shell liquids and σ_{12} is interfacial tension between core and shell liquids. Also $\omega_o^2=8\sigma/\rho R^3$ is normal mode frequency of single droplet derived by Rayleigh[6]. In the frequency conditions, two frequencies must exist. Using two frequencies, we calculate the equations (5)-(7), and finally we obtained surface tension values σ_2 and interfacial tension values σ_{12}. From observed two frequencies, important parameters for determination of interfacial tension values are the ratio of core and shell radius and the density difference between core and shell liquids.

2.2 Numerical simulation of core-shell droplet

In analytical studies of the surface oscillation of core-shell droplet, since core and shell liquids are assumed as the non-viscous fluids, we cannot know how the viscosity affects the surface oscillations. Because viscosity of molten oxides is large and changes with compositions, we performed numerical

Table1 Physical properties and geometrical parameters used in numerical simulations [7]

Physical properties	Iron melt	Molten slag
Density ρ_i[kg/m^3]	7.03×10^3	2.85×10^3
Viscosity μ_i[mPa.s]	5.50	21.4, 107, 214
Surface tension σ_i[N/m]	1.76	0.450
Interfacial tension σ_{12}[N/m]	1.30	
Geometrical parameters		
Equilibrium radius R_i[mm]	0.549, 0.618, 0.706, 0.760, 0.823	0.988
Initial drop shape $(b_i/a_i)_0$[-]	1.20, 1.50	

simulations to know how the surface oscillation changes with changing the viscosity of molten oxides and also the ratio of shell and core liquids[7]. For the numerical simulations, we solved Navier-Stokes equations without external force, and Volume of Fluid (VOF) function was used for the interface motions. Input parameters are listed in **Table 1**. Core part values are used for pure iron values obtained by our experiments using electromagnetic levitations. Shell part of oxides values are not experimental values. We use the average values of literature values of SiO_2-CaO-Al_2O_3, smelting slag. **Figure 1** shows radius change with time for core and shell liquids with different ratio. In this results, viscosity ratio of shell to core liquids is fixed 3.9. **Figure 2** shows the power spectrums of surface oscillations shown in **Fig. 1**. From these power spectrums, we can find two peaks' appearance and also we can find that two peaks' intensity depends on the radius ratio. Through numerical simulations with different conditions of viscosity and radius ratios, we found that optimized conditions to observe precisely two peaks of surface oscillation of core-shell droplet is viscosity ratio below 40 and radius ratio 1.3.[7]

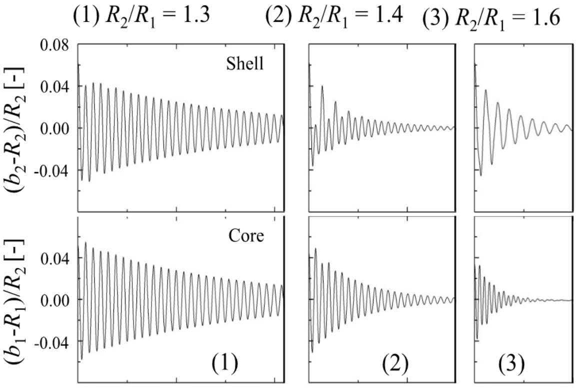

Fig. 1 Effects of radius ratio of shell to core liquids on oscillation behaviors of compound droplets for the case of viscosity ratio of 3.9[7].

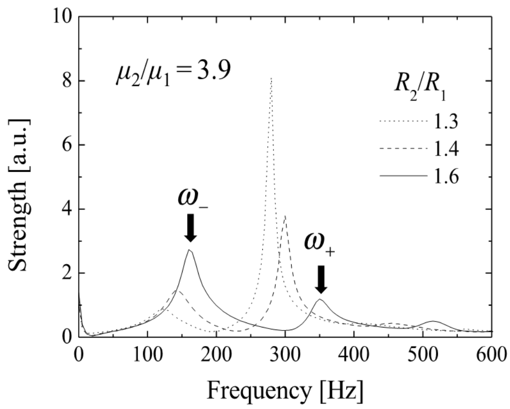

Fig. 2 Effect of radius ratio of shell to core liquids on frequency spectrum of a compound droplet for the case of viscosity ratios of 3.9[7].

3. Short Time Microgravity Experiments by Parabolic Flight

We must confirm formation of core-shell droplet by molten oxides and steel melts under the microgravity conditions before on-orbit experiments in ISS. For the requirements, we observed the formation of core-shell droplet under the microgravity condition by parabolic flight experiments using Gulfstream-II airplane operated by Diamond Air Service (DAS). On the parabolic flight experiments, we cannot use the electrostatic levitation furnace because it is difficult to keep the sample position during parabolic flight due to rapid change of the gravity level. Therefore, we used electromagnetic levitation to

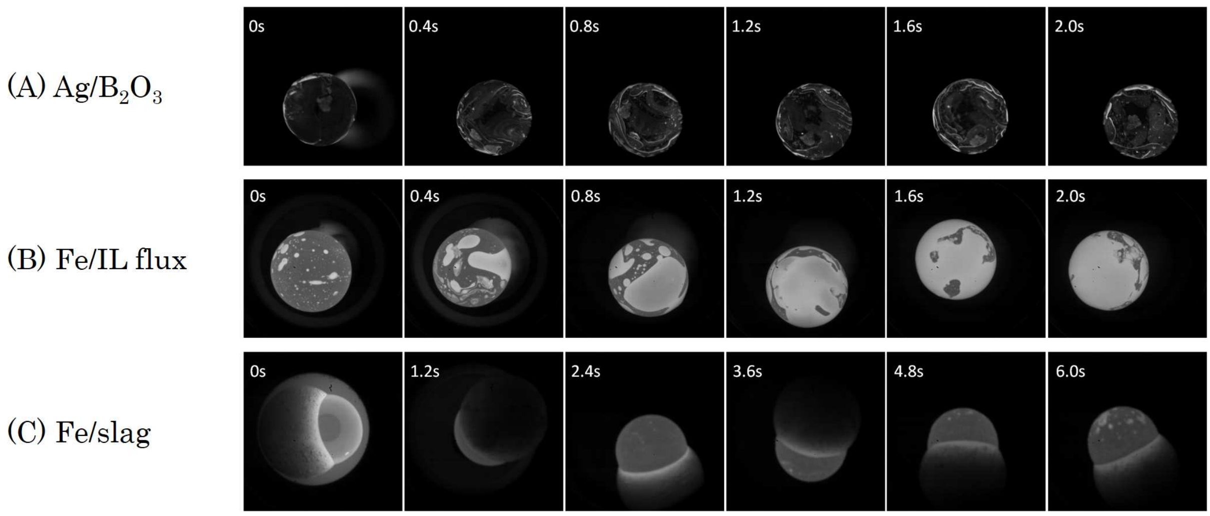

Fig. 3 Top views of electromagnetically levitated metal/oxide sample under μG condition. (A) Ag/B_2O_3, (B)Fe/Ilmenite type welding flux, (C)Fe/Smelting slag (SiO_2-CaO-Al_2O_3 base) [8].

observe the formation of core-shell droplet by molten oxide and steel melts. We newly developed the compact size electromagnetic levitation facilities specialized for the parabolic flight experiments by G-II airplane[8]. Using the facilities, we succeed to observe the formation of core-shell shape droplet by molten oxides and iron melts. **Figure 3** shows time-series images of the formation of core-shell droplet by molten oxide and metal liquids taken by high-speed camera under microgravity during parabolic flight[8]. **Figure 3 (A)** is a reference experiment using molten B_2O_3 and Ag melts. Since melting temperature of these samples is below 1000 K, we used these samples for trial experiments under microgravity conditions by parabolic flight to check out the new facilities. **Figure 3(B) and(C)** are results of molten oxide and Fe melts. Molten oxides in **Fig. 3 (B)** are commercially used welding flux of the ilmenite type, main compositions of Al_2O_3, CaO and Fe_2O_3. On the other hand, molten oxides in **Fig. 3 (C)** are smelting slag compositions of Al_2O_3-CaO-SiO_2. The case of molten oxides by welding flux of ilmenite type covered fully over the iron melts and formed core-shell droplet as shown in **Fig. 3 (B)**. On the other hand, the case of molten oxides of smelting slag covered with the half area of iron melts and therefore did not form core-shell droplet as shown in **Fig. 3 (C)**. The difference is attributed by the interfacial tension differences. Since the interfacial tension of smelting flux to iron melts are relatively high, the free energy contributed from the surface and the interface is large in the present experimental conditions with large volume of iron melt due to the electromagnetic induction efficiency. This means that we must optimize the radius ratio of core-shell droplet by minimizing the free energy contributed from the surface and the interface. From the parabolic flight experiments, we obtained important information of core-shell droplet to prepare the samples for the on-orbital experiments in ISS.

4. Sample Preparation for on-orbital Experiments

Based on these results of core-shell droplet phenomena, we selected 15-types sample for on-orbital experiments in ISS of interfacial tension measurements. Our experiments in ISS have two series period to achieve final results of interfacial tension between molten oxide and steel melts. In first series of experiments, density, viscosity and surface tension of molten oxides will be obtained in order to predict the interfacial tension before on-orbital experiments. The samples of first series on-orbital experiments in ISS are listed in **Table 2**. Molten-oxide samples are selected the refining slag based on Al_2O_3-CaO with small amount of SiO_2. Since the slag viscosity is relatively low, using these slags as molten oxides the conditions of viscosity ratio below 40 will be kept. From the numerical simulations and the parabolic flight experiments, we can decide the volume ratio of oxide and iron for the core-shell droplet. For the first series samples of core-shell droplet using ilmenite-type welding flux based oxide and iron, we prepared the radius ratio of 1.3. The ilmenite-type welding flux based oxides are predicted relatively low viscosity and interfacial tension. Thus the radius ratio was decided mainly from the viscosity effect predicted from the numerical simulations. We prepared 15-type samples in one cartridge, shown in **Fig. 4 (c)**, and three same cartridges for repeating measurements of three times. **Figure 4 (a)** is example of oxide samples and **Fig. 4 (b)** is example of core-shell droplet samples. Our three cartridges containing oxide and core-shell droplet samples succeeded launching on December 6, 2015 using Cygnus CRS OA-4 from Cape Canaveral Air Force Station. On-orbital experiments in ISS of the interfacial tension measurements will start from March, 2016.

5. Conclusion

We started research project of the interfacial tension measurements using core-shell droplet with the electrostatic levitation under long-time microgravity conditions in ISS. The measurement of interfacial tension between molten oxides and steel melts required from the steel industry applications. The

Table 2 Sample list for first series on-orbital experiments

		SiO2	Al2O3	CaO	MgO	MnO	TiO2	FeO
①	Refining Slag_1	10.0	40.0	50.0				
②	Refining Slag_2	10.0	35.0	55.0				
③	Refining Slag_3	10.0	30.0	60.0				
④	Refining Slag_4	14.0	36.0	50.0				
⑤	Refining Slag_5	10.0	44.0	46.0				
⑥	Refining Slag_6	20.0	30.0	50.0				
	Welding Flux							
7	Ilmenite type	25.0		7.0		23.0	18.0	27.0
8		36.0		6.0		21.0	15.0	22.0
9		25.0		7.0		20.0	18.0	30.0
10	High TiO2 Type	27.0		7.0		13.0	53.0	
11		24.0		3.0		9.0	64.0	
12		22.0		12.0		21.0	45.0	
13	Fe･FeO							100.0
14	Fe･Ilmenite-type Flux	25.0		7.0		20.0	18.0	30.0
15	Fe･High TiO2-type Flux	22.0		12.0		21.0	45.0	

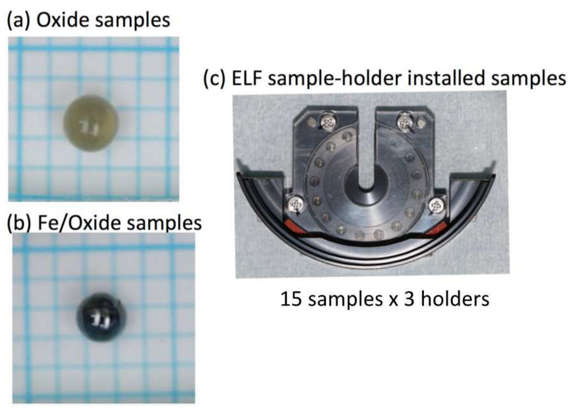

Fig. 4 Samples and sample holder for on-orbital experiments by ELF in ISS, KIBO. (a) Oxide sample, (b) Fe/oxide samples and (c) ELF sample holder.

measurement of interfacial tension between molten oxide and steel-melts uses the modified oscillating drop method using core-shell droplet levitated using the electrostatic levitation furnace in ISS. Since the modified oscillating drop method to measure the interfacial tension is challenging and is not precedent, we must prepare much attention to obtain precise data of interfacial tensions. For the preparation of on-orbital experiments in ISS, numerical simulations and short-time microgravity experiments by parabolic flight have been performed in order to understand the surface oscillation phenomena of core-shell droplet. Through these preparations, the samples of first-series on-orbital experiments in ISS were launched on December 6, 2015. For the first series experiment from March, 2016, we are refining the analysis of the surface oscillation of core-shell droplet combined with numerical simulations in order to obtain precise interfacial tension between molten oxide and steel melts.

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