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Interfacial tension of Fe–Si liquid at high pressure: Implications for liquid Fe-alloy droplet size in magma oceans

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ABSTRACT

The interfacial tension of Fe–Si liquid was measured using *in situ* X-ray radiography at high pressure and temperatures using the sessile drop method. The interfacial tension of Fe–Si liquid decreases (from 665 to 407 mN/m) with increasing temperature (1673–2173 K) at 1.5 GPa. The interfacial tension also decreases gradually with increasing Si content (0–25 at%), suggesting that Si behaves as a "moderately" surface-active element. Comparing the effects of different light elements on the interfacial tension of liquid iron, the most effective elements for reducing the interfacial tension lie in the order S > Si > P, although P has almost no effect. The droplet size of emulsified Fe-alloys in a magma ocean are estimated to be larger for Fe–Si and Fe–P liquids and smaller for Fe–S (S > 10 at%) liquid compared with that for pure Fe liquid. Therefore, if droplets in a magma ocean are enriched in S, chemical equilibrium between droplets and silicate melt is established faster in the magma ocean compared to Fe, Fe–Si and Fe–P liquids.

1. Introduction

Interfacial tension is a fundamental physical property that controls separation and chemical equilibration processes between two immiscible liquids and between liquids and solids. In the deep magma oceans that are considered to form during planetary accretion, liquid Fe-alloy droplets settle through silicate melt due to the density difference. The force balance between interfacial tension of the Fe-alloy droplet and viscous stress from the surrounding silicate melt controls the droplet size (Rubie et al., 2003; Ichikawa et al., 2010; Deguen et al., 2011). The size of Fe-alloy droplets in a magma ocean controls both the reaction rate between Fe-alloy and silicate melt and the time scale of metal/silicate separation.

The interfacial tension between Fe-alloy liquid and solid silicate also controls wetting behavior and thus determines whether liquid Fe-alloy can percolate through crystalline silicates on the grain scale (e.g. Terasaki et al., 2005, 2008a). Therefore, the interfacial tension of liquid Fe-alloy under high pressure is a crucial factor for understanding core formation processes in planetary interiors.

Surface/interfacial tensions of liquid Fe-alloys have been measured at ambient pressure using various techniques, such as the capillary method, the pendant-drop method, the maximum bubble pressure method and the sessile drop method (e.g. Murr, 1975). Because space and geometric configurations are highly limited in sample cell assemblies in high pressure experiments, the sessile drop method, which is based on liquid shape as controlled by a force balance between gravity and surface/interfacial tension (e.g. Rotenberg et al., 1983), is the only way to measure interfacial tension of liquid materials under high pressure (Terasaki et al., 2008b, 2009). In this study, we have measured the interfacial tension of Fe–Si liquid using the sessile drop method at high pressure and high temperature, combined with a synchrotron X-ray radiography technique. The aims were to investigate the effects of Si content and temperature on the interfacial tension of liquid Fe and to determine the droplet size of core-forming metallic liquid in a magma ocean.

2. Experimental

High pressure experiments were performed using a 1500 ton Kawai-type multi-anvil device (SPEED-1500) installed at the BL04B1 beamline of the SPring-8 synchrotron radiation facility in

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Japan (Utsumi et al., 2002). Starting materials were powdered mixtures of Fe (Rare metallic Co. Ltd., 99.9% purity) and FeSi (Kojyundo Chemical Lab. Co. Ltd., 99.9% purity) with the compositions $Fe_{82}Si_{18}$ and $Fe_{71}Si_{29}$ (in at%) that can explain the 10% density deficit of the Earth's core (Poirier, 1994). An Fe–Si pellet, 2.0 mm in diameter, was placed in a boron nitride (BN) capsule and Na-disilicate ($Na_2Si_2O_5$) glass powder was packed around the Fe–Si sample. We have used $Na_2Si_2O_5$ liquid to contain the liquid Fe–Si metal because it does not react with Fe-alloy liquids and its melting temperature is close to the eutectic temperature of the Fe-alloy. Heating was achieved using a cylindrical graphite furnace. Boron–epoxy was located at the X-ray path through the gasket to reduce X-ray absorption. The experimental temperature was monitored using a W3%Re-W25%Re thermocouple with the junction being located just above the BN capsule. Pressure was estimated using measured lattice constants of BN together with its equation of state (Urakawa et al., 1996). 26 mm tungsten–carbide anvil cubes with truncated edge lengths of 12 mm were used.

White X-rays were used for imaging and diffraction. The transmitted X-ray beam from the sample was converted to visible light using a YAG fluorescence screen of 150 μm thickness and was acquired as an X-ray radiographic image using a high-resolution charge-coupled device (CCD) camera (C9300-124, Hamamatsu Co. Ltd.). Exposure times were 300–400 ms and the pixel size of the radiographic images was 4.07–4.09 μm after 2 × 2 binning. In order to identify the state of the sample and to determine pressure, X-ray diffraction from the sample and the BN pressure marker was carried out using a Ge-solid state detector by the energy-dispersive method at a fixed angle of 5.5°. All experiments were performed at a pressure of 1.5 ± 0.3 GPa. X-ray radiography images of the sample were recorded at 100 K steps during heating from 300 to 2173 K. The recovered samples were mounted in epoxy and polished for chemical analysis. Chemical analysis was performed using an electron microprobe (JXA-8800, JEOL) with wavelength dispersive spectroscopy (WDS) installed at Tohoku University. Accelerating voltages of 20 kV for metal and 15 kV for silicate were used.

Details of the utilized sessile drop method are as follows. When liquid Fe-alloy rests on the smooth flat solid surface of a capsule in contact with overlying silicate liquid, the liquid Fe-alloy adopts the form of a rounded drop as a result of force equilibrium between gravity and surface/interfacial tension (Fig. 1). The pressure difference between the inside and outside of the Fe-alloy droplet is given by the classical Laplace equation as follows.

$$\Delta P = \gamma\left(\frac{1}{R_1} - \frac{1}{R_2}\right) \tag{1}$$

where ΔP is pressure difference across an interface, γ is the interfacial tension and R_1 and R_2 are the radii of curvature of the drop (for details see Mungall and Su, 2005). The contact angle (ϕ) between the Fe-alloy droplet and the bottom surface of the capsule is described by Young's equation expressed as follows.

$$\gamma \cos\phi = \gamma_{SB} - \gamma_{MB} \tag{2}$$

where γ_{SB} and γ_{MB} are the interfacial tension between silicate and the capsule base, and Fe-alloy and the capsule base, respectively. Combining these equations, the shape of the liquid droplet can be expressed by the following Bashforth–Adams equation:

$$\frac{R_0}{R} + \frac{R_0 \sin\phi}{x} = 2 + \Delta\rho g z \frac{R_0}{\gamma} \tag{3}$$

where x and z are the coordinates of the horizontal and vertical axes at an arbitrary point A on the interface between the Fe-alloy droplet and the surrounding silicate liquid. R_0 and R are the radii of curvature of the drop at the apex and point A, respectively. $\Delta\rho$, g and γ correspond to the density difference between liquid Fe-alloy and silicate liquid, gravitational acceleration, and the interfacial tension, respectively. Further details of this method are described elsewhere (Rotenberg et al., 1983; Terasaki et al., 2009). From the X-ray radiographic images (Fig. 1a), we obtain the shapes of the liquid Fe–Si droplets at high pressure and temperature (Fig. 1b). Thus, we can calculate the interfacial tension (γ) of Fe–Si liquid using Eq. (3). The contact angle (ϕ) was deduced from the height (H) of the sample droplet and the length (L) of the contact area between the droplet and the base of the BN capsule using the equation (Myochin et al., 1987):

$$\phi = 2\tan^{-1}\left(\frac{2H}{L}\right) \tag{4}$$

3. Results and discussion

A typical radiography image and derived forms of the liquid Fe–Si droplet interface are shown in Fig. 1. The obtained interfacial tensions (γ) and contact angles (ϕ) of Fe–Si liquid in $Na_2Si_2O_5$ liquid are summarized in Table 1 together with the experimental conditions. The pressure measured at the highest temperature

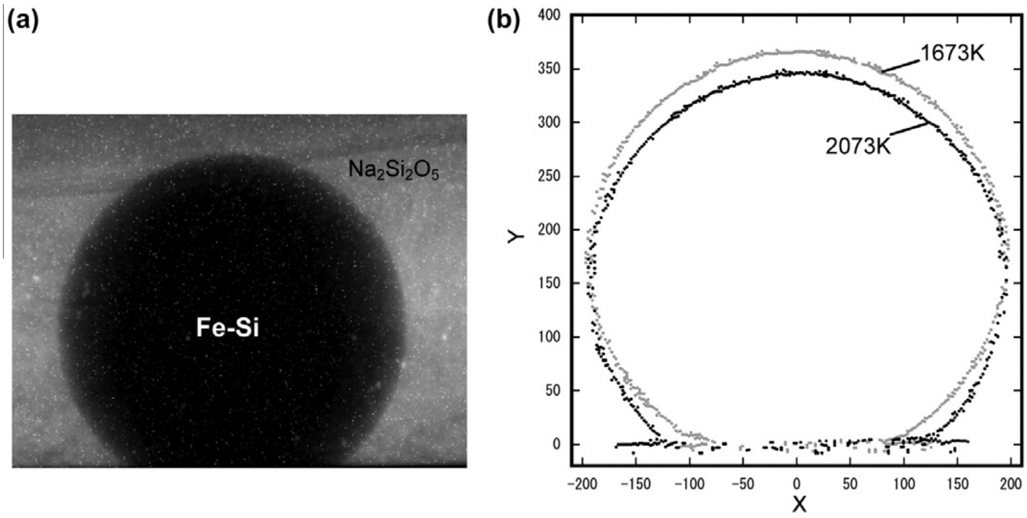

Fig. 1. (a) Typical example of a radiography image of Fe–Si (Si = 25 at%) liquid at 2073 K. (b) Outline plots of interface geometries obtained from radiography images at 1673 and 2073 K.