Note

Repulsive Capillary Force between two Plates Linked by a Concave Molten Slag Bridge

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In the previous papers,^{1,2)} the relaxation tests of molten aluminosilicate slags were carried out to evaluate the mechanical properties of molten slags. Namely, molten slag (35mass%CaO–35mass%SiO₂–30mass%Al₂O₃) drop of about 0.05 g was kept at 1723 K between two Pt disks (6.5 mm dia.) and then was compressed at various compression rates until particular displacement distance, then the compression process was stopped and the stress relaxation with time was measured. The details of the experiment can be found elsewhere.¹⁾ The observed typical stress relaxation test is shown in Fig. 1. The stress relaxation curve was consisted of 3 stages. The force became maximum at maximum stress (Stage I). After this, the force decreased sharply (Stage II). Then the force was decreased gradually and asymptotically approached to the constant value (Stage III). Intriguingly, the molten slag showed the existence of the repulsive residual force even without the compression force.

In the previous paper,¹⁾ we supposed that the origin of this residual force was due to the structural rearrangement difficulty of the aluminosilicate molten slags. Namely the original structure of slags was changed to differently configured structure during the compression process. After the elimination of compress force, it tried to return to the originally configured structure. The structural relaxation should involve breaking and making of the bonds between ions or silicate and aluminate units. The bending or distortion of silicate or aluminate units can be easily restored to the original state, however, the whole structural rearrangement among silicate and alumina units by the interatomic or intermolecular breaking and making of bonds can be difficult. Thus, the deformed structure may stay at the meta-stable configured structure. This meta-stable structure could possess higher energy than that of original structure. This causes the repulsive residual forces. Now we realize, however, that the meta-stable structure may not be the main reason of the repulsive force.

Initially we excluded the effect of surface tension because the molten slags after the compression showed the concave liquid bridge as shown in Fig. 2. Generally speaking, two wetted surfaces can stick together with great strength if the liquid wets them with an angle $\theta < \pi/2$. The θ angle is defined in Fig. 3. Namely, the existence of the

Fig. 1. Schematic description of stress relaxation curve for the molten aluminosilicate slag.

Fig. 2. Meniscus profile observed in the quenched slag bridge after compression.

Fig. 3. Sketch of the liquid bridge geometry, where displayed the parameters involved in the calculations.

concave liquid bridge between two plates suggests the attractive force between two plates. The Laplace pressure within liquid bridge between wetted two plates can be expressed by

$$\Delta P = \gamma \left(\frac{1}{R} - \frac{\cos \theta}{H/2} \right) \dots \dots \dots (1)$$

where θ is the wetting angle, γ is the surface free energy, H is the plate to plate distance, and R is the radius of the liquid bridge, and all these parameters are shown in Fig. 3. If $H \ll R$, ΔP can be simplified to

$$\Delta P \approx \frac{2\gamma \cos \theta}{H} \dots \dots \dots (1)'$$

As long as $\theta < \pi/2$, ΔP can be negative or the force F between the plates can be attractive and the force can be expressed by

$$F = \pi R^2 \Delta P + 2\pi R \gamma \sin \theta \approx 2\gamma \cos \theta / H \quad (H \ll R) \dots (2)$$

The wetting angle θ of less than $\pi/2$ shown in Fig. 2 suggests the existence of the attractive force between the two plates. The observed stress in the previous experiments was repulsive so that we simply discarded the effect of the surface tension on the residual repulsive force in the previous discussion.¹⁾

Under the following conditions, however, the case (1) that the contact angle θ has a large value and the case (2) that the contact angle is small but the value of R is relatively close to that of H (small volume of liquid bridge), the values of $1/R$ and $2 \cos \theta / H$ in the Eq. (1) may be comparable. In these cases, ΔP can be possibly positive or the repulsive force may be developed even a concave liquid bridge is formed.

In Fig. 2, the values of R and H are found to be 0.2 cm and 0.18 cm, respectively. By applying Eq. (2), the force between the two plates can be evaluated by

$$F = \pi R^2 \Delta P + 2\pi R \gamma \sin \theta = \pi R^2 \gamma \left(\frac{1}{R} - \frac{\cos \theta}{H/2} \right) + 2\pi R \gamma \sin \theta = 0.2\pi \gamma (5 - 11.1 \cos \theta + 2 \sin \theta) \dots (2)'$$

The dimensionless capillary force, $5F/\pi\gamma$ as a function of θ is shown in Fig. 4. It is zero at the wetting angle of 53.47° . Namely, if the wetting angle between Pt plate and molten slag was less than 53.47° , the capillary force between two Pt plates can be repulsive. The wetting angle from the shape of meniscus of quenched slag shown in Fig. 2 is likely to be less than 53.47° . Although the meniscus shape at the molten state may be slightly different from that of the quenched sample, it will be small. It means that the observed repulsive residual force in the previous relaxation tests¹⁾ is possibly attributed to the repulsive capillary force.

As we detected the possibility of repulsive capillary forces in a concave liquid bridge, already noticed in literature in both theoretical⁴⁻⁸⁾ and experimental approaches.⁹⁾ To eliminate the effect of surface tension on the stress relaxation tests, the large volume of liquid ($H \ll R$) must be used. Reversely, for the wetting phenomena in a micro-scale, we must think about the existence of the repulsive force in the liquid bridge. Recently Megias-Alguacil and Gauckler⁴⁾ studied the capillary force between two solid spheres linked by a concave liquid bridge. The capillary force was correlated with the relative volume V_{rel} and the wetting angle θ . V_{rel} is the volume of the liquid bridge, V , respect to the volume of the solid sphere, then, $V_{rel} = 3V/(4\pi R^3)$. The result is shown in Fig. 5. Details of the evaluation process can found elsewhere.⁴⁾ From Fig. 5, the capillary force is repulsive for V_{rel} of smaller than 0.03 and wetting angle is below 71.6° . Intriguingly, it suggests that the capillary force can be changed from to attractive to repulsive with decrease of the liquid bridge volume even under the constant wetting angle.

The geometrical condition of a liquid bridge between two plates can correspond to the case that V_{rel} is negligibly

Fig. 4. Map, θ - V_{rel} , displaying the regions of repulsive and attractive capillary forces after Megias-Alguacil and Gauckler.⁴⁾

Fig. 5. The dimensionless capillary force, $5F/\pi\gamma$, as a function of wetting angle, θ .

small. Then, the capillary force will be repulsive from the result⁴⁾ shown in Fig. 5 if the wetting angle is less than 71.6° . This result also support the existence of the repulsive capillary force in the previous stress relaxation tests¹⁾ since the observed wetting angle is likely to be less than 71.6° as shown in Fig. 2.

Summarizing, when dealing with the behavior of a multi-phase system where liquid bridges are present, the evaluation of the Laplace component of the force is very important. Depending on the wetting angle and system's geometry, the capillary force will be attractive and repulsive even if the meniscus keeps concave shape.

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