

To the Editor:

In “Capillary forces between two solid spheres linked by a concave liquid bridge: regions of existence and force mapping,”¹ Megias-Alguacil and Gauckler explore the conditions where liquid bridges may exist and map the capillary forces over ranges of liquid bridge volumes and contact angles. The analyses of the regions of existence and the calculation of liquid bridge volume and relationships between geometrical variables are very interesting and valuable. Unfortunately, there appears to be a sign error in Eq. 14 which affects the trends of the force mapping (Figures 5 to 8). Since we found that this mapping could be very useful, we wanted to present corrected versions of Figures 5 and 6.

With the notation from Reference 1, the capillary force between two spherical particles is given by

$$\frac{F_{\text{cap}}}{\gamma R} = -2\pi \sin \alpha \sin(\alpha + \theta) - \pi R \sin^2 \alpha \left(\frac{1}{\rho} - \frac{1}{L} \right) \quad (1)$$

where γ is surface tension of the liquid, θ is the contact angle of the liquid on the sphere, R is the radius of the sphere, L and ρ are the radii of curvature of the liquid bridge, and α is the half-filling angle (variables are shown in Figure 1 of Reference 1). A negative force indicates attraction. The difference between Eq. 1 here and Eq. 14 in the article¹ is the sign between the first and second terms on the righthand side. A decrease in radius of curvature, ρ , must give rise to a decrease in the pressure in the liquid bridge and an increase in attractive force (i.e., a more negative value of F_{cap} in Eq. 1).

Figure 1 presented here shows the corrected force mapping for a contact angle of $\theta = 20^\circ$ at various values of V_{rel} (the volume of the liquid bridge relative to the volume of a particle) over a range of H/R values where H is the closest distance between particles. Eq. 1 was used to determine forces along with Eqs. 3 to 5 and 7 from Reference 1, which give expressions for α , ρ , L , and V . The range of H/R was dictated by values of H_{min} and H_{break} (given by Eqs. 12 to 13 in Reference 1). Figure 1 shows that when the particles are in contact, the

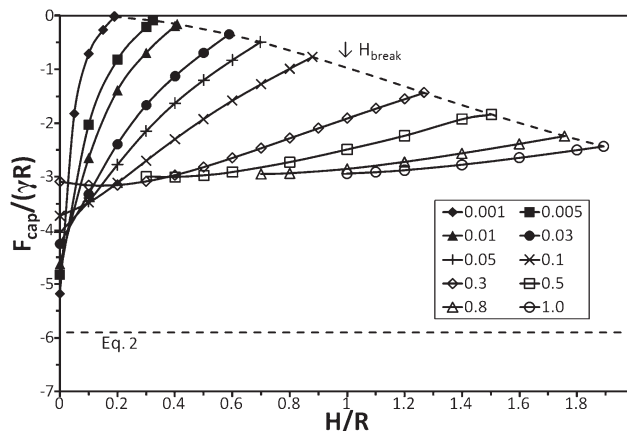


Figure 1. Dimensionless capillary force as a function of the dimensionless separation between particles for the relative liquid volumes indicated in the legend at $\theta = 20^\circ$.

magnitude of the attractive force increases as V_{rel} decreases. The approximate limit is given by the classical estimate for capillary forces²

$$\frac{F_{\text{cap}}}{\gamma R} = -2\pi \cos \theta \quad (2)$$

which assumes that the particles are in contact, α is small, and $L \gg \rho$ (and, thus, is applicable for small V_{rel}). However, as the particle separation increases, the trend is gradually reversed and eventually the attractive force increases as V_{rel} increases. These observations are consistent with calculated results elsewhere (see for example References 3

and 4). Figure 1 also shows for the most part a steady decrease in attractive force as particle separation increases, except at larger values of V_{rel} (above about 0.1 in Figure 1) where the decrease is more gradual and a slight maximum in attractive force may exist. Repulsive (positive) capillary forces are not attained under the conditions plotted since this would require that H exceeds H_{break} .

Figure 2 shows force mapping for $V_{\text{rel}} = 0.01$ at various contact angles over a range of H/R values (we use $V_{\text{rel}} = 0.01$ rather than 0.1 to show the possibility of positive forces). When the particles are in contact, the magnitude of the attractive force increases as con-

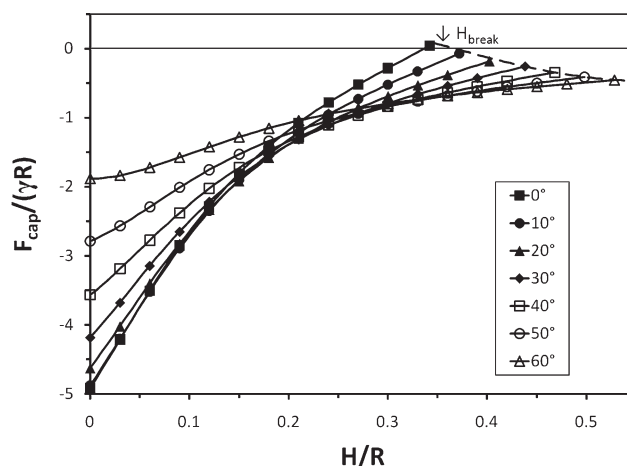


Figure 2. Dimensionless capillary force as a function of the dimensionless separation between particles for the contact angles indicated in the legend at $V_{\text{rel}} = 0.01$.

tact angle decreases. However, similar to Figure 1, as the particle separation increases, the trend is gradually reversed. Note that under certain conditions (small contact angles and small V_{rel}) it is possible to get repulsive forces at sufficient particle separation, and any points where $F_{\text{cap}} = 0$ will be unstable. We did not attempt to map the entire region of repulsive forces as was done by Megias-Alguacil and Gauckler (Figures 7 and 8 in Reference 1). However, Figures 1 and 2 shows that, contrary to the conclusions of Megias-Alguacil and Gauckler, forces will not be repulsive

for small contact angles and V_{rel} values when particles are in contact.

These corrections to the force mapping should help others in predicting experimental behavior and analyzing experimental observations.

Literature Cited

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4. de Lazzar A, Dreyer M, Rath HJ. Particle-surface capillary forces. *Langmuir*. 1999;15:4551–4559.

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