

## Reply:

In our recently published article<sup>1</sup> dealing with capillary forces between spherical particles linked by a concave liquid bridge, there is a sign error in the Laplace component of the force equation (Eq. 14 in Megias-Alguacil and Gauckler<sup>1</sup>), which has been properly pointed out by Bakken et al.<sup>2</sup> in their Letter to the Editor. The correct equation of the capillary force should read as Bakken et al. have noted

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

where  $\gamma$  is the surface tension of the liquid of the bridge,  $R$  is the solid particle's radius,  $\alpha$  is the half-filling angle,  $\theta$  is the wetting angle and  $\rho$ , and  $L$  are the principal radii of the liquid meniscus.<sup>1</sup>

This correction does not affect the method of determining the liquid meniscus shape, neither the discussion of the limiting interparticle distances of the concave liquid bridges as it is shown in Figure 4 of Ref. 1 mapping the  $\theta$ - $V_{rel}$  combinations for the region of existence of such bridges.

The corrected equation of the capillary force (Eq. 1) alters the published<sup>1</sup> shape of the capillary force vs. interparticle distance  $H$ , especially concerning its behavior at very short interparticle distances as indicated by Bakken et al.<sup>2</sup> With the correct equation, repulsive capillary forces are still predicted, but only for the longest possible distances of small liquid amounts.

Revisiting the force character map (original Figure 7 in Ref. 1), which outlines the allowed combinations  $\theta$ - $V_{rel}$  following in attractive and repulsive capillary forces, as shown new in Figure 1, it is observed that repulsive capillary forces will only persist for wetting angles,  $\theta$ , lower than  $\approx 20^\circ$  in a rather limited liquid volume range,  $V_{rel} < 0.023$  (Region III), inside the region of existence. In such a case, repulsive capillary forces exist just for the longest possible distances between particles, practically immediately before the bridge's rupture. All others possible combinations  $\theta$ - $V_{rel}$  (Region I of Figure 4 in Ref. 1) result in attractive capillary forces (Region IV of Figure 1) at any allowed distance between the particles.

The distances  $H_0$  for which  $F_{cap}=0$  are shown in Figure 2. For each possible wetting angle,  $\theta$ , of Region III, the distance of zero capillary force is higher as the bridge's

liquid volume enlarges, but decreases as  $\theta$  increases. Particulate systems linked by concave liquid bridges will shrink if the distance between the particles locates at  $H < H_0$ , and swell if  $H > H_0$ , although in this later case only briefly, because the breaking distance is reached practically immediately.

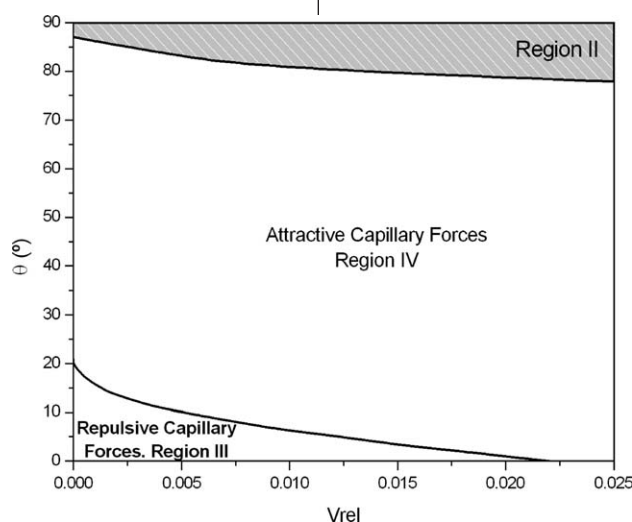
## Acknowledgments

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## Literature Cited

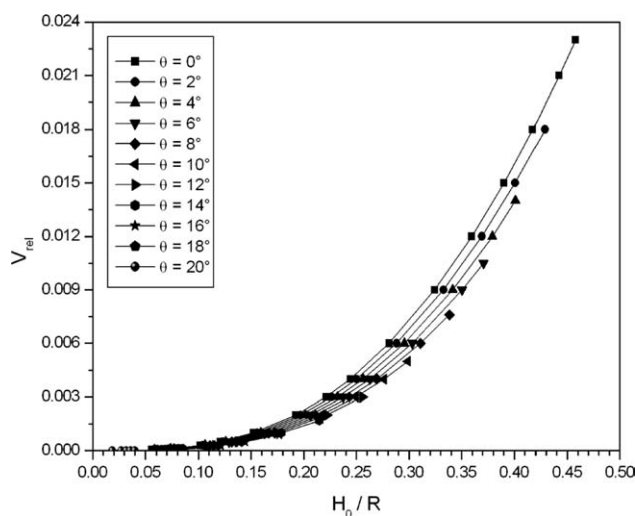
1. Megias-Alguacil D, Gauckler LJ. Capillary forces between two solid spheres linked by a concave liquid bridge: regions of existence and forces mapping. *AIChE J.* 2009;55:1103–1109.
2. Bakken M, McCulfor J, Anklam MR. Letter to the Editor. *AIChE J.* 2010.

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**Figure 1.** Map  $\theta$ - $V_{rel}$  displaying the regions of repulsive (Region III) and attractive (Region IV) capillary forces.

The Region II is shadowed for denoting its exclusion character.



**Figure 2.** Relative volume of liquid of the bridge,  $V_{rel}$ , as a function of the dimensionless interparticle distances  $H_0/R$ , for which  $F_{cap}=0$ , at different wetting angles,  $\theta$ .