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Capillary Forces between Equally Sized Moving Glass-Beads:

An Experimental Study

M. G. Bozkurt¹, D. Fratta² and W. J. Likos³

Abstract: The mechanical response of near-surface unsaturated soils in large-strain environments such as earthquakes, landslides, or debris flows is highly dependent on capillary forces. While the evolution of capillary forces under static loading has been studied in detail, the dynamic response of unsaturated soils associated with the viscous deformation and the rupture of interparticle liquid menisci at large strains is not as well characterized. Particle-scale pullout tests were conducted to achieve better understanding of how separation rates and distances contribute to capillary force evolution and meniscus rupture between two equally-sized glass spheres. Capillary forces evolve non-monotonically in a manner that first increases and then decreases with increasing separation distances and is dependent on the initial meniscus geometry and wettability of the particles. The rate of capillary force reduction and particle separation distance at liquid bridge rupture are functions of the meniscus volume and the rate of particle separation. The two-particle experimental results suggest that the dynamic response of bulk (multiparticle) unsaturated soil systems would depend on processes of drainage and imbibition and provide insight into the evolution of stiffness and the ductility of unsaturated soils undergoing large-strain deformations.

Keywords: unsaturated soils; capillary forces; surface tension; rate of particle separation

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Introduction

Interparticle forces generated within a matrix of unsaturated soils result from the combined effects of net stresses (caused by external body forces and gravity) and matric suction (caused by internal capillary forces and short-ranged adsorptive forces). The interplay between these forces significantly contributes to the behavior of unsaturated near-surface soils and often controls macroscopic engineering behavior such as strength, deformation, and permeability (Edil et al. 1981; Wheeler et al. 2003; Fredlund, 2005). The negative water pressure and the surface tension associated with interparticle pore water menisci in soil systems at low saturation levels increase interparticle forces and produce a rise in strength and small-strain stiffness (Cho and Santamarina 2001; Willett et al. 2003; Pacheco-Vazquez et al. 2012). In particular, the strength and volume change response of coarse-grained materials under low net stress conditions (e.g., in near-surface applications) can be dominated by capillary forces because the effects of gravitational forces are less significant (Santamarina 2001; Lu and Likos 2006).

The forces exerted by water bridges on solid particles have been studied by many researchers (Table 1). These studies have typically included zero or small separation distances between particles. The interpretation of these studies is simplified by assuming that the solid particles have simple geometries (e.g., spheres or plates), that the particles are arranged according to some idealized packing geometry (e.g., simple cubic or tetrahedral), and that menisci between the particles are bounded by a toroid. At low volumetric water content (i.e., within the pendular saturation regime), the interparticle force between particles results from two mechanisms: the surface tension T_s at the fluid (air-water) interface and the negative pressure within the liquid bridge (Rosetti and Simons 2003). The geometry of the meniscus creates a pressure difference

between air and water phases (Fig. 1a). A common model used to calculate the capillary force between two contacting and equally-sized particles with radius R is (Likos and Lu 2004)

$$F_{cap} = \pi R^2 u_a - \pi r_2^2 (u_a - u_w) - 2\pi r_2 T_s \quad [1]$$

where the radius r_2 is the convex curvature of the meniscus, the term $(u_a - u_w)$ is the matric suction, u_a is the air pressure, and u_w is the water pressure (Fig. 1b). If the matric suction term is replaced by $u_a - u_w = T_s \left(\frac{1}{r_1} - \frac{1}{r_2} \right)$ (i.e., Laplace's equation) then the capillary force between the spherical particles becomes:

$$F_{cap} = -\pi r_2 T_s \left(1 + \frac{r_2}{r_1} \right) \quad [2]$$

where the radius r_1 is the concave curvature of the meniscus and the air pressure is set to $u_a = 0$ kPa for most applications. Therefore, the " $\pi R^2 u_a$ " term in the Equation 1 is excluded.

Pitois et al. (2000) extended a formulation of capillary forces between two particles to capture dynamic effects including separation distance between two particles and the viscosity of the wetting fluid:

$$F_{dyn} = F_{cap} + F_{vis} = 2\pi R T_s \cos(\theta) X_v + \left(\frac{3}{2} \right) \pi \eta R^2 \left(\frac{1}{D} \right) V_{el} X_v^2 \quad [3]$$

where $X_v = 1 - \left(1 + \frac{2V}{\pi R D^2} \right)^{-1/2}$, θ is the solid/liquid contact angle, η is the wetting fluid viscosity, D is the separation distance between particles, V_{el} is the rate of particle separation, and V is the liquid volume (Fig. 1c). Equation 3 indicates that, for highly viscous liquids and large rates of particle separation, the contribution of viscous forces is much greater than the contribution of the capillary forces. Fig. 2 summarizes the effects of the parameters in Eq. 3 on the capillary force in terms of capillary number $N_c = \eta \cdot V_{el} / T_s$ and the wettability of the solid mineral/liquid system captured by the solid/liquid contact angle. The figure differentiates the

response into different behavior regimes based on the controlling parameters. For low-viscosity fluids (e.g., deionized water; viscosity $\eta=1.83 \cdot 10^{-5}$ Pa·s and surface tension $T_s=7.2 \cdot 10^{-2}$ N/m at room temperature), the transition to viscous-dominated behavior would only occur at rates of particle separation higher than about $4 \cdot 10^3$ m/s. This rate is extremely high for most phenomena found in geotechnical and earthquake engineering problems, which are limited to values up to around 10^0 m/s (for strains of 10^{-1} in gravel with particle size 0.02 m and frequencies 500 Hz). However, at very low velocities (capillary number $N_c < 1$), the effect of wettability controls the magnitude of the capillary forces.

The effect of gravity on interparticle capillary forces has not been considered by the previous analytical models. The ratio of gravitational forces to capillary forces (Orr et al. 1975) is captured by the Bond number $B_o = \frac{\Delta\rho g L^2}{T_s}$ where $\Delta\rho$ is the difference between the density of the two fluids (water and air in most soil related problems), g is the acceleration of gravity, and L is a representative length. For typical geotechnical engineering problems the density difference is $\Delta\rho \approx 1000$ kg/m³, the acceleration of gravity is $g=9.81$ m/s², the meniscus diameter is on the order $L=0.001$ m, and the surface tension is $T_s=0.072$ N/m. The corresponding Bond number is $B_o=0.13$. Thus, for small particles and meniscus diameters, the effect of the gravity force is less than 7.5 times that of the capillary force and gravity forces may be disregarded. This assumption was experimentally confirmed by Mazzone et al. (1986), which reported a study on how gravity affects capillary force and the rupture distance of interparticle menisci using stainless steel spheres and dibutylphthalate as the bridging liquid. These results indicate that the effect of gravitational forces on the rupture distance is small at low liquid volumes; however, as the liquid volume increases, gravitational effects play an increasingly important role.

The dynamic response of unsaturated soils in high-strain environments such as earthquakes, slope stability failures, and debris flows is controlled by the rapid evolution of menisci and capillary forces due to localized particle movements and strains (Bian and Shahrour 2008; Ravichandran 2009; Orense et al. 2012). While the effects of interparticle capillary forces on static mechanical response have been studied in detail (Scholtes et al. 2008; Hu et al. 2014), the effects of dynamic forces associated with the formation and rupture of menisci at large strain levels are not as well characterized. This paper documents the results of a laboratory experimental study designed to quantify how the separation distance, the rate of particle separation, and the solid-liquid wettability contribute to the capillary force evolution and the rupture between two glass spheres. Experiments were conducted for different liquid bridge volumes and different surface wettability conditions.

Materials and Experimental Program

An experimental program was developed to evaluate the effect of particle size, wettability, liquid bridge volume, and rate of particle separation on the interparticle forces between two equally-sized spherical glass beads. In a stiff frame with displacement control capabilities (Fig. 3), two glass beads were glued to rigid steel rods with cyanoacrylate. The bottom glass bead and its rigid support were placed on a Mettler Toledo-XA 204DR scale with force resolution of 10^{-6} N, while the top glass bead and its rigid support were mounted on an actuator that could be moved up and down at a constant rate of particle separation. The scale was connected to a computer to record the evolution of the capillary force during the separation process while the geometry of the meniscus was monitored with digital microscopy. Two Dino-lite AM413TA digital microscopes

were placed perpendicular from each other and used to align the glass beads in the vertical and horizontal directions, to evaluate when the glass beads were touching, and to capture the evolution of the meniscus geometry during the controlled pullout process. Deionized water was injected between the glass beads with a 0.1-10 μL Cole Parmer precision pipette to create an interparticle menisci of known volume at zero particle separation distance.

The system was prepared by placing a water drop with the precision pipette on top of the bottom glass bead while the beads were separated. The top glass bead was then moved down until the two spheres were in contact. After this point the pullout test begun. Particles initially in contact were separated using rates ranging from $6.83 \cdot 10^{-6}$ m/s to $3.33 \cdot 10^{-4}$ m/s (i.e., $N_c = 9.36 \cdot 10^{-8}$ to $4.57 \cdot 10^{-6}$) until the liquid bridge ruptured. During testing and data interpretation, the effects of gravity forces were disregarded due to the small volume of liquid used to form the meniscus (i.e., low Bond number). This assumption was validated by observations from the microscopes indicating that self-weight deformations of the menisci were negligible.

Two sets of equally-sized glass beads were used in the testing program: 0.8 mm and 3 mm in diameter. The glass beads were washed either with acetone or isopropanol with the intent to render the surface of the glass beads with different levels of hydrophobicity. Fig. 4 shows the effect of the different washes on the affinity of the particles to be wetted by the deionized water drops. This figure shows that the acetone-washed beads yielded a more hydrophilic liquid-solid interaction compared to the isopropanol-washed glass beads. The isopropanol-glass beads yield surfaces with a contact angle of about 90 degrees that should be described as 'less strongly hydrophilic' (Förch et al. 2009). That surface treatment is simply referred as 'less hydrophilic'

surfaces in this study. A suite of tests was conducted to evaluate the evolution of capillary force and the rupture of the liquid bridge for different glass bead sizes, for different levels of hydrophilicity, and for different meniscus volumes and rates of particle separation (Table 2).

Testing Results

Effect of Liquid Bridge Volume and Levels of Hydrophobicity on Capillary Forces

The effect of meniscus volume on the measured capillary force as a function of separation distance was studied for three different meniscus volumes injected between two 0.8-mm radius on less hydrophilic spherical glass beads. Fig. 5 is a plot of the normalized capillary force (i.e., $F/(T_s \cdot R)$ where F is the measured pullout force) evolution as a function of normalized displacement (i.e., D/R) for different liquid volumes. All tests were run at the same rate of particle separation ($V_{el}=2.12 \cdot 10^{-5}$ m/s – $N_c=2.90 \cdot 10^{-7}$). Each test was run three times (i.e., presented as Tests #1, #2, #3 in the figure) to assess repeatability of the experiment. The evolution of the normalized force versus normalized displacements shows that the maximum normalized force does not occur at zero separation distances between the glass beads, but rather after some finite amount of displacement. While this phenomenon was also observed by other researchers (Mazzone et al. 1986; Soulié et al. 2006) it is not captured by the models described by Equations 2 and 3. The location and the magnitude of the maximum normalized force depends on the liquid bridge volume and appears to be a function of the wettability.

The effect of wettability on the location and the magnitude of the maximum normalized force is evident in Fig. 6, which is a plot of the evolution in normalized capillary force versus normalized separation distance for the less hydrophilic and hydrophilic particle surfaces. Results for the

hydrophilic glass bead surfaces show a higher maximum normalized force at smaller normalized separations than the less hydrophilic glass bead surfaces. These results suggest that the initial meniscus geometry is not constrained by a theoretical toroid as described in the models represented by Equations 2 and 3. The reason for this is that when the glass beads are pushed together before the test begins the water bubble is pushed out engaging an advancing contact angle (Yaun and Lee 2013). Then, it requires a certain level of separation between particles to allow the engage the receding contact angle and allow the meniscus to engage in the ideal geometry specified in Equations 2, 3 and Fig. 1 to yield the maximum force. This geometry is engaged at lower separation distances in the case of the glass beads that show strong hydrophilic behavior because the initial meniscus geometry is closer to the geometry captured in Equations 2 and 3. In contrast, the liquid bridge geometry associated with the beads that show less hydrophilic behavior greatly deviates from the idealized toroidal geometry and the maximum pullout force is only achieved at larger particle separation distances when the full receding contact angle is engaged (Yaun and Lee 2013). This is evident in the photos of the initial liquid bridge geometry presented in Fig. 6.

The results shown in Fig. 5 also indicate that the smallest liquid bridge volume ($V=0.2 \mu\text{L}$) yields the highest normalized force and reaches its maximum value at lower separation distances ($D/R=0.18$). While larger liquid volumes ($V=1.0 \mu\text{L}$) yield smaller normalized capillary at higher normalized separation distances ($D/R=0.61$). The increase in liquid bridge volume leads to a maximum normalized force that is stable over a wide range of normalized separation distances. As the volume of the meniscus increases, larger particle separation is required to achieve the ideal geometry associated with the maximum capillary force. After reaching the maximum

pullout force, the normalized forces gradually decrease with increasing normalized separations until the liquid bridge ruptures. Mazzone et al. (1986) also observed that larger liquid bridge volumes yielded larger normalized forces at larger separation distances using dibutylphthalate as the bridging liquid and stainless steel spheres, but did not clearly capture capillary force evolution at very small separation distances.

Effect of Rate of Particle Separation

To determine the effect of rate of particle separation on the measured capillary forces, 5 μL deionized water drops were injected between two 3-mm and two 0.8-mm radius glass beads. The beads were separated using rates of separation ranging from $6.83 \cdot 10^{-6}$ m/s to $3.33 \cdot 10^{-4}$ m/s ($N_c = 9.36 \cdot 10^{-8}$ to $4.57 \cdot 10^{-6}$). Fig. 7 presents the effect of rates of separation for glass beads with different levels of hydrophilicity. Fig. 8 differentiates the effect of bead size and meniscus volume on the normalized capillary force for glass beads that show less hydrophilic response.

The effect of rate of particle separation on the hydrophilicity of glass beads appears to be small (Fig. 7). The most significant effect appears in the rate of decrease of the capillary force after reaching the maximum value and on the normalized displacement at rupture. The maximum normalized force does not appear to be influenced by the rate of particle separation. Maximum normalized force occurs at the normalized separation $D/R=0.046$ for the less hydrophilic beads and at 0.033 for the hydrophilic beads regardless of rate of particle separation. That response is mainly caused by the initial meniscus geometry rather than by dynamic effects. Then, the magnitude of the maximum capillary force and the separation at which it occurs does not depend on the rate of separation, but it depends on the wettability condition and the volume of the

meniscus. Different normalized displacements are required to create the toroid geometry assumed in the models presented in Equations 2 and 3 and for corresponding capillary force to reach a maximum. After the maximum capillary force is reached, the force gradually decreases until the liquid bridge ruptures. This rupture occurs at larger normalized separations for higher rates of separation. The rate of force reduction is also smaller for the higher rates of separation. These effects are greater for the smaller bead sizes and meniscus volumes in the case of the less hydrophilic glass beads (Fig. 8). The rate of capillary force reduction and normalized displacement at rupture for the smaller glass beads ($R=0.8$ mm) and meniscus volume ($V=0.5$ μL) are greatly controlled by the rate of separation. For smaller particle sizes and liquid volumes, there are dynamic effects that sustain the capillary forces and produce more stable response at faster rates of separation, even though the capillary number N_c is very small.

Normalized Displacements at Rupture

The effect of displacement on bridge rupture was investigated for different hydrophilicity conditions. The displacement at rupture is the separation distance between glass beads where the liquid can no longer form a bridge and breaks into two separate liquid volumes. Fig. 9 presents the displacement at rupture as a function of rates of particle separation, where each data point corresponds to a separate test. Results show that up to a certain rate of separation (Fig. 9a - $R=3$ mm and $V=5$ μL), the normalized displacement at rupture increases linearly with the rate of separation. However, when the rate of separation reaches $V_e=1.7 \cdot 10^{-4}$ m/s ($N_c=1.6 \cdot 10^{-6}$), particle displacement at rupture dramatically decreases. Similar results were observed for low rates of separation with smaller glass beads and liquid bridge volume (Fig. 9b - $R=0.8$ mm and $V=0.5$ μL).

The displacement at rupture results were compared against a model presented by Soliue et al. (2005) based on analyses by Lian et al. (1993). The predicted equation for displacement at rupture $D_{rupture}$ is presented as function of the contact angles and the volumes of the menisci:

$$D_{rupture} = \left(1 + \frac{\theta}{2}\right) \sqrt[3]{V} \quad [4]$$

The results obtained with equation 4 for different contact angles are presented in Figure 10 and compared against the rupture displacement results presented in Figures 7a and 8. Note that while the data presented in this study fall within the predicting model, the effect of the rate of particle displacement on the displacement at rupture is not captured by this nor by other published models.

Interpretation

The analysis of the development of capillary forces in unsaturated media is commonly assessed by assuming that the grains are perfectly spherical, arranged according to idealized packing, and menisci geometry has well defined meniscus radii r_1 and r_2 . However, the evolution of meniscus formation is different than what models based on toroidal meniscus geometry assume. Fig. 11 shows that evolution of the meniscus geometry during separation makes it difficult to define concave curvature radius r_1 , especially after necking occurs immediately prior to rupture. Fig. 12 shows a comparison of the measured capillary forces with published theories for idealized two-particle systems. Surface tension for deionized water for the calculations was set to 0.072 N/m. The value for contact angle was measured from the optical images and defined as the angle measured between the liquid-solid interface and the tangent line to the liquid-air interface (the value of contact angle, θ changes from 85° (when two particles in contact) to 20° (just before

rupture – Fig. 12(b)). Although the previously published theories appear to fit well the measured data beyond the maximum pullout force (Soulié et al. 2006), the main difference between the theory and measured capillary force is the location of the maximum force during the pullout deformation. Maximum capillary forces were measured at a nonzero separation distances when the initial meniscus geometry is not constrained by a theoretical toroid. This process can only be predicted if the proper geometry and contact angle is included in the evolution of the pullout process as suggested by Soulié et al. (2006). As summarized in Table 1, Mason and Clark (1965), Mazzone et al. (1986), and Soulié et al. (2006) had also observed the maximum force at non-zero separation distances, and the model by Pitois et al. (2000) captures the capillary force with a separation distance better than other formulations.

In particular, after placing the liquid on top of the bottom glass bead, the top glass bead moved down until two spheres were in contact. During this process, the liquid bridge is compressed creating a “fat” meniscus, particularly in the case of the less hydrophilic glass bead surfaces. During the pullout process the bridge evolves into a well-formed toroidal menisci geometry, thus affecting the location of the maximum force. Hence, it would be expected that during the process of drainage and imbibition in near surface soils, the response would be different. For example, after a large rainfall the menisci volume in the soil matrix would be larger and may require larger deformations to reach maximum strength rupture. Such interpretation provides a new conceptual basis to interpret evolution of capillary forces and the dynamic response of near surface soils. Furthermore, if these results can be translated to 3D arrangements, the evolution of stiffness and strength with strain levels should be re-assessed.

Conclusions

The total capillary force exerted by a liquid bridge between two equally-sized glass beads was measured to assess the effects of separation distance, rate of particle separation, the volume of the liquid bridge, and hydrophilicity. These parameters form the foundation for understanding the effect of partial saturation on the mechanical response of particulate media under large dynamic deformations. The maximum capillary force was found to occur at nonzero particle separation distance. These results contradict models used in the evaluation of pullout tests between particles joined by a capillary bridge. The reason for the mismatch is the assumption in these models that the geometry of the meniscus bridge is always bound by a toroid. In microscopic images it was observed that during the pullout process both the geometry and the contact angle change until the menisci deforms to the point where it is bound by a toroid. At this separation distance, the pullout forces are at a maximum.

The normalized separation distance at the maximum capillary force does not depend on the rate of particle separation. However, the separation distance at the normalized capillary force varies for different liquid bridge volumes. Higher liquid bridge volumes yielded lower normalized capillary forces and reached the maximum normalized capillary forces at higher separation distances. The displacement at rupture was observed to be a function of the rate of particle separation. An increase in rate of particle separation contributed linearly to the displacement at rupture up to a certain rate, after which the displacement at rupture dramatically decreased. The effect of rate of particle separation on the displacement at rupture observed in this study is not yet predicted by current models. This study documents the first set of tests aimed at understanding how the rate of particle separation contributes to the evolution and rupture menisci

and the associated capillary forces in particulate media. These data are required to properly explain large-strain dynamic response of unsaturated soils.

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Figure Captions

Fig. 1. Meniscus between two spheres: (a) two contacting spheres, (b) free body diagram for evaluating interparticle forces, and (c) meniscus between two spheres with separation distance D .

Fig. 2: Effect of viscosity, separation rate, surface tension, and hydrophilicity on the resultant capillary forces as modelled by Equation 3.

Fig. 3. Test setup for the evaluation of the meniscus geometry and capillary force evolution.

Fig. 4: The resultant contact angle depends on the solution used to clean the glass beads. The particles in the images have a particle radius $R=3$ mm and a 4- μ L deionized water drop was injected between them. (a) Acetone-washed hydrophilic glass bead surfaces before and after particles are brought together for pullout test. (b) Isopropanol-washed less hydrophilic glass bead surfaces before and after particles are brought together for pull apart test.

Fig. 5. Effect of meniscus volume on normalized capillary force (displacement rate $V_{el}=7.62 \cdot 10^{-2}$ m/s – $N_c=1.04 \cdot 10^{-3}$ – and $R=0.8$ mm).

Fig. 6. Effect of solid-liquid wettability on normalized capillary force evolution: isopropanol-washed less hydrophilic glass bead surfaces and acetone-washed hydrophilic glass bead surfaces (Radius of the particles $R=3$ mm; rate of particle separation $V_{el}=3 \cdot 10^{-5}$ m/s – $N_c=4.11 \cdot 10^{-4}$).

Fig. 7. Effect of rate of particle separation and wettability on the normalized capillary force: (a) isopropanol-washed less hydrophilic bead surfaces and (b) acetone-washed hydrophilic bead surfaces (Radius of the glass beads $R=3$ mm and meniscus volume $V=5$ μ L)

Fig. 8. Effect of rate of particle separation on normalized capillary force. Isopropanol-washed less hydrophilic bead surfaces for radius of glass beads $R=0.8$ mm and meniscus volume $V=0.5$ μ L.

Fig. 9. Effect of separation velocity on displacement at rupture (a) Low and high displacement rate ranges for isopropanol-washed less hydrophilic glass bead surfaces and acetone-washed hydrophilic glass bead surfaces, $R=3$ mm, $V=5$ μL) and (b) Low velocity range for isopropanol-washed less hydrophilic glass bead surfaces ($R=0.8$ mm, $V=0.5$ μL).

Fig. 10. Displacement at rupture for different contact angles. Square symbols ($\square$): Isopropanol-washed less hydrophilic bead surfaces, radius of glass beads $R=0.8$ mm and meniscus volume $V=0.5$ μL . Circular symbols ($\circ$): isopropanol-washed less hydrophilic bead surfaces, radius of glass beads $R=3$ mm and meniscus volume $V=5$ μL .

Fig. 11. Meniscus evolution with increasing particle separation ($R=0.8$ mm glass beads; $V=0.2$ μL and $V_{el}=1.27$ mm/min). As separation increases toward the point of meniscus rupture, the meniscus geometry r_l becomes difficult to measure due to the necking of the bridge.

Fig. 12. (a) Comparison of test data with analytical models (Rate of particle separation $V_{el}=7.62 \cdot 10^{-2}$ m/s – $N_c=1.04 \cdot 10^{-3}$ – and radius of glass beads $R=0.8$ mm). (b) Data were fitted using the image-measured contact angles.

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Fig. 1. Meniscus between two spheres: (a) two contacting spheres, (b) free body diagram for evaluating interparticle forces, and (c) meniscus between two spheres with separation distance D .

Fig. 2: Effect of viscosity, separation rate, surface tension, and hydrophilicity on the resultant capillary forces as modelled by Equation 3.

Fig. 3. Test setup for the evaluation of the meniscus geometry and capillary force evolution.

Fig. 4: The resultant contact angle depends on the solution used to clean the glass beads. The particles in the images have a particle radius $R=3$ mm and a $4\text{-}\mu\text{L}$ deionized water drop was injected between them. (a) Acetone-washed hydrophilic glass bead surfaces before and after particles are brought together for pullout test. (b) Isopropanol-washed less hydrophilic glass bead surfaces before and after particles are brought together for pull apart test.

Fig. 5. Effect of meniscus volume on normalized capillary force (displacement rate $V_{el}=7.62 \cdot 10^{-2}$ m/s – $N_e=1.04 \cdot 10^{-3}$ – and $R=0.8$ mm).

Fig. 6. Effect of solid-liquid wettability on normalized capillary force evolution: isopropanol-washed less hydrophilic glass bead surfaces and acetone-washed hydrophilic glass bead surfaces (Radius of the particles $R=3\text{ mm}$; rate of particle separation $V_{el}=3\cdot 10^{-5}\text{ m/s} - N_c=4.11\cdot 10^{-4}$).

Fig. 7. Effect of rate of particle separation and wettability on the normalized capillary force: (a) isopropanol-washed less hydrophilic bead surfaces and (b) acetone-washed hydrophilic bead surfaces (Radius of the glass beads $R=3 \text{ mm}$ and meniscus volume $V=5 \text{ }\mu\text{L}$)

Fig. 8. Effect of rate of particle separation on normalized capillary force. Isopropanol-washed less hydrophilic bead surfaces for radius of glass beads $R=0.8 \text{ mm}$ and meniscus volume $V=0.5 \text{ }\mu\text{L}$.

Fig. 9. Effect of separation velocity on displacement at rupture (a) Low and high displacement rate ranges for isopropanol-washed less hydrophilic glass bead surfaces and acetone-washed hydrophilic glass bead surfaces, $R=3$ mm, $V=5$ μ L) and (b) Low velocity range for isopropanol-washed less hydrophilic glass bead surfaces ($R=0.8$ mm, $V=0.5$ μ L).

Fig. 10. Displacement at rupture for different contact angles. Square symbols (□): Isopropanol-washed less hydrophilic bead surfaces, radius of glass beads $R=0.8$ mm and meniscus volume $V=0.5$ μL. Circular symbols (O): isopropanol-washed less hydrophilic bead surfaces, radius of glass beads $R=3$ mm and meniscus volume $V=5$ μL.

Fig. 11. Meniscus evolution with increasing particle separation ($R=0.8$ mm glass beads; $V=0.2$ μL and $V_{el}=1.27$ mm/min). As separation increases toward the point of meniscus rupture, the meniscus geometry r_l becomes difficult to measure due to the necking of the bridge.

Fig. 12. (a) Comparison of test data with analytical models (Rate of particle separation $V_{el} = 7.62 \cdot 10^{-2}$ m/s – $N_c = 1.04 \cdot 10^{-3}$ – and radius of glass beads $R = 0.8$ mm). (b) Data were fitted using the image-measured contact angles.

Table 1: Summary of Relevant Research of Pullout Testing between Particles

	Studied Phenomena	Concluding Remarks
Mason and Clark (1965)	Forces between two spheres and sphere-plate interface with a liquid bridge of a dibutyl phthalate/liquid paraffin mixture were measured.	Force appears to pass through a maximum for small bridge volumes at small separations. Initial attractive force rise was observed as the separation increases.
Schubert et al. (1975)	The strain behavior of a liquid bridge between two and more equal spheres was analyzed.	The adhesion force reached to maximum when two spheres were in contact (zero separation) and adhesion drops with an increase in distance between spheres. The fracture strain was strongly dependent on imbibition or drainage conditions (smaller deformations occurred for drainage conditions).
Mazzone et al. (1986)	Force, geometry and gravity effects on a liquid bridge (made up of dibutylphthalate) between two stainless steel balls were studied.	Gravity had a significant effect on menisci if the Bond number is of the order unity and greater. Maximum force observed at a small but nonzero separation distance. Larger bridge volumes caused larger forces and the rupture of the bridge occurred at larger separation distances.
Pierrat and Caram (1997)	Two components of the maximum tensile force due to a liquid bridge between two non-contact mono-sized spherical particles were formulated.	The separation distance between particles has an important effect on the magnitude of attraction force. The maximum force occurs between two touching particles and the force decreases as the distance between particles increases.
Pitois et al. (2000)	Capillary and viscous forces between two moving ruby spheres were measured for different liquid bridge (made up of PDMS and Brookfield oils) volumes.	The maximum of force has been found for a zero-separation distance. Viscous effects contribute to the increase in the rupture distance, which was found to vary like the square root of the separation velocity.
Cho and Santamarina (2001)	An equation to measure the force between two contacting spheres was provided. Effective stress was predicted as a function of water content, particle size and specific surface using Laplace's equation.	The effective stress increased with decreasing water content and particle size, increasing specific surface and coordination. Decrease in water content caused a dramatic increase in negative pressure in the meniscus however the effective area of the meniscus between spherical particles decreased as well.
Rossetti et al. (2003)	Rupture energy of a silicon oil liquid bridge formed in water between two glass particles was investigated with a micro force balance.	Liquid bridge energy was calculated at rupture distance and at a small but non-zero separation distance due to experimental difficulties to observe the liquid bridge at zero separation. Wettability highly affected the geometry and the capillary pressure of the liquid bridge. Adhesion forces measured during separation are lower than in the case where both particles present a high wettability toward the binder.
Rosetti and Simons (2003)	Interaction of liquid binder between micron-sized particles submerge in a second immiscible liquid was directly measured and observed.	Pressure difference across the bridge interface from the geometry of contact area between the particles and the binder was calculated to predict the force exerted by liquid bridge. Capillary pressure first dropped to a minimum and then started to rise as the concavity increases. The maximum force observed at the point where capillary pressure was minimum.
Willett et al. (2003)	The effects of wetting hysteresis on the properties and behavior of pendular liquid bridges between spherical particles have been investigated both theoretical and experimental.	If contact angle is greater than the receding value, force increases due to pinning as separation distance increases. This will be accompanied by a reduction in the contact angle until the receding value is achieved when the force decreases. If the separation distance decreases after that point, the contact angle increases until advancing value is achieved. That is, pinning will occur and results in a reduction in the force. Wetting hysteresis may cause an extended bridge rupture distance and capillary interactions become dissipative rather than conservative.
Likos and Lu (2004)	Constitutive relationships between water content, matric suction, capillary stress in unsaturated soils were modeled with a theoretical model based on the geometry of the menisci.	Relatively large contact angles result in significantly lower capillary stresses, indicating that the contribution of capillarity to interparticle stress for soils undergoing wetting process may be less than that for soils undergoing drying process.
Rabinovich et al. (2005)	Theoretical and experimental studies were performed to understand the capillary forces between two glass spheres with a fixed liquid bridge volume.	Theoretically, force decreases with an increase in separation for sphere-plate interactions. Experimentally, there is a maximum force at small separation for sphere-sphere interactions however it is not clearly presented.
Soulié et al. (2006)	Capillary forces between two uneven stainless steel balls (ranging diameters 2-8 mm) were measured as a function of geometrical and physical characteristics. Demineralized water was used as a bridge liquid.	Capillary forces increased with an increase in interparticle distance whereas the contact angle decreased at the same time, then capillary forces started to decrease. Experimental test results were in the numerical modelling of radial and axial deformation of arrangement of particles with capillary bridges.

Table 2. Summary of experimental matrix

Surface Wettability	Hydrophilic	Less Hydrophilic	
Cleaning type	Acetone	Isopropanol	
Particle radius, R (mm)	3	0.8	3
Injected liquid bridge volume, V (μL)	5	0.2, 0.5, 1	5
Low rates of particle separation, V_{el} (m/s)	$6.83 \cdot 10^{-6}$, $1.35 \cdot 10^{-5}$, $2.72 \cdot 10^{-5}$, $4.07 \cdot 10^{-5}$	$5.00 \cdot 10^{-6}$, $1.00 \cdot 10^{-5}$, $1.50 \cdot 10^{-5}$, $2.12 \cdot 10^{-5}$, $2.62 \cdot 10^{-5}$	$8.33 \cdot 10^{-6}$, $1.67 \cdot 10^{-5}$, $2.5 \cdot 10^{-5}$, $3.33 \cdot 10^{-5}$
High rates of particle separation, V_{el} (m/s)	$1.67 \cdot 10^{-4}$, $2.00 \cdot 10^{-4}$, $2.33 \cdot 10^{-4}$, $2.67 \cdot 10^{-4}$, $3.33 \cdot 10^{-4}$	-	$1.33 \cdot 10^{-4}$, $2.00 \cdot 10^{-4}$, $2.33 \cdot 10^{-4}$

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