Wettability of particles and its effect on liquid bridges in wet granular materials

Hooman HOORNAHAD ▪ Microlab, Faculty of Civil Engineering and Geosciences,
Delft University of Technology ▪ h.hoornahad@gmail.com
Eduardus A. B. KOENDERS ▪ Institute for Construction and Building Materials,
Technische Universität Darmstadt ▪ koenders@wib.tu-darmstadt.de
Klaas van BREUGEL ▪ Microlab, Faculty of Civil Engineering and Geosciences,
Delft University of Technology ▪ k.vanbreugel@tudelft.nl

Érkezett: 2015. 07. 31. ▪ Received: 31. 07. 2015. ▪ http://dx.doi.org/10.14382/epitoanyag-jsbcm.2015.23

Abstract

This paper provides results of an experimental study on the force-distance relationship between two particles connected by a liquid bridge. This experimental research is based on water bridges and extended to cement paste bridges with the aim to explain and model the rheological behaviour of fresh concrete mixtures, and to create a fundamental basis for developing mixtures with a predefined deformation performance. The research approach is based on a conceptual idea where the mixture is considered as an assembly of mutually interacting "particle-paste-particle" systems and the rheological behaviour of the mixture is related to the inter-particle interactions. The concept of capillary cohesion in wet granular materials, which describes the interaction between two particles connected by a liquid bridge, is considered the first step in the investigations. The effect of the wettability of the particles on the force-distance relation of two particles connected by a liquid bridge is studied. Static and/or quasi-static situations are in focus, i.e. where the cohesion dominates over other effects generated by the liquid, such as viscosity and lubrication. Tests were carried out using spherical stainless steel and glass particles and water was used as the liquid with a surface tension of σ_{in}=0.072 mN/mm. The contact angle between water and glass is about 15° while between water and stainless steel it is about 60°. It is found that the wettability of particles has a great influence on the force-distance relation of the interacting particles. Conclusion is drawn regarding the possibility to use the generated data as a basis for modelling the rheological behaviour of cement-based mixtures.

Keywords: Fresh concrete, rheological model, wet granular material, liquid bridge, wettability, contact angle, interaction force
Kulcsszavak: frissbeton, reológiai modell, nedves szemcsehalmaz, folyadékhíd, nedvesíthetőség, nedvesítési szög, kölcsönhatási erő

Hooman HOORNAHAD
received his PhD from Faculty of Civil Engineering and Geosciences, Delft University of Technology. His field of interest is rheology of paste, mortar, concrete and granular material. He is RILEM member and (co-)author of about 20 conference and journal papers. He was a lecturer and research associate in American Concrete Institute (ACI), Iran Chapter.

Eduardus A. B. KOENDERS
is Full Professor and Head of the Institute for Construction and Building Materials, Faculty of Civil and Environmental Engineering, Technical University of Darmstadt. His research interests include multi-scale modeling of hydration and moisture transport, sustainable and durable materials, recycled aggregates and energy saving material concepts for construction. He is RILEM member and chairman of the RILEM EAC MMC course on multi-scale modeling of concrete.

Klaas van BREUGEL
is Full Professor and Head of the Section Materials and Environment, Faculty of Civil Engineering and Geosciences, Delft University of Technology. His fields of expertise concerns design of concrete containment structures, extreme load conditions, early age concrete and modelling and simulation for materials and structures. He is member of several international organisations ACI, RILEM, IABSE and fib. He has chaired several national and international research committees. In 2013 the Ageing Centre for Materials, Structures and Systems was launched under his supervision.

1. Introduction

Fresh concrete is considered as an intermediate class between cement pastes and granular materials from a rheological perspective [1, 2, 3]. Fresh concrete exhibits a complex behaviour which can be close to either a cement paste or a granular system, depending on the properties and proportion of constituents in the mixture. The aim of this research is to explain and model the rheological behaviour of fresh concrete mixtures and to create a fundamental basis for developing mixtures with a predefined deformational performance [2].

In this study, a mixture is considered as an assembly of mutually interacting "particle-paste-particle" systems (see *Fig. 1.a*), in which the rheological behaviour of the mixture is related to the inter-particle interactions [4, 5]. A system consists of a pair of two-phase elements connected by a paste bridge (see *Fig. 1.b*). Each element consists of an aggregate particle covered with a paste layer. A paste bridge represents the cohesion between the elements and resists increasing distance between two elements [2].

In order to characterize the interaction in a "particle-paste-particle" system the concept of capillary cohesion in wet granular material [6], which describes the interaction between two particles connected by a liquid bridge (see *Fig. 1.c*), is considered the first step in the investigation. First, the interactions in "particle-liquid-particle" systems are studied experimentally. These tests are then extended for "particle-paste-particle" systems. In this paper preliminary results of the experimental work on "particle-liquid-particle" systems are provided.

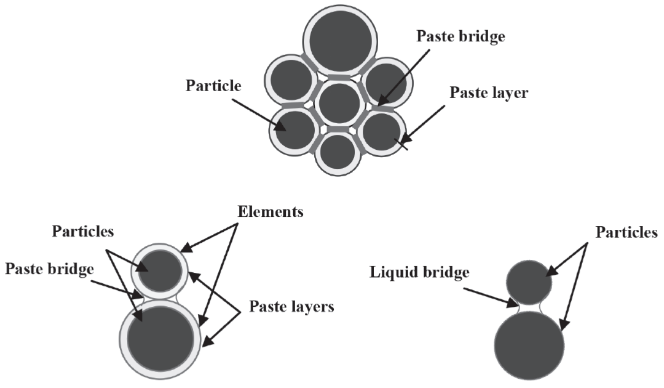

Fig. 1. *Features of two-phase modeling approach [2, 4].*
(a) Scheme of an assembly of mutually interacting "particle-paste-particle" systems
(b) "Particle-Paste-Particle" system
(c) "Particle-Liquid-Particle" system
1. ábra *A két fázisú model alapelvei [2, 4].*
(a) Kölcsönhatásban lévő szemcse-pép-szemcse rendszer sémája
(b) Szemcse-pép-szemcse rendszer
(c) Szemcse-folyadék-szemcse rendszer

2. Interaction between two particles connected by a liquid bridge

In a static or quasi-static situation, where the cohesion dominates over other effects generated by the liquid, such as viscosity and lubrication, the magnitude of the liquid bridge force is related to the geometry of a liquid bridge [6, 7, 8]. The geometry of a liquid bridge depends on 1) the characteristics of the particles (shape, size and wettability), 2) the surface tension of the liquid, 3) the volume of the liquid and 4) the inter-particle distance [2, 6, 9]. In this paper the wettability of solid particles and its effect on the force-distance relation of two particles connected by a liquid bridge is in the main focus.

Wettability is the ability of a solid surface to be wetted when it is in contact with a liquid. Wettability can be characterized by the contact angle θ_o, i.e. the angle at which the liquid–gas interface meets the solid–liquid interface (see *Fig. 2*). When the solid is extremely easy to wet, the contact angle has a low value. In this case a thin film of liquid is formed on the surface (see *Fig. 2.a*). It means that the liquid is strongly attached to the solid surface. With decreasing wettability of solid surface and thus increasing contact angle θ_o the solid–liquid interface decreases (see *Fig. 2.b*). When the solid is extremely difficult to wet, the contact angle θ_o approaches 180°, corresponding to the spherical drop having only one point of contact with the solid [10].

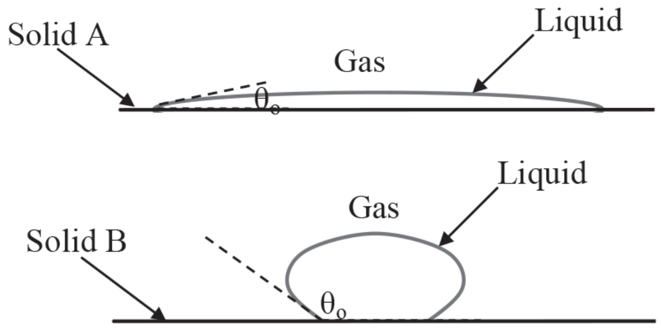

Fig. 2. Scheme of configuration of a liquid on a solid surface.
(a) Solid surface with high wettability (low contact angle θ_o)
(b) Solid surface with low wettability (high contact angle θ_o)
2. ábra Folyadékcsepp elhelyezkedésének vázlata szilárd felületen.
(a) Jól nedvesíthető szilárd felület (kis nedvesítési szög θ_o)
(b) Rosszul nedvesíthető szilárd felület (nagy nedvesítési szög θ_o)

3. Experimental test

In order to study the force-distance relationship between two particles connected by a liquid bridge a displacement controlled tensile test was performed by a specially designed experimental apparatus (see *Fig. 3*) [2]. In the set-up, one of the particles (lower one) is fixed to a support mounted at a highly sensitive balance (10^{-4} g) and the other particle (upper one) is fixed on a cantilever and can move vertically. The movement of this cantilever is controlled by a velocity controlled micrometer screw and its displacement is recorded by a LVDT sensor connected to a computer.

After putting the liquid drop of a given volume on the stationary particle, the two particles are brought into contact. At this stage, the test starts and the distance between the two particles is gradually increased until the point is reached at which rupture of the liquid bridge occurs [2]. At selected inter-particle distances, the magnitude of the interaction force is measured. During the test, a picture of the liquid bridge is recorded for the profile analysis. Temperature and relative humidity in the chamber around the specimen is kept between 22-23° C and between 97-100 %, respectively.

Tests were carried out using spherical stainless steel and glass particles and water was used as the liquid with a surface tension of σ_{in}=0.072 mN/mm. The contact angle θ_o between water and stainless steel is about 60° and between water and glass about 15° (see *Fig. 4*).

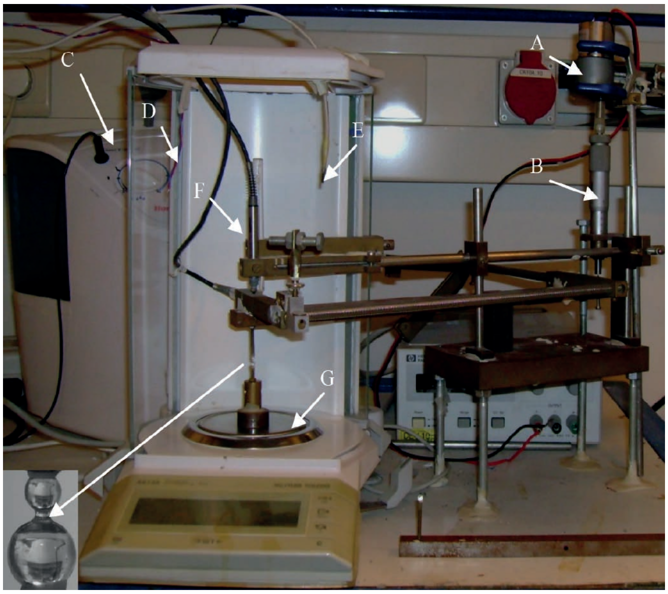

Fig. 3. Test device used for studying the interaction between two particles connected by a liquid bridge or two elements connected by a paste bridge. A: Velocity control armature, B: Micrometer screw, C: Humidifier, D: Temperature sensor, E: Humidity sensor, F: LVDT sensor, G: Scale.
3. ábra Vizsgálóeszköz két szemcse folyadékhíddal vagy két szemcse péphíddal történő érintkezésének vizsgálatához. A: Sebesség szabályzó állványzat, B: Mikrométercsavar, C: Párásító készülék, D: Hőmérő szenzor, E: Páratartalom szenzor, F: Útadó szenzor, G: Mérleg.

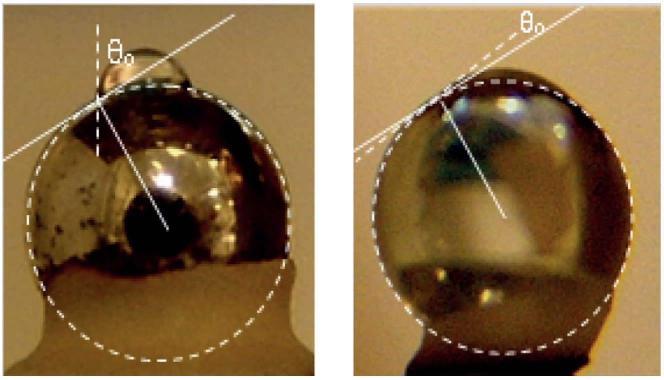

Fig. 4. Contact angle between a water drop and the surface of a particle.
(a) Stainless steel particle (θ_o=60°)
(b) Glass particle (θ_o=15°)
4. ábra Nedvesítési szög vízcsepp és szemcsefelület között.
(a) Rozsdamentes acél szemcse (θ_o=60°)
(b) Üveg anyagú szemcse (θ_o=15°)

4. Results and discussion

The results for 4 mm stainless steel and glass particles for two different volumes of the water bridge (0.1 mm^3 and 0.5 mm^3) are shown in *Fig. 5*. Results show that with increasing inter-particle distance, S the interaction force first increases and then gradually decreases. This behaviour is more noticeable for the stainless steel-water-stainless steel system than for the glass-water-glass system. This behaviour can be explained as follows.

Any force that tries to change the actual shape of the drop in the equilibrium condition causes some instability inside the drop [2]. During the experimental test, it turned out that instability happens in the system when the inter-particle distance becomes less than the height of the drop due to the compressive stress applied to the drop. The applied force tries to increase the solid-liquid interface while the liquid naturally resists against this deformation. This leads to an increase in the contact angle between the profile of the bridge and the solid surface at the intersection of the three-phase interfaces (see θ in *Fig. 6*) [2, 6].

The instability in the system increases by gradually decreasing the inter-particle distance. As the contact angle between water and glass is small (θ_o=15°), water can spread more easily over the surface of the glass particle in comparison with a spread of water on the surface of stainless steel particles, where the contact angle is considerably larger (θ_o=60°) (see *Fig. 4*). Therefore, in the case of glass and water, the drop experiences less instability during the test than in the case of stainless steel and water.

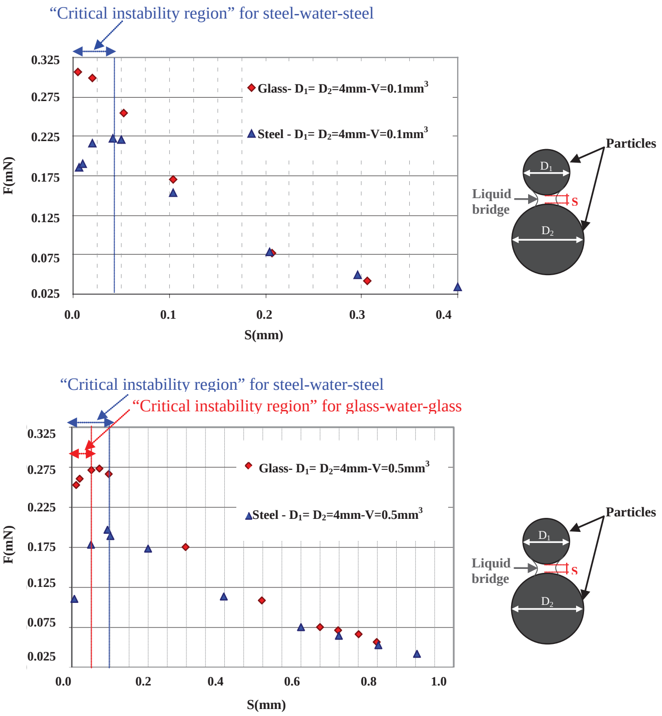

Fig. 5. *Force-distance plot for 4 mm stainless steel and glass particle-pairs.*
(a) Volume of the water bridge V=0.1 mm^3
(b) Volume of the water bridge V=0.5 mm^3

5. ábra *Erő-elmozdulás ábrák 4 mm átmérőjű rozsdamentes acél és üveg anyagú szemcsékre.*
(a) Vízhíd térfogata V=0.1 mm^3
(b) Vízhíd térfogata V=0.5 mm^3

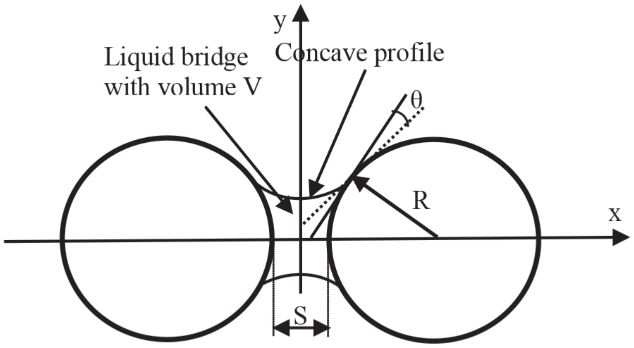

Fig. 6. *Characteristic geometrical scheme of a symmetrical liquid bridge between two particles. θ, contact angle between the liquid bridge and particle surface, is the angle between a line tangent to the gas-liquid interface and a line tangent to the liquid-solid interface at the intersection of three-phase interfaces.*

6. ábra *Két szemcse között kialakuló folyadékhíd jellegzetes geometriai sémája. A nedvesítési szög θ, a gáz-folyadék határfelülethez húzott érintő és a folyadék-szilárd fázis határfelülethez húzott érintő által bezárt szög, a három fázis érintkezési pontjában.*

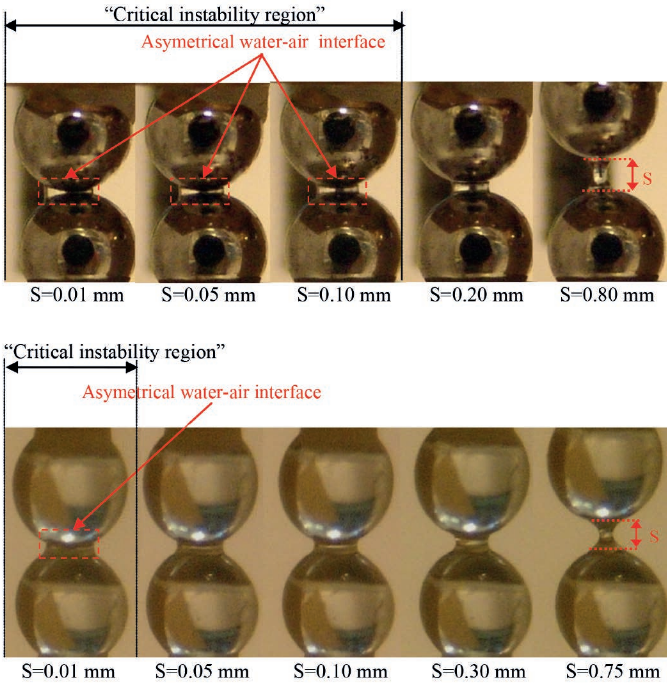

Fig. 7. *Evolution of the shape of the water bridge with the inter-particle distance corresponding to the results indicated in Fig. 5.b. Sc is critical inter-particle distance.*
(a) 4 mm stainless steel particles (θo= 60°, Sc<~0.10 mm)
(b) 4 mm glass particles (θo =15°, Sc<~0.05 mm)

7. ábra *A vízhíd alakjának változása a szemcsék közötti távolság változtatásának hatására, az 5.b ábrán bemutatott eredményekhez kapcsolódóan. Az ábrán Sc a szemcsék közötti kritikus távolság.*
(a) 4 mm átmérőjű rozsdamentes acél szemcsék (θo= 60°, Sc<~0.10 mm)
(b) 4 mm átmérőjű üveg anyagú szemcsék (θo =15°, Sc<~0.05 mm)

The evolution of the shape of the liquid bridges, corresponding to the results of *Fig. 5.b*, is shown in *Fig. 7*. It is observed that the instability effect becomes significant in the "critical instability region", i.e. the region where the water-air interface becomes asymmetrical concave-convex (see *Fig. 7*). In this region the contact angle, θ between the liquid bridge and the particle surface becomes larger than the contact angle between the drop and the particle surface, θ_o [2]. This effect becomes clearer by gradually decreasing the distance between the two particles. The “critical instability region” can be distinguished by the critical inter-particle distance, S_c representing the inter-particle distance below that liquid bridge profile becomes asymmetrical. As it is shown in *Fig. 7* the critical inter-particle distance, S_c depends on the wettability of the particles. Critical inter-particle distance, S_c increases with decreasing wettability of particles.

For the stainless steel-water case, with contact angle of θ_o=60° between the water drop and surface of the particle and with drop volume of $V=0.5\ mm^3$, the instability effect became significant when the distance between the two particles becomes less than about 0.10 mm (see *Fig. 5.b*). In this case the maximum contact angle between the profile of the bridge and the solid surface becomes more than 90° with decreasing inter-particle distance (see *Fig. 7.a*). For the glass-water case, with a contact angle of θ_o=15° between the water drop and surface of the particle and with drop volume of $V=0.5\ mm^3$, the instability effect became significant at a shorter inter-particle distance, S than that for the stainless steel-water case. In this case the critical inter-particle distance, S_c is less than about 0.05 mm (see *Fig. 5.b*). With decreasing inter-particle distance the maximum contact angle between the profile of the bridge and the solid surface approaches to about 40°. Outside the “critical instability region” a quite symmetrical concave profile is observed for the liquid bridge between the particles (see *Fig. 7*). In this region the interaction force decreases with gradually increasing inter-particle distance (see *Fig. 5*). For the stainless steel-water case with drop volume of $V=0.1\ mm^3$, the instability effect became significant for an inter-particle distance less than about 0.05 mm (see *Fig. 5.a*). For the glass-water case with drop volume of $V=0.1\ mm^3$, instability effect was not observed (see *Fig. 5.a*). It is also observed that rupturing of the liquid bridge happens at a larger inter-particle distance with increasing contact angle, θ_o. For the stainless steel-water case the rupture distance is about 20% larger than that of the glass-water case (see *Fig. 5*).

5. Conclusions

Results of an experimental study of the force-distance relation between two particles connected by a liquid bridge have been addressed in this paper. The experimental research on water bridges was the part of the experimental study which has been performed with the aim to create a fundamental basis for developing concrete mixtures with a predefined deformational performance. The effect of the wettability of the particles on the force-distance relation of two particles connected by a liquid bridge in static and/or quasi-static situations was in focus in this paper, where the magnitude of interaction force is related to the geometry of the liquid bridge. Wettability, which is defined as the ability of a solid surface to be wetted when in contact with a liquid, can be characterized by the contact angle θ_o, i.e. the angle at which the liquid–gas interface meets the solid particle–liquid interface. The displacement controlled tensile tests were carried out using spherical stainless steel with low wettability (θ_o=60°) and glass particles with high wettability (θ_o=15°) and water was used as the liquid with a surface tension of σ_{in}=0.072 mN/mm. With respect to the inter-particle distance (S), two regions have been distinguished:

- Region A (critical instability region), with an inter-particle distance $0<S<S_c$, where the profile of the liquid bridge is asymmetrical concave-convex. In this region the interaction force increases with increasing inter-particle distance.
- Region B, with an inter-particle distance $S \geq S_c$, where the profile of the liquid bridge is symmetrical concave. In this region interaction force decreases with increasing inter-particle distance.

It was found that the critical inter-particle distance S_c depends on the wettability of the particles. With decreasing wettability of particles, increasing θ_o, the critical inter-particle distance S_c increases. It is also observed that with decreasing wettability of particles, increasing θ_o, rupturing of the liquid bridge happens at a larger inter-particle distance.

References

[1] Coussot, P. (2005): Rheometry of Pastes, Suspensions, and Granular Materials. *John Wiley & Sons*, New Jersey.

[2] Hoornahad, H. (2014): Toward Development of Self-Compacting No-slump Concrete Mixtures- PhD Thesis. *Ipskamp Drukkers BV.*, Delft.

[3] Hoornahad, H. – Koenders, E. A. B. (2013): Effect of the mix composition on rheological behavior of a fresh granular-cement paste material. *Proc. 1st RILEM Int. Conf. on Rheology and Processing of Construction Materials,* Paris.

[4] Hoornahad, H. – Koenders, E. A. B. (2014): Modelling rheological behavior of fresh cement-sand mixtures. *Proc. 33rd Int. Conf. on Ocean, Offshore and Arctic Engineering,* San Francisco.

[5] Hoornahad, H. – Koenders, E. A. B. (2012): Simulation of the slump test based on the discrete element method (DEM). *Advanced Materials Research,* Vols. 446-449, pp. 3766-3773. http://dx.doi.org/10.4028/www.scientific.net/AMR.446-449.3766

[6] Soulié, F. – Cherblanc, F. – El Youssoufi, M. S. – Saix, C. (2006): Influence of liquid bridges on the mechanical behavior of polydisperse granular materials. *Int.J. for Numerical and Analytical Methods in Geomechanics.* Vol. 30, pp. 213-228. http://dx.doi.org/10.1002/nag.476

[7] Mitarai, N. – Noriz, F. (2006): Wet granular materials. *Advances in Phys.* Vol. 55, No. 1-2, pp. 1-45. http://dx.doi.org/10.1080/00018730600626065

[8] Lambert, P. (2007): Capillary Forces in Microassembly - Modeling, Simulation, Experiments and Case Study. *Springer.* http://dx.doi.org/10.1007/978-0-387-71089-1

[9] Hoornahad, H. – Koenders, E. A. B. – van Breugel, K. (2010): Capillary cohesion between two spherical glass particles. *Key Engineering Materials,* Vols. 417-418, pp. 449-452. http://dx.doi.org/10.4028/www.scientific.net/KEM.417-418.449

[10] Miller, C. A. – Neogi, P. (1985): Interfacial Phenomena Equilibrium and Dynamic Effects Surfactant Science Series 17, *Taylor & Francis Group.*

Ref.:

Hoornahad, Hooman – **Koenders**, Eduardus A. B. – **van Breugel**, Klaas: *Wettability of particles and its effect on liquid bridges in wet granular materials*

Építőanyag – Journal of Silicate Based and Composite Materials, Vol. 67, No. 4 (2015), 139–142. p.

http://dx.doi.org/10.14382/epitoanyag-jsbcm.2015.23