

Rheology of weakly wetted granular materials - a comparison of experimental and numerical data

Rüdiger Schwarze · Anton Gladkyy · Fabian Uhlig · Stefan Luding

Received: date / Accepted: date

Abstract Shear cell simulations and experiments of weakly wetted particles (a few volume percent liquid binders) are compared, with the goal to understand their flow rheology. Application examples are cores for metal casting by core shooting made of sand and liquid binding materials.

The experiments are carried out with a Couette-like rotating viscometer. The weakly wetted granular materials are made of quartz sand and small amounts of Newtonian liquids. For comparison, experiments on dry sand are also performed with a modified configuration of the viscometer. The numerical model involves spherical, monodisperse particles with contact forces

and a simple liquid bridge model for individual capillary bridges between two particles. Different liquid content and properties lead to different flow rheology when measuring the shear stress-strain relations. In the experiments of the weakly wetted granular material, the apparent shear viscosity η_g scales inversely proportional to the inertial number I , for all shear rates. On the contrary, in the dry case, an intermediate scaling regime inversely quadratic in I is observed for moderate shear rates. In the simulations, both scaling regimes are found for dry and wet granular material as well.

Keywords granular material, rheology, DEM, wet contact, liquid bridge, micro-macro transition

R. Schwarze
Institute of Mechanics and Fluid Dynamics,
TU Bergakademie Freiberg
Lampadiusstr. 4, 09596 Freiberg, Germany
e-mail: ruediger.schwarze@imfd.tu-freiberg.de

A. Gladkyy
Institute of Mechanics and Fluid Dynamics,
TU Bergakademie Freiberg
Lampadiusstr. 4, 09596 Freiberg, Germany
e-mail: anton.gladkyy@imfd.tu-freiberg.de

F. Uhlig
Institute of Mechanics and Fluid Dynamics,
TU Bergakademie Freiberg
Lampadiusstr. 4, 09596 Freiberg, Germany
Temporary address during the study:
Multi Scale Mechanics (MSM), Engineering Technology
(CTW) and MESA+, University of Twente
P.O.Box 217, 7500 AE Enschede, The Netherlands

S. Luding
Multi Scale Mechanics (MSM), Engineering Technology
(CTW) and MESA+, University of Twente
P.O.Box 217, 7500 AE Enschede, The Netherlands
e-mail: s.luding@utwente.nl

1 Introduction

Dry granular matter and its flow rheology have been the subject of detailed studies during the last years and slowly their interesting behavior becomes more and more clear [1,2]. The other extreme case are particles suspended in fluids – a field of wide relevance in industrial processes – which nowadays are mostly understood and can be modeled reasonably well [3–7].

However, weakly wetted granular materials have recently attracted new attention, see e.g. Ref. [8–12], even though they were studied earlier [13–16]. Wet as well as dry granular rheology [17,18] plays an important role in geotechnical and geophysical context [19,20], as well as in several technical processes e.g. in growth agglomeration [21], for die-filling [22,23], or in the production of sand cores for casting by core shooting [24]. In the latter example, weakly wetted granular materials are mixtures of a granular matter and few volume-percent of liquid binder. Like dry granular materials, they exhibit non-Newtonian flow behavior, where the relations

between shear stresses and shear rates, for example, can be expressed by nonlinear functions. Since the presence of small amounts of liquid change the rheological behavior of the granular material markedly [25,26], detailed knowledge of the constitutive equations of these materials is of fundamental importance for the control of the corresponding processes. As an example, the filling flow – even in complex core boxes – can be analyzed by CFD simulations [27] when the rheological model of the material is known.

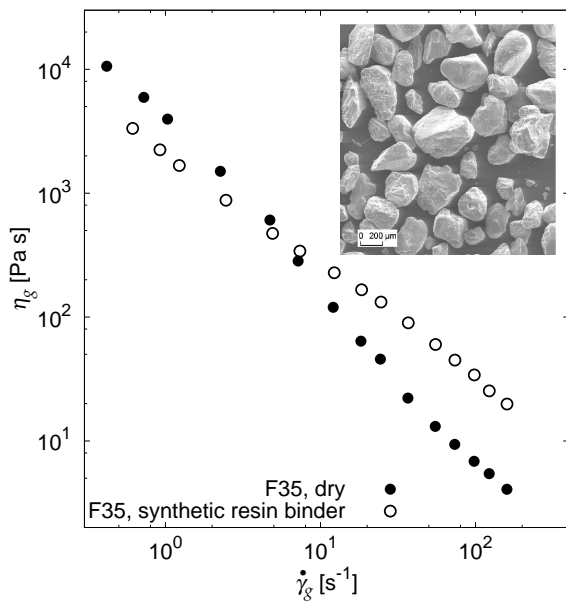


Fig. 1: Characteristics of core shooting material with quartz sand F35 [28]: Rheological data from measurements with dry F35 and with core shooting material with a mass ratio $m_{SR}/m_F = 0.02$ between the mass m_{SR} of the synthetic resin binder and the mass m_F of F35, see section 2 and Ref. [25] for more details. Inset: SEM picture of a sample of F35.

As an example, Fig. 1 shows some rheometer experiment [25] flow curves of a typical core shooting material made of quartz sand F35 (with mean particle diameter $d_P = 0.18$ mm) [28] and synthetic resin binder and the pure, dry F35. The scaling $\eta_g(\dot{\gamma}_g)$ of the apparent viscosity η_g with the shear rates $\dot{\gamma}_g$ differs markedly between the core shooting material and the dry granular F35.

Heuristically, this difference can be explained by the capillary bridges of the liquid binder between individual

sand particles. A more detailed review of the rheological measurements is given in the next section.

Unfortunately, the realization of rheometer experiments involving weakly wetted granular materials is complicated and time consuming. Therefore, alternative and more efficient methods for rheological investigations are highly desirable. In this paper, we use the split-bottom ring shear setup of a rheometer [29] for discrete-element method (DEM) simulations of these partly wet granular materials. Similar DEM simulations of wet granular materials have been performed in order to study the micro-mechanics in cohesive mixing processes [30], the discharge from hoppers [31] or the mixing in a blade mixer [32]. In these papers, explicit capillary forces are added to the contact forces in order to properly describe the interactions between two particles in the wet granular material. As an alternative to the most simple approach pursued below, Grima and Wypych [33] employ an implicit model of the capillary force in their simulations, while Mani et al. [8] explicitly allow for liquid migration between the particles and across the bridges.

Starting from the DEM results, we apply a local micro-macro transition [34–36] in order to obtain rheological flow rules for weakly wetted granular materials. First results indicate, that the numerically determined flow rules exhibit similar differences between dry and wet granular materials as in the experiments. The device used to measure the stress-strain relations is the split-bottom ring-shear cell as invented by Fenistein et al. [29] and used by others for dry, frictional and (van der Waals) elasto-plastic adhesive particle systems [34–42].

2 Previous rheological measurements

Rheological data of weakly wetted granular materials (core shooting materials) have been recently measured in a Searl-type rotating viscometer [25] with a fixed bottom and outer cylinder wall and a rotating inner cylinder wall. Sand particles are glued to the fixed and rotating cylinder walls in order to define proper wall shear stress conditions and reduce wall-slip. In these shear cell experiments, a shear band width w_{sb} of about 10 - 20 particle diameters d_P was expected.

For measurements on dry sand, the gap width $b_{dry} = 2$ mm $\simeq 10 d_P \simeq w_{sb}$ between the inner and outer cylinder was fitted to the assumed width of the shear zone in the granular material. With this setup, all the material in the gap should be sheared. Here, the upper annulus of the viscometer gap was open.

For measurements on core shooting materials, the upper annulus was closed by a movable circular ring

| p_e [kPa] | F35, synthetic resin $m_{SR}/m_F = 0.01$ | | F35, sodium silicate $m_{SS}/m_F = 0.02$ | |
|-------------|---|-------|---|-------|
| | 4 kPa | 8 kPa | 4 kPa | 8 kPa |
| K | 2084 | 3291 | 1807 | 2603 |
| α | 0.90 | 1.07 | 1.11 | 1.10 |

Table 1: Parameters of the flow rules for core shooting materials of Fig. 2 in Eq. (2). Values of K and α are obtained from a least square fit of Eq. (2) to the wet data, for $0.4\text{s}^{-1} < \dot{\gamma}_g < 200\text{s}^{-1}$. Here, values of K are mere fit-parameters since their units are non-linearly dependent on the values of α and are thus not given.

in order to superimpose an external pressure p_e to the weakly wetted granular material. Without p_e , fissures arose in the shear zone, which induced the disruption of the measurements. In all measurements, p_e exceeded the pure hydrostatic pressure level $p_h = \rho g h \simeq 1\text{ kPa}$ of the core shooting material, with bulk density $\rho \simeq 1400\text{ kg/m}^3$ and fill level $h \simeq 70\text{ mm}$ in the viscometer gap. The gap width $b_{wet} = 10\text{ mm} \simeq 50d_p \simeq 5w_{sb}$ between the inner and outer cylinder was increased for these experiments. We assume, that parts of the material in the gap were not sheared in this configuration. Therefore, the exact width of the shear zone remains unknown, which induces an uncertainty in the absolute values of the shear rate. However, the scaling $\eta_g(\dot{\gamma}_g)$ should have been correctly resolved in these measurements, too, only amplitudes of η_g are maybe somewhat shifted.

In Fig. 2, some results of the rheometer measurements on two core shooting materials with F35 as basic sand but different liquid binders are displayed. For both materials, the apparent shear viscosity

$$\eta_g = \frac{|\tau_g|}{\dot{\gamma}_g} \quad (1)$$

of the core shooting material exceeds the values of the dry granular F35 markedly for shear rates $\dot{\gamma}_g > 10\text{ s}^{-1}$. Here, the quantities τ_g and $\dot{\gamma}_g$ are *globally* defined, i.e. they describe the dynamics of the bulk granular material and set-up: The mean shear stress $\tau_g = F_{t,o}/A_o$ is the quotient of the tangential force $F_{t,o}$ at the outer cylinder wall, which is measured by a force transducer, and the outer cylinder wall area A_o . The mean shear rate $\dot{\gamma}_g = U_i/b$ is approximated by the quotient of the velocity U_i of the rotating inner cylinder wall and the gap widths $b = b_{dry}$ or $b = b_{wet}$.

Problems in the realization of the measurements are indicated by the large uncertainty in the results for the core shooting material with synthetic resin binder and $p_e = 8\text{ kPa}$.

The flow rules of the core shooting materials in the measured shear rate intervals can be described as a first

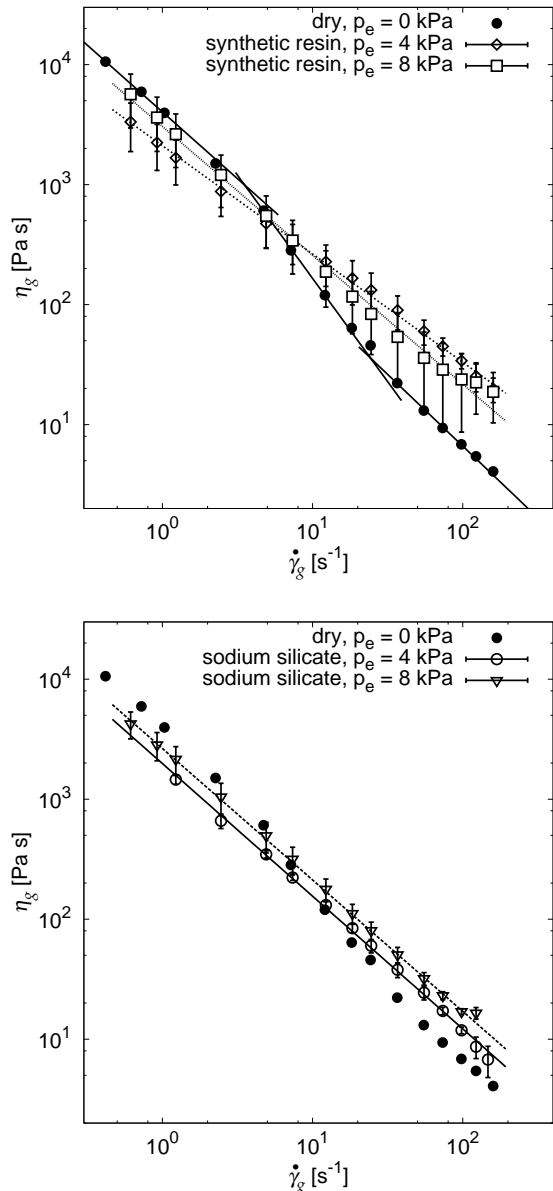


Fig. 2: Apparent shear viscosity $\eta_g(\dot{\gamma}_g)$ for core shooting material made of F35 and synthetic resin binder with $m_{SR}/m_F = 0.01$ (top), and made of F35 and sodium silicate binder with $m_{SS}/m_F = 0.02$ (bottom); error bars indicate the standard deviation of the experimental data. Results of dry F35 are also given for comparison, performed with smaller gap b . Measurements are made with different external pressure levels p_e . Continuous and dashed lines indicate the scaling between η_g and $\dot{\gamma}_g$, obtained from least square fits of Eq. (2) to the wet data, for $0.4\text{ s}^{-1} < \dot{\gamma}_g < 200\text{ s}^{-1}$.

approximation by a power law

$$\eta_g(\dot{\gamma}_g) = K |\dot{\gamma}_g|^{-\alpha} \quad (2)$$

with the consistency factor K and a power law index α . (Note that the nonlinear power-law spoils the units and has to be phrased better using dimensionless quantities as discussed below.) Table 1 summarizes the values of K and α for the different core shooting materials for different levels of p_e .

All measurements indicate shear-thinning behavior of the core-shooting materials with power laws close to $(\dot{\gamma}_g)^{-1}$. However, some interesting differences are also found: At the same levels of p_e , the parameters K and α differ markedly between the two core shooting materials with the same basic sand but different liquid binders. Obviously, the presence of small amounts of different liquids in the same basic granular material affects the flow behavior significantly. For increasing p_e , the power law index α in case of core shooting material made of F35 with synthetic resin binder decreases, whereas α rises for core shooting material made of F35 with sodium silicate binder.

The proposed scaling $\eta_g \propto p_e/\dot{\gamma}_g$ of Jop et al. [17] is only found in the core shooting material with sodium silicate binder, where the continuous and dashed curves are nearly parallel. In case of the core shooting material with synthetic resin binder, where the continuous and dashed curves intersect, the scaling seems to be more complex.

The rheological behavior of dry sand in the same rheometer (but with smaller gap b) seems to be more complex; we fail to fit the experimental data by a simple power law ansatz, Eq. (2), with constant K and α . Instead, we found three distinct regimes for low, medium and high shear rates. The scaling is $\eta_g \propto \dot{\gamma}_g^{-1.1}$ for low, $\eta_g \propto \dot{\gamma}_g^{-1.7}$ for medium, and $\eta_g \propto \dot{\gamma}_g^{-1.2}$ for high shear rates, see Fig. 2(top). These different regimes are not found for the weakly wetted granular materials.

On the way to a better theoretical description of this complex rheology, it is advisable/necessary to use a dimensionless shear rate:

$$I_g = \frac{\dot{\gamma}_g d_P}{\sqrt{p_g/\rho}} \quad (3)$$

as defined in [1]. This number describes the ratio of inertial and confining stress, i.e. pressure, of the sheared material. For the evaluation of the different measurements, the global pressure level p_g is estimated by $p_g \simeq p_e$, because $p_e \gg p_h$ as explained above. The quantities $\dot{\gamma}_g$, p_g , and I_g are *global* parameters, i.e. are obtained from an external measurement that implies an average over the whole shear cell. In case of the dry sand, where

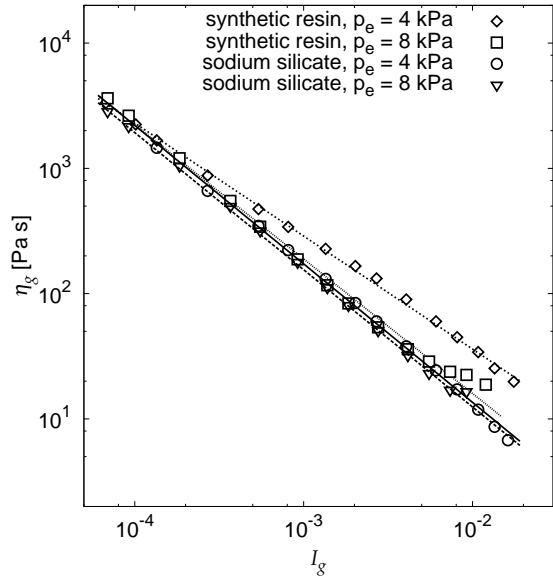


Fig. 3: Apparent shear viscosity as a function of dimensionless shear rate for the two core shooting materials and the two external pressure levels p_e of Fig. 2. Continuous and dashed lines indicate the scaling $\eta_g \sim I_g^{-\alpha}$, with α from table 1.

no external pressure p_e was applied, I_g cannot be properly evaluated for these measurements.

Fig. 3 summarizes the observed apparent shear viscosities $\eta_g(I_g)$ for the different core shooting materials and the different levels of p_e . In this semi-dimensional representation, all measurements follow roughly the same shear-thinning behavior of the core-shooting materials with scaling as $\eta_g \propto I_g^{-\alpha}$ with $\alpha \simeq 1$, which are indicated by continuous and dashed lines as in Fig. 2.

Interestingly, the synthetic binder is sensitive to the confining stress, whereas the sodium silicate binder is not. The former displays considerably higher apparent viscosity for smaller p_e . The more systematic investigation of these subtle differences are ongoing and will be published elsewhere.

3 Model fundamentals

The flow of a sheared, weakly wetted granular material is now investigated by means of DEM simulations.

3.1 Equations of motion

The DEM model is based on Newton's equations of motion for the translational and rotational degrees of free-

dom of a spherical particle

$$m_i \frac{d^2 \underline{r}_i}{dt^2} = \sum_{\{i,j\}=c} \underline{f}_{ij} + m_i \underline{g} \quad (4)$$

$$J_i \frac{d\omega_i}{dt} = \sum_{\{i,j\}=c} \underline{l}_{ij} \times \underline{f}_{ij} \quad (5)$$

with mass m_i , position \underline{r}_i , moment of inertia J_i , and angular velocity ω_i of particle i . The right hand side terms in Eq. (4) are the sum of the inter-particle forces \underline{f}_{ij} due to contacts c with particles j and volume/body forces, here from gravity \underline{g} . The right hand side in Eq. (5) is the torque arising from the contacts c with the branch vector \underline{l}_{ij} , i.e., the distance vector from the particle center to the contact point of the two particles i and j .

The inter-particle forces \underline{f}_{ij} are modeled by well-known force-overlap relations in combination with capillary forces, which are induced by liquid bridges between the interacting particles. Details of the contact force modeling are given in the next subsection.

3.2 Contact force law

Since the realistic modeling of the deformation of two interacting particles, e.g. in a core shooting material, is much too complicated, the inter-particle force is described by a force-overlap relation. The modeling of the contact force is based on the quantities/symbols given in Figs. 4 and 5.

The force \underline{f}_{ij} on particle i , from particle j , at contact c , can be decomposed into a normal and a tangential part as

$$\underline{f}_{ij} = f_{ij}^n \hat{n}_{ij} + f_{ij}^t \hat{t}_{ij} + f_{ij,c} \hat{n}_{ij}. \quad (6)$$

with normal \hat{n}_{ij} and tangential unit vector \hat{t}_{ij} at the contact point c . The normal force f_{ij}^n , the tangential force f_{ij}^t and the capillary force $f_{ij,c}$ due to a liquid bridge between particle i and j are specified below.

Two dry particles i, j with radius R , which are moving with velocities \underline{v}_i and \underline{v}_j and rotating with angular velocities ω_i and ω_j , interact, if the normal overlap δ is positive

$$\delta = 2R - (\underline{r}_i - \underline{r}_j) \cdot \hat{n}_{ij} > 0. \quad (7)$$

The *normal contact force* involves a linear repulsive and a linear dissipative force,

$$f_{ij}^n = k_n \delta + \gamma_n v_{ij}^n, \quad (8)$$

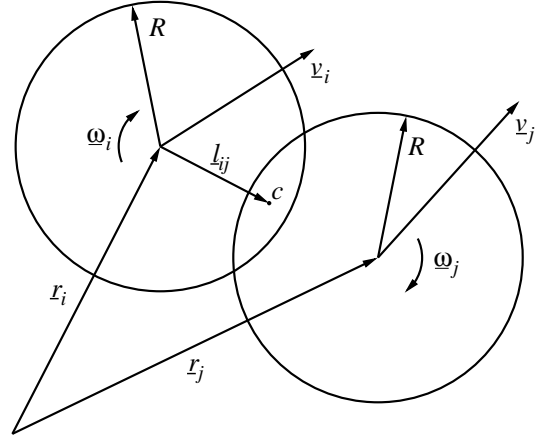


Fig. 4: Sketch of the contact between two particles i, j .

with normal spring stiffness k_n , normal viscous damping γ_n and normal velocity v_{ij}^n .

Our model is able to capture friction forces and torques, as well as rolling and torsion torques, as described in Ref. [43]. For the sake of brevity we did set the latter interactions to zero and focus on friction only, i.e. the tangential friction force is

$$f_{ij}^t = k_t \chi + \gamma_t v_{ij}^t, \quad (9)$$

with tangential spring stiffness k_t , tangential viscous damping γ_t and tangential velocity v_{ij}^t , where χ is the integral of v_{ij}^t over time, adapted such that the tangential force is limited by Coulomb sliding friction

$$f_{ij}^t \leq \mu_C f_{ij}^n, \quad (10)$$

with Coulomb's coefficient of friction μ_C .

Note, that f_{ij}^n and f_{ij}^t give only non-zero contributions to \underline{f}_{ij} , when the two particles are in contact, $\delta > 0$. The *capillary force* $f_{ij,c}$ is also active when two particles separate after a contact. Details of the modeling of $f_{ij,c}$ are given in the next subsection.

3.3 Capillary forces

The shapes of liquid bridges between individual particles of a granular medium depend strongly on the amount of the added liquid, see e.g. Refs. [14,16]. For core shooting materials with low mass ratios between binder and dry sand or sand-like materials we expect, based on the findings in [16], that the grains are connected by individual capillary bridges. The relevant parameters of such a bridge are indicated in Fig. 5.

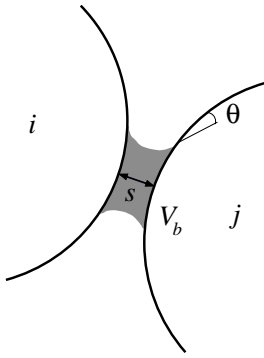


Fig. 5: Sketch of a liquid bridge between particles i and j with the bridge length s , the bridge volume V_b and the contact angle θ .

The liquid volume of the bridge is V_b . The length $s = -\delta$ of the bridge is given by the surface distance of the two particles, which are connected by the bridge. Finally, the equilibrium contact angle $\theta < 90^\circ$ is found at the bridge-particle contact line.

With these parameters, we approximate the interparticle force $f_{ij,c}$ of the capillary bridge according to the proposal of Willett et al. [15], see also [16,32], who calculated $f_{ij,c}$ with the gorge method at the bridge neck

$$f_{ij,c} = \frac{2\pi\gamma R \cos(\theta)}{1 + 1.05\hat{s} + 2.5\hat{s}^2}. \quad (11)$$

with surface tension, γ and dimensionless $\hat{s} = s\sqrt{R/V_b}$. Note, that $f_{ij,c}$ exists only during and past a contact between particle i and j , providing a non-zero contribution to \underline{f}_{ij} until the total distance s between i and j rises above the critical bridge length $s > s_{crit}$. Then, the bridge ruptures and $f_{ij,c}$ becomes zero. Several authors have proposed correlations between s_{crit} and other parameters of the capillary bridge [13,14]. We use the approximation of Willett et al. [15]

$$s_{crit} = R \left(1 + \frac{1}{2}\theta \right) \left[\left(\frac{V_b}{R^3} \right)^{\frac{1}{3}} + 0.1 \left(\frac{V_b}{R^3} \right)^{\frac{2}{3}} \right]. \quad (12)$$

The model equations presented above were implemented into the open-source DEM software package LIGGGHTS, version 2.3.2. The simulations were carried out at the HPC cluster CVC at the University Computer Center of the TU Bergakademie Freiberg.

3.4 Capillary force model validation

For the validation of the capillary force calculation, we perform DEM simulations according to the experiments described in Willett et al. [15]. There, the separation

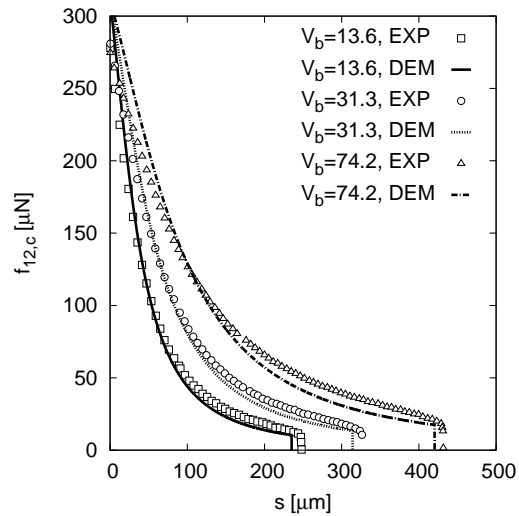


Fig. 6: Forces in a capillary bridge with contact angle $\theta = 0^\circ$ between two equal-sized particles; lines indicate results of the DEM simulation, points give the experimental data of Willett et al. [15]. Liquid bridge volumes V_b are given in nano-liters ($10^{-9}\text{l}=\text{nl}$).

process of two equal-sized particles with $R = 2.4\text{ mm}$ connected by a capillary bridge has been investigated. The liquid surface tension was $\gamma = 20.6\text{ mN/m}$. The particles were perfectly wetted by the liquid, i.e. $\theta = 0^\circ$. Experiments with different liquid bridge volumes $V_b = 13.6, 31.3$ and 74.2 nl have been carried out.

In the DEM simulations, we track the capillary force $f_{12,c}$ due to Eq. (11) when the particles separate with velocity $v_{12}^n = 0.001\text{ m/s}$ from the distance $s = 0$ (at the end of the mechanical contact between the two particles) until $s = s_{crit}$ (the rupture distance, Eq. (12)).

Fig. 6 gives a comparison of the DEM results and the experimental data of Willett et al. [15], showing the good agreement between our data and the measurements.

3.5 Setup of the numerical rheometer

The flow and rheology of dry and wet granular materials in a three-dimensional split bottom shear cell [29,34–36] are investigated with the DEM model. Basic parameters of the shear cell geometry are sketched in Fig. 7, with the values: $r_i = 14.7\text{ mm}$, $r_s = 85\text{ mm}$, $r_o = 110\text{ mm}$, and $U_o = 6.9\text{ mm/s}$.

In contrast to previous studies, where only a quarter of the rheometer was modeled, see Refs. [34–36], here we study the full ring-geometry. The rheometer is filled with $n_P \simeq 210000$ particles, which all have the same diameter $d_P = 2\text{ mm}$, with density $\rho_P = 2000\text{ kg/m}^3$. We

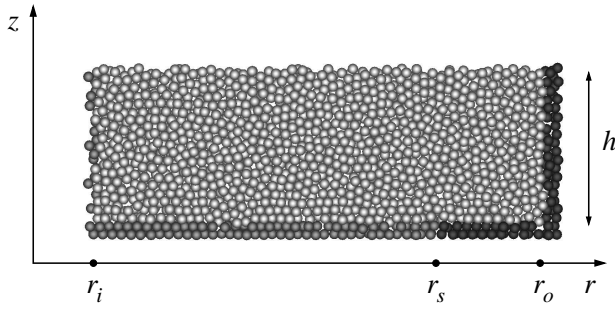


Fig. 7: Setup of the numerical rheometer, light gray: sheared granular material, medium gray: static, inner part of the shear cell, dark gray: rotating, outer part of the shear cell (the medium and dark gray particles are part of the rough walls of the shear cell and thus displayed as particles).

assume, that polydispersity has in a first approximation little influence on the macroscopic flow behavior of the granular material, similar to the findings in Refs. [44, 45]. The expected small effects of polydispersity should be investigated in more detail in the future, but are not subject of the present study.

The mechanical parameters of the particles are chosen as $k_n = 110 \text{ N/m}$, $\gamma_n = 2 \cdot 10^{-3} \text{ kg/s}$, $k_t = 12 \text{ N/m}$, $\gamma_t = 0.5 \cdot 10^{-3} \text{ kg/s}$, and $\mu_C = 0.01$ (if not mentioned otherwise) in order to match those of the numerical simulations by Luding [34, 36] for straightforward validation purposes.

An adaption of the contact model to real sand F35 is presently not possible and must be postponed since the material parameters are actually unknown. However, this is no essential constriction, because details of the contact model, including stiffness, have been found to cause only small differences for granular flows in the collisional and dense regimes, as long as the particles are not too soft, see e.g. Refs. [46, 47], where the effect of softness of the material was studied in more detail.

The results of our calculations with dry granular material ($f_c = 0$) are used to verify and validate the DEM code and force model implementation, in comparison with previous results [34]. The simulations with wet granular materials should reveal if the influence of the liquid bridges (the model of which was validated in subsection 3.4) on the apparent shear viscosity is the same as in the experiments.

For the simulations of wet granular materials, the equilibrium contact angle θ was varied between 0° and 20° , whereas the surface tension $\gamma = 20.6 \text{ mN/m}$ was kept constant. The simulations were conducted for two different bridge volumes V_b (4.2 nl, 42 nl) for each capillary bridge. Assuming an average number $n_{c,i} = 6$ of

capillary bridges per particle, this corresponds approximately to a mass ratio $m_l/m_{ap} = 0.15\%$, 1.5% between the liquid and the dry granular material, respectively. Here, the total mass $m_l = \sum_{i=1}^{n_P} \rho_l V_b n_{c,i}/2$ of the liquid is calculated with an arbitrary fixed value $\rho_l = 1000 \text{ kg/m}^3$, because ρ_l is not applied in the DEM model. The total mass of all dry particles is $m_{ap} = \sum_{i=1}^{n_P} m_i$. Note, that m_l/m_{ap} was found to change during the simulation, because $n_{c,i}$ varies in the interval $6 \lesssim n_{c,i} \lesssim 7$, however this small effect was ignored and $n_{c,i} = 6$ was kept constant for the calculation of V_b .

The influence of other, more complex liquid distributions is analyzed in ongoing simulations and will be reported elsewhere.

4 Results

4.1 Micro-macro transition

As in Refs. [34–36], continuum quantities like the solid-fraction ϕ , the velocity-field \underline{u} or the stress-field $\underline{\sigma}$ are computed by a micro-macro transition method from the DEM results, e.g.,

$$\phi(\underline{r}) = \frac{1}{\Delta t \Delta V} \int_{\Delta t} \sum_{i \in \Delta V} V_i dt, \quad (13)$$

$$\underline{u}(\underline{r}) = \frac{1}{\Delta t \Delta V} \int_{\Delta t} \left(\sum_{i \in \Delta V} V_i \underline{v}_i \right) dt \times \frac{1}{\phi(\underline{r})}, \quad (14)$$

$$\underline{\sigma}(\underline{r}) = \frac{1}{\Delta t \Delta V} \int_{\Delta t} \left(\sum_{i \in \Delta V} m_i \underline{v}'_i \otimes \underline{v}'_i + \sum_{c \in \Delta V} \sum_{\{i,j\}=c} \underline{f}_{ij} \otimes \underline{l}_{ij} \right) dt, \quad (15)$$

with fluctuation velocity $\underline{v}'_i = \underline{v}_i - \underline{u}(\underline{r})$, averaging time intervals of typically $\Delta t = 5 \text{ s}$, a discrete averaging time-step $dt = 0.05 \text{ s}$, and particle volume V_i , together with the (ring/torus-shaped) averaging volume ΔV at various positions $\underline{r} = (r, z)$ in the system (with cylindrical coordinates). Further parameter-fields like strain rate magnitude, shear stress magnitude, hydrostatic pressure, apparent viscosity and inertial number can be cal-

culated from these variables as:

$$\dot{\gamma} = \frac{1}{2} \sqrt{\left(\frac{\partial u_\varphi}{\partial r} - \frac{u_\varphi}{r}\right)^2 + \left(\frac{\partial u_\varphi}{\partial z}\right)^2}, \quad (16)$$

$$|\tau| = \sqrt{\sigma_{r\varphi}^2 + \sigma_{z\varphi}^2}, \quad (17)$$

$$p = \frac{1}{3} (\sigma_{rr} + \sigma_{zz} + \sigma_{\varphi\varphi}), \quad (18)$$

$$\eta = \frac{|\tau|}{\dot{\gamma}}, \quad (19)$$

$$I = \frac{\dot{\gamma} d_P}{\sqrt{p/\rho}}. \quad (20)$$

In contrast to the experimental setup, these parameters can be investigated *locally*, i.e., at arbitrary positions \underline{r} anywhere in the filled measurement volume (gap) of the rheometer.

All simulations which are discussed afterwards run for 20 s. For the average, only the period between 15 s and 20 s are into account. Therefore, the system is examined in quasi-steady state flow conditions (the transient behavior at the onset of shear is disregarded). However, it cannot be excluded, that long-time relaxation effects may have an impact on our findings, which is not adequately resolved by our relatively short simulations.

4.2 Dry granular material

Fig. 8 compares the shear stress intensity $|\tau|/p = \mu_m$ in our DEM simulations to the previous findings of Luding [34,36], with the same macroscopic friction value $\mu_m = 0.14$ for $\mu_C = 0.01$ as reported earlier.

In order to allow for a more quantitative analysis of the dynamic behavior of the dry granular material, its shear band structure is analyzed. The shear bands in dry granular matter have been intensively studied in experiments and DEM simulations, see e.g., Refs. [29, 34]. Among other details, as will be discussed elsewhere, the profiles of the velocity field are well approximated by error functions:

$$\omega(r) = A + B \operatorname{erf}\left(r - \frac{R_c}{W}\right) \quad (21)$$

where the dimensionless amplitudes are $A \simeq B \simeq 0.50$, R_c is the center of the shear band, and W is its width. The strain rate as function of r and z and the shear band structure in the rheometer gap are shown in Fig. 9.

Obviously, our results for both the shear stress intensity and the shear band structure agree well with the findings in [34], which were obtained with other software for DEM simulation and analysis. Therefore we

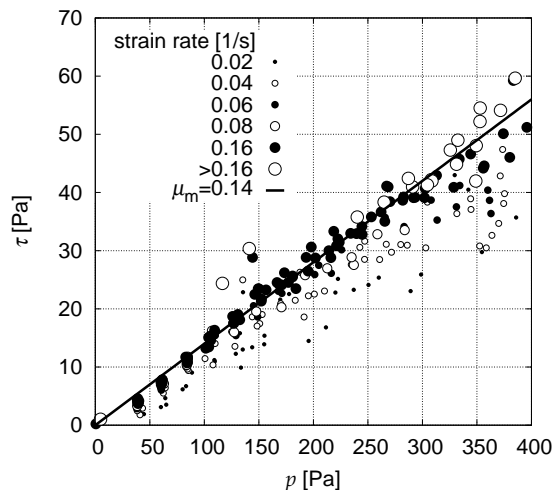


Fig. 8: Local shear stress $|\tau|$ plotted against local pressure p . Different symbol sizes indicate the magnitude of the strain rate $\dot{\gamma}$ at different locations within the rheometer gap, with $\dot{\gamma}$ given in the inset in units of s^{-1} ; larger symbols correspond also to larger $\dot{\gamma}$. The solid line represents the function $|\tau| = \mu_m p$, with the macroscopic friction coefficient $\mu_m = 0.14$.

conclude, that there are no implementation errors in the basic LIGGGHTS code and our dry contact model implementation, for the parameters used here.

4.3 Wet granular material

4.3.1 Macroscopic friction and cohesion

In Fig. 10, the dry results are compared to two simulations with liquid bridges. The addition of the liquid bridge forces leads to larger shear stress magnitudes so that the macroscopic yield stress at critical state (the termination locus, reached after large shear strain) is shifted upwards. The offset on the vertical axis is referred to as the macroscopic cohesion c . While the dry data are fitted well by the macroscopic line fit with $c = 0$, the wet data for small $p < 100$ Pa considerably drop below the fit result with constant $c > 0$.

4.3.2 Local shear viscosities

Next, the local shear viscosities η are compared for dry and different wet granular materials. Figs. 11 and 12 give the correlations $\eta(I)$, which are found from several DEM simulations with different liquid bridge volumes $V_b = 4.2$ nl, 42 nl and contact angles $\theta = 0^\circ, 10^\circ$ and 20° . The DEM simulations are evaluated locally in the

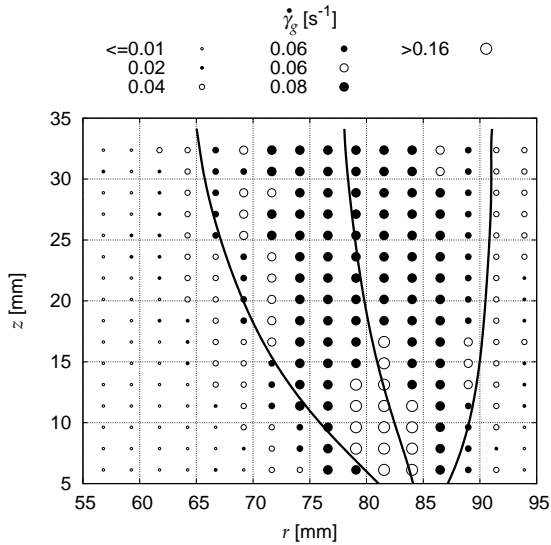


Fig. 9: Strain rate $\dot{\gamma}$ as function of radial and vertical position within the rheometer gap. As in Fig. 8, different symbol sizes indicate the magnitude of $\dot{\gamma}$. The lines indicate the center location R_c (middle line) and the width W of the shear bands (outer lines), as obtained from the fit function Eq. (19).

granular medium. Therefore, in contrast to the experiments, a single DEM simulation provides many data-points at various bulk-densities, shear-stresses, pressures and shear-rates that are plotted as different symbols for different V_b and θ .

As in the experiments, the inverse proportional dependence of the apparent viscosity on I is evidenced for the dry and wet material, and, as to be expected, the addition of small amounts of liquid increases the viscosity by about 10 to 20%. (The change appears not that large only due to the logarithmic vertical axis in the plots). The scaling $\eta \sim I^{-\alpha}$ changes from $\alpha = 0.9$ to $\alpha = 2$ in all materials, in different regimes. The influence of the contact angle and the liquid bridge volume V_b on the rheology of the wet granular materials is presently investigated in more detail and results will be published elsewhere.

4.3.3 Shear bands

Next, the influence of the liquid on the dynamic behavior of the granular material is investigated. The shear bands for dry and wet materials are given in Fig. 13. Variations in V_b and θ induce noticeable changes in the shear band structure. With increasing liquid content, its center position moves inwards to smaller radial distances. For small liquid content, the shear band width decreases, whereas it increases for the larger liquid con-

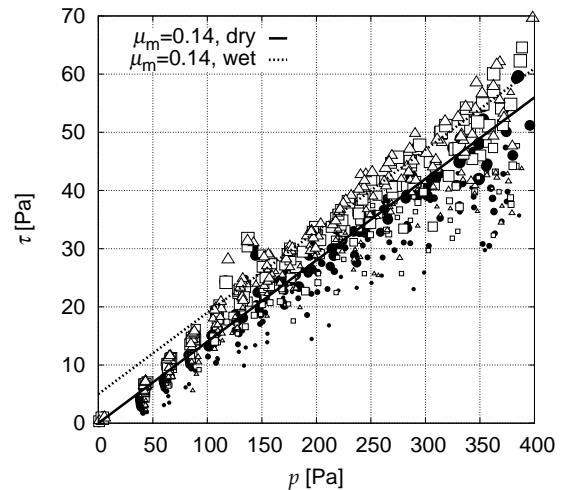


Fig. 10: Shear stress $|\tau|$ plotted against pressure p . Different symbols indicate results from dry material (\bullet), and from two wetted materials, which contain liquid bridges with (i) $\theta = 0^\circ$, $V_b = 4.2$ nl (\square) and (ii) $\theta = 20^\circ$, $V_b = 42$ nl (\triangle). The magnitude of the strain rate $\dot{\gamma}$ is indicated by the size of the symbols similar as in Fig. 8. The solid and the dashed lines represent the function $|\tau| = \mu_m p + c$, with the macroscopic friction coefficients μ_m for the dry ($c = 0$) and the wet ($c = 5$ Pa) materials, respectively.

tent simulations. The correlation for changing contact angle θ is more complex. Due to the high liquid content, the shear band moves inwards. Not surprisingly, the shift is largest for lowest contact angle $\theta = 0^\circ$. But against expectation, the lowest shift is found for $\theta = 10^\circ$ and not for $\theta = 20^\circ$. Additionally, the qualitative change of the shear band as reported for very strong van der Waals type adhesion [36] is not reproduced here, possibly since the liquid bridge forces never become strong enough to have a similar effect. These interesting findings are presently investigated in more detail, results will be published elsewhere.

5 Conclusions

Shear experiments are complemented by a numerical rheometer study with core shooting materials as application in mind, but with a much more general perspective concerning concepts and methods. The simple DEM contact and liquid bridge model is validated using previous results from a three-dimensional split bottom ring shear cell for dry materials, and with two-particle collision data from more advanced numerical and experimental studies for weakly wet materials.

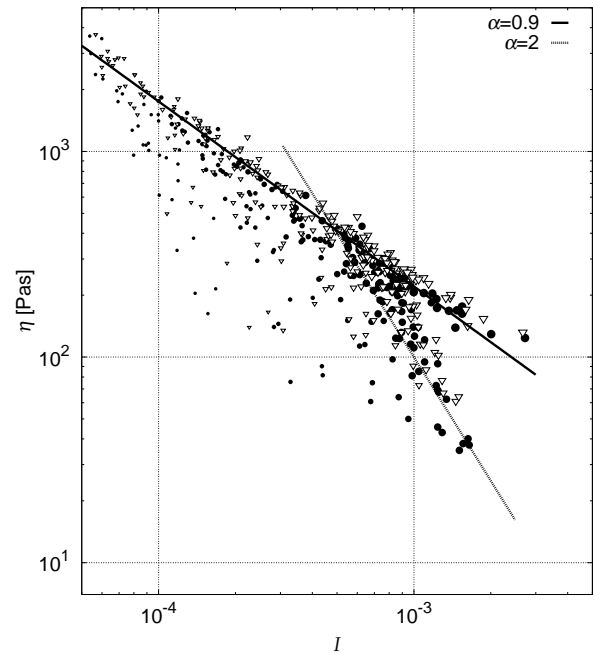
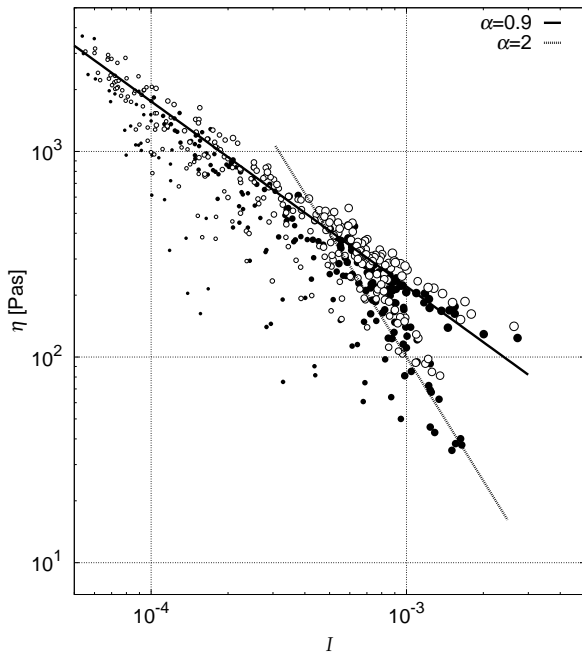
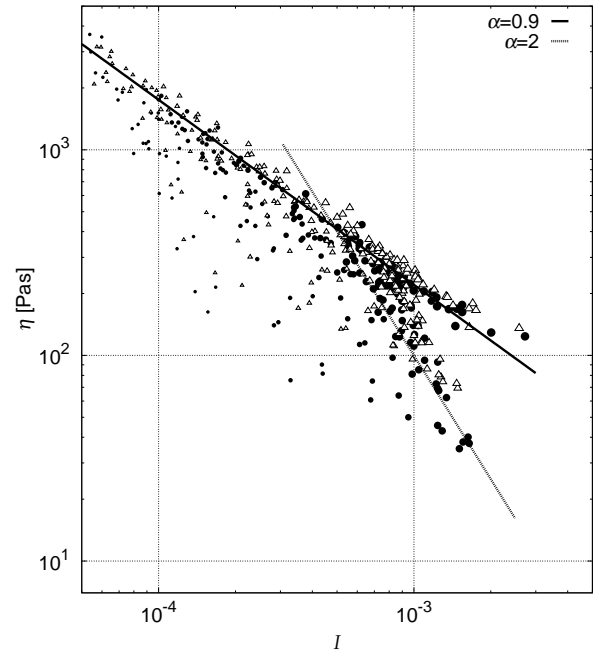
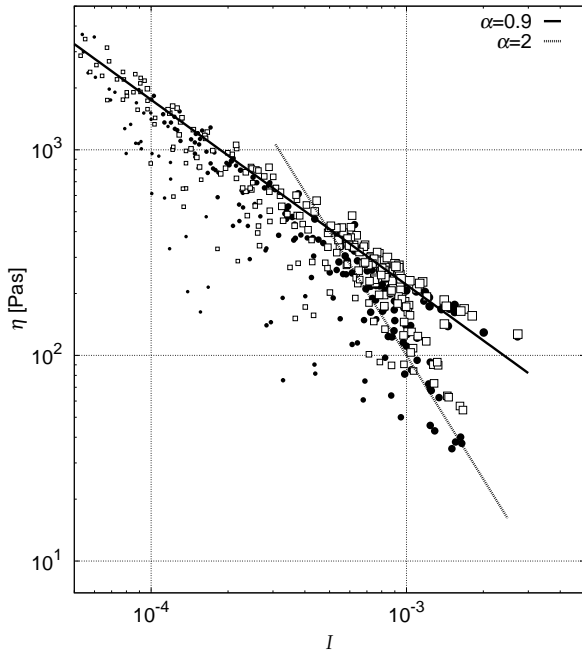


Fig. 11: Local shear viscosity $\eta(I)$ from the DEM simulations. Different symbols indicate results from dry (\bullet) and wet material (open symbols). The wet material contains liquid bridges with (top) $V_b = 4.2$ nl (\square) and (bottom) $V_b = 42$ nl (\circ), whereas $\theta = 0^\circ$ is the same for both cases. The magnitude of the local strain rate $\dot{\gamma}$ is indicated by the size of the symbols similar as in Fig. 8.

Fig. 12: Apparent shear viscosity $\eta(I)$ from the DEM simulations. Different symbols indicate results from dry (\bullet) and wet material (open symbols). The wet material contains liquid bridges with (top) $\theta = 10^\circ$ (\triangle) and (bottom) $\theta = 20^\circ$ (∇), whereas $V_b = 42$ nl is constant in all cases. The magnitude of the strain rate $\dot{\gamma}$ is indicated by the size of the symbols similar as in Fig. 8.

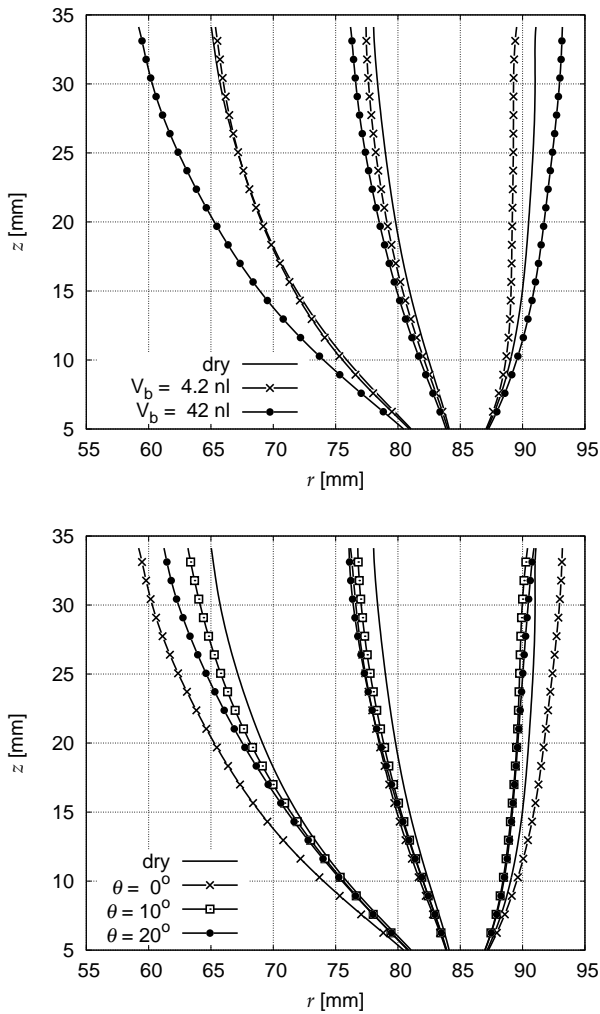


Fig. 13: Shear bands in dry and wet material, see Fig. 9. Different line styles indicate results from dry (continuous) and wet material (dashed). Here, points serve only for distinction but don't represent flow data. In the upper subfigure, the wet material contains liquid bridges with (i) $V_b = 4.2$ nl and (ii) $V_b = 42$ nl, whereas $\theta = 0^\circ$ is constant in both cases. In the lower subfigure, the wet material contains liquid bridges with (i) $\theta = 0^\circ$, (ii) $\theta = 10^\circ$ and (iii) $\theta = 20^\circ$, whereas $V_b = 42$ nl is constant here.

The DEM simulations of wet granular material show that the internal structures of the sheared material, i.e., the shear bands, are qualitatively the same as for dry materials. However, they move inwards with increasing liquid content and while getting a little narrower for small liquid content, then become wider for very high liquid content. Finally, changing contact angles influence shear bands in non-trivial manner.

The apparent local shear viscosity of the granular material significantly increases when only small amounts of liquid are added to the material, representing well the trend as seen from the experiments.

Future studies will involve a more quantitative study of the constitutive relations that describe the rheology of the material and their implementation into CFD or FEM codes to predict large scale core shooting flow behavior. The simulations and the experiments have to be performed in a more comparable way between wet and dry configurations, i.e. using the same gap width; the relation between the local and global viscosities (due to local and global shear rates) has to be better understood. An open question is how much a pendular liquid bridge contact model could be simplified, i.e. which details and non-linearities are important and which are not. Nevertheless, more complex capillary bridge models have to be used, which allow the description of other than the pendular bridge regimes, too.

Acknowledgments

We acknowledge the support of the ERASMUS program which allowed us to host FU during his Master study at the University of Twente. Helpful discussions with T. Weinhart, A. Singh, V. Magnanimo are appreciated. RS acknowledges the German Science Foundation (DFG) for funding parts of the work under project no. SCHW 1168/6-1, and SL acknowledges the NWO/STW, VICI grant 10828, and the DFG, project SPP1482 B12, for partial financial support. Finally, we acknowledge the constructive criticism of the referees of the paper.

References

1. GDR MiDi: On dense granular flows. *Eur. Phys. J. E* 14, 341-365 (2004)
2. Nakagawa, M., Luding, S.: *Powders and Grains 2009*. American Institute of Physics, AIP Conference Proceedings 1145 (2009)
3. Zhu, H.P., Zhou, Z.Y., Yang, R.Y., Yu, A.B.: Discrete particle simulation of particulate systems: Theoretical developments. *Chem. Eng. Sci.* 62, 3378-3396 (2007)
4. van der Hoef, M.A., van Sint Annaland, M., Deen, N.G., Kuipers, J.A.M.: Numerical simulation of dense gas-solid fluidized beds: A multiscale modeling strategy. *Ann. Rev. Fluid Mech.* 40, 4770 (2008).
5. Kafui, D.K., Johnson, S., Thornton, C., Seville, J.P.K.: Parallelization of a Lagrangian-Eulerian DEM/CFD code for application to fluidized beds. *Pow. Tech.* 207, 270-278 (2011)
6. Voigtman, T.: Yield stresses and flow curves in metallic glass formers and granular systems. *Eur. Phys. J. E* 34, 106 (2011)
7. Moorcroft, R.L., Cates, M.E., Fielding, S.M.: Age-Dependent Transient Shear Banding in Soft Glasses. *Phys. Rev. Lett.* 106, 055502 (2011)

8. Mani, R., Kadau, D., Herrmann, H.J.: Fluid depletion in shear bands. *Phys. Rev. Lett.* 109, 248001 (2012)
9. Harireche, O., Faramarzi, A., Alani, A.: A toroidal approximation of capillary forces in polydisperse granular assemblies. *Gran. Mat.* (2013). doi:10.1007/s10035-013-0425-9
10. Liu, P.Y., Yang, R.Y., Yu, A.B.: The effect of liquids on radial segregation of granular mixtures in rotating drums. *Gran. Mat.* (2013). doi:10.1007/s10035-013-0392-1
11. Hsiau, S.S., Liao, C.C., Tai, C.H., Wang, C.Y.: The dynamics of wet granular matter under a vertical vibration bed. *Gran. Mat.* (2013). doi:10.1007/s10035-013-0412-1
12. Zakerin, M., Kappl, M., Backus, E.H.G., Butt, H.J., Schonfeld, F.: Capillary forces between rigid spheres and elastic supports: the role of Young's modulus and equilibrium vapor adsorption. *Soft Mat.* 9, 4534-4543 (2013)
13. Lian, G., Thornton, C., Adams, M.J.: A theoretical study of the liquid bridge forces between two rigid spherical bodies. *J. Colloid Interface Sci.* 161, 138-147 (1993)
14. Weigert, T., Rippberger, S.: Calculation of the liquid bridge volume and bulk saturation from the half-filling angle. *Part. Part. Syst. Char.* 16, 238-242 (1999)
15. Willett, C.D., Adams, M.J., Johnson, S.A., Seville, J.P.K.: Capillary bridges between two spherical bodies. *Langmuir* 16, 9396-9405 (2000)
16. Herminghaus, S.: Dynamics of wet granular matter. *Adv. Phys.* 54, 221-261 (2005)
17. Jop, P., Forterre, Y., Pouliquen, O.: A constitutive law for dense granular flows. *Nature* 441, 727-730 (2006)
18. Goddard, J.D.: A dissipative anisotropic fluid model for non-colloidal particle dispersions. *J. Fluid Mech.* 568, 1-17, (2006)
19. Gabrieli, F., Lambert, P., Cola, S., Calvetti, F.: Micromechanical modelling of erosion due to evaporation in a partially wet granular slope. *Int. J. Num. Anal. Meth. Geomech.* 36, 918-943 (2012)
20. Hayman, N.W., Ducloue, L., Foco, K.L., Daniels, K.E.: Granular controls on periodicity of stick-slip events: kinematics and force-chains in an experimental fault. *Pure Appl. Geophys.* 168, 2239-2257 (2011)
21. Pietsch, W.: Agglomeration processes. Wiley-VCH, Weinheim (2002)
22. Guo, Y., Wu, C.Y., Thornton, C.: The effects of air and particle density difference on segregation of powder mixtures during die filling. *Chem. Eng. Sci.* 66, 661-673 (2011)
23. Guo, Y., Wu, C.Y., Kafui, K.D., Thornton, C.: 3D DEM/CFD analysis of size-induced segregation during die filling. *Pow. Tech.* 206, 177-188 (2011)
24. Beeley, P.R.: Foundry technology. Elsevier Butterworth-Heinemann, Oxford (2001)
25. Schwarze, R., Rudert, A., Tilch, W., Bast, J.: Rheological behavior of sand-binder mixtures measured by a coaxial cylinder rheometer. I. *Foundry Res.* 60(3), 2-6 (2008)
26. Liao, C.-C., Hsiau, S.-S.: Experimental analysis of dynamic properties in wet sheared granular matter. *Pow. Tech.* 197, 222-229 (2010)
27. Rudert, A., Schwarze, R., Tilch, W., Bast, J.: Computational fluid dynamics of the core shooting process. *Foundry Trade J. I.* 185, 147-151 (2011)
28. Quarzwerke Frechen, Austria
www.quarzwerke.at/datenblaetter/Quarzsand_F32-F36.pdf
29. Fenistein, D., van de Meent, J.W., van Hecke, M.: Universal and wide shear zones in granular bulk flow. *Phys. Rev. Lett.* 92, 094301 (2004)
30. McCarthy, J.J.: Micro-modeling of cohesive mixing processes. *Pow. Tech.* 138, 63-67 (2003)
31. Anand, A., Curtis, J.S., Wassgren, C.R., Hancock, B.C., Ketterhagen, W.R.: Segregation of cohesive granular materials during discharge from a rectangular hopper. *Gran. Mat.* 12, 193-200 (2010)
32. Radl, S., Kalvoda, E., Glasser, B.J., Khinast, J.G.: Mixing characteristics of wet granular matter in a bladed mixer. *Pow. Tech.* 200, 171-189 (2010)
33. Grima, P.W., Wypych, P.W.: Development and validation of calibration methods for discrete element modelling. *Gran. Mat.* 13, 127-132 (2011)
34. Luding, S.: The effect of friction on wide shear bands. *Part. Sci. Technol.*, 26, 33-42 (2008)
35. Luding, S.: Constitutive relations for the shear band evolution in granular matter under large strain. *Particuology* 6, 501-505 (2008)
36. Luding, S., Alonso-Marroquin, F.: The critical-state yield stress (termination locus) of adhesive powders from a single numerical experiment. *Gran. Mat.* 13, 109-119 (2011)
37. Sadrekarimi, A., Olson, S.M.: A new ring shear device to measure the large displacement shearing behavior of sands. *Geotech. Test. J.*, 32, 197-208, (2008)
38. Börzsönyi, T., Unger, T., Szabo, B., Wegner, S., Angenstein, F., Stannarius, R.: Reflection and exclusion of shear zones in inhomogeneous granular materials. *Soft Mat.* 7, 8330-8336 (2011)
39. Dijkstra, J.A., Wortel, G.H., van Dellen, L.T.H., Dauchot, O., van Hecke, M.: Jamming, yielding, and rheology of weakly vibrated granular media. *Phys. Rev. Lett.* 107, 108303 (2011)
40. Börzsönyi, T., Szabo, B., Törös, G., Wegner, S., Török, J., Somfai, E., Bien, T., Stannarius, R.: Orientational order and alignment of elongated particles induced by shear. *Phys. Rev. Lett.* 108, 228302 (2012)
41. Wang, X., Zhu, H.P., Yu, A.B.: Microdynamic analysis of solid flow in a shear cell. *Gran. Mat.* 14, 411-421 (2012)
42. Slotterback, S., Mailman, M., Ronaszegi, K., van Hecke, M., Girvan, M., Losert, W.: Onset of irreversibility in cyclic shear of granular packings. *Phys. Rev. E* 85, 021309 (2012)
43. Luding, S.: Cohesive frictional powders: Contact models for tension. *Gran. Mat.* 10, 235-246 (2008)
44. Göncü, F., Duran, O., Luding, S.: Constitutive relations for the isotropic deformation of frictionless packings of polydisperse spheres. *Compt. Rend. Mec.* 338, 570-586 (2010)
45. Shaebani, M.R., Madadi, M., Luding, S., Wolf, D.E.: Influence of polydispersity on micromechanics of granular materials. *Phys. Rev. E* 85, 011301 (2012)
46. Luding, S., Clément, E., Blumen, A., Rajchenbach, J., Duran, J.: Anomalous energy dissipation in molecular dynamics simulations of grains. *Phys. Rev. E* 50, 4113-4122 (1994)
47. Otsuki, M., Hayakawa, H., Luding, S.: Behavior of pressure and viscosity at high densities for two-dimensional hard and soft granular materials. *Prog. Theo. Phys. Suppl.* 184, 110-133 (2010)