



Modeling adhesive force distributions on highly rough surfaces



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ABSTRACT

Adhesive forces between particles have a major impact on many processes. The calculation of these forces is possible in the case of ideal smooth surfaces. Nevertheless real surfaces are rough which typically leads to decreased forces. Furthermore the forces are distributed over several orders of magnitude. In the presented paper, a new model to calculate the force distributions is developed. The model is based on surface roughness parameters which can be measured by atomic force microscopy. It is valid for van der Waals forces as well as for hydrophobic forces.

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1. Introduction

Adhesive forces between particles or particles and a plate influence a wide range of applications in the field of process engineering and particle technology. The determination of these forces is important to understand processes like deep bed filtration, agglomeration comminution etc. on a microscopic scale. Possible apparatuses to measure adhesive forces are for example the Surface Force Apparatus (SFA), centrifuges and the Atomic Force Microscope (AFM) which is used in this study [1]. Independent of the used apparatus the surface of the used solids has to be cleaned carefully and if a surrounding liquid is used, this liquid should have a high purity. This is caused by the high impact of adsorbed impurities on the measured forces. Furthermore, especially under dry conditions the adsorption of water from the surrounding humidity on the surfaces influences the measured forces [1–3].

One further general problem is that the measured force is always the total of all possible acting forces. This causes an understanding of possible forces like van der Waals forces, electrostatic forces, hydration forces, capillary forces and so one for the interpretation of the receipt force distance plots.

Most of the interacting solids have rough surfaces which decrease the interacting forces dramatically compared to smooth surfaces [3–10]. It is also shown in literature that rough surfaces lead to highly distributed forces in the range of several orders of magnitude [11–14].

One of the first approaches to describe the roughness influence on the van der Waals (vdW) forces was published by Rumpf [15]. Thereby,

the roughness is modeled by a small hemispherical asperity added concentrically onto the particle surface which interacts with a plate. By this, the effective particle to plate distance is increased and the adhesive force decreases. Further developments of this approach use spherical caps instead of hemispheres which is closer to reality [16]. For the case of nano-scale roughness, Rabinovich et al. connect the height of the spherical caps with the root-mean-square (rms)-roughness of the surface [10,17,17,18]. The rms-roughness can be measured for example by atomic force microscopy [6,11,18]. Thus, a calculation of adhesive forces by measuring the topography of a surface becomes possible.

Parsons et al. published a different model based on interacting infinitesimal surface areas which have randomly distributed deviations from the mean height. The adhesive forces are calculated by the integration of the energy of interaction over the interacting surfaces [19].

All presented models are not able to calculate an adhesive force distribution for a given surface roughness. You and Wan close this gap by assuming a distributed rms-roughness of the interacting surfaces [20,21]. In our opinion this is contradictorily to the characteristic property of the rms-roughness. The rms-roughness is a characteristic value of the topography profile which should be fixed value for one surface.

If the interacting particles are surrounded by water also non-vdW forces have to be considered. In the case of poor wetting conditions between the liquid and solids this force is called hydrophobic force [1,22]. This force occurs due to preferred orientation and structuring of water molecules near the hydrophobic solid surfaces and can be calculated by the theory published by van Oss [23,24]. As shown further in the results part, the hydrophobic force increases the adhesive force significantly and has to be considered therefore. Models to calculate the influence of surface roughness on hydrophobic forces are not present in literature up to now.

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Hence, the aim of this paper is to present a modified model to calculate the distribution of vdW and hydrophobic forces between interacting rough surfaces.

2. Materials and methods

2.1. Atomic force microscopy

Atomic force microscopy (AFM) is used to determine the adhesive forces between the particle and the substrate directly. As AFM a Park XE-100 from Park Systems, South Korea, is used. For the adhesive force measurements, tipples All-In-One Cantilevers Type A from Budget Sensors are used. The spring constant of the cantilever is determined with aid of the resonance frequency and the calculation published by Butt et al. [25]. To measure the adhesive forces directly between the particle and the substrate the colloidal probe technique as described elsewhere is used [11,26,27]. Therefore, a spherical alumina particle with a diameter of about 25 μm is glued onto the end of the cantilever. As counterpart a non-polished alumina substrate is used. Furthermore coated alumina particles and as well substrates are used to quantify the effect of hydrophobicity to the adhesive forces. The coating is done with Dynasylan F8261 from Evonik, Germany which leads to a non-polar hydrophobic surface due to their PTFE-like functional group [14]. The adhesive force measurements are carried out in dry and wet conditions at 20 °C. Before every measurement the particles as well as the substrates are cleaned with deionized water and isopropanol to avoid impurities at the surface.

For each set of conditions, adhesive forces are measured on five different randomly chosen locations on the substrate. At each location 256 measurements in a $15 \times 15 \mu\text{m}$ matrix are done. The obtained force-distance curves are analyzed using a Matlab routine. Thereby the minimum force during the retrace is detected and used as adhesive force. The Matlab routine is also used to exclude adhesive forces due to capillary forces which can occur especially at hydrophobic surfaces [11,28].

The AFM is also used to make topography scans of the used substrate. Therefore, a $15 \times 15 \mu\text{m}$ surface area is scanned using contact mode AFM. Fig. 1 shows an example topography measurement of the used alumina substrate. To obtain a reliable *rms*-roughness value for further calculations topography scans are repeated 4 times at different locations.

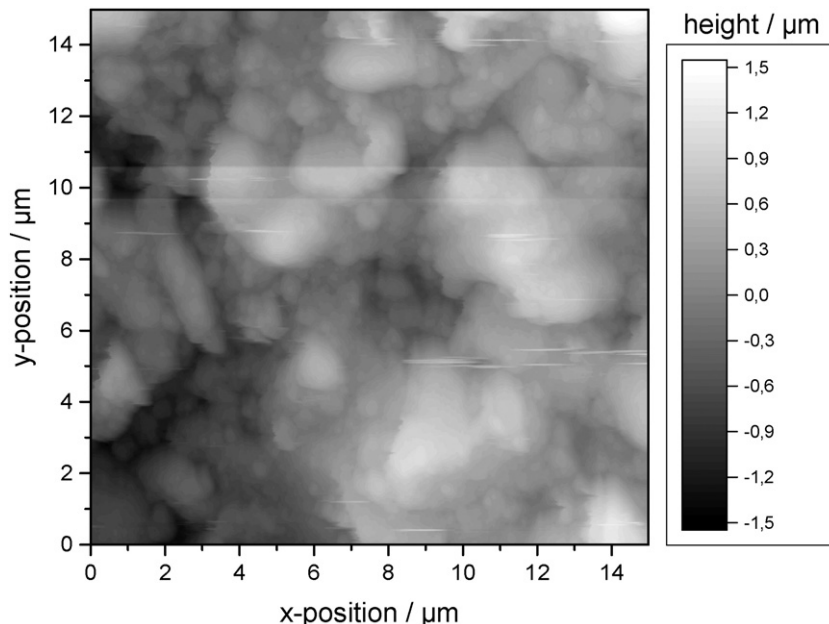


Fig. 1. Topography measurement of the used alumina substrate obtained from contact mode AFM.

Besides the scanning of the substrates, also the topography of the used particles is measured. Fig. 2 illustrates the topography and an example line profile of the scanned particle. One can see, that the surface of the particle is smooth compared to the macroscopic roughness of the substrates. Consequently, the particle roughness is neglected during the force modeling.

2.2. Contact angle measurements

Contact angle measurements are carried out to determine the polar energy of interaction between the solid surfaces in water. This energy of interaction is connected to the hydrophobic adhesive force by the theory published by van Oss [23,29]. Thereby, contact angle measurements with three different liquids are carried out. For the liquids, the dispersive as well as the positive and negative polar part of the surface energy is known (Table 1). For the calculation the mean value of 10 contact angle measurements is used to calculate the required energy values.

The surface energy of the coated alumina is calculated by Eq. (1).

$$1 + \cos\theta = \frac{2}{\gamma_l} \left(\sqrt{\gamma_s^d \gamma_l^d} + \sqrt{\gamma_s^+ \gamma_l^-} + \sqrt{\gamma_s^- \gamma_l^+} \right) \quad (1)$$

Because of the three unknown surface energy parameters γ_s^d , γ_s^+ and γ_s^- of the solid, three different liquids have to be used and Eq. (1) has to be solved three times simultaneously. The result for the coated alumina shows a nearly non-polar surface with a surface energy of 19.5 mJ/m^2 which is in the range of polytetrafluoroethylene (PTFE) [30]. This result is plausible, because the functional group of the used silane has a PTFE-like structure. Eq. (2) can now be used to calculate the free polar energy of interaction which is further used to calculate the hydrophobic force.

$$w_{phob} = 4 \cdot \left(\sqrt{\gamma_s^+ \gamma_s^-} + \sqrt{\gamma_l^+ \gamma_l^-} - \sqrt{\gamma_s^- \gamma_l^+} - \sqrt{\gamma_s^+ \gamma_l^-} \right) \quad (2)$$

For the case of two coated alumina surfaces surrounded by water, the free polar energy of interaction is 77 mJ/m^2 . The polar interaction for the non-coated alumina is neglected. Due to the polar surface of the non-coated alumina no hydrophobic interaction can occur.

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