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Method of quantifying surface roughness for accurate adhesive force predictions

Casey Q. LaMarche[a], Stuart Leadley[b], Peiyuan Liu[a], Kevin M. Kellogg[a], Christine M. Hrenya[a,*]

[a] Department of Chemical and Biological Engineering, University of Colorado, Boulder, CO 80303, USA
[b] Dow Corning Europe SA, Rue Jules Bordet, Park Industrial – Zone C, B-7180 Seneffe, Belgium

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ABSTRACT

The van der Waals force between contacting surfaces depends strongly on the surface roughness. Theories that allow for estimating the adhesion force, or the force to separate surfaces from contact, with simple, single-equation theories (i.e., by considering roughness asperities as submerged-spheres) can be easily instituted in discrete element method simulations of many-particle systems, but require inputs that rely on quantification of the surface roughness. In this work, Atomic Force Microscope (AFM) topographical surface maps reveal that two scales of roughness characterize the surfaces of particles examined, similar to prior studies. Previously, the separation of the two roughness scales and determination of the associated wavelength, which are necessary for predicting adhesion forces, relied on visual selection. Here, an objective methodology to separate the scales of surface roughness and calculate the wavelength of each scale is established. Two artifacts are identified when using the new methodology that negatively impact adhesion force predictions if not eliminated, namely the Gibbs artifact and an "atomic-scale-noise" artifact. Procedures to overcome these artifacts are developed. The resulting surface roughness characterizations are employed in a new theory, the predictions of which are in excellent agreement with AFM pull-off force measurements. The new theory extends a current van der Waals theory, which treats surface roughness as submerged spheres, by accounting for two rough surfaces instead of one.

1. Introduction

The van der Waals force between solids varies greatly over short separation distances. Specifically, the force is inversely proportional to the square of the separation distance. For rough particles, surface asperities act as spacers that separate surfaces and serve to reduce the contact area, because the asperity itself is smaller in size than the native surface (i.e., asperity-asperity contact for rough surfaces vs. surface-surface contact for smooth surfaces). Consequently, surface roughness reduces the force needed to separate surfaces in contact, or the adhesion force, by orders of magnitude compared to smooth surfaces (Fuller and Tabor, 1975). Real surfaces are not perfectly smooth, and even nanometer scale roughness can affect the adhesion force (Rabinovich et al., 2000b). For example, Royer et al. (2009) found that roughening particle surfaces with a nanoparticle coating leads to an attenuation of particle clustering in granular jets. Accurately quantifying the effect of surface roughness on adhesion impacts many practical applications, such as colloid retention (Torkzaban and Bradford, 2016), bio-inspired adhesives (Chen et al., 2015), formation of biofilms (Preedy et al., 2014), particle packing (Feng and Yu, 2000), hopper flow (Anand et al., 2009), fluidized beds (Galvin and Benyahia, 2014), and rotating drums (Quintanilla et al., 2006a). Accordingly, a method of characterizing the surface roughness is necessary for accurate force predictions as well as other phenomena that are impacted by surface roughness, such as battery efficiency (Tabakovic et al., 2015; Su et al., 2015), corrosion resistance (Zhao et al., 2015), thermal conductivity of nanowires (Lim et al., 2012), contact angle (Zhenyu et al., 2016), heat transfer (Morris et al., 2015; Peterson and Fletcher, 1990) capillary condensation (Rabinovich et al., 2002; Chai et al., 2014), etc.

For smooth surfaces, two types of theories have been used to predict the adhesion force: the Hamaker (1937) theory, in which the van der Waals force is a function of surface separation distance, and the JKR/DMT (Johnson et al., 1971; Derjaguin et al., 1975) theory in which the surface energy of the compressing surfaces is used to determine the adhesion force. The choice of JKR (Johnson et al., 1971) or DMT (Derjaguin et al., 1975) is determined, in part, by the material properties of the system under investigation, namely, JKR for soft materials and DMT for hard. Not surprisingly, when applied to even mildly rough surfaces, these (smooth-surface) theories are

* Corresponding author.
E-mail address: hrenya@colorado.edu (C.M. Hrenya).

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reported to over-predict the measured adhesion force by orders of magnitude (Chai et al., 2014; Schaefer et al., 1995; Götzinger and Peukert, 2003; Li et al., 2006). For example, Segeren et al. (2002) found that the measured adhesion force for a silica particle interacting with a roughened silicon wafer was over-predicted by as much as an order of magnitude with the Hamaker theory for smooth surfaces (i.e. using intermolecular separation distances). Similarly, Schaefer et al. (1995) measured the adhesion force of glass, polystyrene and tin particles to be 50 times less than predicted with the JKR theory. Due to the uncertainties associated with the surface energy and contact area required as inputs to the JKR/DMT theory (Rabinovich et al., 2000b; Segeren et al., 2002; Jones et al., 2002), the focus of this work is on Hamaker-based models (details on JKR/DMT based models can be found elsewhere, e.g., (Schaefer et al., 1995; Prokopovich and Starov, 2011; Prokopovich and Perni, 2010, 2011; Perni and Prokopovich, 2015; Persson and Scaraggi, 2014; You and Wan, 2013, 2014; Quintanilla et al., 2006b; Cheng et al., 2002)).

To account for surface roughness, the Hamaker theory has been modified to incorporate asperities of different shapes. For example, several groups treated surface roughness as spherical asperities (Rabinovich et al., 2000b; Schaefer et al., 1995; Katainen et al., 2006; Jallo et al., 2011; Matope et al., 2012). Rumpf (1990) and Rabinovich et al. (2000a, 2000b) developed two of the prominent Hamaker-based theories that account for roughness. The Rumpf (1990) theory assumes asperities are hemispherical. The Rumpf theory has been found to under-predict the measured force by an order of magnitude for surfaces (Rabinovich et al., 2000b; Götzinger and Peukert, 2003; Israelachvili, 2011; Yang et al., 2007). This under-prediction can be traced to the hemispherical asperity assumption (i.e., the asperity height equals its half-width so the adhesion force only depends on the asperity height measurement), as several groups have found that the adhesion force does not correlate to the asperity height alone, and thus a lateral measurement (width) of surface roughness is also necessary (Rabinovich et al., 2000a, 2000b; Schaefer et al., 1995; Butt et al., 2005; Kim and Russell, 2001) (the hemispherical geometry may be more appropriate for nanoparticle-coated surfaces, as indicated in Chen et al. (2008), because the asperity height is correlated to the width, or diameter, of particles used for coating). Accordingly, to overcome the large errors predicted by the Rumpf model, Rabinovich et al. (2000a, 2000b) extended the Rumpf theory by treating asperities as submerged spheres, which removes the hemispherical restriction and allows for a wider range of asperity geometries, such as short and squatty (Rabinovich et al., 2000b; Rumpf, 1990; Beach et al., 2002).

For both the Rumpf and Rabinovich et al. theories, surface deformation is not considered and therefore contact is assumed to occur at a single point, which is referred to as single-asperity contact. Multiple-asperity contacts, on the other hand, can affect adhesion forces for surfaces that are soft and/or flat (Katainen et al., 2006), and for some nanoparticle-coated surfaces (Quintanilla et al., 2006b; Chen et al., 2009). The surfaces used here are hard (Young's modulus ~70 GPa), represent a sphere-plate interaction (i.e. not flat-flat interaction), and the surfaces are not coated with nanoparticles. Accordingly, singe-asperity contact is assumed for the surfaces of interest here, an assumption justified by calculations below. However, it is worth noting that in some systems that exhibit multiple-asperity contacts, the mean adhesion force, but not its full range, is predicted well using the singe-asperity contact assumption (Cooper et al., 2001b).

The modified Hamaker theories described above (i.e. Rumpf and Rabinovich et al. theories) take the form of a single analytical expression. However, a different approach can be used to numerically integrate the van der Waals force between surfaces (Cooper et al., 2001a, 2001b; Eichenlaub et al., 2006, 2004; Jaiswal et al., 2009; Chen et al., 2010). These computational approaches, which reproduce measured surface roughness as discretized meshes using Fourier transforms, for example (Jaiswal et al., 2009), and use Hamaker constants fitted from measured force-displacement curves (which agree with the often wide, factor of two, range reported in literature) (Jaiswal et al., 2009), predict adhesion forces in good quantitative agreement with measurements (Cooper et al., 2001a, 2001b; Eichenlaub et al., 2006, 2004; Jaiswal et al., 2009; Chen et al., 2010). Such computational approaches (Cooper et al., 2001a, 2001b; Eichenlaub et al., 2006, 2004; Jaiswal et al., 2009; Chen et al., 2010) are a good option for predicting adhesion forces between surfaces with irregular shapes and/or multiple-asperity contacts. Nonetheless, a new series of simulations is required for each set of interacting surfaces due to the phenomenological nature of the computational expression for adhesion force (Cooper et al., 2001a, 2001b; Eichenlaub et al., 2006, 2004; Jaiswal et al., 2009; Chen et al., 2010). On the other hand, the simple, analytical expression under consideration here can be applied to any spherical particle in which single-asperity contacts dominate (Katainen et al., 2006; Matope et al., 2012; Laitinen et al., 2013). Because such particles are common in a wide range of applications (paints, fluidized beds, gasifiers, etc.), this streamlined approach remains an important alternative to the strictly computational method.

All of the Hamaker-based theories for rough surfaces require quantification of the roughness. For example, Rabinovich et al. (2000a) related the asperity height and radius to the roughness wavelength (λ; peak-to-peak distance between asperities) and the Root Mean Square (RMS) of the roughness height. Rabinovich et al. (2000b) further advocated separating the roughness into large- and small-scale components, each described with separate λ and RMS height, as they found the small-scale roughness dominated the adhesion force for the materials investigated. While determining the RMS height of an individual scale is relatively straightforward, separating roughness scales and quantifying wavelengths is convoluted and previous studies relied on visual determination (Rabinovich et al., 2000b; Chai et al., 2014; Katainen et al., 2006; Beach et al., 2002).

Several previous investigators implemented the Rabinovich et al. theory and found predictions compared well to adhesion force measurements (Rabinovich et al., 2000b; Chai et al., 2014; Schaefer et al., 1995; Katainen et al., 2006; Matope et al., 2012; Beach et al., 2002). For instance, Rabinovich et al. (2000b) improved upon the order of magnitude under-prediction from the Rumpf theory to a factor of two under-prediction of the adhesion force with the Rabinovich et al. theory for the rougher surfaces in their studies. Additionally, Beach et al. (2002) arbitrarily separated the roughness scales and visually selected asperities to measure λ for several flat surfaces with varied roughness levels, and found their adhesion forces measured with hard particles were under-predicted by a factor of four, or less, by the Rabinovich et al. theory (Beach et al., 2002). Chai et al. (2014) stated that selecting asperities is arbitrary, and chose a representative value of λ for surfaces with increasing roughness, and under-predicted the measured adhesion force by a factor of two to six (except for their roughest surfaces where predictions improved). Furthermore, Katainen et al. (2006) used the curvature at the apex of arbitrarily-selected asperities as the asperity radii and height in the Rabinovich et al. theory, instead of using the roughness λ and RMS height, but still under-predicted the measured adhesion forces by approximately half, or less. While these visual and arbitrary methods of quantifying roughness have improved predictions from over the order-of-magnitude differences noted above to typically within a factor of two or four of measured values (Rabinovich et al., 2000b; Chai et al., 2014; Schaefer et al., 1995; Segeren et al., 2002; Katainen et al., 2006; Beach et al., 2002), there is still considerable room for improvement. Accordingly, the focus of this work is on developing a method to quantify surface roughness that does not rely on visual means.

Finally, it is also worth noting that the Rumpf (1990) and Rabinovich et al. (2000a, 2000b) theories were developed for contacts between one rough surface and one smooth surface, and are appropriate for the controlled adhesion-force measurements between relatively smooth particles and rough wafers typically made via Atomic