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Adhesion as an interplay between particle size and surface roughness

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Abstract

Surface roughness plays an important role in the adhesion of small particles. In this paper we have investigated adhesion as a geometrical effect taking into account both the particle size and the size of the surface features. Adhesion is studied using blunt model particles on surfaces up to 10 nm root-mean-square (RMS) roughness. Measurements with particles both smaller and larger than surface features are presented. Results indicate different behavior in these areas. Adhesion of particles smaller than or similar in size to the asperities depend mainly on the size and shape of the asperities and only weakly on the size of the particle. For large particles also the particle size has a significant effect on the adhesion. A new model, which takes the relative size of particles and asperities into account, is also derived and compared to the experimental data. The proposed model predicts adhesion well over a wide range of particle/asperity length scales.

Keywords: Adhesion; Surface roughness; Pull-off force; Atomic force microscope

1. Introduction

Hardly any real surface is smooth at a submicroscopic level. Practical surfaces often possess significant roughness and even highly polished silicon wafers in semiconductor industry are rough in subnanometer scale [1]. Surface roughness plays an important role in adhesion since it reduces the contact area between the bodies leading to significantly reduced interaction [2–4]. Surfaces may posses roughness in several length scales, but due to the short range of the van der Waals interaction, roughness in nanoscale ultimately determines the strength of adhesion.

Invention and progress of colloidal probe technique have boosted the studies of adhesion and the effects of nanoscale roughness [5]. In order to explain experimentally observed adhesion on rough surfaces, both analytical [6–8] and computational [9–11] approaches have been used. While computational methods produce good agreement with experiments [11,12], they are complex and thus not easily applied for fast estimation of the adhesion for specific systems. On the other hand, analytical equations rely on only few parameters which are easily determined from the surface topography. This ensures that they can be easily used for multiple systems to estimate the adhesion [13,14].

Recent models are based on approach where the asperities are assumed to be hemispherical caps on a smooth substrate [6]. Rabinovich et al. [7,8] have suggested that the adhesion on surfaces exhibiting asperities should be written as a combination of sphere–sphere and sphere–surface interactions in the form

$$F_{\mathrm{adh}} = \frac{\mathcal{A}_{\mathrm{H}} R}{6H_0^2}\left[\frac{r}{r+R} + \frac{1}{(1+y_{\max}/H_0)^2}\right], \qquad (1)$$

where $\mathcal{A}_{\mathrm{H}}$ is the Hamaker constant, R is the radius of the adhering particle, H_0 is the equilibrium distance, r is the radius of the asperity on the surface, and $y_{\max}$ is the height of the asperity. The first term is the adhesion between the asperity and the particle, separated by H_0 and the second term the contribution of the particle–substrate interaction. Fig. 5a illustrates the contact geometry used in deriving Eq. (1). This approach overestimates the adhesion because the last term takes the whole interacting surface into account as a plane, resulting in double counting of the contribution of the substrate under the asperity. To reduce the effect of double counting, a condition $R \gg r$ must apply in-

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dicating that the adhering particle is much larger than a single asperity.

When the particle and the substrate are of different materials, the Hamaker constant can be calculated from

$$\mathcal{A}_{H_{12}} = \sqrt{\mathcal{A}_{H_{11}}\mathcal{A}_{H_{22}}}. \qquad (2)$$

In this work we have demonstrated that although the model by Rabinovich et al. gives reasonable estimates for pull-off forces with particles comparable in size with the asperities on the surface, it underestimates the adhesion if the particles are much larger than the asperities. We have derived a new model for this region which takes into account multiple contacts with the surface. The new model has been tested by comparison to experimental data over a wide range of particle/asperity sizes. The comparison illustrated that both models are needed to cover the whole experimental range.

2. Experimental methods

2.1. Surfaces

Polycrystalline TiO_2 thin films prepared by atomic layer deposition (ALD) were used as the studied surfaces. ALD is a gas phase thin film deposition method which produces uniform and high quality thin films with good reproducibility [15]. The films were grown on borosilicate glass substrates using a process described earlier [16]. The deposition temperature was 620 K resulting in anatase structure. Films with three different surface geometries were prepared by varying the film thickness.

Table 1
Properties of the rough TiO_2 films. Radius of curvature and height represent properties of the typical asperity on the surface

Film thickness (nm)	Rms roughness (nm)	Density of asperities ρ ($1/\mu m^2$)	Radius of curvature r (nm)	Height y_{max} (nm)
10	0.65	880	30 ± 13	1.5 ± 0.5
130	6.2	270	10 ± 2	15 ± 4
500	11	120	13 ± 2	26 ± 9

The surfaces were imaged using a Digital Instruments Nanoscope III with extender electronics. The main properties of the films are presented in Table 1. The density of asperities is determined from the number of grains on the 1 μm^2 topographic AFM images. The local maxima were identified from the images, and maxima that do not exceed a preset height level were excluded. The limit was determined using the height distribution of the surface and set half width at half maximum (HWHM) above the mean surface level. A paraboloid was fitted to each accepted local maxima. Each paraboloid was allowed to find an optimal shape and location. The radii of curvature at the apexes of the fitted paraboloids were used as the radii of the asperities. The heights of the grains were determined as the difference between the apex of the fitted parabola and an estimated value for the surface level around the grain. The surface level was calculated individually for each asperity. Distributions of radii of curvatures and heights are presented in Fig. 1 for all three surfaces used in the experiments. Since the acceptance limit for asperities is chosen arbitrarily, the number of grains is more indicative than absolute. In addition to these accepted maxima there were also a number of low local maxima, which we expect not to contribute to the adhesion. Also, it should be noted that the asperity curvature of the two roughest surfaces is very close to the standard AFM tip apex curvature, indicating very sharp asperities. This is due to the crystallinity of the coatings.

2.2. Particles

Eroded silicon tips exhibiting flat apexes with native oxide were used as blunt silica particles. The flat apex of different size were made by eroding fresh tips against a silicon sample. Approximate scanning parameters during erosion were: scan size 4 µm, rate 4 Hz, normal force 50 nN, and angles 0°, 45°, 90°, and 135°. The apex size was controlled by the erosion time, though also some variations on the normal force were used. Single crystal silicon cantilevers were calibrated by the Sader et al. method [17] by the manufacturer (CSC17-F, Mikro-

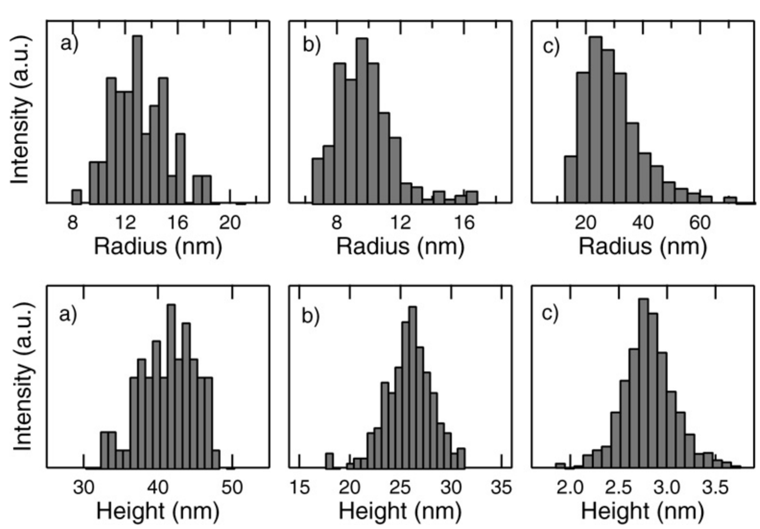

Fig. 1. Radius (top) and height (bottom) distributions of the asperities on the samples. (a) 500-, (b) 130-, and (c) 10-nm thick TiO_2 coating on borosilicate glass.