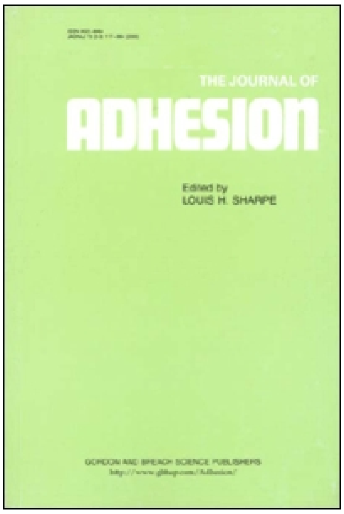

The Journal of Adhesion
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713453635

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To cite this Article Ding, W. , Zhang, H. and Cetinkaya, C.(2008) 'Rolling Resistance Moment-Based Adhesion Characterization of Microspheres', The Journal of Adhesion, 84: 12, 996 — 1006
To link to this Article: DOI: 10.1080/00218460802577084
URL: http://dx.doi.org/10.1080/00218460802577084

The Journal of Adhesion, 84:996–1006, 2008
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ISSN: 0021-8464 print/1545-5823 online
DOI: 10.1080/00218460802577084

Rolling Resistance Moment-Based Adhesion Characterization of Microspheres

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A microsphere-surface adhesion characterization technique based on the rolling resistance moment of the adhesion bond is reported and demonstrated. With a nanomanipulation system, an increasing lateral pushing force is applied to a polymer microsphere adhered on a silicon substrate in the ambient environment, and the rolling displacement of the particle in response to the external force is recorded. The work of adhesion for polymer microspheres to the silicon substrate is extracted from the force-displacement curves. Unlike the traditional colloid probe technique where particles are glued to the cantilever beam and the normal detachment force as a function of the out-of-plane displacement is measured, in the current work the work of adhesion is directly determined from the measured rolling resistance moment and the proposed approach requires no prior knowledge of the particle diameter. In addition to the work of adhesion for the set of microspheres, the critical angles of rolling prior to rolling are estimated. The reported results form further experimental evidence for the existence of a rolling resistance moment.

Keywords: Emulsion aggregation (EA) toner; Microspheres; Particle detachment; Rolling resistance moment; Toner adhesion; Work of adhesion

INTRODUCTION

Micro-sized spherical particles are fabricated and processed in large quantities in a number of industries from printing to pharmaceuticals. The knowledge of the particle adhesion properties is critical for unit processes such as compaction, coating, printing, polishing, and cleaning. As a result, over the years, the adhesion of micro-spherical

Received 5 May 2008; in final form 2 October 2008.

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particles to flat surfaces has attracted tremendous research attention. Many contact mechanics-based adhesion theories have been proposed to understand the stiffness and strength of the particle-substrate bond [1–3], and various experimental techniques have been proposed to characterize the particle-substrate adhesion [4–10].

Moment balance of forces acting on a particle in obtaining a detachment criterion has been traditionally used for predicting the onset of rolling-based detachment of the particle. This criterion assumes no resisting moment to rolling at the adhesion bond prior to rolling. In recent years, it has been demonstrated both analytically [11] and experimentally [12] that the adhesion bond between a microparticle and a surface indeed creates a resistance against the rolling initiation of the particle. Recently, the rolling resistance moment-based microparticle adhesion study in the vacuum chamber of a scanning electron microscope (SEM) with a custom-made nanomanipulator was reported [13]. This rolling resistance is analogous to the static friction force observed prior to sliding in linear friction experiments. Unlike the nanoscale particle pushing studies performed with an atomic force microscope (AFM) (*e.g.*, see Sitti [14]), the pushing experiments performed in the SEM chamber allowed direct visual observation of the rolling and sliding motions of the particles.

In the present study, it is demonstrated that the adhesion bonds between polymer microspheres and a silicon substrate can be characterized in the ambient environment by employing the rolling resistance moment of the adhered particle. The rolling resistance moment-based lateral pushing test configuration is depicted in Fig. 1. Under a lateral pushing force (F), the stress distribution in the particle-substrate contact area becomes non-uniform, which creates a restoring moment (rolling resistance moment, M_y) to rolling motion and this moment is proportional to the angle of rotation (θ). With a custom-made positioning system, a series of lateral pushing tests are conducted under an optical microscope. The response of the adhered microparticle to the lateral pushing force is recorded, and the work of adhesion between the polymer microparticle and silicon substrate is determined from the pre-rolling slope of the force-displacement curve. Compared with other particle adhesion characterization techniques such as the colloid probe technique [4], the electric field detachment method [7], and the centrifugal detachment method [7,15], the current technique has several advantages. The current technique provides the adhesion properties of an individual particle to a substrate, while the electric field detachment and centrifugal detachment methods provide the (bulk) adhesion properties for a large group of particles. Compared with the colloid probe technique, the current

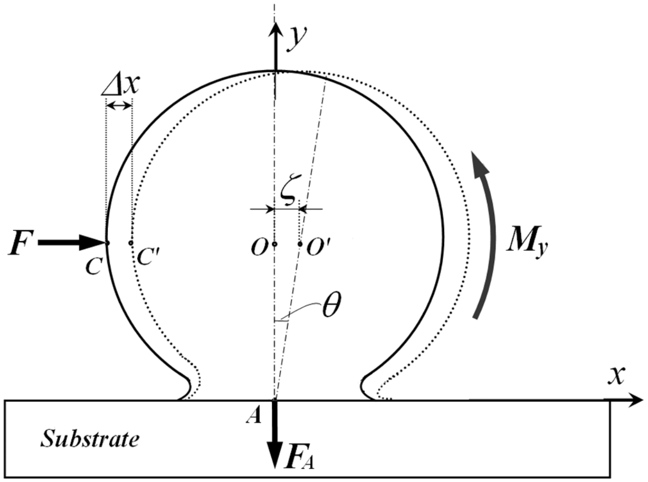
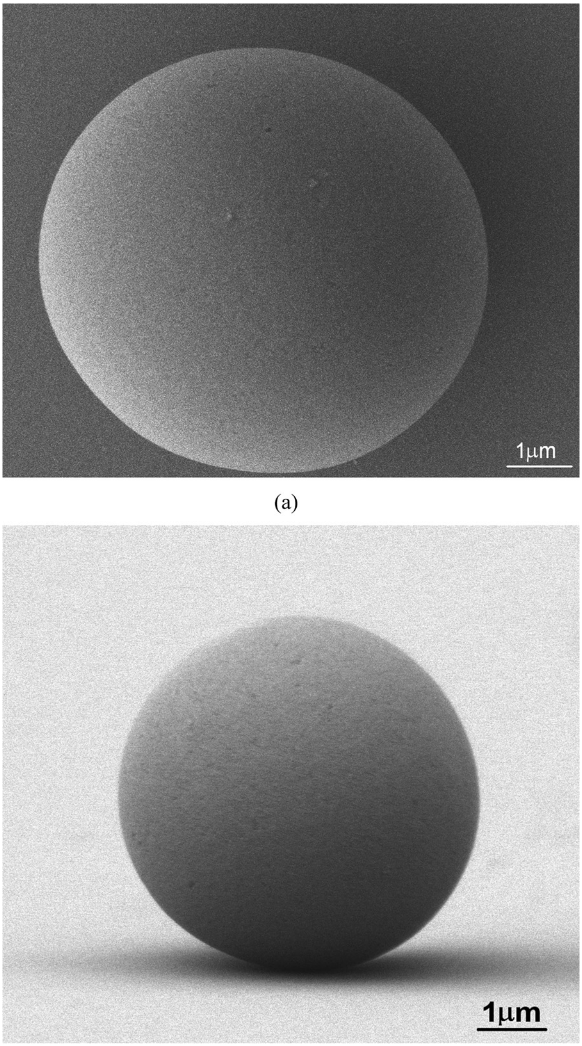


FIGURE 1 A moment (M_y) resisting (rolling resistance moment) the rolling of a micro-sphere subjected to a lateral force (F) at a lateral translation of (ξ) (contact area is not to scale).

technique does not involve the laborious process of gluing a particle to the cantilever beam, is essentially non-destructive for the particles, and does not require the particle diameter information to determine the work of adhesion between the particle and substrate. Compared with our previous study in the SEM [13], the current technique has lower accuracy in force and displacement measurements because the spatial resolution of an optical microscope is inferior to that of an SEM. However, performing measurements in the ambient avoids the complication associated with the charging of the nonconductive polymer particles by the SEM electron beam. In addition, it provides the flexibility to explore the influence of certain parameters such as the relative humidity and meniscus formation on particle-substrate adhesion which is impossible to investigate inside the chamber of a high vacuum SEM.

EXPERIMENTAL PROCEDURE

The particles investigated in the current study are polymer microspherical toner produced by the emulsion aggregation (EA) method [16] with a nominal particle diameter of 9.0 µm (experimental toner particles provided by Xerox Corporation, Webster, NY, USA). The outer layer of the toner particle is made of polystyrene with other ingredients such as pigment and wax enclosed in the shell. SEM analysis reveals that these polymer particles are virtually perfectly spherical and have fairly smooth surfaces at the nanometer length-scale (Fig. 2). For particle-substrate adhesion study, the polymer



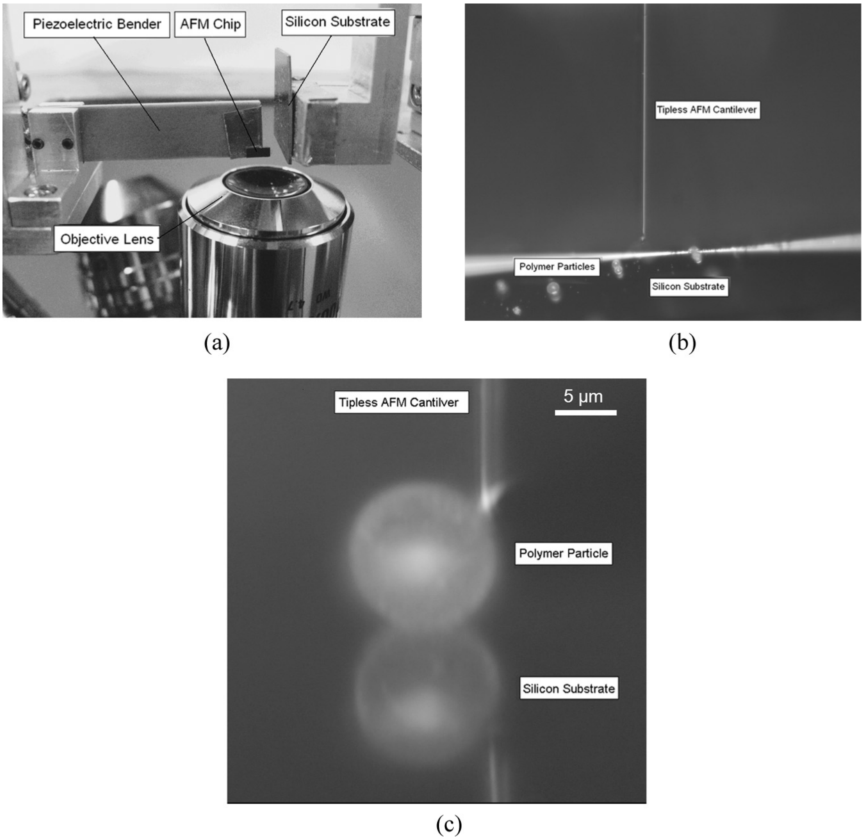
(a)

(b)

FIGURE 2 SEM images of a polymer microsphere coated with a 10 nm gold layer for charge dissipation during SEM observation: (a) top view; (b) side view.

particles are dry-deposited on a plasma cleaned single-crystal silicon substrate (p-type doped (100) oriented wafer, University Wafer, Boston, MA, USA) immediately before the execution of the reported experiments. The experiments are performed with a nanomanipulation system composed of two opposing *xyz* linear positioning stages (122-1135/1155, OptoSigma Inc., Santa Ana, CA, USA) mounted on top of an inverted optical microscope (Epiphot 200, Nikon, Japan). The positioning stages are driven by piezoelectric actuators (MRA 8351, New Focus, Inc., San Jose, CA, USA) that provide linear motion with a motion resolution of approximately 30 nm. A piezoelectric bender (CMBP 05, Noliac A/S, Denmark) that provides fine positioning at sub-nanometer resolution is mounted on the top of one of the linear positioning stages. The positioning and particle pushing processes are monitored through a high-resolution digital camera (DXM 1200, Nikon, Japan) attached to the optical microscope, which has a pixel resolution of around 35 nm. The experimental setup for pushing experiments is depiected in Fig. 3a. An AFM chip with a tipless cantilever beam (CSC 12, length 350 μm, nominal force constant 0.03 N/m, MikroMasch, Inc., Wilsonville, OR, USA) is attached to the free end of the piezoelectric bender, and the silicon substrate with polymer particles deposited is mounted on the opposing positioning stage. The nanomanipulation system allows the precise positioning of the cantilever beam with respect to the polymer particles on the substrate while the positions of the cantilever beam and the particle are monitored through the high-resolution digital camera (Figs. 3b,c).

During a pushing test, an increasing *dc* voltage incremented in discrete steps was applied to the piezoelectric bender to displace the free end of the bender. The AFM cantilever beam (attached to the free end of the piezoelectric bender) was thus actuated to move towards the particle. Since the particle was adhered on the substrate, the AFM cantilever beam was deflected and a lateral pushing force was applied to the particle. The corresponding force increase in the test was 10–20 nN per step and the time interval between each pushing step was approximately 30 seconds. The response of the particle to the lateral pushing force was recorded by acquiring a digital image at each pushing step. From the recorded images, the pushing force (F) and particle displacement (Δx) were determined through image analysis as follows: (1) Particle displacement determination: the displacements of the particle in the x-direction (Δx_n) at each pushing step were obtained from the processing of recorded images by tracking the pixel positions the particle. (2) Pushing force determination: the AFM cantilever beam served as the force sensing element, and the applied force (F) was calculated from the cantilever deflection (D) using the linear



(a)

(b)

(c)

FIGURE 3 (a) Optical manipulation system setup; (b) Lateral pushing test experimental setup; (c) Close-up of the pushing of an adhered polymer particle with a tipless cantilever.

bending stiffness of the cantilever beam (K), $F = K \times D$. The stiffness constants of the AFM cantilever beams (K) were calibrated in the ambient after the experiments with a resonance method developed by Sader *et al.* [17] and are listed in Table 1. However, direct determination of the cantilever deflection (D) from the recorded digital image was a non-trivial task because the fixed end of the cantilever beam was out of the view field of the camera at high magnification (Fig. 3c). We utilized a method to calculate the cantilever deflection based on the calibration of the piezoelectric bender response [18]. As previously mentioned, the AFM cantilever beam was displaced by actuating the piezoelectric bender. The applied *dc* voltage was increased in constant steps, which, consequently, induced an approximately

TABLE 1 Key Adhesion Data for the Six Polymer Microspheres in the Pre-Rolling and Rolling Phases of the Rolling Experiments

Particle no.	Cantilever beam stiffness (N/m)	Particle diameter (μm)	Pre-rolling stiffness (N/m)	Rolling stiffness (N/m)	Critical distance (on set of rolling) (nm)	Critical angle of rolling ($\times 10^{-3}$ rad)	Work of adhesion ($\times 10^{-3}$ J/m^2)
1	0.030	8.1	0.29	0.10	245	60	15
2	0.019	7.6	0.20	0.07	105	64	11
3	0.030	7.5	0.47	0.16	175	47	25
4	0.030	7.3	0.32	0.06	70	19	17
5	0.031	8.2	0.81	0.10	105	26	43
6	0.031	7.4	0.42	0.08	175	47	22

constant increase of the AFM chip displacement. At each pushing step, the cantilever deflection (ΔD_n) was obtained by subtracting the displacement of the free end of the cantilever beam (ΔT_n) from the displacement of the fixed end of the cantilever beam (ΔC_n), *i.e.*, $\Delta D_n = \Delta C_n - \Delta T_n$. The displacements of the free end of the cantilever beam (T) were obtained from analysis of recorded images. The displacements of the fixed end of the cantilever beam (C), the same as the displacements of the AFM chip, were obtained by the following procedure: after each pushing test, the same sequence of *dc* voltage steps was applied to the piezoelectric bender, and the displacements of the unloaded cantilever beam were recorded, which were experimentally confirmed to be the same as the displacements of the fixed end of the cantilever beam (C) during the pushing test.

Following the determination of the pushing force (F_n) and the particle displacement (Δx_n) at each pushing step, a force-displacement (F-Δx) curve is constructed for each particle tested. The work of adhesion between the particle and substrate can then be extracted from the response of the particle to the lateral pushing force [13]. For the lateral pushing test experimental configuration (Fig. 1), the slope of the force-displacement curve (k) can be approximated in a displacement range corresponding to the pre-rolling phase of motion as

$$k = \frac{F}{\Delta x} = \frac{4M}{\theta d^2}, \tag{1}$$

where M is the moment generated by the pushing force with respect to the particle-substrate contact area, θ is the angle of rotation of the particle with respect to the particle-substrate bond, and d is the diameter of the spherical particle. According to Dominik and Tielens [11], the rolling resistance moment as a function of the angle of rotation, θ, can be approximated as

$$M \approx 6\pi W_A (d/2)^2 \theta, \tag{2}$$

where W_A is the work of adhesion between the particle and substrate. Therefore, from Eqs. (1) and (2), the work of adhesion is directly proportional to the slope of the force-displacement curve, *i.e.*,

$$W_A = \frac{k}{6\pi}. \tag{3}$$

It is noteworthy that, in the current approach, no knowledge of the particle diameter is required for determining the work of adhesion between the particle and substrate.

EXPERIMENTAL RESULTS

The experimental procedure detailed above was applied to six polymer particles. The force-displacement (F-Δx) curves extracted from the pushing tests are presented in Fig. 4a. With increasing pushing force, the displacement of the particle increases. The initial portions of the six curves are presented in Fig. 4b. Following the first few loading steps, the slope of the curves often decrease significantly. This behavior is consistent with our previous observations with the SEM [13], and we believe that at the first few loading steps there exists resistance against the rolling initiation of the particles. Therefore, the initial particle displacement is due to the pre-rolling motion of the particle. The sudden slope change after the initial displacement indicates that the adhesion bond between the particle and substrate yields (breaks) and loses its ability to resist rolling. As a result, the particle starts rolling, possibly without slip in the initial phase of rolling, on the substrate. The maximum particle displacements prior to rolling are observed to be in the range of 70–245 nm. These values are comparable with the contact radius between the polymer particle and the substrate (100–300 nm) estimated based on the Johnson-Kendall-Roberts (JKR) adhesion model [1]. With further increase of the lateral pushing force, the particle may experience rolling with slip, stick-slip, or sliding motions on the substrate (Fig. 4a), and such behavior was also observed in our previous study in the SEM [13].

The lateral pushing test results are summarized in Table 1. The diameters of individual particles are measured from the recorded images, and the work of adhesion between the polymer particle and silicon substrate is calculated from the pre-rolling slope of the

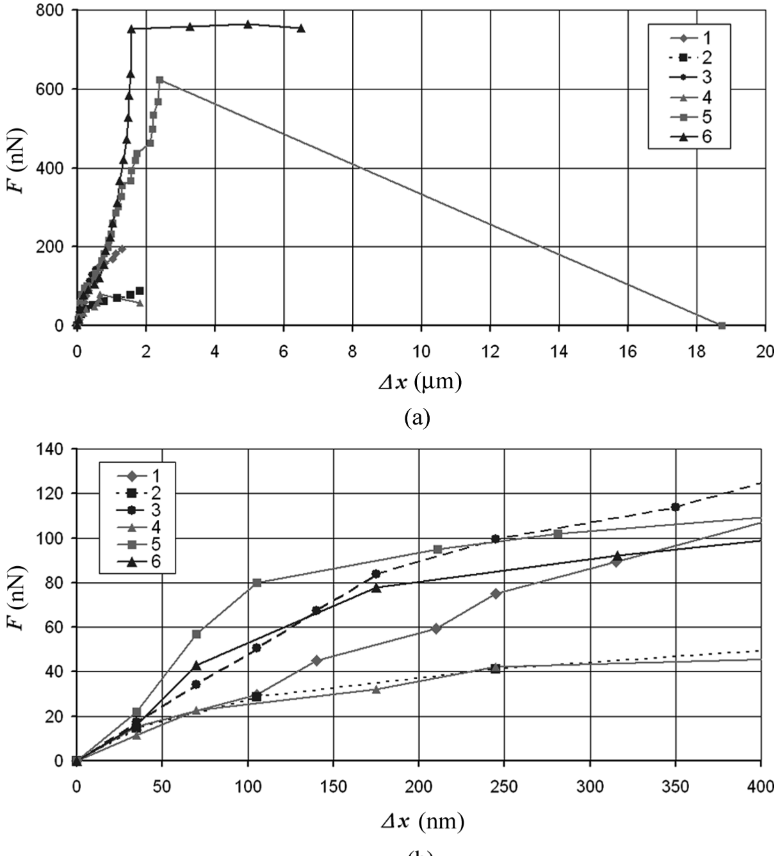


FIGURE 4 (a) Force-displacement curves of the six polymer microspheres under lateral pushing force; (b) Close-up of the initial portions of the curves.

force-displacement curve (Fig. 4b). For these experimental EA toner particles with a polystyrene shell, it is determined that the measured work of adhesion values are in the range of 11–43 $\mathrm{mJ/m^2}$. While there is no other direct experimental data on such toner particles, the work of adhesion values obtained were in the same range as those of other toner particles reported in the literature [19]. Moreover, the measured work of adhesion between the polymer particle and the silicon also agrees well with that between a polystyrene latex microsphere and silicon from both the theoretical prediction [12] and our previous experiments in the SEM [13]. The corresponding critical angles of rolling (θ^*) are estimated from the pre-rolling portion of the force-displacement curve using the approximation $\theta \approx \tan\theta \approx \Delta x/(D/2)$ [13]. As reported in Table 1, the corresponding critical angles of rolling are in the range of 19–64 miliradians.

CONCLUSIONS

In summary, we have demonstrated that the rolling resistance moment of an adhered polymer microparticle can be measured in the ambient environment, and that the work of adhesion of the bond can be extracted from this information. Lateral pushing tests on the toner particles adhered to the silicon substrate were performed with a custom-made manipulation system under an inverted optical microscope. The response of the particle to the lateral force was obtained, and the particle-substrate adhesion property was deduced from the slope of the corresponding force-displacement curve. From the pre-rolling slope the force-displacement curve, the work of adhesion of the polymer particle on a silicon substrate was determined in the ambient environment, which ranged from 11 to 43 $\mathrm{mJ/m^2}$. The corresponding critical angles of rolling were estimated to be 19–64 milliradians. The adhesion measurement in the ambient demonstrated in this work allows electrical charge-free characterization of polymer particle-substrate adhesion. It requires no permanent attachment of a particle to the cantilever beam for testing and, therefore, is non-destructive for the particles. Also, in the current approach, no knowledge of the particle diameter is needed for the work of adhesion determination.

ACKNOWLEDGMENTS

This work was supported by grants from Xerox (Xerox Corporation University Affairs Committee Award, 2007–2010) and from the Coulter School of Engineering at Clarkson University (Seed Research Grant for Exploration of Micro/Nano-Scale Intermolecular Surface Interactions, 2007–2008).

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