Rolling resistance moment of microspheres on surfaces

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In the current work, for the first time, the existence of a rolling moment of resistance of an adhesion bond between a microsphere and flat surface subjected to external dynamic force has been experimentally demonstrated. The rotational motion of spherical particles deposited on a wafer is excited in the 0–3.5 MHz range using a piezoelectric transducer. The approach is based on (i) the observation that the contribution of the rotational (rocking) motion to the axial displacement of the particle are few orders of magnitude higher than those of the purely axial motion and (ii) the existence of a relationship between the rotational natural frequency of the adhesion bond and the work of adhesion. The natural frequency of the rotational (rocking) motion is extracted from the low frequency components of the transient response of the particle in the axial direction, which is measured by a laser interferometer. The existing theoretical adhesion models for rolling resistance moment are evaluated using the experimental results. Good agreement between the theoretical predictions and experimental values is found.

1. Introduction

The rolling resistance moment exhibited by a spherical particle bonded to substrate is an important measure for studying interfacial forces and particle removal processes, as well as measuring the work of adhesion. Many adhesion theories have been proposed to understand the axial stiffness and strengths of particle–surface bonds. Among these theories, the Johnson–Kendall–Roberts (JKR) theory [1] and Derjaguin–Muller–Toporov (DMT) theory [2] have gained wider acceptance [3]. The JKR model is considered to be a good approximation for soft, small particles, while the DMT model for hard, large particles. In Johnson and Greenwood [4], a unifying framework for existing theories (namely, Hertz, JKR, DMT, M-D [5], and Bradley [6]) has been proposed and a transition between them has been established for ranges of external loads and an elasticity parameter. In these one-dimensional theories the pressure field in the contact area is assumed symmetric with respect to planes crossing the surface normal. To study rolling of a particle, adhesion models must be

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modified to take the asymmetric pressure fields generated at the contact area into consideration.

The JKR theory is a one-dimensional adhesion model. It assumes that a particle in contact with a flat substrate induces short-range forces (adhesion and elastic forces) between the substrate and the particle, and this leads to deformation of the particle and the substrate at the contact area. The study of particle rolling requires a two-dimensional analysis. The rolling resistance moment is a moment that opposes the rotational motion of the particle on the surface. In deriving an expression for this rolling moment, an external force is applied at the centre of the spherical particle [7]. Due to this external force, which creates a moment with respect to the bond zone, the pressure distribution in the contact area becomes asymmetric, resulting in pressure variations in the contact area at the leading edge and at the trailing edge of the contact with the surface. The resulting asymmetric pressure field in the contact area creates a restoring moment in small rotational angles. When excitation is present, this restoring force, along with the rotational inertia of the particle, results in free oscillatory vibrations of the particle with respect to its contact.

The present work is an experimental investigation for demonstrating the existence of the rolling resistance moment in rotational motion of particles on surfaces. The vibrational motion of a particle is induced by subjecting the particle–substrate system to base excitation. The particle tends to make simultaneous axial and rotational (rolling) vibrational motion if the amplitude of the rotational motion is small compared to a characteristic length-scale associated with the initiation of purely rolling motion (crack propagation at the leading edge and crack closing at the trailing edge of the contact area). To measure the rolling and the axial modes of these vibrations in a non-contact manner, a laser interferometer is employed. It is determined that, since the axial bond is much more stiff than the rotational bond, rotational motion dominates the purely axial bond motion at low frequencies, therefore the axial motion measured at the top of the particle is mainly due to the rolling vibrations of the particle. The natural frequencies of the rolling motion of the particle are obtained by subsequent spectrum analysis. The resonance frequencies of these rocking vibrations are related to the work of adhesion of the particle–substrate system. The experimentally obtained values of the work of adhesion are compared with the theoretical values of work of adhesion from the literature.

2. Axial and rocking motions of a microsphere

The JKR theory [1] is a one-dimensional (axial) adhesion model for a spherical particle attached to a flat substrate. The JKR model states that the external force–axial displacement relation consists of two components (contact (Hertzian) and adhesion, respectively):

\[
F = \frac{1}{\beta} r^{3/2} \delta^{3/2} - \frac{2\alpha}{\beta} r^{3/4} \delta^{3/4}
\]

where \( \delta \) is the axial displacement of the centre of the particle with respect to the surface of the substrate due to the applied external force, \( F \), and \( \beta = r/K \) where \( K \) is
the stiffness coefficient of the adhesion bond given by

\[ K = \frac{4E_1E_2}{3(E_2(1-v_1^2) + E_1(1-v_2^2))} \]  

(2)

where \( E_1 \) and \( E_2 \) are the Young’s modulus and \( v_1 \) and \( v_2 \) are the Poisson’s ratios of the substrate and particle, respectively, and \( \alpha = 3\pi W_d r \). The stiffness expression, equation (1), is a nonlinear relationship between the applied external force, \( F \), and axial displacement, \( \delta \). The stiffness expression is linearized at the stable equilibrium point at \( \delta^* = (2\alpha \beta)^{3/2}/r \) to determine the axial natural frequency of the particle–substrate system in the neighbourhood of its stable equilibrium:

\[ K^* = \frac{dF}{d\delta} \bigg|_{\delta=\delta^*} = \frac{3r\alpha^{1/3}}{2^{5/3}\beta^{2/3}} \]  

(3)

Thus, the natural frequency of the adhesion bond in the axial direction (normal to the flat substrate) is obtained as

\[ \omega_N = \sqrt{\frac{K^*}{m}} = \frac{3\alpha^{1/6}}{2^{11/6}r\sqrt{\pi\beta} \beta^{1/3}} \]  

(4)

where \( m \) is the mass of the particle. The axial frequency for a 21.4 \( \mu \)m polystyrene latex (PSL) particle on a silicon substrate is calculated as \( f_n = 1.83 \) MHz from the stiffness coefficient \( K^* = 707.4 \) N m\(^{-1}\) while, on copper substrate, it is determined as \( f_n = 1.85 \) MHz from \( K^* = 718.6 \) N m\(^{-1}\).

Besides its axial motion, the particle can have a rotational degree of freedom due to radial modes of motion of the base and can make rotational vibrations on a flat surface (figure 1a) with respect to the centre of the contact area. Unlike the axial motion, in order to model this rotational motion of the particle, a two-degree-of-freedom adhesion theory needs to be utilized. It is known that the non-uniform stress distribution in the contact area during the rotational motion creates a restoring moment (also referred to as resistance moment) to rolling motion (figure 1b) [7], which is proportional to the angle of rotation and leads to angular free-vibrations of the particle.

The pressure distribution \( p(x, y) \) for a spherical particle on a flat substrate, according to the JKR adhesion model, has to be cylindrically symmetric when no external rotational moment is exerted on the particle. The moment of resistance in the case of symmetric pressure distribution is, therefore, \( M_y = \iint xp(x, y) \, dx \, dy = 0 \). However, if an external shear force or horizontal displacement field is applied to the substrate, the moment associated with rotational (rocking) motion of the particle results in shifting of contact area. In other words, the contact area is no longer centered around the point which is located at the centre of the original contact circle and the pressure distribution becomes asymmetric. The contact radius of the 21.4 \( \mu \)m PSL particle on a silicon substrate is approximately 231 nm and the force of adhesion is 1.18 \( \mu \)N while on a copper substrate is about 235 nm and the force of adhesion is 1.24 \( \mu \)N.

The pressure distribution \( p(x, y) \) and the moment associated with the rocking motion can be calculated from the following assumptions [7]: (i) the true shifted contact area is approximated by decomposing the contact area into two circles of different radii \( a + \xi \) and \( a - \xi \) where \( \xi \) is the shift in the contact area due to rocking motion; (ii) the half circle \( x < 0 \) is one half of a symmetrical contact with contact
Figure 1. (a) Base excitation method diagram for measuring work of adhesion (not to scale). (b) Rocking motion of particle subject to base excitation.
radius \( a + \xi \) and pressure distribution \( p(r, a + \xi, \delta_2) \); (iii) the half circle \( x > 0 \) has a smaller contact radius \( a - \xi \) and a corresponding pressure distribution \( p(r, a - \xi, \delta_2) \). The total moments due to both the half circles is calculated. The resulting distribution is discontinuous at \( x = 0 \). From the derivations reported by Dominik and Tielens [7], an approximation for \( M_y \) is given as

\[
M_y \approx 2\xi(-E^*\delta_2 + 3P_c\hat{\alpha}^3).
\] (5)

Utilizing Johnson’s solution [8] for \( \delta_2 \), corresponding to the contact radius \( a \), the resulting moment simplifies to

\[
M_y \approx 4P_c\hat{\alpha}^{3/2}\xi
\] (6)

This expression indicates that the moment associated with the pressure asymmetry is proportional to the pull-off force \( P_c \) and the shift of the centre of contact area. For a spherical particle in contact with a flat surface, \( P_c = (3/2)\pi W_A r \) where \( W_A \) is the work of adhesion, and if the normal forces stay within the range of \(-P_c < P < P_c\), the factor \( \hat{\alpha}^{3/2} \) varies in the range 0.5–1.2 and assuming \( \hat{\alpha} = 1 \), the rolling resistance moment for a particle on a flat substrate is further simplified from equation (6) to \( M_y \approx 6\pi W_A \xi r \). Utilizing this restoring rotational moment, the equation of motion of a spherical particle in free-rotational oscillation on a flat surface is obtained as

\[
I \ddot{\theta} + 6\pi W_A \xi r = 0,
\]

where \( \xi = r\theta \) is the shift in contact area due to the asymmetric pressure. The resonance frequency of this oscillator for the rocking motion can be determined as

\[
\omega_n = \frac{1}{r^{3/2}} \sqrt{\frac{45 W_A}{4 \rho}}
\] (7)

where \( \rho \) is the mass density of the particle material. Clearly, the rocking natural frequency is dependent on the radius of the particle, work of adhesion of the particle–substrate system and the density of the particle. It is noteworthy that the elastic properties of the particle and substrate material appear to play no role in the rocking motion while the natural frequency of the axial motion is a function of the elastic properties of the particle–substrate system. From equation (7), the rocking frequency for a 21.4 \( \mu \)m PSL on a silicon substrate is calculated as \( f_n = 72.5 \) kHz and for PSL particles on a copper substrate as \( f_n = 74.32 \) kHz.

3. Experimental determination of rocking resonance frequencies and work of adhesion

To verify the relationship between the rocking natural frequency and adhesion properties, the resonance frequencies of a microsphere on surfaces of substrates made of different materials were experimentally determined. To measure the rocking frequency of a particle on a flat substrate, a set of experiments were designed and conducted. The experimental technique is based on a measurement scheme for axial motion of microparticle with a laser interferometer. The schematic of the experimental set-up and instrumentation diagram is depicted in figure 2.

Dry 21.4 \( \mu \)m spherical polystyrene latex (PSL) particles (Duke Scientific Inc.) were used in all the experiments reported in this study. They were deposited on pieces of metal-coated wafer surfaces. The wafer piece was then placed on a
3.5 MHz transducer (Panametrics, V682). Coupling gel was applied between the wafer sample and the transducer for good acoustic transmission. The transducer was mounted on a xy-translation stage of an optical microscope. A CCD camera was attached to the optical microscope to monitor the experiments. A laser Doppler vibrometer (LDV) [Polyteck], vibrometer controller unit (OFV 3001), fibre interferometer unit (OFV 511) and ultrasonic displacement decoder (OVD 030) was integrated with an optical microscope; the laser beam of the fibre interferometer unit is transmitted through the microscope objective. The size of laser spot of the fibre interferometer unit can theoretically be reduced to 0.5 μm using a 100× objective of the optical microscope. The laser spot is directed to the top of the particle on the surface (figure 3). The transducer was excited by a square pulse from pulser/receiver (Panametrics, Model 5077PR). The particles on the wafer performed a complex motion due to the combination of vibrational modes (axial and radial) of the irregular wafer piece. The axial response of the particle to this complex vibration field was measured using the LDV. The frequency band of the displacement decoder is specified as 50 kHz–30 MHz. The LDV measures the axial motion of the particle by comparing the frequency and phase difference of the emitted and the reflected laser beam. This difference in phase is decoded into displacement by the

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**Figure 2.** Schematic of the experimental setup for work of adhesion measurement and instrumentation diagram.

**Figure 3.** Positioning the interferometer laser spot (a) on a PSL particle deposited on a substrate (b) on the substrate.
displacement decoder while the difference in frequency is decoded into velocity by velocity decoder. The decoders generate a waveform in time domain. The trigger line to the digitizing oscilloscope (Tektronix TDS 3052) was provided from the pulser/receiver for synchronizing data acquisition. The laser spot of the fibre interferometer unit was focused onto the particle. This procedure was repeated to measure the response of the surface of the wafer piece to characterize the base motion.

A procedure for measuring the displacements of the rocking particles is developed as follows. Dry 21.4 μm PSL particles were deposited on a small piece of silicon wafer and sufficient time was allowed for the particles to relax and adhere to the wafer. As the expected axial motion frequencies are in the MHz range (from equation (4), 1.83 MHz for the PSL–silicon system and 1.85 MHz for the PSL–copper system) and the expected rocking motion frequencies are in the kHz range (from equation (7), 72.5 kHz for PSL–silicon and 74.3 kHz for PSL–copper), there is a good frequency band separation and, thus, the components of the motion of particles subjected to base excitation can be identified easily. This separation occurs because the stiffness of the axial bond is much higher than that of the rolling resistance. The transducer was excited with a pulse of amplitude 400 V and 100 Hz PRF from the pulser/receiver unit. It was observed that some of the particles on the wafer tend to agglomerate and form clusters, while some others tend to oscillate as single particles. These single oscillating particles on the wafer were located for rocking frequency measurements. The laser spot of the fibre interferometer unit was focused on a particle using 100× magnification of the optical microscope as shown in figure 3a. The transient axial motion of the particle was recorded from the oscilloscope for signal processing. The xy-translation stage was adjusted to focus the laser spot on the wafer (figure 3b) and the transient surface response was recorded. This procedure was repeated for different particles on the substrate and the corresponding waveforms were digitized and stored for signal processing. The experiment was repeated for various particles on copper and tantalum-sputtered silicon wafers and the results were compared.

4. Results of experimental investigation

The axial displacement waveforms of the 21.4 μm PSL particle on tantalum substrates are depicted in figure 4. The axial displacement (δ) measured by the interferometer is related to the angular motion of the particle (θ) (figure 1b) via \( \delta = 2\rho(1 - \cos \theta) \). The contribution of the axial extension of the adhesion bond to the measured axial displacement \( \delta(t) \) is negligible compared to the measured axial displacement due to the rocking mode of vibration. The maximum rocking angle (θ) of the PSL particle on a silicon substrate is, for instance, measured as approximately 0.16°. The corresponding shift in contact area for the PSL–silicon system is, therefore, calculated as \( \xi = 29.96 \) nm. It is instructive to compare this shift to the radius of the contact area that is approximately \( a = 231 \) nm.

The amplitude of the axial motion component can be approximated from the measured transient surface acceleration data. The maximum surface acceleration of the silicon and copper substrates can be approximated from the axial displacements waveforms. The amplitudes of maximum surface acceleration for silicon and copper substrates are estimated from the displacement amplitude and frequency data (\( a_{\text{max}} \sim d_{\text{max}}(2\pi f)^2 \sim 10^5 \text{ m s}^{-2} \)). The maximum force \( F_{\text{max}} \) at the surface of
the substrate is estimated from $F_{\text{max}} = ma_{\text{max}}$. The corresponding maximum extension of the particle–surface bond in axial mode of vibration is, therefore, in the order of $\delta_{\text{max}} = F_{\text{max}}/K^*$, where $K^*$ is the stiffness coefficient of adhesion bond at the stable equilibrium point. From equation (3), the stiffness coefficient for silicon and copper substrates are calculated as $K^* = 707.4 \text{ N m}^{-1}$ and $K^* = 718.6 \text{ N m}^{-1}$, respectively. The numerical value of the axial motion amplitude for PSL particle on silicon and copper substrates is approximately 0.9 nm. Since the amplitudes of the measured axial motion is in the order of $O(10) \text{ nm}$, the axial motion amplitude is nearly two orders of magnitude lower than that reported in the current experiments. In consequence, it is established that the obtained amplitudes are predominantly due to the rocking motion of the particle.

From the transient response of PSL particles on a tantalum substrate, it is clear that the wafer response due to the base motion diminishes faster than that of the particle (figure 4). The attenuative nature of the particle rocking motion in figure 4 is attributed to the viscoelastic properties of the PSL materials [9]. The temporal response of PSL particles on silicon, copper and tantalum substrates are transformed into spectral domain using the Fast Fourier Transform (FFT) algorithm to determine the natural frequencies of the rocking motion as depicted in figure 5. The frequency

![Figure 4](image-url)

Figure 4. (a) Time domain response of PSL-tantalum system. The dashed line corresponds to particle response while the solid line corresponds to wafer response. (b) Close-up of waveforms in (a).
Figure 5. Frequency spectra of the response of the PSL particles on silicon (a), copper (b) and tantalum (c) substrates. The dash lines represent the calculated natural frequencies ($f^* = 72.4$ kHz and $f^* = 74.3$ kHz) based on adhesion properties of silicon (a) and copper (b) from Visser [10], respectively. No adhesion data on tantalum were found in the literature.
Table 1. Work of adhesion measurement experimental results. The expected work of adhesion values are calculated from the reported Hamaker constants of the particle and substrate materials [10]. The density, Young’s modulus and Poisson’s ratio of PSL particles are given as 
\[
\rho = 1040 \text{ kg m}^{-3}, \quad E = 2.77 \text{ GPa}, \quad v = 0.33, \text{ respectively.}
\]

<table>
<thead>
<tr>
<th>Particle–substrate system</th>
<th>Rocking natural frequency (kHz)</th>
<th>Experimental work of adhesion (mJ m(^{-2}))</th>
<th>Expected work of adhesion (mJ m(^{-2}))</th>
<th>Expected rocking and axial natural frequency (MHz)</th>
<th>Substrate density (kg m(^{-3}))</th>
<th>Young’s modulus (GPa)</th>
<th>Poisson’s ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>PSL–silicon</td>
<td>64.1, 79.3, 94.1, 109.0, 123.0, 136.6</td>
<td>18.4, 28.1, 39.6, 53.1, 67.6, 83.4</td>
<td>23.5</td>
<td>72.5 (kHz)</td>
<td>2329</td>
<td>127</td>
<td>0.28</td>
</tr>
<tr>
<td>PSL–copper</td>
<td>69.4, 136.0, 180.0, 218.0, 251.0</td>
<td>21.5, 82.7, 144.9, 212.5, 281.7</td>
<td>24.7</td>
<td>74.3 (kHz)</td>
<td>8833</td>
<td>114.7</td>
<td>0.34</td>
</tr>
<tr>
<td>PSL–tantalum</td>
<td>55.6, 69.0, 98.2, 116.1, 142</td>
<td>13.8, 21.3, 43.1, 60.3, 90.2</td>
<td>n/a</td>
<td>n/a</td>
<td>16650</td>
<td>186</td>
<td>0.34</td>
</tr>
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</table>
spectrum indicates that the particle vibrates with multiple frequencies and hence there could be substantial variation in the work of adhesion with the direction and plane of the rocking motion. The observed natural frequencies are related to the work of adhesion as according to equation (7). The experimental values for work of adhesion, obtained from the rocking frequencies, are compared to the work of adhesion values that are estimated from the properties of materials reported in the literature [10]. The mechanical properties of the particle and substrate materials and the results of the analysis of experimental results are tabulated in table 1.

5. Conclusions

The existence of a rolling resistance moment in a microsphere on a flat surface is, for the first time, experimentally demonstrated by a series of experiments based on laser interferometry technique. First, it is determined that the contribution of the axial mode of vibration from the axial displacement of the particle is negligible compared to the contribution of the rolling motion to the axial displacement measured by the laser interferometer. Thus, it is concluded that the measured resonance frequencies correspond to the rotational oscillation component of the particle motion. The resonance frequencies of the rocking motion are determined from spectrum analysis of the axial displacement waveforms and then related to the work of adhesion of the particle–substrate system. The experimental work of adhesion of PSL particles on three different substrates is calculated and the obtained numerical values are compared to the values reported in the literature. There is good agreement of the experimental data with analytical predictions for work of adhesion. Thus, it is experimentally established that the rolling resistance moment on a microsphere–substrate system exists when an external rotational moment is applied to the microsphere.

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