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Adhesive contact of randomly rough surfaces: experimental and numerical investigations

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Abstract. The contact mechanics of soft matters is strongly affected by short-range adhesive interactions, which can lead to large deformations and contact instabilities.

In this work, we present both experimental and numerical investigations of the adhesive contact between soft elastic bodies with a Greenwood and Williamson (GW)-like roughness. To investigate the coalescence of neighbour contact spots, surfaces have been designed with overlapping spherical asperities. Normal contact experiments are carried out by using a home-built device. Numerical simulations are performed with the Interacting and Coalescing Hertzian Asperities (ICHA) model, conveniently modified to take account of adhesion according to the Johnson, Kendall & Roberts (JKR) theory.

1. Introduction
Adhesive contact of soft elastic bodies is of great interest in several engineering applications, ranging from bionispired adhesives [1, 2], soft robots [3, 4], stretchable electronics [5, 6].

Surface roughness is known to widely affect the interfacial properties of materials. Modeling adhesion between rough soft elastic media is a tricky challenge. The tribological properties of soft matter are strongly characterized by complex phenomena, e.g., the elastic instabilities raising at the jump into contact [7].

The classical contact mechanics theories are used to describe the simple case of the contact between two elastic spheres. Johnson, Kendall & Roberts (JKR) theory is commonly used to estimate adhesion between compliant soft bodies with short-range adhesive interactions [8]. On the other hand, Derjaguin, Muller & Toporov (DMT) theory is widely used to characterize the contact between hard bodies with long-range adhesive interactions [9].

Several models have been developed in the framework of JKR theory. Fuller & Tabor (FT) model [10] is an extension of the classical Greenwood & Williamson (GW) multiasperity theory [11], in which adhesion is modeled according to JKR theory. GW model is based on the assumption that all the asperities have the same radius of curvature and elastic coupling is negligible. Refs. [12, 13] showed that neglecting lateral interaction of asperities and coalescence of merging contact spots can lead to a significant underestimation of the contact area even at very small load.

Moreover, Persson [14] developed a theory of adhesion between an elastic solid and a hard randomly rough substrate taking into account that partial contact may occur between the solids.
at all length scales. He found that "even when the surface roughness is so high that no adhesion can be detected in a pull-off experiment, the area of real contact may still be several times larger than when the adhesion is neglected."

Degrandi-Contraires et al. [15] showed that the contact behavior between a flat PDMS surface patterned with cylindrical pillars and a spherical indenter can be accurately described by applying JKR theory with an effective adhesion energy depending on the roughness of the substrate. However, recently Acito et al. [16], performed classical JKR tests on rubber samples with a GW-type roughness. They demonstrated that the findings of Degrandi-Contraires et al. [15] cannot be extended to their surfaces because "the separation of length scales between the microasperity contacts and the macroscopic contact region is insufficient to allow for the use of a JKR model-based approach with an effective adhesion energy."

In the present paper, we experimentally investigate the adhesive contact of a smooth glass lens and a rubber substrate with a well-defined rough surface. Roughness is textured by a set of spherical microasperities with identical radius of curvature, like in a GW-type surface. A similar study has been performed in Refs. [16, 17], but we have removed the non-overlapping constraint between the asperities.

A numerical study is then performed by exploiting two multiasperity models. The first one is a discrete version of FT model, in which the geometry and the position of each single summit is determined rather than using the statistics of the asperities distribution. The second model is the Interacting and Coalescing Hertzian Asperities (ICHA) model [18], which takes account of both elastic coupling and coalescence of merging asperities. Moreover, both models are conveniently developed to consider adhesion according to a slightly modified JKR theory [19], which considers the jump into contact instabilities.

Recently, a version of the ICHA model incorporating adhesion according to DMT theory has been presented in Refs. [20, 21]. Such model proved to be very accurate in the assessment of the main contact quantities when contact occurs between rough hard elastic bodies with low surface energy. However, for soft materials the assumption of neglecting surface deformations due to adhesion is no longer valid. In this case, a JKR-type approach is certainly more appropriate [22].

2. Adhesion of surfaces with GW-like roughness

Here we present the results of an experimental campaign aimed at investigating the adhesive normal contact between a spherical smooth indenter and a nominally flat rubber substrate with roughness described by a GW-type geometry. Specifically, spherical identical microasperities are randomly distributed both spatially and in height. Differently from previous works [16, 17], adjacent spherical bumps can overlap and coalescence of merging contact spots occurs during loading experiments.

In Ref. [16], the effect of lateral interaction between the asperities has been numerically investigated for the sphere-on-flat adhesive contact. Specifically, it was observed that the contribution of the elastic coupling becomes less important when increasing the standard deviation of the asperities height. In such case, a classical discrete FT multiasperity model is accurate in describing the area-load relation.

In this work, we instead want to investigate both elastic coupling and coalescence effects. For this reason, different patterned surfaces have been numerically designed and PDMS samples has been accordingly modeled. Experimental measurements of the contact area and microcontacts distributions are then compared with the numerical predictions of a discrete version of the FT and ICHA models.
2.1. Experimental details
Classical JKR-like tests have been carried out between an optically glass lens and a rubber substrate. The glass indenter, which is assumed to be smooth, has a radius of curvature of 103.7 mm. The rubber substrate is made of PolyDiMethylSiloxane (PDMS). One PDMS sample has been manufactured by cross-linking at 70 °C for 24 hours a mixture of Sylgard 184 and Sylgard 527 liquid silicones, with a 0.35:0.65 weight ratio.

The top of PDMS sample is textured with three patterned rough surfaces. In particular, roughness is characterized by spherical microasperities having the same radius of curvature. Patterned surfaces have been obtained by moulding PDMS in PolyMethylMethAcrylate (PMMA) forms milled using ball-end mills with a radius of 100 µm. Smoothening of the spherical cavities of the PMMA molds has been performed by exposing them to a saturated CHCl₃ vapor for 30 minutes. A part of the PDMS sample has been kept smooth to allow measurements of the elastic modulus and the adhesion energy.

Three surfaces have been generated with a squared nominal area of 8 mm², where asperities are randomly distributed with a density of 2.9 × 10⁷ m⁻². Asperities heights follow a Gaussian distribution with standard deviation of 5 µm. For the first surface, asperities are collocated with a non-overlapping constraint, which has been removed for the other two surfaces. For these surfaces, coalescence phenomena may occur during loading experiments.

Indentation tests of the glass lens on the smooth part of the PDMS sample were performed and the contact radius vs load data were fitted according to JKR theory. Figure 1 shows the contact radius vs applied load relation. The fitting line allows to evaluate the values of the composite elastic modulus ($E^* = 1.21$ MPa) and adhesion energy ($\Delta \gamma = 0.037$ J/m²).

Normal contact experiments have been performed under controlled load conditions. In particular, the glass indenter is fixed to a vertical translation stage by a double cantilever beam of known stiffness (290 N/m). The contact load is measured from the deflection of the cantilever, which is detected by means of a high resolution optical sensor (Philtec D64-L).

Contact pictures are recorded at fixed load steps, ranging between 0.01 – 0.05 N, with an incremental step of 0.01 N. Both the indenter and the PDMS sample are transparent. During the contact loading, illuminating in transmission with a white LED diffusive panel, microcontacts appear as bright disks (figure 2). The measure of the area of each microcontact is performed by using standard image thresholding techniques. The total contact area $A$ is then obtained by summing up all microcontact areas.

Further details about the experimental setup can be found in Refs. [16, 17].

2.2. Experimental results and comparison with numerical predictions
Contact measurements have been performed on three different patterned surfaces. One (pattern A) presents a random spatial distribution of asperities with a non-overlapping constraint, the others (pattern B and pattern C) are instead characterized by the presence of overlapping asperities.

In the design of patterns B and C, we introduced a constraint regarding the minimum distance between the apex of two neighboring asperities. The minimum distance is equal to 0.8 and 0.6 times the radius of the microasperities for patterns B and C, respectively.

A useful quantity to characterize the surface roughness is the correlation length $\xi$, which gives the average distance that one has to travel on the surface topography (and in a random direction) in order to no longer be on the same bump. Specifically, $\xi$ is related to the variogram $H(d)$, being $d$ a horizontal lag value. $H(d)$ can be defined as the squared difference in height between two random points at a distance $d$

$$H(d) = \frac{1}{2N} \sum_{i=0}^{N-1} [h(x_i) - h(x_i + d)]^2,$$  (1)
Figure 1. The contact radius $a^{3/2}/R$ as a function of the applied load $F/a^{3/2}$. Results are referred to normal contact experiments between the glass spherical indenter ($R = 103.7$ mm) and the smooth PDMS sample. Experimental measurements are denoted with markers, while the solid line is obtained by fitting data with JKR theory.

Figure 2. Image in transmission of the contact between the spherical indenter and a patterned rough surface (compressive load $F = 0.05$ N). The inset shows the coalescence of merging contact spots.
Figure 3. The variogram \( H(d) \) for the three patterned surfaces. The correlation length is calculated in \( x \) (solid line) and \( y \) (dashed lines) directions. We obtained \( \xi = 120 \ \mu m \) for the pattern A and \( \xi = 100 \ \mu m \) for the patterns B and C.

where \( h(x_i) \) is the height of a point located at a random position \( x_i \) and \( N \) is the total number of measurements. In figure 3, \( H(d) \) is shown for the three patterned surfaces; calculations have been performed on rough profiles extracted from the surface topography in \( x \) and \( y \) directions, respectively. The correlation length \( \xi \) is equal to the distance at which \( H(d) \) is no longer dependent on the value of \( d \). For our surfaces, we find that \( \xi \) is always of the same order of magnitude of the microasperities radius. However, the presence of overlapping bumps (patterns B and C) leads to a little decrease in \( \xi \).

Figure 4 shows the contact area \( A \) as a function of the applied load \( F \) for the three patterned surfaces. Numerical calculations have been performed with the FT-discrete and ICHA models.

For each load level, five repetitions of the test were performed on each patterned surface and at the same location on the pattern. Data were obtained close to adhesive equilibrium as after each load increment the system was kept at rest for a long time (1000 s). Curves show non-linearity and, at given load, higher contact areas are detected for the surfaces B and C, where coalescence occurs. We notice that exposition of the spherical cavities of the PMMA molds to a saturated CHCl\(_3\) vapor causes a little increase in the nominal radius of curvature of the asperities resulting in slightly larger values of the contact area at the higher loads.
In all cases, numerical and experimental data are in good agreement. The inclusion of the elastic coupling in the ICHA model, leads to greater values of the contact area with respect to the FT-discrete model. This is in agreement with previous findings [12, 20].

For pattern A, the predictions of the ICHA and FT-discrete models are very close, especially in the range of small loads. For patterns B and C, greater differences are observed due to coalescence of merging asperities. In the range of loads considered in the experiments, differences are not significant as the number of overlapping contact regions is quite small. However, we remember that the effect of coalescence is instead very significant when roughness develops on several length scales [20].

Figure 5 shows the contact spots at increasing load steps \( F = 0.01, 0.03, 0.05 \) N for pattern C. The first three images (a-c) refer to the contact regions observed during the experiments (we removed the background and highlighted contact spots in blue). The other figures (d-f) show the contact spots distribution predicted by the ICHA model. The inset in the figure shows a superposition of the numerical contact areas (red circles) with the experimental ones (blue disks) at the load value \( F = 0.05 \) N. The coalescence of microcontacts is well detectable, so as the replacement of the merging contact spots with an equivalent macro-region that occurs in the ICHA model.

3. Conclusions
In this work, we present experimental investigations of the adhesive contact between a ‘rigid’ smooth sphere and nominally flat surfaces with GW-like roughness. To capture the coalescence of contact spots, surfaces were designed with overlapping spherical bumps of identical radius of curvature.

Experimental data are then compared with the numerical predictions of an *ad-hoc* multiasperity model, which takes account of both elastic interactions between asperities and coalescence of merging contact regions. The model is based on JKR theory, which is conveniently modified to consider jump into contact instabilities.

The observed agreement is very good; however, at this initial state, we performed tests and calculations only for the loading phase; therefore, further studies and developments are required to test the reliability of the model.

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Appendix A. Numerical model
The *Interacting and Coalescing Hertzian Asperities* (ICHA) model is an advanced multiasperity model aimed at studying the contact between rough surfaces. It showed to be quite accurate and efficient in predicting the main contact quantities in adhesionless cases [23, 12] and in presence of adhesion modeled with a DMT-type approach [13, 20]. In the present work, we have instead used a different version of the ICHA model, in which adhesion is modeled according to the JKR theory [22]. Specifically, following the JKR formalism, the normal displacement \( w_i \) of the elastic half-space at the location of the asperity \( i \) can be written as

\[
    w_i = \frac{a_i^2}{R_i} - \sqrt{\frac{2\pi a_i \Delta \gamma}{E^*}} + \hat{w}_i
\]  

(A.1)
Figure 4. The true contact area $A$ ($m^2$) as a function of the applied load $F$ (N). Results refer to normal contact experiments between a spherical indenter and three different patterned rough surfaces. Five contact repetitions have been performed in the same location of the patterns. Experimental measurements are denoted with circular markers. The FT-discrete and ICHA models predictions are shown with blue dotted line and red solid line respectively.
Figure 5. Experimental (blue disks) and numerical (red circumferences) microcontact spots. Results are shown for the pattern C, at loads $F = 0.01$ N (a, d), $0.03$ N (b, e) and $0.05$ N (c, f). The inset shows a superposition of the measured contact spots with the numerical one predicted at the same level of applied load. The length of the rectangle in the inset is equal to 1 mm.

where

$$\hat{w}_i = \sum_{j=1,j\neq i}^{n_{ac}} \frac{a_j^2}{R_j \pi R_j} \left( \sqrt{\frac{r_{ij}^2}{a_j^2}} - 1 + \left( 2 - \frac{r_{ij}^2}{a_j^2} \right) \arcsin \left( \frac{a_j}{r_{ij}} \right) \right)$$

$$- \frac{1}{\pi a_j E^*} \sqrt{8 \pi a_j^3 E^* \Delta \gamma} \arcsin \left( \frac{a_j}{r_{ij}} \right)$$

(A.2)

is the displacement due to the elastic coupling. The term $\hat{w}_i$ takes into account that the displacement on the asperity $i$ depends on the contributions of all the contacting asperities $n_{ac}$. In equation (A.2), $r_{ij}$ is the distance between the asperities $i$ and $j$.

The classical JKR model predicts the first contact occurs when the approach is equal to zero. However, Wu [7] found that jump-on occurs at a critical gap

$$\Delta_{ON} = \left( 1 - 2.641 \mu^{3/7} \right) \varepsilon$$

(A.3)

where $\varepsilon$ is the range of attractive forces and $\mu = \left( \Delta \gamma^2 R / E^* \right)^{1/3}$ / $\varepsilon$ is the so-called Tabor parameter [24].

On the base of the Wu’s findings, Ciavarella et al. [19] suggested to add the effect of van der Waals interactions in the JKR theory, by using equation (A.3) for the jump-on critical distance. Therefore, one can assume that a contact raises when the gap between an asperity and the half-space becomes smaller than the jump-on distance.

The first estimate of the asperity contact radius is done by inverting the JKR relation

$$\delta = \frac{a_j^2}{R} - \sqrt{\frac{2 \pi a_j \Delta \gamma}{E^*}}$$

Then, after a further increase in the penetration $\eta_i$, the contact radius
is increased by the quantity
\[
\Delta a_i = \frac{\eta_i}{2a_i/R_i - \sqrt{\pi \Delta \gamma/(E^*a_i)}}
\]  \hspace{1cm} (A.4)

We further recall that, according to the method developed in Ref. [18], asperities with overlapping contact spots are replaced with a new equivalent one, which maintains the same total contact area of the suppressing asperities. The position of the new asperity is defined by keeping unchanged the volume centroid, and its radius of curvature \( R_{eq} \) is empirically assumed equal to \( (R_i^2 + R_j^2)^{1/2} \). Finally, the height \( h_{eq} \) of the new equivalent asperity is defined so that the contact radius of the corresponding contact spot returns effectively the area \( \pi a_{eq}^2 \) at the given separation.

The total contact area and the total load are obtained by summing up the contributions of all the asperities in contact. In this respect, we recall that the contact load \( F_i \) exerted on the spot of radius \( a_i \) is calculated with the JKR formula
\[
F_i = 4 \frac{E^* a_i^3}{R_i} - \sqrt{8\pi E^* \Delta \gamma a_i^3},
\]

where \( R_i \) is the radius of the asperity \( i \), \( E^* \) is the composite elastic modulus and \( \Delta \gamma \) is the interface energy of adhesion.

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