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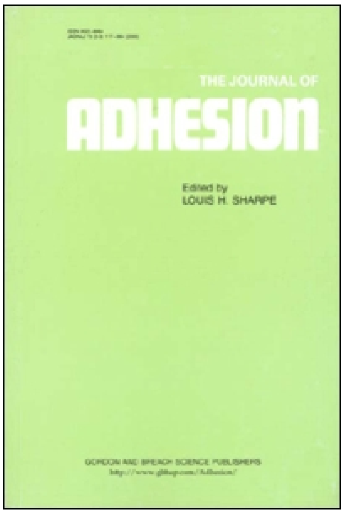

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Finite Element Modeling of Particle Adhesion: A Surface Energy Formalism

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The adhesion of particles is modeled with finite element analysis using an energy approach comparable with that used in the JKR formalism. The strain energy of a cylindrically symmetric system, comprising a particle adhering to a surface with a fixed contact size, is computed as a function of contact size and then added to an energy term that is linearly proportional to the contact patch area. These computations also include contributions from the potential energy of a body force comparable with that which might be applied by a centrifuge. The results show regions of stability (adhesion) where a local energy minimum exists and regions of release where separation of the particle from the surface leads to a continuous decrease in the energy of the system. The effect of the deformation of the particle is included implicitly as a result of the FEM which provides details of the strains and stresses within the system. Discussion concentrates on the physical meaning of the behaviors and the significance of JKR-like theories that use an effective surface energy to represent electrostatic and van der Waals contributions to the adhesion. Modeling the effects of surface roughness of particles and the plastic deformation of particles through an effective surface energy is considered.

Keywords: Particle adhesion; Finite element method; Surface energy; Numerical contact mechanics

INTRODUCTION

The adhesion of particles is well known to be the direct result of the electrical interactions between molecules. These can be either

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electrodynamic, such as in the case of van der Waals interactions between electrically neutral species, or electrostatic, as in the case of direct attraction between oppositely-signed charged species. Some researchers favor explanations such as the charged-patch model [1, 2] dominated by electrostatic behavior while others believe the electrostatic attraction is primarily a far-field effect with the near-field behavior controlled by van der Waals interactions [3–6]. Computations have been made that include both contributions to explain the spatial dependence of the force measured between a particle mounted on an AFM probe and a surface [7].

The charged-patch model of particle adhesion and the uniformly charged approach both focus on the distribution of charge on the surface of the particle and the attractive forces that are developed as a result of the electric charge interactions. Alternatively, the Lifshitz–Hamaker approach [8–10] is taken to include the van der Waals contribution. These two approaches have generated a dichotomy of thought regarding particle adhesion. Those favoring the Lifshitz–Hamaker approach tend to view adhesion in terms of surface energy contributions, such as those discussed in the JKR model [11]. Alternatively, those favoring the electrostatic models tend to view adhesion and particle detachment in terms of balancing the attractive and applied (detachment) forces. Both the electrostatic and van der Waals contributions include action at a distance, namely the direct attraction of opposite sign charge *via* the $1/r^2$ law or the attraction of induced dipoles that lead to the $1/r^6$ dependence, sometimes broadly referred to as the London dispersion force [12]. While it has been shown for simple geometries [13] that the van der Waals forces, as modeled by the Lennard–Jones potential, can give rise to an apparent surface energy, the detailed mechanics of the attraction cannot be explained by a surface energy alone [14, 15]. Rather, the spatial rearrangement of the atoms that occur as a result of the interaction forces must be included. This rearrangement of atoms gives rise to larger forces than would be expected for van der Waals interactions between static atomic groupings and leads to a hysteresis in the load-displacement behavior [16, 17].

It seems clear that the use of the surface energy paradigm is a simplification of the physics described above, with the intent that the important aspects of the adhesive behavior would be captured using

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the surface energy representation. Indeed, the wide-spread acceptance of the JKR theory [11] suggests that much of the behavior of the particle and surface interaction has been captured with the use of an effective surface energy. In the larger perspective, the use of the surface energy under the rubric of an energy method is well known to represent the energetics of a situation without regard to the detailed mechanisms involved. Indeed, this is one of the beauties of the energy approach to mechanics problems, in that the messy mechanistic treatment can sometimes be avoided. The downside of the energy approach is that its result provides no information as to the mechanism or its details.

Experimental approaches to the study of particle adhesion have shown that the mechanical properties of the particles play a substantial role in the apparent adhesion to surfaces [18]. Rimai *et al.*, have shown that the relationship between contact-patch size and particle size varies depending on the mechanical properties of the particles and substrates. Theoretical treatments that involve plasticity [19] have also shown that the permanent deformation that occurs as a result of electrostatic and van der Waals interactions leads to a reduction in the stored strain energy of the system. Since this stored energy is recovered upon removal of an elastic particle, the plasticity serves to enhance the apparent adhesion by reducing the elastic energy that can be recovered during particle removal. Analyses of this type are difficult to perform for the commercially-important irregular geometries, hence, the benefit of finite element methods. In this paper, we use finite element methods on simple geometries to establish the overall approach. In subsequent papers, it is our intent to use more complex geometries and surface textures that more closely model the interaction of irregularly-shaped particles and particles with surface addenda.

In order to lend credibility to the finite element approach to the study of adhesion from a mechanistic point of view, the present paper takes a critical approach by examining the implications of the use of an effective surface energy. This approach has two direct benefits. First, it allows an independent assessment of the assumptions and analytical procedures of the JKR theory by computing the strain energies associated with the deformation process by an independent means. Second, it allows us to examine the influence of changes in how the loads are applied, such as the use of body forces rather than

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equipollent loading in the far field. This is of particular significance when the results are to be compared with particle experiments where forces are applied *via* centrifugation. On a more far-reaching scale, this approach provides a mechanism to apply the JKR formalism to particles of irregular geometry and particles where large scale deformations can occur. In addition, the effect of additional long-range interactions on particle adhesion can be determined. Such long-range interactions are not allowed in the JKR formalism, which assumes that all interactions occur strictly within the area of contact.

ANALYTICAL BACKGROUND

In the field of particle adhesion, the dominant theory is that first proposed in 1971 by Johnson, Kendall and Roberts (JKR) [11]. It has withstood the test of time and has gained wide acceptance, having been found to predict adhesional behavior accurately under a variety of circumstances [20]. Earlier theoretical works by Derjaguin *et al.* (DMT) [21], while providing similar phenomenology, do not represent the experimental data as well and are drifting out of favor. Subsequent work that expands on the JKR theory by incorporating plasticity, such as that of Maugis and Pollock (MP) [22], or that allow deformations of larger degree [23, 24], have yet to gain the degree of overall acceptance of the JKR theory. There are numerous works which leverage the initial application of the energy balance between elastic strain energy and surface energy first proposed by Johnson *et al.* [11] and the interested reader is referred to broader reviews in the area of contact mechanics [25].

Beautiful in its simplicity, the JKR formalism takes a continuum mechanical approach to describing the adhesion of particles to other particles and to substrates. Specifically, the JKR theory calculates an equilibrium contact radius, a, from the sum of the elastically stored energy, the energy associated with the surface forces, and the mechanical potential energy of the externally-applied load. The resulting relationship is given by

$$a^3 = \frac{R}{K}\left\{P + 3w_A\pi R + \left[6w_A\pi RP + (3w_A\pi R)^2\right]^{1/2}\right\} \qquad (1)$$

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where R, P, and w_A represent the particle radius, the applied force on the particle, and the work of adhesion between the particle and substrate, respectively, and K is an effective stiffness related to the Young's moduli and Poisson's ratios of the system. For the case where one material is considered rigid relative to the other, the effective stiffness is dominated by the modulus and Poisson's ratio of the more compliant material. Consequently, from the experimental point of view, a hard spherical particle with an assortment of compliant substrates provides the same equilibrium contact patch dimensions as the more difficult-to-fabricate assortment of compliant spherical particles on rigid substrates.

Examining the above expression provides specific relations between contact and particle radii and the applied forces. For example, if $P = 0$, the zero load contact radius is given by

$$a_0 = \left[\frac{9w_A\pi(1-\nu^2)}{2E}\right]^{1/3} R^{2/3}. \tag{2}$$

When $P > 0$ so that the particle is pressed into the surface, a increases. Alternatively, when $P < 0$, so that the load is pulling the particle from the surface, a decreases. Johnson *et al.* [11] have argued that, because the solutions to the JKR equation must be real, separation must occur when

$$P = -\frac{3}{2}w_A\pi R, \tag{3}$$

which corresponds to a finite contact radius at separation given by $a_s \approx 0.63a_0$. There are no constant load solutions for $a < 0.63a_0$.

Implicit in the JKR formalism are several basic assumptions, including that of small strains, as required by the analytical methods, and the assumption that all interactions occur within the zone of contact. This latter assumption is a consequence of the use of a surface energy and suggests that the forces resulting from changes in surface energy must make themselves felt near to the points or areas of contact. However, the requirement that interactions vanish at the circumference of the contact zone implies an infinite stress at that location, resulting in locally large strains. The implications of these assumptions have been discussed in detail by Tabor [26]. The assumption regarding force location is not consistent with the action-at-a-distance concept usually associated with electrostatic and electrodynamic interactions.

The stress singularity that occurs at the edge of the contact patch has been shown to have the same general properties as the singularity at the tip a sharp crack, leading to the application of fracture mechanics [27] to the problem of adhesion. This allows a substantial expertise in the area of fracture mechanics to be applied to the adhesion problem and the interested reader is advised to seek out references in this area as well [28 – 30]. The earliest fracture mechanics work goes back to the energy balance of Griffith [31] and his use of the stress analysis of Inglis [32]. Early in his work, Griffith qualitatively argued that the elastic strain energy liberated by the growth of a crack could be used to balance the work needed to create new surfaces based on thermodynamic arguments. However, he did not publish his theory until he could provide a mathematical justification, which, as it turns out, relied on the work of Inglis [32] published seven years earlier. Although somewhat more difficult to see, the tendency of an adhesive joint to spread by wetting of one surface by another has the same mechanics. The energy liberated by the wetting of a curved surface by a planar surface is used to deform both bodies until the rate of energy liberation by advancement of the contact area just balances the rate of storage of energy in the strain fields of the bodies. When external loads are applied to separate the adherends, the problem becomes a fracture problem. In the area of fracture mechanics, it is well known that surface energy alone does not control the macroscopic fracture behavior. Rather, an effective surface energy that is many times the actual surface energy is needed to rationalize and explain the observed behavior. The reason for the additional energy is that far-field deformations occur during the growth of a crack. This deformation consumes energy far in excess of the intrinsic surface energy and dominates the overall fracture behavior, particularly when plastic deformation occurs. Even so, however, a simple scalar number is used to characterize fracture behavior. This is called the critical crack driving force or the critical stress intensity but, in the final analysis, it is equivalent to an effective surface energy. With the same physics controlling the removal of particles, the critical surface energy per unit distance of crack advance will exceed that attributable to surface energies by substantial margins.

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Since effective surface energies control both fracture and adhesion, then particle adhesion can be thought of as a fracture mechanics problem with specific geometric features that enable stresses to be developed without external loads. The question of whether or not a particle adheres to a surface can be discussed in terms of whether or not it is energetically favorable for a crack to propagate along the particle-substrate interface when loads are applied. In that sense, a change in the JKR contact patch size with applied load is merely a manifestation of the Griffith's criterion. The JKR separation condition expresses a loss of stability where a decrease in contact patch size reduces the overall system energy for all loading conditions.

Although the JKR model does accurately describe numerous aspects of particle adhesion, it also raises many questions. For example, why does a model that assumes small strains work despite the fact that the strains can be quite large near the edge of the contact zone? Why does separation of the particle from the substrate occur at a specific finite radius? Are there differences in the stresses (and, therefore, the removal forces) if the load encompasses the entire particle (as would be exerted by an ultracentrifuge), as opposed to a point (*e.g.*, the particle attached to an AFM cantilever). In this study, preliminary results addressing these questions using finite element modeling are presented. The successes of these methods allow us to look forward to the use of the FEM to answer questions regarding what happens when the materials respond either plastically or hyperelastically.

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COMPUTATIONAL DETAILS

The JKR formalism of particle adhesion was modeled as a large-strain, linear-elasticity problem using ANSYS 5.3, a commercial finite element modeling package. In this modeling, a particle, having an elastic modulus in the range of 5 MPa and a Poisson's ratio of 0.495, was adhered to a rigid substrate for a specific contact radius by judicious application of boundary conditions normal to the surface of the substrate. Situations with larger and smaller contact radii were modeled by changing the number of nodes that were required to be in normal contact with the rigid surface. Motion at the constrained nodes in the plane of the rigid surface was allowed, providing for lateral expansion and contraction of the two contacting surfaces. It was assumed, and subsequently verified, that the stresses would be localized within the half of the particle actually nearest the substrate, thereby allowing us to concentrate the nodes of the finite element package within that region. Computations were performed with arbitrary dimensions, thereby enabling the results to be scaled for any size particle.

Each calculation began by applying boundary conditions to the undeformed axisymmetric representation of the lower half of the particle. Displacement boundary conditions pulled the nodes on the surface of the spherical particle down into a common plane representing the rigid substrate while not restricting radial motions. Body force boundary conditions were applied as accelerations corresponding to specific numbers of "g" forces as would occur in a centrifuge. The boundary conditions were ramped in 10 substeps with converged intermediate solutions using the large deformation (non-linear geometry) option. Output files were queried for stress tensors, strain tensors, total loads at the displacement boundary conditions, total strain energy of the particle, radius of the contact patch, and motion of the center of the particle. Motion of the center of mass was computed from the strained locations of the nodes.

By varying the specific nodes bonded to the surface and the magnitude of the body force, the total energy of the system is computed as a function of the equilibrium contact patch dimension. Following the JKR model, this total energy was obtained as a sum of the strain energy of the system, the surface energy of the system, and the mechanical potential of the exterior forces.

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RESULTS

Figure 1 shows a plot of the total energy (assuming no externally-applied load) as a function of the diameter of the contact patch for a 5 μm radius particle in contact with the rigid substrate (assuming $w_A = 0.17\,\mathrm{J/m^2}$ and $E = 4\,\mathrm{MPa}$). The equilibrium contact radius, as determined by the location of the minimum in the total energy, is found to be at $a = 2.0\,\mu\mathrm{m}$. Experimental data by Rimai and DeMejo [20] report a contact radius for similar-size glass particles on a

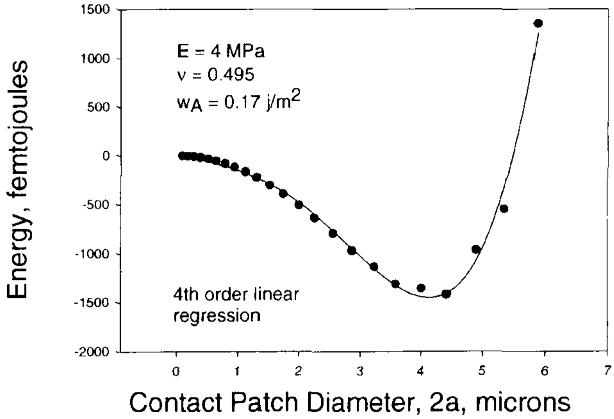

FIGURE 1 Energy as a function of contact patch diameter for a spherical particle on a rigid substrate. Work of adhesion between particle and substrate is shown along with properties of the spherical particle.

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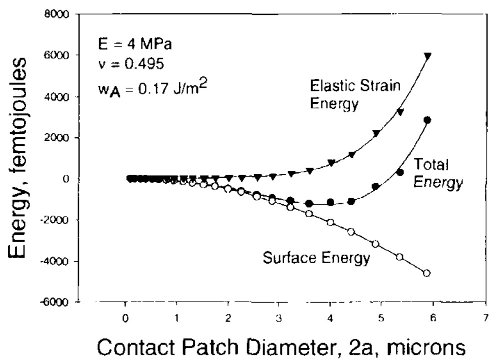

FIGURE 2 Energy as a function of contact-patch size showing surface energy and elastic energy contributions compared with total system energy.

polyurethane substrate ($E \approx 4\,\text{MPa}$) of approximately $2\,\mu\text{m}$, in good agreement with the finite element results.

Figure 2 presents the elastic strain energy and the surface energy contributions to the total energy as a function of contact patch diameter for the same conditions of $E = 4\,\text{MPa}$ and $w_A = 0.17\,\text{J/m}^2$ discussed above. The total energy is shown as well. It is clear from Figure 2 that the reason for the contact-patch-size-dependent minimum in energy, and, thus, a stable value of the contact patch, is the competition between the decrease in energy due to the surface wetting and the increase in system energy stored in elastic strain.

As previously indicated, the JKR model predicts that particle-substrate separation occurs during the application of a negative load when the contact radius decreases to approximately $0.63a_0$. The behavior obtained with finite element modeling suggests a similar behavior, which is illustrated in Figure 3. Here we note that the negative of the slope of the energy *vs.* contact patch size represents a generalized restoring force akin to the crack-driving force in fracture mechanics. The positive slope shown in Figure 3 for increases in contact patch size represents a restoring force that is negative, characterizing a compression in the contact zone, that resists displacing the system to larger contact sizes. That is, the system resists an increase in contact patch size due to compressive loads because the energy of the system rises when the strain energy contributions exceed the energy reduction associated with the surface energy term.

When attempts are made to pull the particle off and, thus, decrease the contact patch size, Figure 3 suggests a local tension that increases at first but then reaches a maximum value at $\sim 0.63a_0$. With further decreases in contact radius, the derivative, or generalized restoring force, decreases smoothly from its maximum to zero. This implies that the restoring force exerted by the substrate on the particle, as a result

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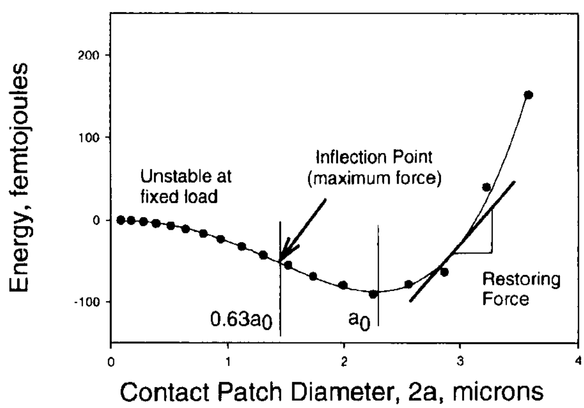

FIGURE 3 Energy *vs.* contact-patch diameter illustrating the generalized restoring force that results from excursions in contact-patch size. The instability at $\sim 0.63a_0$ is shown with an arrow.

of the balance between surface energy reduction and elastic strain energy increase, is a smooth continuous function with a local maximum. Naturally, this translates into a force normal to the substrate surface during various stages of removal that reaches a maximum and then decreases to zero if the particle is not allowed to respond to the forces, a situation akin to displacement control. In load control, however, the system has no solutions for values of $a < 0.63a_0$. The JKR model appeals to the need for a positive real value of the square root in Eq. (1), that is, a real solution, to explain why no solutions are available for $a < 0.63a_0$ and why the particle must detach. Here it is seen that, for load control, the restoring force that is generated by the deflected shape in terms of strain and surface energy contributions is everywhere less than the force applied to get it into this position, leading to an unstable separation of the particle from the surface.

Whereas the schematic representations of the energy *vs.* contact-patch size behaviors examined thus far help us conceptualize the physics that generate the restoring forces, any energy contributions associated with applied loads must be included as well. This energy due to loading is the potential energy of the load and it represents the ability of the applied loads to do work if and when the system deforms to a new geometry. Figure 4 shows the total energy of the particle-substrate system with the inclusion of the mechanical

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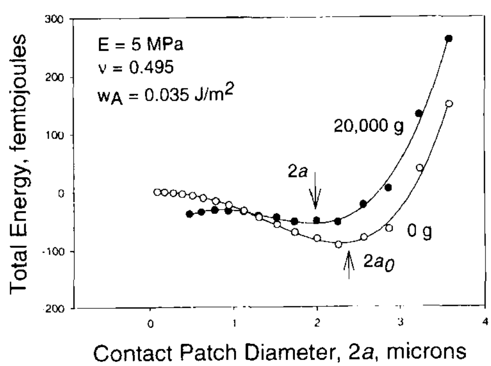

FIGURE 4 Comparison of total system energies at 0 g and 20,000 g accelerations, including the potential energy of the mechanical forces due to acceleration. Note the work of adhesion is about five times smaller, leading to a smaller value of a_0.

potential energy of an exterior body force. This represents a load-control situation where a centrifuge is used to apply tensile loading to the particle by applying 20,000 g of acceleration. In these calculations, the interaction energy is substantially smaller because we have changed the parameters E and w_A to match physical experiments where particle removal can be achieved by centrifugation. The physical parameters of the 5-micron particle are given in Figure 4, which illustrates that the minimum energy position has shifted. As shown in Figure 4, increasing the loading to the equivalent of 20,000 g causes the energy curve to tilt. This causes the local minimum to shift to the left, leading to a smaller equilibrium contact patch, designated by the symbol $2a$ as compared with the initial contact patch size of $2a_0$.

Figure 5 shows the individual contributions to the total system energy. As before, the elastic strain energy increases with contact-patch size and the surface energy contribution decreases with contact-patch size. In addition, there is a mechanical potential energy term resulting from the acceleration. This mechanical potential energy of

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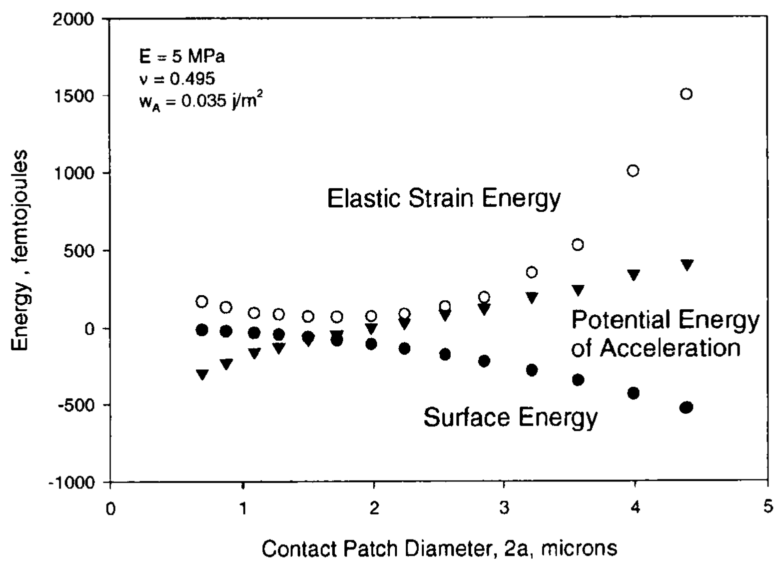

FIGURE 5 Energy components for the system shown in Figure 4. Note that the potential energy of the acceleration is nearly linear with contact-patch size. See text for implications of this linearity. The applied force is $P = -1.284\,\mu N$ where negative sign indicates tension.

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the body force caused by acceleration increases approximately linearly with contact-patch size. Referring once again to Figure 4, if one imagines a marble settling into the local energy minimum like a ball in a bowl, as the mechanical potential is applied, the linearity makes the bowl appear to tilt. Larger values cause even more tilt until a point is reached where the marble rolls out of the local minimum and down the energy curve. At this point, there is a critical value of the contact-patch size which is unstable against further displacements to smaller contact-patch sizes. This is the physical reason why there are no load solutions for contact patches sizes less then $\sim 0.63a_0$. It is energetically favorable for the system to separate and there is no local minimum in energy.

Conceptualizing the problem in this way, it can also be observed that there is a local energy maximum created by the mechanical potential term. The change in energy from the local minimum to the local maximum is a measure of the energy which must be supplied to remove the particle while it is being centrifuged at 20,000 g. It provides a measure of the stability of the system against fluctuations in the critical parameters that control adhesion. To remove the particle, the acceleration (or other applied forces) must be increased until the energy change needed to decrease the contact size either vanishes or can be supplied by random motion of the particle, perhaps as available from Brownian motion of the surrounding medium.

Figures 6a–d show a series of plots of total energy *vs.* contact patch size for accelerations of 0, 5000 g, 20,000 g and 50,000 g, respectively. The four graphs of Figure 6 are on the same scale for ease of comparison. As the force due to the acceleration is increased, the graphs tilt progressively in a way that produces smaller contact patch sizes and decreased stability. At the largest acceleration of 50,000 g, the lowest energy position has clearly slid off the graph to the left indicating a zero contact-patch size is appropriate.

Figure 7 shows a parametric plot of the total energy expressions given in the original JKR theory development [11] as a function of P_c, the critical load for particle removal sometimes referred to as the pull-off, separation, or detachment force. At $P=0$, the curve shows a distinct minimum at $2a_0$ and, as expected, gradually changes shape until, at $P=1.2P_c$, a curve is generated that has a negative slope throughout, comparable with Figure 6d. Unexpectedly, the case for $P=1.0P_c$ clearly shows a negative slope at all values of contact-patch

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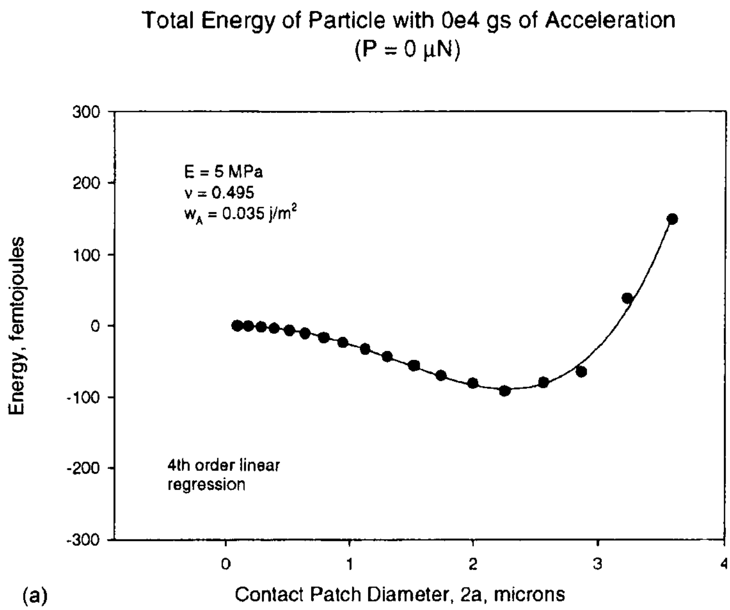
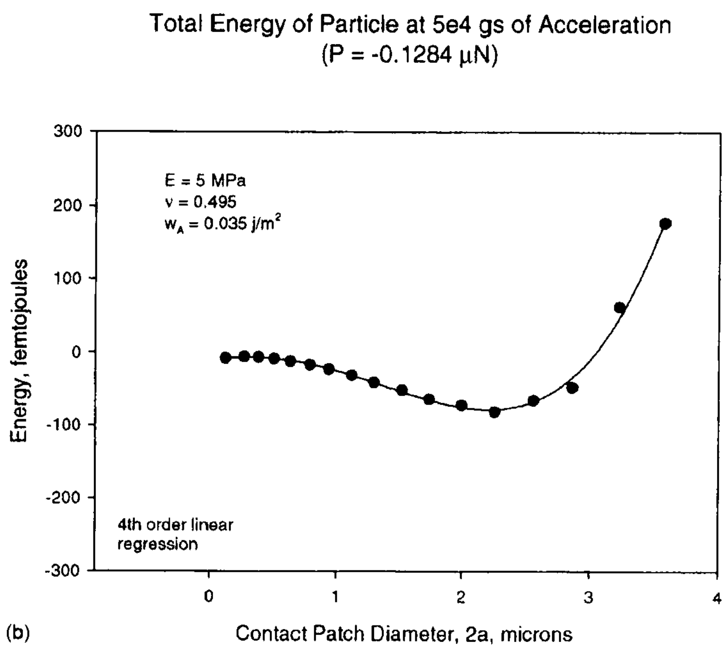
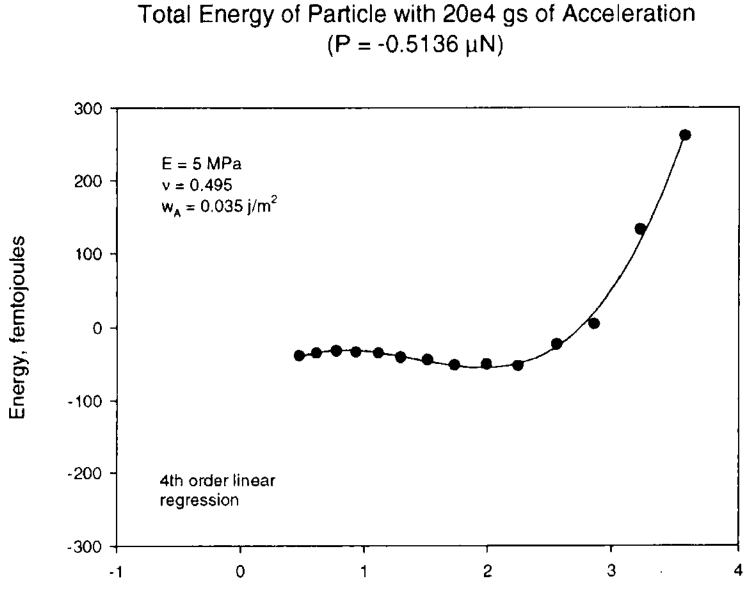


(a)

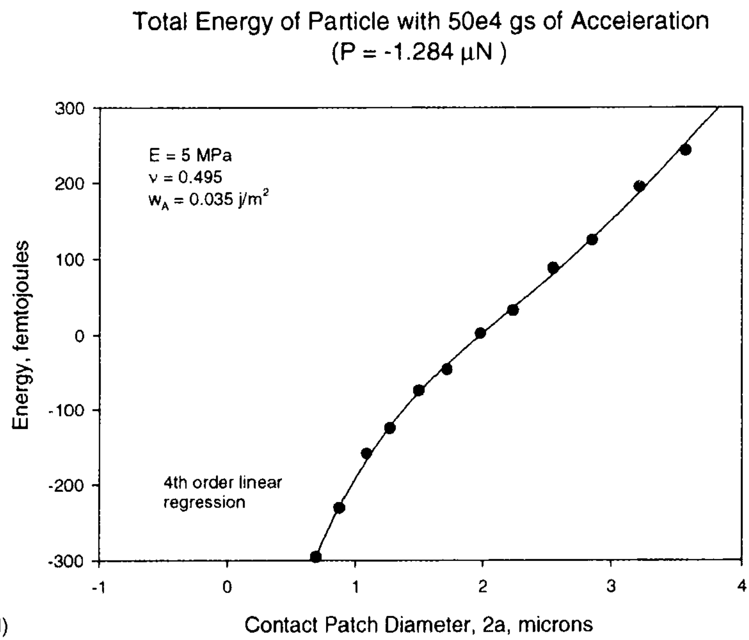


(b)

FIGURE 6 Comparison of energy *vs.* contact patch size for various applied accelerations that might be applied in an ultra-centrifuge. (a) 0 g; (b) 5,000 g; (c) 20,000 g; (d) 50,000 g. Note that the energy minimum shifts left and disappears as the acceleration increases.



(c)



(d)

FIGURE 6 (Continued).

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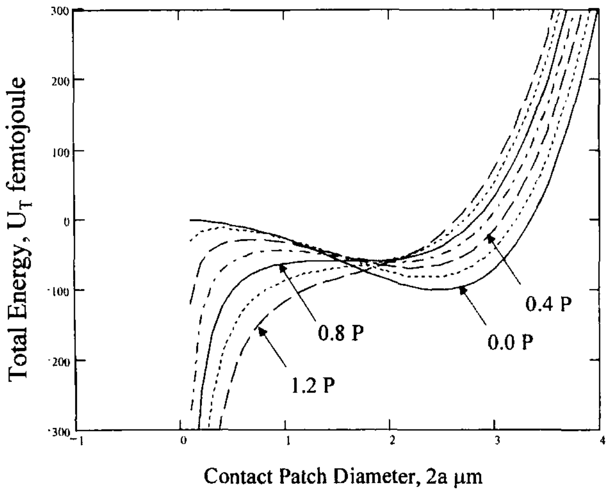

FIGURE 7 Parametric plots of total energy expressions from JKR theory development [11] as a function of the critical load needed for particle removal, P_c.

size, rather than the level behavior anticipated. The level behavior seems to occur at smaller values closer to $0.8P_c$ instead. The present authors do not have an explanation for this observation. Overall, however, the results of the finite element analysis and the results obtained analytically by Johnson, Kendall and Roberts [11] are in reasonable qualitative agreement in light of the differences in loading methods used in the two analyses.

In addition to the contact radii and energies, the stresses and strains were also calculated using finite element modeling. The results are very similar to the analytical results obtained from Maugis and Pollock [19], suggesting that the finite element analysis is capable of solving problems of this type in a reliable fashion. The method may now be applied to irregular geometries, hollow particles, biological cell structures, particles with surface roughness, particles with coatings and property gradients, and particles with distinct surface charges.

CONCLUSIONS

Many features and assumptions in the JKR adhesion formalism can be modeled and tested using finite element methods. Finite element

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analysis is able to illustrate the physics behind the predictions of the JKR theory, and helps shed light on the implications of the assumptions used in that theory. Further work into topics such as the consequences of the large stresses encountered at the contact zone, as well as the effects of yielding and hyperelasticity, is envisioned. This method also allows for the possibility of examining the consequences of assuming the existence of surface energy in other cases where analytical complexity precludes traditional analysis methods, such as irregular geometries, geometries with gradients in properties, or situations where localized space charges cause electrostatic effects.

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