Adhesive depth-sensing indentation tests: Slopes of the force - displacement curves.

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Abstract

Depth-sensing indentation experiments are a very important tool for estimating mechanical properties of modern materials. A review of various aspects of contact problems that used as the theoretical basis for interpretation of depth-sensing nanoindentation experiments is presented. Usually, analytical treatment of the indentation tests is based on analysis of the slopes of the force-displacement curves according to the the non-adhesive Hertz contact theory. However, molecular adhesion is crucially important for many physical processes at the micro/nano-scales. Here, depth-sensing indentation techniques are reviewed and analyzed using the recent results obtained for adhesive contact problems. Fundamental relations for adhesive nanoindentation tests are derived for both frictionless and no-slip boundary conditions within the contact region. It is argued that the adhesive effects may be very important for treatment of the nanoindentation tests because the slopes of the force - displacement curves may considerably differ from the slopes derived using the non-adhesive contact theory.

Keywords: adhesive contact, depth-sensing indentation, evaluation of elastic modulus

1. Introduction

Depth-sensing indentation (DSI) techniques are widely used for estimating mechanical properties of small or thin samples of materials. DSI means that the force-displacement ($P - \delta$) curve is continuously monitored where P is the load applied to a probe (indenter) and δ is the displacement of a rigid indenter or the approach between the distant points of the deformable probe and the sample. If the maximum depth of indentation is below the micrometre scale, then the DSI techniques are referred to as depth-sensing nanoindentation. The DSI techniques were introduced by Kalei in his PhD thesis [45] prepared under the supervision of M.M. Khrushchov. Currently, the mechanical properties of many materials are extracted mainly from the DSI experiments that involve devices having nominally sharp pyramidal indenters. The DSI techniques are quite popular because they allow the researchers to estimate the reduced elastic (contact) modulus of contact pair of materials using the slopes of the unloading branches of the $P - \delta$ curves using the so-called BASh (Bulychev-Alekhin-Shorshorov) formula [24, 25]) or its modifications (see, e.g. a very popular method introduced by Oliver and Pharr [57]). However, the DSI techniques that are commonly used were developed to study mechanical characteristics relatively hard elastic-plastic materials. Indeed, the force-displacement curves obtained by the use of sharp indenters have an elastic-plastic behaviour at the loading branch and it is assumed that the unloading branch is purely elastic. The evaluation of elastic moduli is based on analysis of the slopes of the force-displacement curves according to the the non-adhesive Hertz contact theory. Due to popularity of such DSI techniques, there are attempts to extend the areas of their application. For example, the DSI by sharp indenters were applied to coals [3, 33, 51], biological materials (see references in [42]), and polymers [66]. The DSI techniques that use of sharp indenters, have both advantages and drawbacks (see discussions [21, 28, 38]). Here we concentrate on one of these drawbacks: the BASh formula neglects the molecular adhesion, while forces of molecular adhesion may be very important at the nanometer scale [48]. It is especially important for soft materials.

Even in the pioneering paper by Kalei [46] it was mentioned that the adhesion may influence the experimental results. Nevertheless, the common analytical treatment of the force-displacement indentation curves ignores the adhesion between contacting solids (see, e.g. discussions in [20, 8]). The compressed loads are considered in contact mechanics as positive and respectively the tensile forces are negative. Note that in materials science community, the depth of indentation is traditionally denoted not by δ but by h. Because common analysis of DSI experiments is based on the non-adhesive contact theory, the force-displacement curves of nanoindentation tests are usually recorded only for positive (compressive) loads. However, the nanoscale may be characterized as the 'sticky universe' [48], and the DSI tests employing Atomic Force Microscopes (AFM) demonstrate clearly the adhesive part of $P - \delta$ curves, i.e. the tested material sticks to the AFM probe and separates from it only at some negative (tensile) critical load P_c. Because contact probing of soft and biological materials is a very timely research field (see e.g. [42, 49, 56, 63]), it is important to consider molecular adhesion between the probe and the tested materials. Here, the indentation contact mechanics is reviewed for various interfacial conditions: frictionless, no-slip and with molecular adhesion. The theoretical analysis of adhesive DSI experiments is novel. It is argued that the influence of molecular adhesion on slopes of the force - displacement curves may be quite significant.

2. Preliminaries.

We will consider the contact between a convex indenter and a flat elastic sample. If it is assumed that there is no adhesive forces, then the indenter and the sample contact initially at one point. Both Cartesian $x_1 = x, x_2 = y, x_3 = z$ and cylindrical r, ϕ, z, coordinate frames, where $r = \sqrt{x^2 + y^2}$ and $x = r\cos\phi, y = r\sin\phi$, will be employed. The the point of initial contact between the indenter and the half-space $x_3 \geq 0$ is taken as the origin (O) of at coordinate systems.

2.1. Frictionless Hertz-type contact problems.

Originally, H. Hertz [41] considered three-dimensional ($3D$) problem of frictionless contact between two isotropic, linear elastic solids. The shapes of the solids were approximated as elliptic paraboloids, while the boundary conditions for the contacting solids are formulated as for elastic half-spaces. A list of assumptions of the Hertz contact theory is given by [8].

It is assumed that an indenter is pressed by the force P to a boundary of the sample. After the contact is established, displacements u_i and stresses σ_{ij} are generated. In a geometrically linear formulation of the contact problem, the boundary conditions are formulated on the boundary of the half-space, i.e. on the plane $z = 0$. If an isotropic linear elastic half-space is characterized by the Young's modulus E and the Poisson ratio ν then its contact (reduced) modulus E^* is defined as

$$E^* = \frac{E}{1 - \nu^2}. \tag{1}$$

If the elastic bodies having contact moduli E_1^* and E_2^* respectively are in contact between each other then the Hertz-type contact problem is mathematically equivalent to the problem of contact between an isotropic elastic half-space with contact modulus E^*

$$\frac{1}{E_I^*} = \frac{1}{E_1^*} + \frac{1}{E_2^*} \tag{2}$$

and a rigid body whose effective shape function f is equal to the initial distance between the surfaces, i.e. $f = f_1 + f_2$ where f_1 and f_2 are the shape functions of the solids. Hence, the equation of the axisymmetric indenter surface given by a function f, can be written as $x_3 = -f(r, \phi), \quad f \geq 0$.

We are especially interested in indenters whose shapes are described by monomial (power-law) functions

$$f(r, \phi) = B_d(\phi) r^d, \tag{3}$$

where B_d is the shape function, $[B_d] = L^{1-d}$, $[\cdot]$ denotes the physical dimension of a variable and L denotes physical dimension of length. Note that pyramidal indenters are particular case of power-law shaped indenters, when $d = 1$. The Hertz-type contact problems for power-law shaped indenters are self-similar if the constitutive relations between stresses and strains in the material are homogeneous functions of strains. This property was initially discovered by Galanov [35, 36] for isotropic media. Independently, self-similarity of non-adhesive Hertz-type contact problems for isotropic elastic materials was discovered in [5]. Later it was shown that these contact problems are self-similar even if the solids are anisotropic (see, e.g. [6, 7, 8]).

Further we will consider only the axisymmetric contact problems. This means that nothing depends on ϕ and, hence, (3) for axisymmetric indenters may be written as

$$z = -f(r), \tag{4}$$

and if f is a monomial (power law) function, then

$$z = -B_d r^d, \tag{5}$$

where B_d is the shape constant. Note that the Hertz approximation of a spherical indenter of radius R is a particular case of (5) when $d = 2$ and $B_2 = (2R)^{-1}$.

A cone of semi-vertical angle $\pi/2-\alpha$, is also a power-law shaped indenter, when $d = 1$, and $f(r) = B_1 r$. For a linearized formulation of the Hertz-type problem for a rigid cone, it is required that α is small compared with 1 and, therefore, $B_1 = \tan\alpha \approx \alpha$.

The $P-\delta$ relations for the frictionless Hertz-type contact problems were obtained by Hertz [41] for $d = 2$, Love for [52] for $d = 1$, Shtaerman [62] for $d = 2m$ where m is a natural number, and eventually by Galin [39, 40] for arbitrary body of revolution $f(r)$, in particular for arbitrary d. Note that the Boussinesq solution [23] for a flat-ended cylindrical indenter may be also considered as a solution for a power-law shaped indenter where $d = \infty$. The Galin solutions to the Hertz-type contact problems for arbitrary shaped, indenter of revolution $f(r)$, $f(0) = 0$ are

$$P_H = 2E^* \int_0^a \rho \Delta f(\rho) \sqrt{a^2 - \rho^2} d\rho, \tag{6}$$

$$\delta_H = \int_0^a \rho \Delta f(\rho) \text{arctanh}(\sqrt{1 - \rho^2/a^2}) d\rho, \tag{7}$$

where a is the radius of contact region and Δ denotes the two-dimensional Laplace operator

$$\Delta = \frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2} = \frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r}. \tag{8}$$

Here and henceforth the index H denotes that the value is attributed to the corresponding non-adhesive Hertz-type contact problem.

Rostovtsev [61] derived an alternative form of the Galin expressions (6) and (7)

$$P_H(a) = 2E^* \int_0^a \frac{\rho^2 f'(\rho) d\rho}{\sqrt{a^2 - \rho^2}} \tag{9}$$

and

$$\delta_H(a) = \int_0^a \frac{f'(\rho)}{\sqrt{1 - \rho^2/a^2}} d\rho. \tag{10}$$

In particular, for power-law shaped indenters, the $P - \delta$ relation is

$$P_H = \left[\frac{(E^*)^d}{C(d) B_d}\right]^{\frac{1}{d+1}} \left(\frac{2d}{d+1}\right) \delta_H^{(d+1)/d} \tag{11}$$

where

$$C(d) = \frac{d^2}{d+1} 2^{d-1} \frac{[\Gamma(d/2)]^2}{\Gamma(d)}$$

and Γ is the Euler gamma function.

If one uses the contact radius a as the external characteristic parameter of the Hertz-type contact problem for the power-law shaped indenters, then the corresponding load $P_H(a)$ is defined by the following Galin relations [39]

$$P_H(a) = C(d) E^* B_d a^{(d+1)}, \tag{12}$$

while the displacement $\delta_H(a)$ is given by

$$\delta_H(a) = B_d C(d) \frac{d+1}{2d} a^d. \tag{13}$$

Employing (9) and (10), one can take derivatives dP_H/da and $d\delta_H/da$ and obtain the following expression for slopes S_H of the frictionless Hertz-type contact problems

$$S_H = \frac{dP_H/da}{d\delta_H/da} = \frac{dP_H}{d\delta_H} = 2E^* a. \tag{14}$$

It was suggested to re-write (14) as the following BASh relation [24]

$$S_H = \frac{2\sqrt{A}}{\sqrt{\pi}} E^* \tag{15}$$

where A is the area of the contact region.

Bulychev et al. [25] argued that "*an important practical property of the curve of the elastic unloading of the plastic imprint is the independence of its slope at the initial stage of the unloading from the character of the pressure distribution under the imprint*". They concluded that if (15) is applied not to axisymmetric indenters but to sharp pyramidal indenters, then the error will be small. Nowadays the BASh relation is the the cornerstone of the nanoindentation test interpretations. It was found that the BASh formulae or its modifications provide quite reasonable estimations of the elastic constants of tested materials. It gives also a possibility to do a detailed study of elastic characteristics of components of inhomogeneous materials (see, e.g. [33]). All these features are advantages of the use of sharp indenters in combination with the BASh approach. However, the techniques based on the use of the original form of the BASh relation have several drawbacks ([21, 28, 38]).

2.2. No-slip Hertz-type contact problems.

As it has been mentioned, the derivation of the main formulae of the above models is based on the assumption that the material points within the contact region can move along the punch surface without any friction. However, if one intends to use the Hertz-type problems to study the influence of the molecular adhesion, then it would be more natural to assume that a material point that came to contact with the punch sticks to its surface, i.e. to assume that the non-slip boundary conditions are valid. The Hertz-type problems with no-slip boundary conditions were studied in classic papers by Mossakovskii and Spence [54, 55, 64]. The no-slip condition can be written as

$$\frac{\partial u_r}{\partial P}(r, 0, P) = 0, \quad dP > 0. \tag{16}$$

i.e. the values of the radial displacements u_r within the contact region do not change with augmentation of the external parameter of the problem (the external load). The schematic descriptions of the trajectories of surface material points near and within the contact regions with no-slip condition (16) were presented by Spence [64] for a paraboloid indenter and by Chaudhri [27] for conical indenter.

The compressing normal stresses σ_{zz} in the Boussinesq-Mossakovskii problem, i.e. the stresses under a flat-ended punch of the radius a under (16) condition are

$$\sigma^0_{zz}(r, 0, a) = K\delta_0 \frac{1}{r}\frac{d}{dr} I_M(r). \tag{17}$$

Here δ_0 is the depth of the punch,

$$I_M(r) = \int_0^r \sin\left(\beta \ln \frac{a-x}{a+x}\right) \frac{x}{\sqrt{\rho^2 - x^2}} dx$$

and

$$\beta = \frac{1}{2\pi}\ln(3 - 4\nu), \qquad K = \frac{4E(1-\nu)}{\pi(1-2\nu)(1+\nu)\sqrt{3-4\nu}}.$$

The formula (17) was presented by Mossakovskii [54]. Later its correctness was checked by Keer [47] and Spence [64]. However, the final expression for the integral of the stresses over the contact region was presented by Mossakovskii [54] with a misprint. The correct $P-\delta$ expression for a flat-ended indenter of radius a is

$$P = 2E^* C_{NS} a\delta \tag{18}$$

where

$$C_{NS} = \frac{(1-\nu)\ln(3-4\nu)}{1-2\nu} \tag{19}$$

is the parameter introduced by Borodich and Keer [20] to characterize the no-slip contact problems. They derived also the following $P - \delta$ relation for power-law shaped indenters (5) [21]

$$P_H = E^* C_{NS} \frac{d}{d+1} \left[\frac{4I^*(d)}{B_d} \frac{\Gamma(d)}{[\Gamma(d/2)]^2}\right]^{1/d} \delta_H^{\frac{d+1}{d}} \tag{20}$$

where

$$I^*(d) = \int_0^1 t^{d-1} \cos\left(\beta \ln \frac{1-t}{1+t}\right) dt.$$

The expression (20) can be also written as

$$\delta_H = \left[\frac{B_d C(d)}{dI^*(d)(E^* C_{NS})^d}\right]^{\frac{1}{d+1}} \left(\frac{d+1}{2d}\right) P_H^{d/(d+1)}. \tag{21}$$

If one uses the contact radius a as the external characteristic parameter of the Hertz-type contact problem with no-slip boundary conditions for the power-law shaped indenters, then the corresponding load $P_H(a)$ is defined by the following Borodich-Keer relations [21]

$$P_H(a) = \left(\frac{C(d) C_{NS} E^* B_d}{dI^*(d)}\right) a^{(d+1)}, \tag{22}$$

while the displacement $\delta_H(a)$ is given by

$$\delta_H(a) = B_d C(d) \frac{d+1}{2d^2 I^*(d)} a^d. \tag{23}$$

If the material is incompressible, i.e. $\nu = 0.5$ then $C_{NS} = 1, \beta = 0$, and $I^*(d) = 1/d$. Hence, the Borodich-Keer no-slip relations coincide with the Galin frictionless relations.

Substituting $d = 1$, one obtains the known solution for for conical indenters [64]

$$P = E^* C_{NS} A_C a^2, \quad \delta = A_C a, \quad A_C = \frac{\pi B_1}{2I^*(1)}, \tag{24}$$

where

$$I^*(1) = \frac{\ln(3-4\nu)\sqrt{3-4\nu}}{2(1-2\nu)}. \tag{25}$$

Substituting $d = 2$, one obtains the known solution for for spherical indenters [55] (see a discussion in [21]).

It was also shown [20] that the slopes S_H of the no-slip Hertz-type contact problems for axisymmetric indenters of arbitrary profile are

$$S_H = dP_H/d\delta_H = 2C_{NS} E^* a. \tag{26}$$

It is clear that (26) may be written as $S_H = 2C_{NS} E^* \sqrt{A}/\sqrt{\pi}$, i.e. as a no-slip modification of the BASh relation (15).

3. Adhesive contact problems and DSI

3.1. The Derjaguin ideas and the JKR formalism.

Apparently the first paper on adhesive contact between elastic solids was published by Derjaguin [29]. Derjaguin suggested to take into account deformations of solids in order to estimate the adhesion between them. He suggested also to calculate the total energy of between contacting solids, and he introduced the so-called Derjaguin approximation that is employed explicitly or implicitly in overwhelming majority of modern models of adhesive contact.

Using Derjaguin's idea of calculation of the total energy of between contacting solids, Sperling [65] developed a model of adhesion between elastic spheres that is usually is referred to as the JKR (Johnson-Kendall-Roberts) or JKRS model of adhesive contact. In fact, the model was independently rediscovered in [44] using another approach. According to Kendall [48], they combined Derjaguin's idea of calculation of the total energy of between contacting solids and Johnson's idea of superposition of the Hertz and Boussinesq solutions [43]. This allows to solve adhesive contact problems as a formal combination of solutions to non-adhesive Hertz-type and Boussinesq-type contact problems, is the classical Johnson-Kendall-Roberts formalism [9, 58]. The JKR formalism was used to extend the JKR model and to solve the problem of adhesive contact between the power-law shaped indenters [37, 26].

It was shown that the JKR formalism may be used to solve the problems of adhesive contact for anisotropic solids whose materials have rotational symmetry of their elastic properties [34, 8, 15, 16]. It was also shown that the JKR formalism allows the researchers to estimate the influence of no-slip boundary conditions within the contact region [18, 19, 8]. In fact, it was shown [19] that the difference between no-slip and frictionless solutions is quite small.

Later Borodich [8] reinforced the JKR formalism by the use of the properties (14) and (26) of the slopes S_H for frictionless and no-slip cases respectively. This allowed us to apply the JKR formalism to problems of adhesive contact for arbitrary shaped indenters of revolution $f(r)$, $f(0) = 0$. In particular, for isotropic elastic solids, the frictionless JKR theory leads to the following expressions

$$P = P_H - \sqrt{8\pi w E^* a^3}, \tag{27}$$

and

$$\delta = \delta_H - \sqrt{\frac{2\pi w a}{E^*}} \tag{28}$$

where w is the work of adhesion (the specific energy (per area) or the energy per unit area needed to separate the surfaces from contact to infinity), P_H and δ_H are the solutions (6) and (7) of the appropriate Hertz-type contact problem.

Using the JKR formalism, similar expressions were obtained in problems of adhesive contact for thin stretched membranes [11], for thin [17] and thick [1] layers, for bilayers [32], and for a special case of a toroidal indenter [2]. An accurate and clear formulation and derivation of the 'reinforced' JKR formalism were presented in [58]. In particular, the solutions to the JKR-type adhesive contact problems may be written as the following parametric expressions

$$P = P_H - \sqrt{\frac{4\pi w a S_H^2(a)}{S'_H(a)}}, \tag{29}$$

and

$$\delta = \delta_H(a) - \sqrt{\frac{4\pi w a}{S'_H(a)}}. \tag{30}$$

These expressions unify the previously obtained results (see a discussion in [9]).

3.2. Slopes of the JKR force-displacement curves.

Note that

$$S_{JKR} = \frac{dP}{d\delta} = \frac{dP}{da} / \frac{d\delta}{da}. \tag{31}$$

Therefore, one can calculate the slopes of the adhesive force-displacement curves.

Further consider the slope of the $P - \delta$ curve for the branch where $P \geq P_c$ and $\delta \geq \delta_m$, i.e. $S_{JKR} = dP/d\delta \geq 0$. Here P_c is the force of adhesion, i.e. $dP/d\delta = 0$ at $P = P_c$, and δ_m is the displacement corresponding to this force. In the classic case of contact between a spherical indenter of radius R and a flat surface of a sample, we have

$$\delta_m = -\frac{a_c^2}{3R}, \quad P_c = -\frac{3}{2}\pi w R.$$

The radius of contact region a_c that $P(a_c) = P_c$ is

$$a_c = \left(\frac{9\pi w R^2}{8E^*}\right)^{1/3}. \tag{32}$$

Let us denote

$$A_{ad} = (2\pi w / E^*)^{1/2}$$

and take the derivatives of P and δ using (27) and (28). Then we have

$$\frac{dP}{da} = \frac{dP_H}{da} - 3E^* A_{ad} a^{1/2}$$

and

$$\frac{d\delta}{da} = \frac{d\delta_H}{da} - 0.5 A_{ad} a^{-1/2}$$

Taking into account that

$$\frac{dP_H}{da} = 2E^* a \frac{d\delta_H}{da},$$

we obtain

$$\frac{dP}{d\delta} = 2E^* a \left\{ \frac{d\delta_H/da - 1.5A_{ad}a^{-1/2}}{d\delta_H/da - 0.5A_{ad}a^{-1/2}} \right\}.$$

It follows from the Galin solution (see, e.g. [21]) that

$$\frac{d\delta_H}{da} = \int_0^a \frac{r\Delta f(r)}{\sqrt{a^2 - r^2}} dr,$$

therefore, we obtain the result announced earlier [12]

$$S_{JKR} = 2E^* a \left\{ \frac{\int_0^a \frac{r\Delta f(r)}{\sqrt{a^2-r^2}} dr - 1.5A_{ad}a^{-1/2}}{\int_0^a \frac{r\Delta f(r)}{\sqrt{a^2-r^2}} dr - 0.5A_{ad}a^{-1/2}} \right\}. \quad (33)$$

It can be rewritten as

$$S_{JKR} = 2E^* a \left\{ 1 + \frac{2}{1 - 2a^{1/2}\delta'_H(a)/A_{ad}} \right\}. \quad (34)$$

Evidently, if the work of adhesion $w = 0$ then $A_{ad} = 0$. In this case, (33) or (34) reduced to (14).

3.3. Adhesive indentation of power-law shaped indenters.

It was shown earlier that the indenter shapes near the tip have some deviation from their nominal pyramidal shapes. It is convenient to approximate the shape function of the probe near the tip by a monomial function of radius for both nanoindenters [35, 36, 22, 50] and AFM tips [4].

Note that if the external load $P = 0$, then according to the JKR theory of adhesive contact neither the displacement $\delta(0)$ nor the radius of the contact region $a(0)$ are equal to zero. In the coordinate system (P, δ) , the point $(0, \delta(0))$ belongs to the branch $P \geq P_c$ and $\delta \geq \delta_m$, i.e. $S_{JKR}[0, \delta(0)] \geq 0$.

It is convenient to write the JKR-type relations in dimensionless form using as units some characteristic parameters of the adhesive contact problems. Evidently, the values of characteristic parameters may be chosen rather arbitrary (see a discussion in [19]). Here we will take the radius $a(P)$ of the contact region at $P = 0$ as a characteristic radius a^*. Substituting $P = 0$ into $P(a)$ expression for the case of power-law shaped axisymmetric indenters (5), one can obtain

$$a^* = a(0) = \left[\frac{8\pi w}{E^* C^2(d) B_d^2} \right]^{1/(2d-1)} \quad (35)$$

in the case of frictionless contact, and

$$a^* = a(0) = \left[\sqrt{\frac{8\pi w}{E^* C_{NS}}} \frac{dI^*(d)}{C(d)B_d} \right]^{\frac{2}{2d-1}} \quad (36)$$

in the case of no-slip contact.

Now we need to choose the expressions for other two characteristic parameters P^* and δ^*. For frictionless JKR contact problems, we take

$$P^* = \left\{ \frac{(8\pi w)^{d+1}(E^*)^{d-2}}{[C(d)B_d]^3} \right\}^{\frac{1}{2d-1}} \quad (37)$$

and

$$\delta^* = \left[\frac{2^{d+1}}{C(d)B_d} \left(\frac{\pi w}{E^*} \right)^d \right]^{\frac{1}{2d-1}}. \quad (38)$$

For the non-slip JKR adhesive contact

$$P^* = \left\{ \frac{(8\pi w)^{d+1}(E^* C_{NS})^{d-2}}{[C(d)B_d/(dI^*(d))]^3} \right\}^{\frac{1}{2d-1}}, \quad (39)$$

and

$$\delta^* = \left[\frac{2^{d+1} dI^*(d)}{C(d)B_d} \left(\frac{\pi w}{E^* C_{NS}} \right)^d \right]^{\frac{1}{2d-1}}. \quad (40)$$

The above choices of the characteristic parameters lead to the following form of the dimensionless solutions for the power-law shaped indenters

$$P/P^* = (a/a^*)^{d+1} - (a/a^*)^{3/2} \quad (41)$$

and

$$\frac{\delta}{\delta^*} = \frac{d+1}{d} \left(\frac{a}{a^*} \right)^d - \left(\frac{a}{a^*} \right)^{1/2}. \quad (42)$$

Now we can derive expressions for slopes of the $P-\delta$ curves. Using (31), one obtains

$$S_{JKR} = \frac{P^*}{\delta^* a^*} a \left[1 + \frac{2}{1 - 2(d+1)(a/a^*)^{(2d-1)/2}} \right]. (43)$$

It follows from (35), (38) and (37) that for frictionless JKR problem

$$\frac{P^*}{\delta^* a^*} = 2E^*$$

and it follows from (36), (40) and (39) that for no-slip JKR problem

$$\frac{P^*}{\delta^* a^*} = 2E^* C_{NS}.$$

The classic frictionless JKR problem was solved for a spherical indenter of radius R. For this solution, one can write the expression for S_{JKR} and obtain the result announced earlier [12]

$$S_{JKR} = 2aE^* \left[\frac{1 - 3\sqrt{(\pi R^2 w)/(8E^* a^3)}}{1 - \sqrt{(\pi R^2 w)/(8E^* a^3)}} \right]. \quad (44)$$

or

$$S_{JKR} = 2aE^* \left[1 + \frac{2}{1 - \sqrt{(8E^* a^3)/(\pi R^2 w)}} \right]. \quad (45)$$

On the other hand, for a spherical indenter of radius R, one has $d = 2$, $B_2 = 1/(2R)$, $C(2) = 8/3$, $f(r) = B_2 r^2$. Then $P^* = 6\pi w R$ and other two characteristic parameters are

$$a^* = \left(\frac{9\pi w R^2}{2E^*} \right)^{1/3}, \ \delta^* = \left(\frac{6\pi^2 w^2 R}{(E^*)^2} \right)^{1/3}. \quad (46)$$

It is possible to substitute these values into (43) and write

$$S_{JKR} = 2aE^* \left[1 + \frac{2}{1 - 6(a/a^*)^{3/2}} \right]. \quad (47)$$

One can check the correctness of the expressions (44) and (47). If one substitutes $a = a_c$ as (32) into (44) and (47), then one obtains from both formulae that $S_{JKR} = 0$ as it should be because $P(a_c) = P_c$. In fact, one could derive (45) directly from (34). Indeed, it follows from (23) for $d = 2$ that $\delta_H(a) = a^2/R$ and $\delta'_H(a) = 2a/R$, while $A_{ad}^{-1} = (2\pi w/E^*)^{-1/2}$. Substituting these values into (34), one obtains (45).

Hence, neglecting of the adhesion will produce the following error E_{er}

$$E_{er} = |(S_{JKR} - S_H)/S_H| = \left| \frac{2}{1 - 6(a/a^*)^{3/2}} \right| \quad (48)$$

where S_H is the slope of the $P - \delta$ curve of the Hertz-type contact problems defined by (14). Evidently, that at the moment of attachment, i.e. if $a = a^*$, one has $E_{er} = 40\%$.

As an example, let us consider polyvinylsiloxane (PVS) material. Its exact characteristics depend on many factors that may vary in process of preparation of the sample. However, we may take as typical values of the contact modulus and work of adhesion the following values $E^* = 4$ MPa and $w = 0.1$ N/m extracted by our method [10] (see, for details [13, 14, 60, 59]). One can take as a typical radius of modern AFM spherical probe $R = 4\mu m$. Then for $a = 2\mu m$, one has $\delta_H = 1\mu m$ and

$$E_{er} = \left| \frac{2}{1 - 6(2/1.78)^{3/2}} \right| \approx 32.5\%.$$

Thus, one can see that for soft materials, there is a quite significant difference between the slopes of the non-adhesive force-displacement curve S_H and the adhesive JKR curve S_{JKR}. Because the current approaches to DSI tests are based on the use of the slopes to experimental $P - \delta$ curves, the ignorance of the adhesion may lead to a significant errors in calculations of elastic characteristics of materials.

4. Conclusion.

A review of various aspects of contact problems that are used as the theoretical basis for interpretation of modern depth-sensing nanoindentation techniques is presented. It is argued that the common approaches to DSI experiments are based on the use of the non-adhesive Hertz contact theory, while it is important to consider molecular adhesion between the probe and the tested materials. The DSI curves have been analyzed using the recent results obtained for adhesive contact problems. Although the studies were based on the use of the reinforced JKR formalism, the theory cover almost all practically interesting cases for soft materials [9].

The general expressions for slopes of the force-displacement curves have been derived for both frictionless and no slip boundary conditions within the contact region. It has been shown that the adhesive effects may be very important for treatment of the nanoindentation tests. Indeed, the difference between the slopes of the non-adhesive force-displacement curves and the adhesive JKR curves may be quite significant.

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