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Indentation Load-Displacement Relations for the Spherical Indentation of Elastic Film/Substrate Structures

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Abstract: The spherical indentation of elastic film /substrate structures is analyzed using the finite element method. The load-displacement curves of the film /substrate structures of various configurations are obtained and analyzed. A generalized power law relation is established, which can be used to analyze the load-displacement curve of elastic film /substrate systems under spherical indentations. The indentation load is dependent on the modulus ratio of the film to the substrate and film thickness. A semi-analytical expression for the power of the power law relation is also obtained as a function of the normalized film thickness and normalized film modulus, which can be used to determine the modulus of the film from a spherical indentation test.

Keywords: Nanoindentation, thin film, finite element method.

1 Introduction

Layered materials (film /substrate systems) have been increasingly used in microelectronics, optoelectronics and as protective coatings of engineering structures. This has promoted the study of the mechanical behavior of layered structures for improving the structural integrity and stability. The low-load/low-depth indentation, or nano-indentation, has become an attractive tool to characterize the mechanical properties of films and multi layers. When a thin film is deposited on a substrate, the deformation and stress field in the layered structure becomes much complicate due to the presence of the substrate. The classical Hertz contact theorem is no longer valid in describing the load-depth curve for the indentation of a layered material. Considerable efforts have been devoted to analyzing the indentation of film-substrate systems through the methods of experimental testing, analytical modeling, and finite element analysis in order to understand the mechanical behavior of the layered structures.

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The early experimental work on the indentation of thin films may be due to Waters (Waters, 1965), who indented rubber sheets adhered to glass substrates using spherical balls of various sizes (spherical indentation). The indentation displacements were recorded with a standard dial gauge and then plotted as a function film thickness. A master curve was found for the relation between the indentation displacement and film thickness, which was fitted with an empirical equation. Substantial amount of experimental work has been conducted recently on the indentation of thin films using various shaped indenters (King, 1987; Mencik et al., 1997; Korsunsky, et al., 1998; Tsui and Pharr, 1999; Geng et al., 2008; Geng et al., 2007; Geng et al., 2005). It is generally believed that the indentation depth should be less than one tenth of the film thickness to avoid the substrate effects. Several analytical approaches have been developed for characterizing the indentation of thin film-substrate systems (Harding and Sneddon, 1945; Spence, 1968; Johnson, 1985; Yu et al., 1990; Gao et al., 1992; Yang, 1998; Hsueh and Miranda, 2004; Yang, 2006; Yang, 2003). These include the uses of functional invariants, the Wiener–Hopf method, integral transforms, and complex variables methods, among others.

The finite element method (FEM) has recently been used to analyze the indentation of film /substrate structures. Djabella and Arnell used the finite element method to investigate the contact stresses due to Hertzian indentation of coating/substrate systems consisting of hard coatings on a relatively low modulus substrate (Djabella and Arnell, 1992). The effect of film thickness on the stress magnitudes was examined by Page and Hainsworth by modeling the Zirconia-stainless steel couple (Page and Hainsworth, 1993). Cai and Bangert performed the finite element modeling of several coating/substrate systems and examined the effect of critical ratio of indentation depth to film thickness on the hardness measurement (Cai and Bangert, 1995). The substrate effects and the critical penetration depth in film /substrate systems were further studied by a number of researchers (Chen and Vlassak, 2001; Lichinchi, et al., 1998; Sun, et al., 1995; Xu and Rowcliffe, 2004). The roundness or truncation of the indenter tip is crucial for the measurement of thin films using nanoindentation, based on the finite element analysis conducted by Tang and Arnell (Tang and Arnell, 1999).

Most of the experimental/analytical studies have focused on a limited number of film-substrate systems. A solution to the indentation problem of all possible film /substrate configurations remains yet to be developed. Although the finite element analysis may have the ability to explore a large range of film-substrate systems, most work as reported has been focused on the simulations of indentation processes and stress distributions and fewer has been focused on the characterizations of load-displacement curves. The purpose of this work is to use the finite element method to analyze the indentation of elastic film /substrate systems of a wide range of configurations and then establish a generalized power-law relation to describe the indentation load-displacement responses.

2 Boundary Value Problem

Consider the indentation of an elastic thin film adhered to an elastic substrate by a rigid spherical indenter (Figure 1). Without any body force, the mechanical equilibrium conditions are assumed valid during indentation,

$$\frac{\partial \sigma_{ij}^{(m)}}{\partial x_j} = 0 \tag{1}$$

where σ_{ij} $(i,\ j = r,\ \theta,\ z)$ are the components of the stress tensor, x_i are the components of the position vector of a material point, and the superscript (m) $(m = 1,\ 2)$ represents the film $(m = 1)$ and the substrate $m = 2$, respectively.

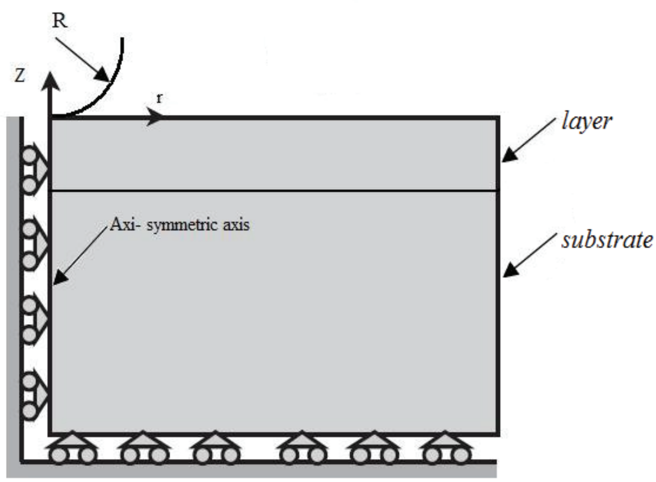


Figure 1: Schematic of the axisymmetric indentation of a semi-infinite film-substrate material with a rigid spherical indenter.

As a rigid, spherical indenter is pressed onto the surface of a semi-infinite elastic material. The contact boundary conditions in a cylindrical coordinate $(r,\ \theta,\ z)$ are

$$\sigma_{rz}^{(1)}(r,0) = 0 \text{ for } r < a \tag{2}$$

$$u_z^{(1)}(r,0) = f(r) - \delta \text{ for } r < a \tag{3}$$

where $u_z^{(1)}$ is the displacement component in the film along the loading direction, $f(r)$ is the surface profile of the indenter tip, δ is the displacement of the indenter, and a is the radius of the contact area to be determined in the simulation. Equation (2) represents the condition of frictionless contact. Outside the contact area, the surface is at stress-free state, i.e.

$$\sigma_{rz}^{(1)}(r,0) = \sigma_{zz}^{(1)}(r,0) = 0 \text{ for } r > a \tag{4}$$

The far field condition requires, $\sigma_{rz}^{(1)}(r,z) \to 0$, $\sigma_{zz}^{(1)}(r,z) \to 0$, $u_r^{(1)}(r,z) \to 0$, and $u_z^{(1)}(r,z) \to 0$ as $r \to \infty$.

At the film-substrate interface, the continuities in the displacements and stresses are required

$$u_r^{(1)}(r,-h) = u_r^{(2)}(r,-h) \text{ and } u_z^{(1)}(r,-h) = u_z^{(2)}(r,-h) \tag{5}$$

$$\sigma_{rz}^{(1)}(r,-h) = \sigma_{rz}^{(2)}(r,-h) \text{ and } \sigma_{zz}^{(1)}(r,-h) = \sigma_{zz}^{(2)}(r,-h) \tag{6}$$

where h is the thickness of the film.

The indentation load applied to the indenter can be calculated as

$$F = -2\pi \int_0^a \sigma_{zz}^{(1)}(r,0) r dr \tag{7}$$

For a spherical indentation of homogeneous, elastic half-space, the relationship between the indentation load and the indentation depth is (Hertz, 1882)

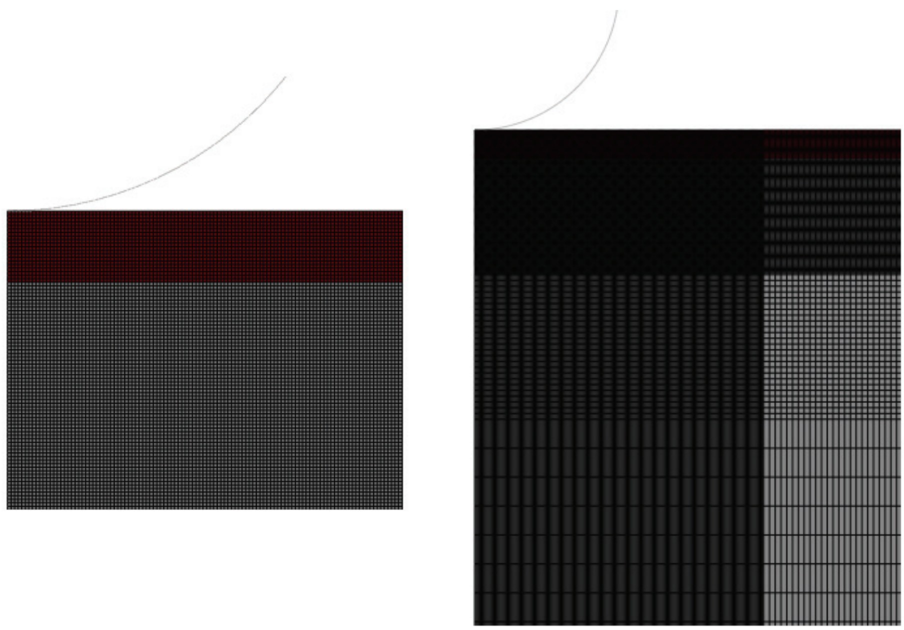
$$F = \frac{4}{3} \frac{E\sqrt{R}}{1-\nu^2} \delta^{3/2} \tag{8}$$

where R is the radius of the spherical indenter, and E and ν are the Young's modulus and Poisson ratio of the material. Equation (8) indicates that the indentation load is a power function of the indentation depth with a power of 1.5 for the indentation of homogeneous materials.

3 Numerical Simulation

The finite element simulations of the spherical indentation of the film /substrate structures were conducted, using the commercial finite element code of ABAQUS 6.8 (Dassault Systèms Simulia Corp. Providence, RI) (ABAQUS, 2008). The overall size of the finite element model was taken large enough to ensure the characteristic of half space. The ratio of the radius of the rigid, spherical indenter to the length of the geometrical model was 1:12 along both the loading and radial directions to allow for considerable decrease in deformation before reaching the constrained boundaries. The nodes along the axisymmetric axis were constrained in the radial direction to simulate axisymmetric nature of the problem; and the nodes on the bottom side of the model were constrained in the z-direction.

Figure 2 shows the finite element mesh used in the simulation. Fine mesh was used just adjacent to the indenter, and the element size gradually increased away from the indenter. The element CAX4R (axi-symmetrical element with four nodes and quadrilateral cross-section) was used in the simulation. There were two degrees of freedom at each node, one in the radial direction and the other in the vertical direction. The indenter was modeled as an analytically rigid surface. Contact formulation was applied between the analytically rigid indenter and the top surface of the material. The frictionless contact was incorporated in the contact formulation of the indenter and the specimen.



(a) (b)

Figure 2: Finite element model of the spherical indentation of a film-substrate structure; (a) coarse mesh, (b) fine mesh.

The radius (R) of the spherical indenter used in the analysis was 50 μm. The thickness (h) of the film used in the finite element modeling was normalized with respect to the indenter radius and h/R = 0.2, 0.4, 0.6, 0.8, 1.

In the present analysis, the Young's modulus and the Poisson ratio of the substrate were a modulus E_2=69 GPa and ν_2=0.33, which were kept as constants. The film properties were changed and normalized with the properties of the substrate, as summarized in Tables 1 and 2.

Table 1: Modulus of the films used in the finite element analysis.

$\mathbf{E_1/E_2}$
0.010
0.025
0.050
0.075
0.10
0.25
0.50
0.75
1
10
25
50
75
100

Table 2: Poisson's ratio of the films used in the finite element analysis.

ν_1/ν_2
0.10
0.25
0.50
0.75
1.25
1.50

4 Results and Discussion

To validate the finite element model, the indentation was first performed on a homogeneous structure by letting E_1=E_2=69 GPa and ν_1=ν_2=0.33. The indentation load-displacement was obtained and then compared with the Hertzian contact theory (Equation 8), as shown in Figure 3. It is seen that the FE solution is in excellent agreement with the Hertzian contact result. Also included in the figure are the indentation load-displacement curves of two typical film /substrate structures (E_1/E_2=0.1 and E_1/E_2=10). It is observed that for the layered structures, the Hertzian relation can not accurately describe the indentation load-displacement responses of elastic layered structures.

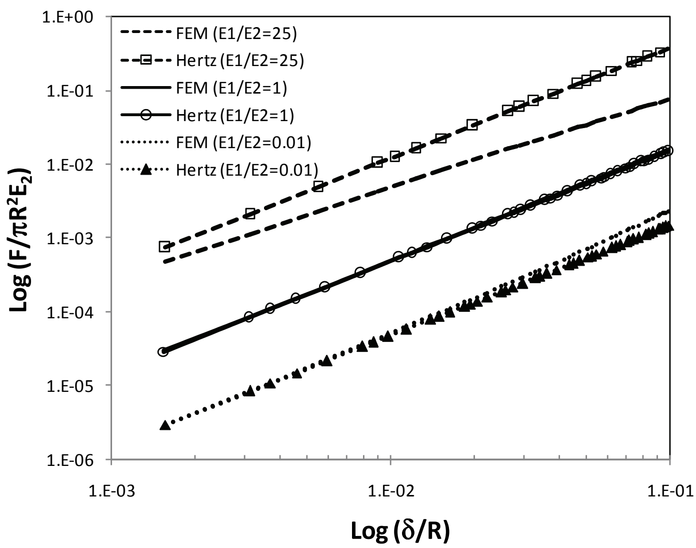


Figure 3: Comparisons of the finite element results and the Hertz contact theory in the indentation of film-substrate structures. As E_1/E_2=1, the film-substrate structure reduces to a homogeneous one.

The indentation deformation of a film-substrate medium depends upon the film thickness and film properties (assuming the substrate is known). As mentioned above, the present study is limited to the elastic film /substrate systems. The film-substrate systems of all possible configurations (various film thicknesses and various film properties (modulus and Poisson's ratio)) were modeled, as given in Tables 1-2. A total of approximately 350 analyses were conducted. Figure 4 shows the typical load-displacement curves for the indentation of thin films of various moduli, with a film thickness of 0.2R and a Poisson's ratio of 0.33. The maximum indentation depth (δ_{max}) in the present study was controlled to 12% of the indenter radius. The indentation load and indentation displacement were normalized with respect to $\pi R^2 E_2$ and R, respectively. It is seen that the reaction force increases

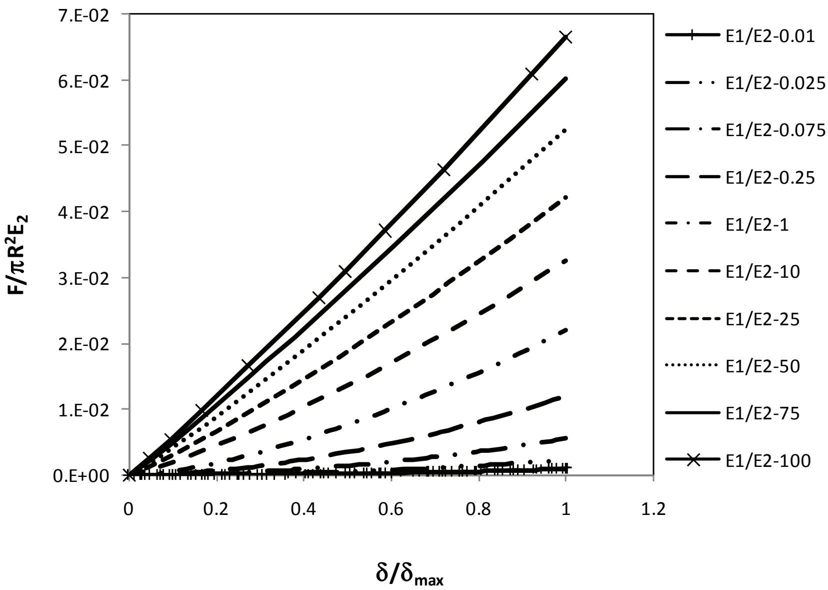


Figure 4: ypical indentation load-displacement curves for the film-substrate structures. The film thickness h/R=0.2.

with increasing the film modulus (E_1/E_2), as expected. The maximum indentation force was a functions of both film modulus and film thickness, which are illustrated in a 3D plot as shown in Figure 5. In general, the indentation force increases with increasing film modulus (E_1/E_2), while it decreases with increasing film thickness (h/R) for $E_1/E_2<1$ (soft film-hard substrate) and increases with increasing film thickness (h/R) for $E_1/E_2>1$ (hard film-soft substrate). As $E_1/E_2=1$, the indentation force remains constant with respect to h/R, since the structure now becomes homogeneous.

The indentation of the film-substrate structure with various film Poisson's ratios was also analyzed. The mechanical properties of the film-substrate system (E_1/E_2) were varied in the range of 0.01 to 100, and the Poisson's ratio of the film (ν_1/ν_2) varied in the range of 0.10 to 1.5. The indenter was pushed to a maximum depth of δ_{max}=0.12R and the maximum reaction forces are calculated, as shown in Figure 6. Overall, the Poisson's ratio is seen to have little effect on the indentation response. Similar finding also was reported earlier (Mesarovic and Fleck, 1999; Chen et al., 2007). Thus, it is reasonable to conclude that a small change in Poisson's ratio of the film may have negligible impact on the indentation load-displacement response. It is realized when the Poisson's ratio of the film approaches extreme values, such as

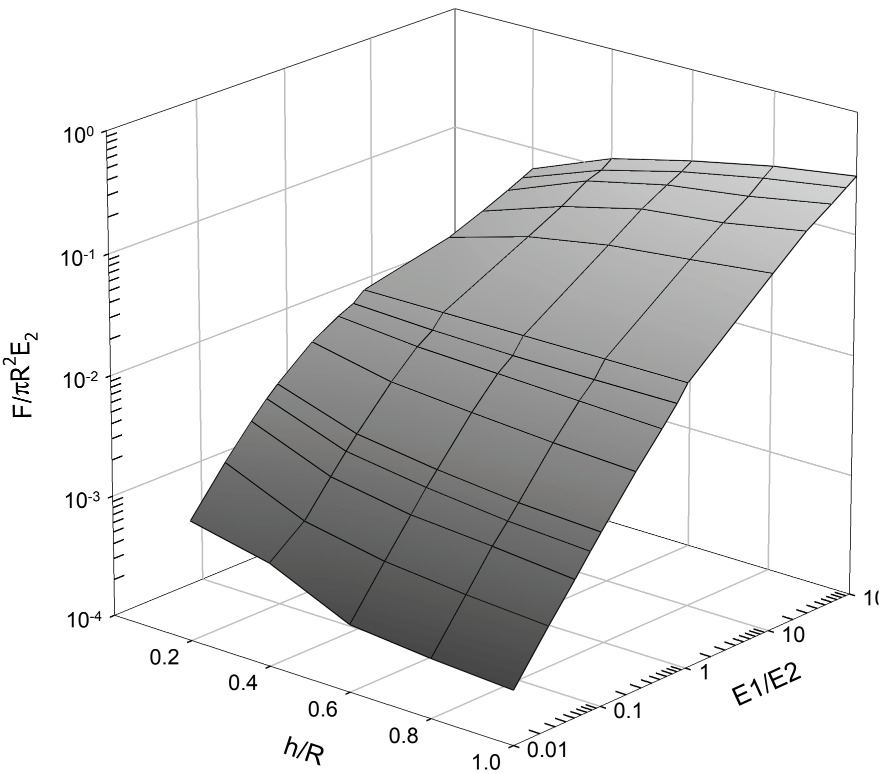


Figure 5: Maximum indentation load as functions of normalized film modulus (E_1/E_2) and normalized film thickness (h/R). The film Poisson's ratio is E_1/E_2=1.

$\nu_1/\nu_2 \geq 1.5$ or $\nu_1 \geq 0.495$, the film becomes incompressible and its properties will be affected by any small change in Poisson's ratio. This will be the special case which is beyond the scope of this paper.

For all the possible film-substrate configurations, the indentation load-displacement curves has been generated and analyzed with power law functions. Combining all the power-law relations, a generalized power-law equation is established as

$$F = CE_2\delta^p \tag{9}$$

where F is the indentation load and δ the indenter displacement. C and P are two

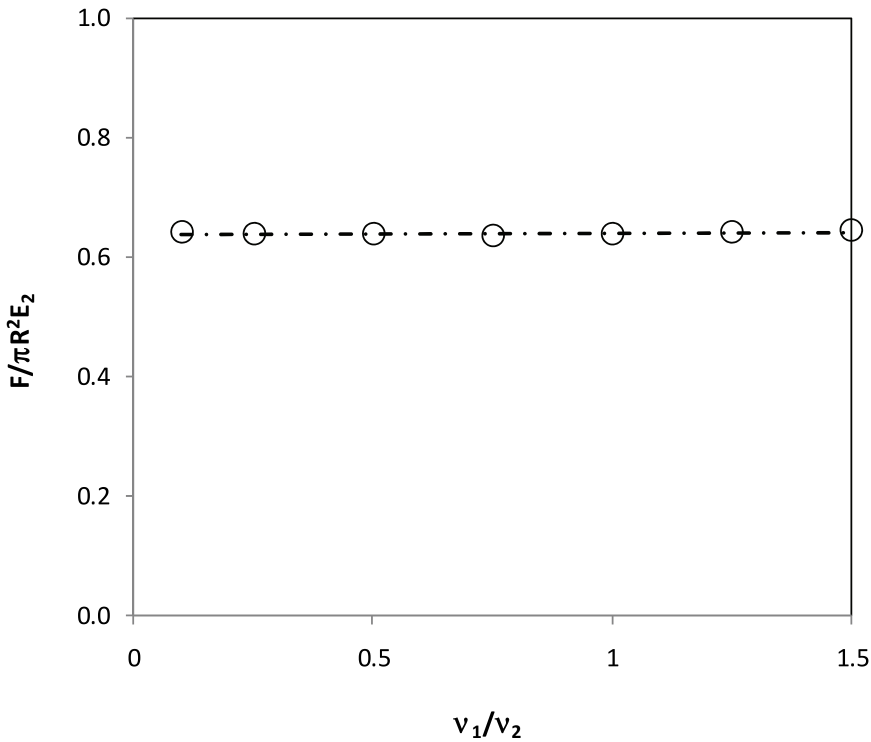


Figure 6: Maxmium indentation load as a function of film Poisson's ratio.

parameters and defined as

$$\log C = C_0 + C_1 \log(E_1/E_2) + C_2[\log(E_1/E_2)]^2 + C_3[\log(E_1/E_2)]^3 \tag{10a}$$

where C_0 = 0.9892-0.02725 $(\frac{h}{R})$ +0.068268 $(\frac{h}{R})^2$ -0.03375 $(\frac{h}{R})^3$
C_1 = 0.2939+1.7397 $(\frac{h}{R})$-1.9264$(\frac{h}{R})^2$ +0.77333$(\frac{h}{R})^3$
C_2 = -0.11509+0.064477 $(\frac{h}{R})$ +0.00013037 $(\frac{h}{R})^2$ -0.013021$(\frac{h}{R})^3$
C_3 = 0.038188-0.15565 $(\frac{h}{R})$ + 0.18009 $(\frac{h}{R})^2$ -0.072568 $(\frac{h}{R})^3$

$$\log P = P_0 + P_1 \log(E_1/E_2) + P_2[\log(E_1/E_2)]^2 + P_3[\log(E_1/E_2)]^3 + P_4[\log(E_1/E_2)]^4 \tag{10b}$$

where P_0 = 0.18478 -0.010549 $(\frac{h}{R})$ + 0.0081517 $(\frac{h}{R})^2$ -0.0017708 $(\frac{h}{R})^3$

$P_1 = -0.11958 + 0.15183\,(\frac{h}{R}) - 0.082706\,(\frac{h}{R})^2 + 0.013552\,(\frac{h}{R})^3$
$P_2 = -0.0000081967 - 0.081956\,(\frac{h}{R}) + 0.11148\,(\frac{h}{R})^2 - 0.047344\,(\frac{h}{R})^3$
$P_3 = 0.0089133 - 0.0059241\,(\frac{h}{R}) - 0.010213\,(\frac{h}{R})^2 + 0.0081615\,(\frac{h}{R})^3$
$P_4 = 0.0006394 + 0.0070813\,(\frac{h}{R}) - 0.010569\,(\frac{h}{R})^2 + 0.0045875\,(\frac{h}{R})^3$

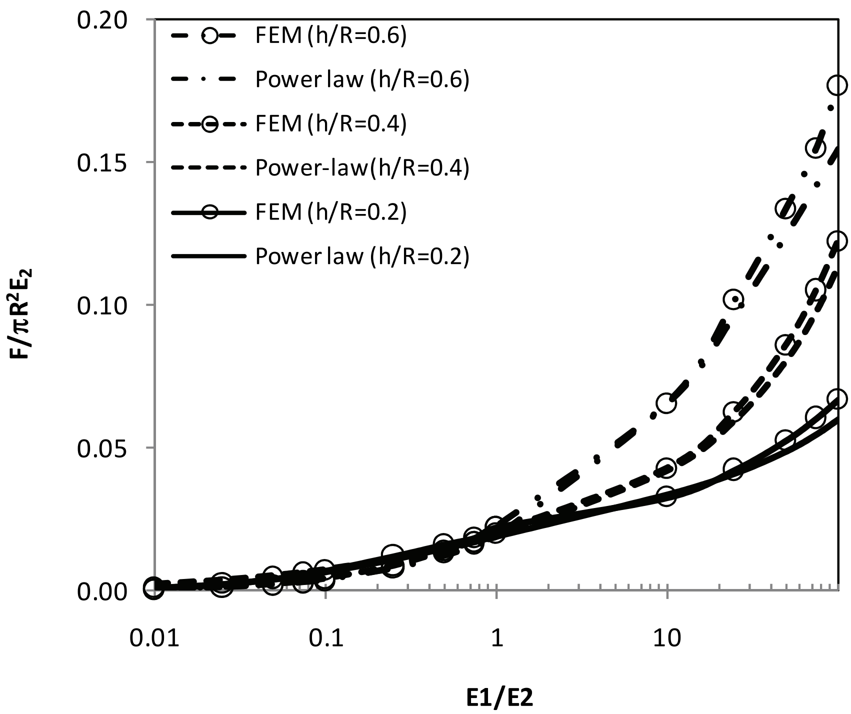


Figure 7: Comparisons of the FEM results and the generalized power law predictions: maxmium indentation force vs. film modulus.

Both *C* and *P* are dependent of only the material properties and the geometrical configuration of the indenter and the film-substrate system. Equation 9 suggests that the load-displacement curve fro an indentation of an elastic film /substrate material depends explicitly upon the film properties (modulus E_1/E_2) and film thickness (h/R). The effect of the film Poisson's ratio becomes negligible as explained in previous section. The generalized power-law equation is used to predict the load-displacement curve of each film-substrate configuration and then compared with the simulation results from the FEM. In all cases, good correlations have been obtained. The detailed results are not shown here due to the large amount of data.

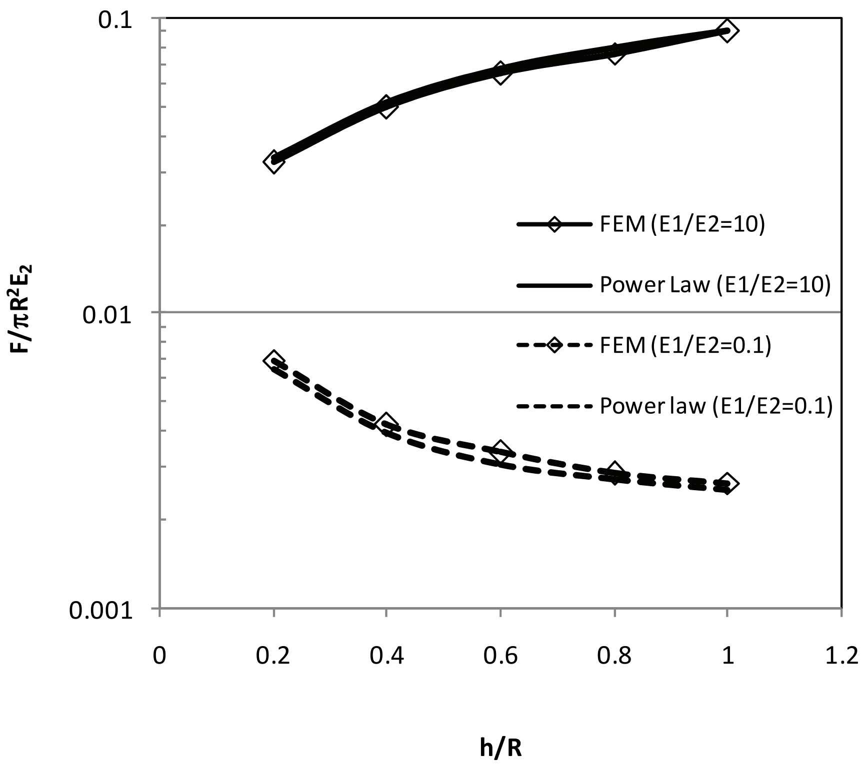


Figure 8: Comparisons of the FEM results and the generalized power law predictions: maxmium indentation force vs. film thickness.

Figure 7 and Figure 8 show the reaction forces at the maximum indentation depth (δ_{max}=0.12R) as functions of the film modulus (E_1/E_2) and film thickness (h/R). Both have demonstrated good agreement between the generalized power law and the FEM results. Therefore, the generalized power law relation can be used to describe the load-displacement curve for the spherical indentation of an arbitrary film /substrate structure.

Equation 10(b) describes the power, p, in the generalized analytical equation as a function the ratios of E_1/E_2 and h/R. Figure 9 depicts the dependence of p as a function of film modulus (E_1/E_2). The power (p) generally increases with decreasing the film modulus. Overall, the value of p is found in the range of 1.07 $\sim$ 2.28. At E_1/E_2=1, the bi-layer film /substrate structure reduces to a homogeneous material and the corresponding power p$\approx$1.5, which is consistent with the Hertz equation (Equation 8). This result demonstrates that generalized power law relation is valid. The magnitude of p depends also on the film thickness (h/R), which generally decreases with increasing the film thickness when $E_1/E_2<1$ (Figure 10). As the film thickness reaches beyond one indenter radius ($h/R \geq 1$), the effect of substrate become insignificant and p approaches 1.5 – the film /substrate structure behaves like a homogeneous one.

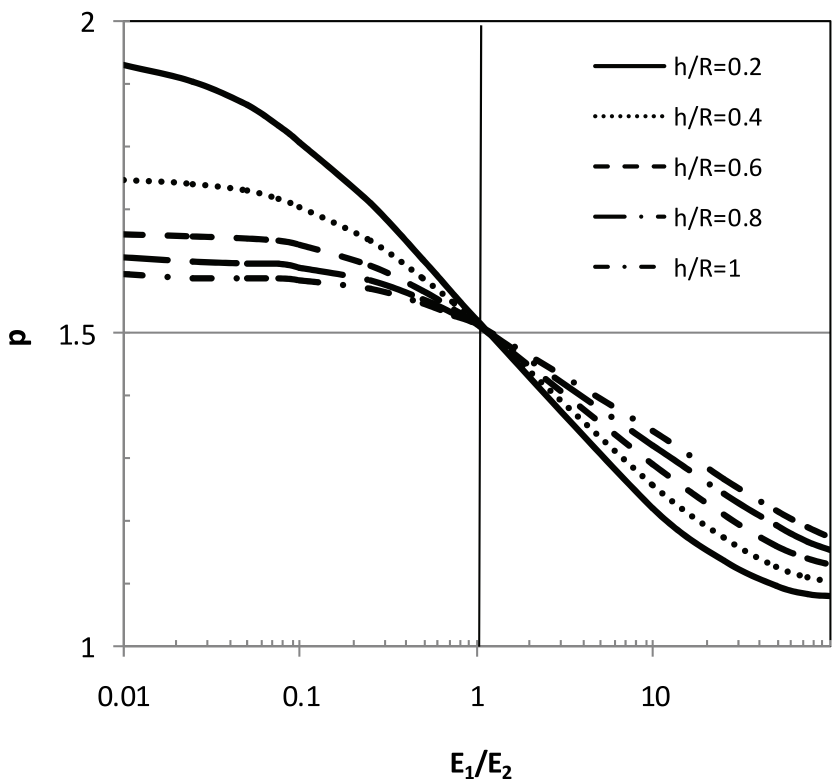


Figure 9: Variation of the power coefficient, p, as a function of film modulus (E_1/E_2).

The practical application of the present generalized power law equation can be understood as follows. For an elastic film of thickness h deposited on a rigid substrate, the load-displacement (F-δ) can be obtained for a spherical indentation by a rigid, spherical indenter of radius R. By plotting log(F) vs log(δ), the power, p, can be estimated as the slope of the liner plot (based on Equation 9). Once p is known, the film modulus E_1 can be readily computed through Equation 10(a), provided that

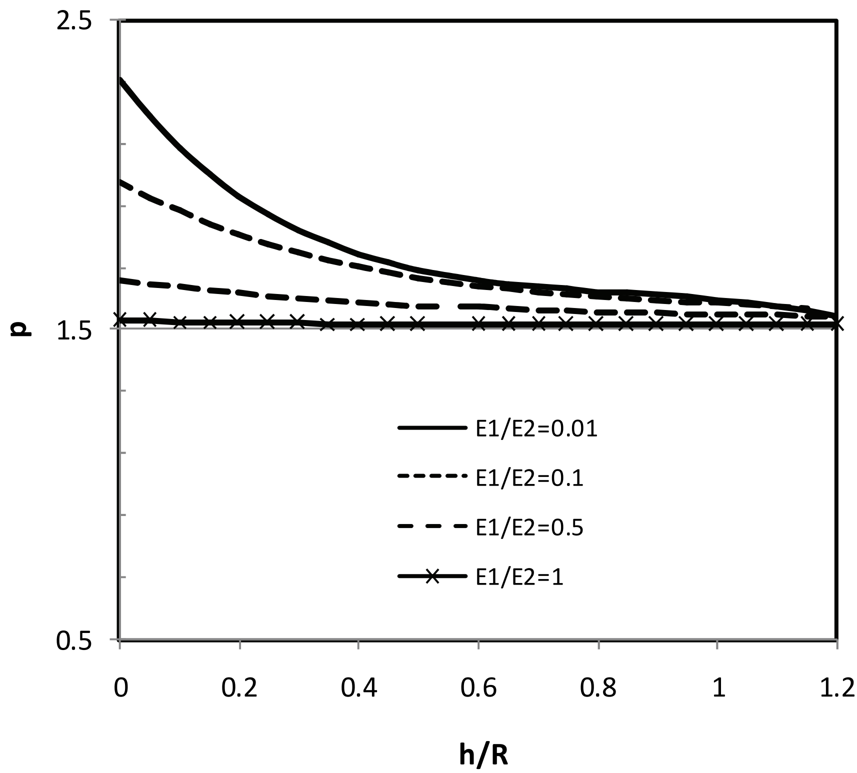


Figure 10: Variation of power coefficient, p, as a function of film thickness (h/R).

the substrate modulus E_2 is known.

From Eq. (9), one can calculate the contact stiffness, S,

$$S = \frac{dF}{d\delta} = CpE_2\delta^{p-1} \tag{11}$$

which is only a power function of the indentation displacement.

5 Summary

The indentation of elastic film /substrate structures by a rigid, spherical indenter was studied using the finite element method. The indentation load-displacement curves of the film /substrate systems of various physical configurations (film geometry and film properties) were obtained and analyzed. A generalized power law equation was established which can be used to fit the indentation load-displacement response of an arbitrary elastic film /substrate material. The indentation force is found to increase with increasing film modulus (E_1/E_2), and to increase with decreasing film thickness (h/R) for $E_1/E_2<1$ and with increasing film thickness (h/R) for $E_1/E_2>1$. The power, p, in the generalized power law equation was determined as a function of the film thickness and film modulus. From the point view of applications, the power p can be determined from the slope of the indentation load-displacement curve, log (F) vs. log(δ), of the film /substrate structure. Then, the film modulus E_1 can be readily estimated provided the substrate modulus (E_2) is known.

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