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Determining the Elastic Modulus of Compliant Thin Films Supported on Substrates from Flat Punch Indentation Measurements

M.J. Wald · J.M. Considine · K.T. Turner

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Abstract Instrumented indentation is a technique that can be used to measure the elastic properties of soft thin films supported on stiffer substrates, including polymer films, cellulosic sheets, and thin layers of biological materials. When measuring thin film properties using indentation, the effect of the substrate must be considered. Most existing models for determining the properties of thin films from indentation measurements were developed for metal and dielectric films bonded to semiconductor substrates and have been applied to systems with film-substrate modulus ratios between 0.1 and 10. In the present work, flat punch indentation of a thin film either bonded to or in contact with a substrate is examined using finite element modeling. A broad range of film-substrate modulus ratios from 0.0001 to 1 are investigated. As the substrate is effectively rigid compared to the film when the film-substrate modulus ratio is less than 0.0001, the results are also useful for understanding systems with lower film-substrate modulus ratios. The effects of the contact radius, film thickness, elastic properties, and friction between the film and the substrate on the measured stiffness were quantified using finite element modeling in order to understand how the elastic properties of the film can be extracted from indentation measurements. A semi-analytical model was developed to describe the finite element modeling results and facilitate the use of the results to analyze experimental measurements. The model was validated through analysis of indentation measurements of thin polyethylene sheets that were supported on substrates of various stiffness.

Keywords Indentation · Thin films · Finite elements · Substrate effects · Soft materials

Introduction

There are many applications that require the measurement of mechanical properties of thin films with elastic moduli from several kPa to hundreds of MPa. These films are often bonded to or supported on substrates that are substantially stiffer and have moduli of tens to hundreds of GPa. Examples include polymer films, cellulosic sheets, and thin layers of biological materials on glass and metal substrates. Instrumented indentation is an attractive method for characterizing these films as it requires minimal sample preparation and multiple measurements can typically be obtained from a single specimen. The challenge in testing films supported on substrates is that the measured indentation stiffness is often a function of both the elastic properties of the film and the substrate. In order to determine the properties of the film alone, an appropriate mechanics model and knowledge of the elastic properties of the substrate are required. The goal of the present work is to develop a model that allows the elastic modulus of a thin film to be extracted from flat punch indentation measurements made on thin film-substrate systems in which the ratio of the elastic modulus of the film to that of the substrate is between 10^{-4} and 1.

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A cylindrical flat punch indentation test is considered here because it is has numerous advantages for measuring low modulus materials. First, the large contact area of the flat punch indenter results in higher loads in tests that are easier to measure. Second, the contact area is constant with indentation depth, eliminating the need for characterizing an area function for the indenter. Finally, the large contact area is useful for determining the average properties of materials which are porous or have heterogeneous microstructures [1]. The primary drawbacks of flat punch indentation are the need for careful indenter-sample alignment and the fact that the large contact radius usually leads to the substrate influencing the measured indentation stiffness when testing thin films. The issue of tip-sample alignment can be addressed through proper design of the measurement instrument. Substrate effects can be addressed through mechanics analysis of the film-substrate system, such as the one presented in this work.

Background

Instrumented indentation is a common method for characterizing the elastic properties of thin films supported on substrates (Fig. 1). This technique is well-suited for characterizing small volumes of material at low loads and small displacements. Compared to traditional tension or compression tests, indentation allows for simple sample preparation, but has a more complicated stress-state that can make proper analysis challenging. Oliver and Pharr [2] describe a widely accepted method for determining the elastic modulus of an elastic halfspace from indentation tests using Sneddon's solution for axisymmetric indentation of an elastic halfspace [3]. This approach relates the elastic modulus of the specimen to the stiffness measured in indentation tests using the following relationship (assuming a rigid indenter):

$$E^* = \frac{E}{1-\nu^2} = \frac{\sqrt{\pi}}{2} k \frac{1}{\sqrt{A}} \tag{1}$$

where E^* is the reduced modulus, E is Young's modulus, ν is Poisson's ratio, k is the measured stiffness, and A is contact area at the load at which the stiffness is measured.

A common rule of thumb states that the Oliver–Pharr method can be applied to determine the properties of a thin film on a substrate if the thickness of the film is at least ten times greater than the indentation depth [4]. However, the validity of this approximation depends on the geometry and dimensions of the indenter relative to film thickness [5]. When the film is not sufficiently thick, and equation (1) is used to analyze the data, E^* is an effective reduced modulus, E^*_{eff}, that is a function of Young's moduli of the film, E_1, and substrate, E_2, Poisson's ratios of the film, ν_1, and the substrate, ν_2, indenter contact radius, a, and film thickness, t. To extract the elastic properties of the film alone from E^*_{eff}, the properties of the substrate and an appropriate mechanics model of the system are needed.

Several models have been proposed to describe the relationship between E^*_{eff} and the elastic properties of the system, indenter geometry, and film thickness in order to allow film modulus to be extracted from data collected from indentation tests on film-substrate systems [5–13]. Doerner and Nix [6] developed an empirical model with a fitting parameter to account for the effect of a substrate in indentation measurements of thin tungsten films on silicon substrates using a Berkovich indenter. King [7] used a numerical analysis to extend the Doerner–Nix model to various flat punch geometries. Saha and Nix [8] subsequently modified King's model to apply it to non-flat indenter geometries. Gao et al. [9] used a perturbation method based on Sneddon's solution [3] to develop a closed form solution to find E_1 from E^*_{eff} data measured at various a/t ratios using a cylindrical flat punch indenter for E_1/E_2 values between 0.5 and 2. An analysis by Rar et al. [10] extended the Gao model to larger modulus mismatch. Results were presented for E_1/E_2 from 0.1 to 10 and a/t between 0.1 and 10. Sakai et al. [5, 11] developed an analytical model for axisymmetric geometries using Boussinesq's Green's function solution. Similar to the results of Gao et al. [9] and Rar et al. [10], the Sakai model employs weight functions. Results were presented for E_1/E_2 from 0.2 to 10 and a/t from 0.01 to 10.

An analytical model proposed by Bec et al. [12] approximates the film-substrate system during flat punch indentation as two springs in series. The stiffness of one spring is determined by the compression of the film under uniform displacement by a rigid flat punch indenter. The other spring

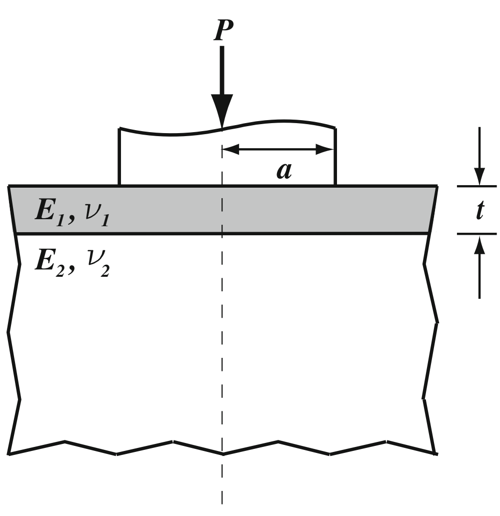

Fig. 1 Schematic of indentation of a thin elastic film with thickness t on an elastic substrate with a cylindrical flat punch indenter of radius a

has a stiffness defined by the response of an elastic half-space with the substrate's elastic properties under uniform displacement. Combining these springs gives the following relationship:

$$\frac{1}{k} = \frac{t}{f\left(\frac{a}{t}\right)\pi a^2 E_1^*} + \frac{1}{f\left(\frac{a}{t}\right)2aE_2^*}, \tag{2}$$

where k is the measured stiffness, and E_1^* and E_2^* are the reduced moduli of the film and substrate, respectively. Bec et al. used $f(\frac{a}{t}) = 1 + \frac{2t}{\pi a}$. Cases where E_1/E_2 varied from 0.1 to 10 and *a/t* varied from 0 to 3 were considered.

Finally, the limiting case of the indentation of a thin elastic film bonded to a rigid substrate was examined by Yang [14]. For a cylindrical flat punch, the measured stiffness is related to the elastic properties of the film as follows [14]:

$$k = \frac{(1-\nu_1)\pi a^2 E_1}{(1+\nu_1)(1-2\nu_1)t}. \tag{3}$$

As this simple result essentially considers compression of the film under the indenter, it is only valid when the contact radius is much larger than the film thickness.

Objective and Scope

Existing models for understanding the indentation response of film-substrate systems were largely developed to describe systems with E_1/E_2 between 0.1 and 10. While useful for studying thin metal and dielectric films on semiconductor substrates, these models are not applicable for testing soft thin films, such as thin polymer layers and biological materials, supported on stiff substrates. The rigid substrate solution presented in [14] is applicable for investigating soft films, but is limited to cases where the substrate is very stiff and the contact radius is large relative to the film thickness. Little work has been done to understand the relationship between E^*_{eff} and the elastic properties of the system, the contact radius, and the film thickness when E_1/E_2 is less than 0.1, when the substrate is not considered rigid, and when *a/t* is less than 10.

Most thin film indentation modeling also assumes the film is perfectly bonded to a substrate. While this is a suitable approximation for many applications in which the thin film is cast or deposited on a substrate, thin polymer films and biological materials may be simply resting on the substrate during testing. Supporting thin films on a substrate is an attractive option for testing certain materials, however little work has been done to investigate E^*_{eff} for cases when the thin film is in frictional contact, but not bonded to a substrate.

The overall objective of this work is to develop a method for determining the elastic modulus of thin, low modulus films supported on thick substrates from cylindrical flat punch indentation measurements. To accomplish this, a finite element model is employed to quantify the effect of E_1/E_2 and *a/t* on E^*_{eff} for thin elastic films bonded to a substrate or in frictional contact with a substrate for E_1/E_2 from 10^{-4} to 1 and *a/t* from 1 to 100. The roles of Poisson's ratio of the thin film and the friction coefficient between the film and the substrate (for non-bonded cases) are examined. A semi-analytical model is presented to describe the results from the finite element simulations to facilitate application of the results to experimental data. Finally, indentation measurements of a polyethylene sheet supported on substrates with various stiffness are reported and compared to the modeling results.

Finite Element Simulations

Model Geometry

A finite element (FE) model was developed to investigate the effects of E_1, E_2, ν_1, ν_2, a, t, and the bonding or coefficient of friction, μ, between the film and substrate, on the effective modulus, E^*_{eff}, measured using flat punch indentation. 31720 axisymmetric eight node elements were used to mesh the model of a thin film-substrate system (Fig. 2). The length of the film and substrate were set at $100a$. The thickness of the substrate was $1{,}000t$. The number of elements and model dimensions were determined from a convergence study. The indenter was modeled as a rigid body in frictionless contact with the film. For the case of a thin film bonded to a substrate, the coincident nodes at the interface of the thin film and the substrate were coupled in all degrees of freedom. To examine a thin film supported by a substrate,

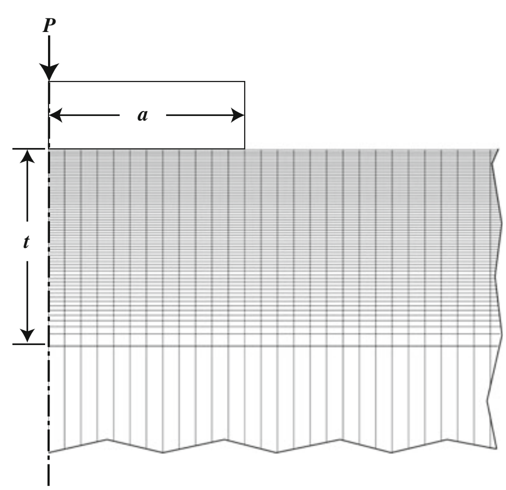

Fig. 2 Image of a portion of the finite element mesh used to study the indentation of a thin film on a substrate with a rigid flat punch. The width of the film and substrate is $100a$. The height of the substrate is $1{,}000t$

frictional contact elements were used to model the interaction between the film and the substrate. The model was solved using the commercial finite element package Abaqus 6.7 (SIMULIA, Providence, RI).

The stiffness of the system is the slope of the load-indentation depth response, which is constant with respect to depth for cylindrical flat punch indentation [3]. The effective modulus of the system is calculated from the FE determined stiffness using the Oliver–Pharr relationship, which simplifies to the following for a cylindrical flat punch indenter:

$$E^*_{\mathrm{eff}} = \frac{1}{2a}k. \tag{4}$$

Simulations of a thin film bonded to a substrate were run for $E_1/E_2 = 10^{-4}$ to 1, $a/t = 1$ to 100, $t = 80$ to 100 µm, $E_1 = 100$ MPa, $\nu_1 = 0.3$ to 0.49, and $\nu_2 = 0.3$. For a thin film in contact with a substrate, simulations were run for $E_1/E_2 = 10^{-4}$ to 1, $a/t = 1$ to 100, $t = 100$ µm, $E_1 = 100$ MPa, $\nu_1 = 0.3$ to 0.49, and $\nu_2 = 0.3$. To investigate the effect of friction between the film and substrate, the friction coefficient, μ, was varied from 0 to 0.4.

Results of Parametric Studies

FE calculations of a thin elastic film bonded to an elastic substrate were used to map the effects of E_1, E_2, a, and t on the effective modulus of the film-substrate system. As expected, when the film and substrate had the same properties, E^*_{eff}/E_1 did not depend on the indenter contact radius or film thickness. For cases in which the substrate modulus is larger than the film modulus, E^*_{eff} is a function of *a/t* and E_1/E_2 (Fig. 3), suggesting the substrate is effectively rigid compared to the film. For low values of E_1/E_2, E^*_{eff} is independent of E_1/E_2 (Fig. 3). The specific value of E_1/E_2 below which the substrate can be considered rigid depends on *a/t*.

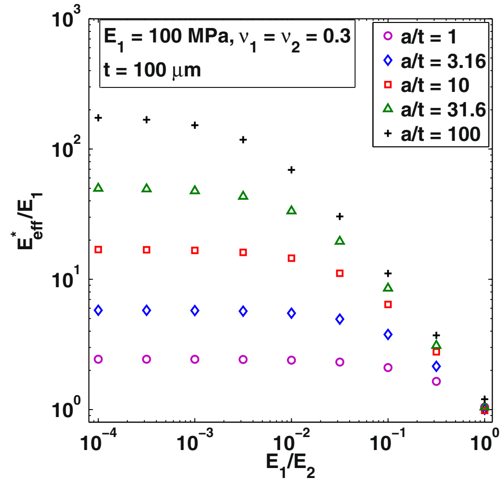

Fig. 4 Normalized E^*_{eff} calculated using FE analysis for a thin film in frictionless contact with a substrate

The indentation of a thin film in frictionless contact with a substrate using a cylindrical flat punch indenter was also examined. The relationship between E^*_{eff}/E_1 and E_1/E_2, shown in Fig. 4, is similar to that for the bonded thin film-substrate system, shown in Fig. 3. However, E^*_{eff} for the frictionless contact cases is generally less than those of the bonded thin film system (Fig. 3). The percent difference in E^*_{eff} between the bonded and frictionless cases varies as a function of E_1/E_2 and *a/t*, as shown in Fig. 5. In general, the difference in E^*_{eff} between the bonded and frictionless cases decreases as E_1/E_2 increases, with the exception of the case where E_1/E_2 is 1 and *a/t* is 10. When E_1/E_2 is 1,

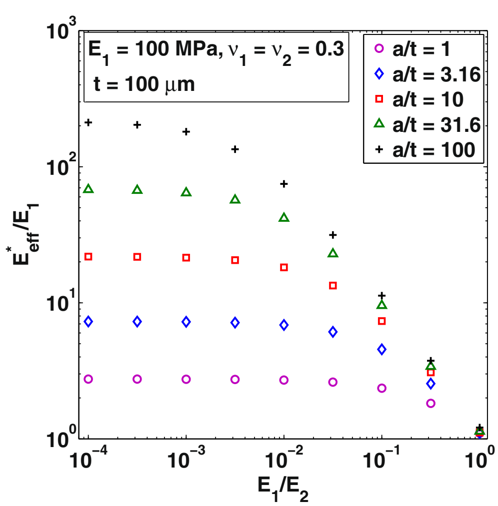

Fig. 3 Normalized E^*_{eff} calculated using FE analysis for a film bonded to a substrate

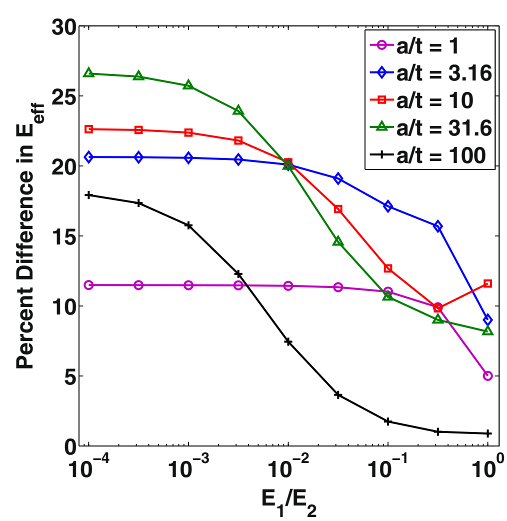

Fig. 5 Percent difference in E^*_{eff} for a film in frictionless contact with a substrate compared with a film bonded to a substrate

$\nu_1 = 0.3$, and *a/t* is 100, there is less than 2 % difference between the bonded and frictionless cases. However, when the substrate is much stiffer, E^*_{eff} is up to 26 % lower in the frictionless case. E^*_{eff} for the frictionless case is less than the bonded case because the film is able to expand in the transverse direction as it is compressed by the indenter. In the bonded case, the film is laterally constrained.

The effects of Poisson's ratio of the film and the coefficient of friction between the film and substrate on E^*_{eff} were also investigated. For E_1/E_2 from 10^{-4} to 1, *a/t* from 1 to 100, $t = 100$ µm, $E_1 = 100$ MPa, and $\nu_2 = 0.3$, ν_1 was varied from 0.3 to 0.49 and μ was varied from 0 to 0.4. For thin films in frictionless contact with a substrate, E^*_{eff} increases as ν_1 increases, reaching a maximum of 22.7 % relative to the $\nu_1 = 0.3$ case when ν_1 was 0.49, E_1/E_2 was 10^{-4}, and *a/t* was 10. The coefficient of friction was varied while Poisson's ratio was fixed at 0.3. E^*_{eff} is most sensitive to changes in the friction coefficient when *a/t* is small. The decrease in E^*_{eff} relative to a bonded thin film varies from 12 % when μ is 0 to less than 4 % when μ is 0.4. As *a/t* increases, the effect of friction becomes less significant and the change in E^*_{eff} is similar to that shown in Fig. 4. The results of these calculations are not shown here in plot form, but are described by the semi-analytical model (section Semi-analytical Model).

Sensitivity Studies

The FE model was also used to investigate the sensitivity of E^*_{eff} to a change in E_1 for the bonded and frictionless film cases. Figure 6(a and b) show the change in E^*_{eff} for a 10 % increase in E_1 for the bonded and frictionless cases, respec-

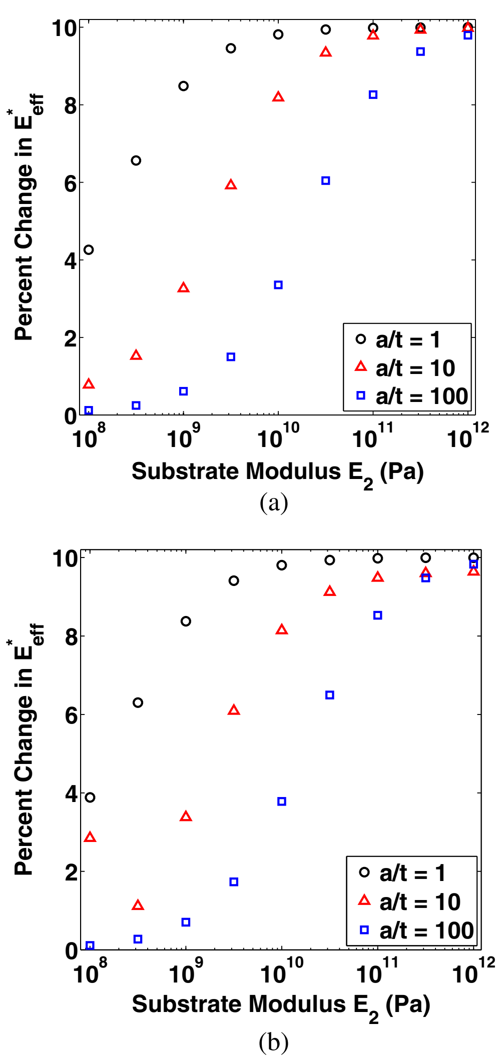

Fig. 6 Sensitivity of E^*_{eff} to a 10 % increase in E_1 ($E_1 = 100$ MPa, $\nu_1 = \nu_2 = 0.3$) for (**a**) a thin film bonded to a substrate and (**b**) a thin film in frictionless contact with a substrate

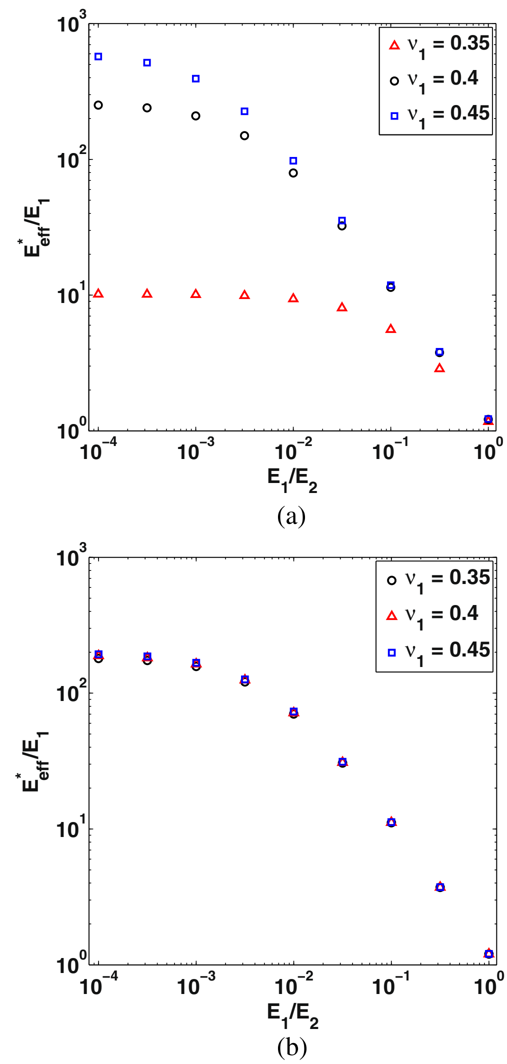

Fig. 7 Sensitivity of E^*_{eff} to changes in ν_1 for (**a**) a thin film bonded to a substrate and (**b**) a thin film in frictionless contact with a substrate. The cases shown are for $E_1 = 100$ MPa and $\nu_2 = 0.3$

tively. In both cases, when the film and the substrate have similar properties, there is little difference in E^*_{eff}. However, at higher E_2 values, E^*_{eff} increases by about 10 % for a 10 % increase in E_1.

The effect of Poisson's ratio, ν_1, of a film bonded to a substrate and a film in frictionless contact with a substrate on E^*_{eff}, was also examined. For E_1/E_2 from 10^{-4} to 1, $a/t = 100$, $t = 100$ μm, $E_1 = 100$ MPa, and $\nu_2 = 0.3$, ν_1 was varied from 0.35 to 0.45. The results are shown in Fig. 7(a and b). For a thin film bonded to a substrate (Fig. 7(a)), E^*_{eff} is insensitive to changes in ν_1 when the film and substrate have similar properties. However, when the substrate is much stiffer than the film, E^*_{eff} changes significantly for different values of Poisson's ratio. For $E_1/E_2 = 10^{-4}$, E^*_{eff} decreases 24 % when ν_1 decreases from 0.4 to 0.35 and increases 72 % when ν_1 increases from 0.4 to 0.45. E^*_{eff} is less sensitive to changes in ν_1 when the film is in frictionless contact with the substrate (Fig. 7(b)). In this case, when $E_1/E_2 = 10^{-4}$, E^*_{eff} decreases 4 % when ν_1 decreases from 0.4 to 0.35 and increases 3 % when ν_1 increases from 0.4 to 0.45.

Semi-analytical Model

Derivation

A closed-form solution for the indentation of a thin elastic film on an elastic substrate is not available. However, the system lends itself to development of a simple analytical model that can be calibrated with the results from the FE simulations. The indentation stiffness is a function of the elastic properties of the system, the film thickness, and the indenter radius. Here, we take a similar approach to that of Bec et al. [12] and represent the film and substrate as two springs combined in series. However, we include two constants in the model that are determined by fitting the model to results from the FE calculations.

The stiffness of the first spring, k_1, approximates the stiffness of the film beneath the indenter as that of a cylinder with radius, a, and thickness, t, under uniform compression. The cylinder is free to expand in the transverse direction in this simple picture. However, in the real system, the transverse expansion of the thin film is constrained by the material surrounding the cylinder as well as friction or bonding between the film and substrate. The degree of constraint depends on indenter contact radius, film thickness, Poisson's ratio of the film, and bonding or the coefficient of friction between the film and the substrate. To account for this constraint, a fitting parameter, C_1, is included in k_1, which is given as:

$$k_1 = \frac{C_1 \pi a^2 E_1}{t}. \tag{5}$$

C_1 is expected to be a function of *a/t*, ν_1, and μ.

The stiffness of the second spring, k_2, is approximated as an elastic halfspace with a uniform pressure applied over a circular region. The uniform pressure is due to stress transmitted to the substrate by the film. The radius of the region of applied pressure depends on a and t and in general will be slightly larger than the indenter contact radius. Similar to the thin film stiffness, the substrate stiffness will be affected by constraints on the transverse expansion of the substrate due to ν_2 and μ. To account for these assumptions, a fitting parameter, C_2, which is a function of *a/t*, ν_2, and μ, is introduced into the following equation for the stiffness of an elastic halfspace [15]:

$$k_2 = \frac{C_2 \pi a E_2}{2}. \tag{6}$$

The stiffness of the film-substrate system is found by combining the stiffnesses in equations (5) and (6) in series:

$$k = \frac{k_1 k_2}{k_1 + k_2} = \frac{C_1 C_2 \pi a^2 E_2 \xi}{2 C_1 a \xi + C_2 t}, \tag{7}$$

where $\xi = E_1/E_2$.

Fitting Parameters

C_1 and C_2 are assumed to depend on *a/t*, ν_1, ν_2, and μ, and are determined by fitting the semi-analytical model to the FE results using a nonlinear least squares method. An equation for a normalized effective modulus, which was obtained by rewriting equation (7), is used in the fitting process:

$$\frac{E^*_{eff}}{E_1} = \frac{\pi C_1 C_2}{2\left(2 C_1 \xi + C_2 \frac{t}{a}\right)}. \tag{8}$$

C_1 and C_2 values are given in Tables 1, 2, and 3 for the cases of a thin film bonded to a substrate and for a thin film supported by a substrate where the Poisson's ratio of the film and the coefficient of friction between the film and substrate are varied. C_1 and C_2 are not monotonic with respect to *a/t* when *a/t* is 100. This suggests that a different model may be needed to fully capture the behavior at large *a/t*.

Table 1 C_1 and C_2 for a thin film on a substrate ($\nu_1 = 0.3$)

a/t	$\mu = 0$		$\mu = 0.2$		$\mu = 0.4$		Bonded	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	1.54	2.18	1.62	2.18	1.68	2.20	1.69	2.38
3.16	1.16	1.40	1.29	1.50	1.33	1.64	1.39	1.66
10	1.08	1.31	1.13	1.32	1.17	1.32	1.30	1.48
31.6	1.01	1.29	1.03	1.29	1.04	1.29	1.29	1.45
100	1.12	1.45	1.13	1.45	1.14	1.45	1.28	1.53

Table 2 C_1 and C_2 for a thin film on a substrate ($\nu_1 = 0.4$)

a/t	$\mu = 0$		$\mu = 0.2$		$\mu = 0.4$		Bonded	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	1.59	2.32	1.69	2.32	1.80	2.33	2.13	2.27
3.16	1.16	1.43	1.52	1.46	1.70	1.50	2.04	1.59
10	1.17	1.32	1.32	1.33	1.43	1.33	2.12	1.40
31.6	1.12	1.31	1.17	1.31	1.21	1.31	2.16	1.38
100	1.22	1.45	1.24	1.45	1.25	1.45	2.18	1.46

A comparison between the FE and semi-analytical models in Fig. 8(a and b) shows that the semi-analytical model describes the FE model well for the range of *a/t* and E_1/E_2 values examined. For the case of a film bonded to a substrate (Fig. 8(a)), the difference between the semi-analytical model and FE results is ±2 % for all *a/t* examined when E_1/E_2 is greater than 0.1. The error reaches a maximum of 6.6 % when *a/t* is 100 and E_1/E_2 is 10^{-4}. For a thin film in frictional contact with a substrate, agreement between the models is best when the substrate is stiffer than the film (Fig. 8(b)). The error is within ±2 % for all *a/t* examined when E_1/E_2 is between 10^{-4} and $10^{-1.5}$. For a frictionless film on a substrate the error reaches a maximum of 8 % when E_1/E_2 is 1, *a/t* is 3.16, and ν_1 is 0.3. For cases where the coefficient of friction is 0.2 and 0.4, the maximum error is 11 % where E_1/E_2 is 1 and *a/t* is 31.6. Figure 8(a and b) show there is no increase in error when *a/t* is 100 suggesting that while C_1 and C_2 are not monotonic with respect to *a/t*, the semi-analytical model still described the FE model well when *a/t* is 100. A significant increase in C_1 for a bonded film occurs as ν_1 approaches 0.5. When ν_1 is near 0.5, the film becomes incompressible, resulting in a significant increase in the indentation stiffness when it is constrained in the transverse directions. This behavior is not observed when the film is in frictionless contact with the substrate, because the lack of constraints allows the film to expand in the transverse directions.

Table 3 C_1 and C_2 for a thin film on a substrate ($\nu_1 = 0.49$)

a/t	$\mu = 0$		$\mu = 0.2$		$\mu = 0.4$		Bonded	
	C_1	C_2	C_1	C_2	C_1	C_2	C_1	C_2
1	1.78	2.27	1.85	1.51	1.89	2.72	2.97	2.25
3.16	1.40	1.43	1.88	1.46	2.49	1.49	4.52	1.57
10	1.32	1.33	1.68	1.34	2.01	1.34	9.88	1.34
31.6	1.29	1.33	1.41	1.33	1.52	1.33	14.90	1.34
100	1.28	1.43	1.33	1.42	1.36	1.45	16.73	1.45

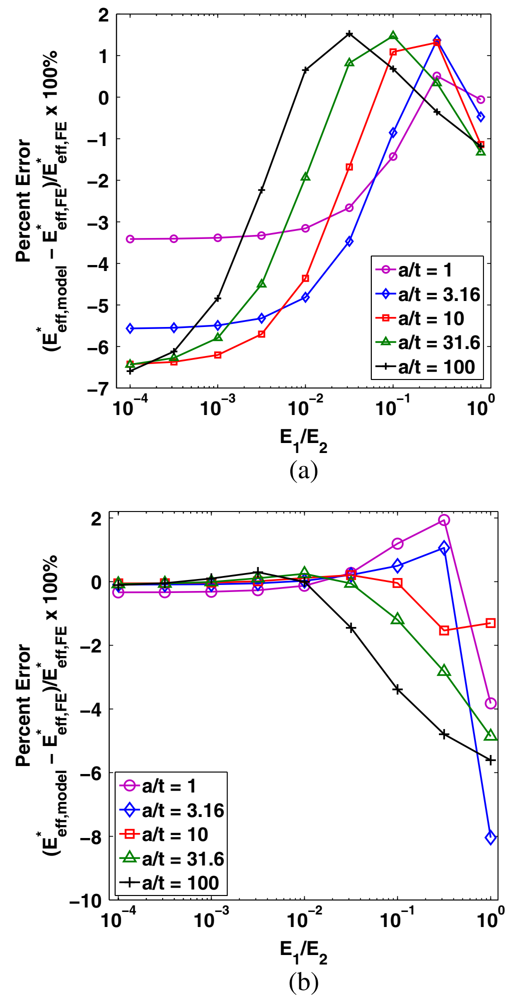

Fig. 8 Percent error between the semi-analytical model and the results from the FE simulations for (**a**) a thin film bonded to a substrate ($E_1 = 100$ MPa, $\nu_1 = \nu_2 = 0.3$) and (**b**) a thin film supported by a substrate ($E_1 = 100$ MPa, $\mu = 0$, $\nu_1 = \nu_2 = 0.3$)

Rigid Substrate Model

Yang presented asymptotic solutions for axisymmetric indentation of a compressible thin film perfectly bonded to a rigid substrate (i.e., $E_1/E_2 = 0$) [14]. This result is valid for cases when the contact radius is much larger than the film thickness. The FE results and semi-analytical model reported here were compared with Yang's solution for a thin film bonded to a rigid substrate (equation (3)).

Equation (3) can be rewritten in terms of the normalized effective modulus:

$$\frac{E^*_{\text{eff}}}{E_1} = \frac{\pi}{2} \frac{1 - \nu_1}{(1 + \nu_1)(1 - 2\nu_1)} \frac{a}{t}. \tag{9}$$

Using equation (9), Yang's model and results from the FE model are compared (Fig. 9). As expected from assumptions made in development of the rigid substrate model, the agreement is best when *a/t* is large and the substrate is much

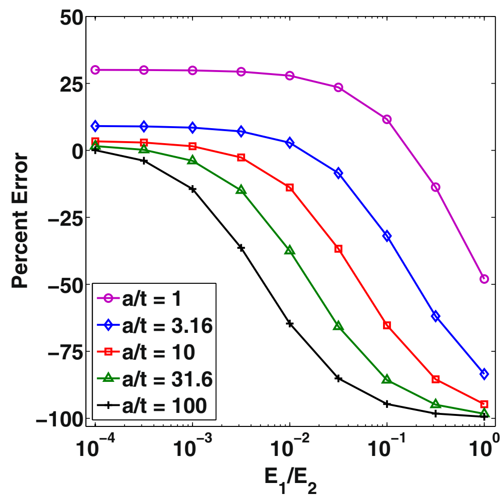

Fig. 9 Comparison of E^*_{eff} from FE calculations and Yang's model of a thin film on a rigid substrate ($E_1 = 100$ MPa, $\nu_1 = 0.3$)

stiffer than the film. When E_1/E_2 is 10^{-4}, the error is about 0.1 %. As E_1/E_2 increases and *a/t* decreases, the FE results diverge from the rigid substrate limit and Yang's solution is no longer applicable.

The agreement between Yang's model and our semi-analytical model for the limiting case of a film on a rigid substrate was investigated by evaluating equation (7) as $E_1/E_2 \to 0$. The semi-analytical model reduces to

$$k_{\mathrm{rigid}} = \frac{C_1 \pi a^2 E_1}{t}. \tag{10}$$

Setting equation (10) equal to equation (3) shows that

$$C_1 = \frac{1 - \nu_1}{(1 + \nu_1)(1 - 2\nu_1)}, \tag{11}$$

when E_1/E_2 is small and *a/t* is large. From the semi-analytical model fitting discussed previously, C_1 is 1.28 for a thin film bonded to a substrate where ν_1 is 0.3 and *a/t* is 100 (Table 1). Using equation (11), C_1 is 1.35 for a thin film bonded to a rigid substrate when the Poisson's ratio of the film is 0.3 and *a/t* is large. This suggests that the semi-analytical model describing the FE results agrees with Yang's solution when the substrate is much stiffer than the film and *a/t* is large.

Experimental Methods and Results

Microscale cylindrical flat punch indentation experiments were used to validate the modeling results. The effective modulus of a low density polyethylene (LDPE) film on various substrates was determined from force and displacement measurements acquired during indentation tests. Indentation experiments were performed using a TA Q800 DMA System (TA Instruments, New Castle, DE, USA) with a steel cylindrical flat punch indenter with a radius of 225 μm. The frame compliance was determined by finding the difference between the measured compliance of fused silica and the known material compliance of fused silica. The mean frame compliance was 1.01 μm/N with a standard deviation of 0.02 μm/N.

To compare with the results of the finite element study, 6.35 mm thick low density polyethylene, polymethylmethacrylate (PMMA), Aluminum 6061, and Stainless Steel 304 substrates were selected to support an LDPE film with a thickness of 76 ± 6 μm. The elastic properties of the substrate materials are given in Table 4. Because the properties of LDPE and PMMA vary depending on manufacturing process and structure, the substrate moduli were calculated from indentation tests using the method proposed by Oliver and Pharr for an elastic halfspace [2].

The stiffness of an LDPE film on various substrates was determined by taking the slope of the force-displacement data recorded during unloading near the maximum load. The indenter load was increased at a rate of 10 mN/s to the maximum load. A maximum load of 400 mN was chosen such that the indentation depth would be sufficient to ensure complete contact between the indenter and specimen. The maximum load was then held for 1 min. The specimen was then unloaded at a constant rate of 10 mN/s. The unloading portion of the experimental data was fit using the following relationship:

$$P = c(h - h_f)^m, \tag{12}$$

where c, h_f, and m are fitting parameters [16]. The stiffness is the slope of the unloading segment of applied load-indentation depth data evaluated at the maximum depth, $h_{\max}$, or

$$k = mc(h_{\max} - h_f)^{m-1}. \tag{13}$$

After accounting for frame compliance, the effective modulus is calculated using equation (4).

The measured normalized effective modulus of an LDPE film on various substrates is shown as a function of substrate modulus in Fig. 10. As expected from the FE model results, the effective modulus initially increases as substrate modulus increases, but becomes independent of substrate modulus when the substrate is much stiffer than the film. The mean E^*_{eff} measured on the aluminum substrate is not expected to be greater than that of the steel substrate and

Table 4 Substrate material properties

Material	E	ν
LDPE	226 MPa	0.43 [18]
PMMA	2.3 GPa	0.4 [19]
Aluminum 6061	69 GPa [20]	0.33 [20]
Stainless Steel 304	193 GPa [20]	0.27 [20]

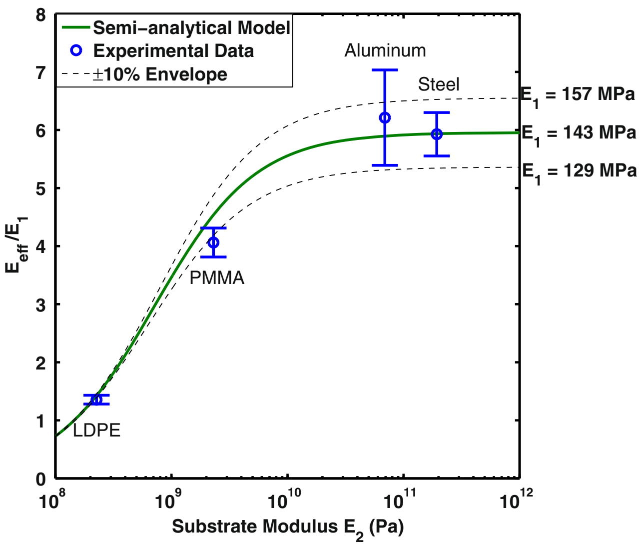

Fig. 10 Measurements of effective modulus of an LDPE film on substrates of different stiffness. The semi-analytical model was fit to the experimental data using E_1 as an unknown and $E_1 = 143$ GPa was obtained. The dashed lines show ±10 % boundaries for E_1

the cause of the large variability in the data collected on the aluminum substrate is unknown.

By rewriting equation (8), the semi-analytical model can be used to determine E_1 by fitting to the experimental data in Fig. 10 using

$$E_1 = \frac{2\left(2C_1\xi + C_2\frac{t}{a}\right)E^*_{\text{eff}}}{\pi C_1 C_2}. \tag{14}$$

Because *a/t*, E_2, ν_1, and ν_2 are known, appropriate C_1 and C_2 values can be chosen from the tables and the model can be fit to data collected from indentation experiments on a thin film supported by substrates with various moduli.

The semi-analytical model was used to determine the modulus of the LDPE film. C_1 and C_2 were found for $a/t = 2.96$ using linear interpolation of the data presented in Table 2 for a thin film ($\nu_1 = 0.4$, $\nu_2 = 0.3$) in frictionless contact with a substrate. For this case, C_1 was 1.28 and C_2 was 1.51. Using these values of C_1 and C_2 and treating E_1 as unknown, the model was fit to the experimental data to determine the modulus of the film. The elastic modulus of the film calculated using this method was 143 MPa. The fitted model is shown in Fig. 10 along with lines showing the model for film moduli which are ±10 % of the fitted modulus. The calculated film modulus is within the range reported for LDPE [17] and the model describes the trend of the data well.

Discussion

The results from the parametric studies of a thin film bonded to a substrate and a thin film in frictional contact with a substrate show E^*_{eff} depends on both E_1/E_2 and *a/t*. As the substrate becomes much stiffer than the film, the limiting case of a rigid substrate is approached and E^*_{eff} is independent of E_1/E_2. The E_1/E_2 at which this occurs decreases as *a/t* increases. However, we note that since E^*_{eff} becomes nearly independent of modulus ratio at E_1/E_2 values less than $5(10^{-4})$ for all *a/t* values studied, the results presented can be used to understand systems with moduli ratios less than 10^{-4} as well. There is a significant difference in E^*_{eff} between the bonded and frictionless film cases. This suggests that accounting for the interface between the film and substrate is important for accurately determining E_1.

The effect of varying ν_1 and μ on E^*_{eff} is a function of E_1/E_2 and *a/t*. Increasing ν_1 can cause a significant increase in E^*_{eff} when the film is bonded to the substrate. When *a/t* is 10 and E_1/E_2 is 10^{-4}, a Poisson's ratio of 0.49 resulted in a 22.7 % increase in effective modulus relative to $\nu_1 = 0.3$. While the effect of the Poisson's ratio of the film can be minimal in some cases, in general, it must be accounted for to accurately determine E_1. The friction coefficient between the thin film supported on a substrate also affects E^*_{eff}. However, relative to the case of a bonded film on a substrate, the percent change of E^*_{eff} between the frictionless cases and the case where μ is 0.4 was less than 10 % over the range of E_1/E_2 and *a/t* examined. As *a/t* and E_1/E_2 increased, the change between the cases with friction and those without friction became smaller. Thus, unless the friction between the film and the substrate is very large, the contact can generally be assumed to be frictionless if the film is not bonded to the substrate.

The sensitivity analysis showed that the change in E^*_{eff} due to a 10 % increase in E_1 depends on E_1/E_2 and *a/t*. From an experimental standpoint, these data suggest that it may be beneficial to test thin films on substrates that are significantly stiffer than the film to maximize the sensitivity to small differences in the film modulus. Furthermore, for both the bonded and frictionless film cases, it appears that the range of substrate moduli where the sensitivity to E^*_{eff} to small changes in E_1 is near 10 % decreases as *a/t* increases. This suggests that the range of substrate moduli over which E^*_{eff} is sensitive to changes in E_1 is largest when the contact radius and thickness are selected so *a/t* is near 1.

The FE results also showed that variations in ν_1 affect E^*_{eff}. For the cases examined, ν_1 had the largest impact on E^*_{eff} for a thin film bonded to a substrate when *a/t* was 100 and the substrate was much stiffer than the film. ν_1 had a less significant effect on E^*_{eff} when the thin film was in frictionless contact with the substrate. These results suggest that it is important to have an accurate estimate of ν_1 because the validity of the semi-analytical model depends on knowing ν_1 to chose appropriate C_1 and C_2 values. For films in which ν_1 is not well known, it may be advantageous to indent the film when it is supported but not bonded to a substrate. Because E^*_{eff} is much less sensitive to changes in ν_1 in this case, errors in the approximation of ν_1 will have less impact on E^*_{eff} than if the film were bonded to the substrate.

The comparison between the FE model and Yang's solution for a thin film on a rigid substrate gives insight into the range of E_1/E_2 and *a/t* where the rigid substrate assumption is valid. For the cases examined, the agreement is very good when E_1/E_2 is 10^{-4} and *a/t* is between 10 and 100, which quantifies Yang's assumption that the contact radius of the indenter must be much greater than the film thickness. As E_1/E_2 increases, the substrate is no longer considered rigid and Yang's model diverges from the FE results. When E_1/E_2 is 1, the error between Yang's model and the FE results is between 25 and 100 % for the cases examined. Also, as *a/t* decreases, the error between the rigid substrate solution and the rigid limit suggested by the FE model increases, but the range of E_1/E_2 over which the rigid substrate assumption is valid increases.

A semi-analytical model that consists of two springs combined in series was developed to describe the relationship between the measured indentation stiffness and the elastic properties of a thin film on a substrate, the contact radius, and the film thickness. Fitting parameters, C_1 and C_2, were calibrated using results from the finite element study. These parameters depend on *a/t*, ν_1, ν_2, and μ. The semi-analytical model described the FE model well, with less than 11 % error between the two models for the entire range of parameters examined and much smaller errors (< 2 %) over a large range. Also, for a thin film bonded to a substrate, C_1 approaches the value predicted for a thin film on a rigid substrate, showing good agreement with the model developed by Yang [14].

The high indentation stiffness observed for the bonded film on a substrate when *a/t* and ν_1 are large suggests it may be challenging to perform meaningful indentation measurements in these cases. The FE simulations suggest that E^*_{eff} values calculated from experimental data would be much larger than the film's elastic modulus. Because of this, it may be difficult to decouple the effects of ν_1 from the desired elastic modulus. It may be advantageous to conduct experiments with a *a/t* ratio near 1 when testing materials with high Poisson's ratio.

The validation experiments that consisted of indentation measurements of an LDPE film on substrates of various moduli demonstrated that the model developed in this work can accurately capture the dependence of E^*_{eff} on substrate moduli varying over about three orders of magnitude. The modulus of the film determined by fitting the model to data collected on four different substrates is within the range expected for LDPE. The fit matches the data reasonably well, as shown in Fig. 10, however the model does deviate from the experiments at certain points. These differences may be due to imperfect alignment between the specimen and the indenter. The increase in standard deviation of E_{eff} when the substrate is much stiffer than the film may be due to increased sensitivity to misalignment of the indenter due to the high stiffness of the substrate. Nevertheless, the measurements obtained generally match the model predictions.

Conclusion

Instrumented indentation is a widely used technique for determining the elastic properties of thin films on substrates. However, if the contact radius is large relative to the film thickness, the stiffness measured using indentation is related to an effective modulus that depends on the elastic properties of the thin film and substrate, the indenter contact radius, and the film thickness. Several models have been developed previously to determine the elastic properties of a thin film on a substrate and have been shown to be effective when E_1/E_2 is between 0.1 and 10. These models have not been shown to be applicable for systems with a larger modulus mismatch between the film and substrate. In the present work, cylindrical flat punch indentation of thin films on elastic substrates, where the substrate has a modulus greater than or equal to that of the thin film (E_1/E_2 from 10^{-4} to 1) and the indenter contact radius is equal to or larger than the thickness of the thin film (*a/t* from 1 to 100), were investigated.

A 2-D axisymmetric linear elastic finite element model was developed to investigate the effective modulus as a function of E_1/E_2 and *a/t*. The cases of a film bonded to a substrate and a thin film in frictional contact with a substrate were examined. A semi-analytical model was developed to describe FE results and present them in a form that can easily be used to analyze experimental measurements (equation (14)). The system was modeled as two springs in series where the stiffness of the springs corresponded to that of the thin film and substrate. The model was calibrated to the FE model results using two fitting parameters. The fitting parameters, summarized in Tables 1–3, depend on *a/t*, ν_1, and the type of contact between the thin film and the substrate.

Validation experiments were conducted on an LDPE film on various substrates to validate the FE and semi-analytical models. C_1 and C_2 for the *a/t* used in the experiments were calculated from Table 2 using linear interpolation. Using these values and treating E_1 as unknown, equation (14) was fit to the experimental data. The calculated film modulus was within in the range expected for LDPE and the semi-analytical model describes the trend in the data well.

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References

1. Soons J, de Baere I, Dirckx J (2011) New double indentation technique for measurement of the elasticity modulus of thin objects. Exp Mech 51(1):85–95
2. Oliver W, Pharr G (1992) An improved technique for determining hardness and elastic-modulus using load and displacement sensing indentation experiments. J Mater Res 7(6):1564–1583
3. Sneddon I (1965) The relation between load and penetration in the axisymmetric Boussinesq problem for a punch of arbitrary profile. Int J Eng Sci 3(1):47–57
4. Pharr G, Oliver W (1992) Measurement of thin-film mechanical-properties using nanoindentation. Mater Res Bull 17(7):28–33
5. Sakai M, Zhang J, Matsuda A (2005) Elastic deformation of coating/substrate composites in axisymmetric indentation. J Mater Res 20(8):2173–2183
6. Doerner M, Nix W (1986) A method for interpreting the data from depth-sensing indentation instruments. J Mater Res 1(4):601–609
7. King R (1987) Elastic analysis of some punch problems for a layered medium. Inter J Solids Struct 23(12):1657–1664
8. Saha R, Nix W (2002) Effects of the substrate on the determination of thin film mechanical properties by nanoindentation. Acta Mater 50(1):23–38
9. Gao H, Chiu C, Lee J (1992) Elastic contact versus indentation modeling of multilayered materials. Int J Solids Struct 29(20):2471–2492
10. Rar A, Song H, Pharr G (2002) Assessment of new relation for the elastic compliance of a film-substrate system. Mater Res Soc Symp Proc 695:431–6
11. Sakai M (2006) Elastic and viscoelastic contact mechanics of coating/substrate composites in axisymmetric indentation. Philos Mag 86(33-35):5607–5624
12. Bec S, Tonck A, Georges J, Georges E, Loubet J (1996) Improvements in the indentation method with a surface force apparatus. Philos Mag A 74(5):1061–1072
13. Zhou B, Prorok B (2010) A discontinuous elastic interface transfer model of thin film nanoindentation. Exp Mech 50:793–801
14. Yang F (2006) Asymptotic solution to axisymmetric indentation of a compressible elastic thin film. Thin Solid Films 515(4):2274–2283
15. Johnson K (2003) Contact mechanics, 9th edn. Cambridge, New York
16. VanLandingham M (2003) Review of instrumented indentation. J Res Nat Inst Stand Technol 108(4):249–265
17. Brandrup J, Immergut E, Grulke E, Abe A, Bloch D (2003) Polymer handbook, 4th edn. Wiley, New York
18. Willett J (1994) Mechanical-properties of LDPE granular starch composites. J Appl Polym Sci 54(11):1685–1695
19. Throne J (1996) Technology of thermoforming. Hanser Publishers, New York
20. Boresi A, Schmidt R (2003) Advanced mechanics of materials, 6th edn. Wiley, New York