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A Technique for Deducing In-Plane Modulus and Coefficient of Thermal Expansion of a Supported Thin Film

A technique for determining the in-plane modulus and the coefficient of thermal expansion (CTE) of supported thin films has been developed. The modulus and CTE are calculated by solving two coupled equations that relate the curvature of film samples deposited on two different substrates to the thermal and mechanical properties of the constituents. In contrast with the conventional method used to calculate modulus and CTE, which involves differentiation of the thermal stress in the film, this new technique does not require the differentiation of the thermal stress, and can also provide the temperature-dependence of the in-plane CTE and elastic modulus of supported thin films. The data reduction scheme used for deducing CTE and elastic modulus is direct and reliable.
[DOI: 10.1115/1.1448522]

Introduction

The in-plane elastic modulus and coefficient of thermal expansion (CTE) for a thin film residing on a substrate are important in many applications. One question arises as to whether the physical and mechanical properties of supported thin films in applications can be significantly different from the properties of chemically identical bulk materials. There have been many methods developed to measure these properties of thin films for the purpose of understanding the relation between the microstructure and behavior of material in the bulk and material in thin films coated on a substrate. There are many test methods available to probe the elastic modulus and/or CTE of supported thin films. The basic concept among them is that these properties can be deduced from the response of a film/substrate system perturbed either mechanically, thermally, acoustically, or optically through the corresponding governing equations [e.g., [1–5]].

Retajczyk and Sinha [6] proposed the two-substrate concept and developed a method for deducing CTE and elastic modulus of supported thin films. Their method is to measure the curvature as a function of temperature for identical films on two different substrate materials (two coated bimaterial circular plates), and the in-plane stress of the film is calculated from the curvature through Stoney's equation [7]. The slope of the curve of stress as a function of temperature is simply related to two unknowns, namely the bimodulus and in-plane CTE of the film, through an algebraic equation. Accordingly, these two unknowns are calculated from two coupled algebraic equations created using the two different substrates. In this method (referred to as the stress approach), the elastic modulus and CTE were assumed to be constant over the temperature range of interest in order to solve the coupled algebraic equations. This assumption that the properties are independent of temperature often might not be the case, especially for organic films. Many versions of the characterization method for thin film elastic modulus and in-plane CTE have been developed based on this two-substrate concept with the stress approach [e.g., [8–11]].

In this study, we developed a new approach (referred to as the strain approach) for deducing the elastic modulus and CTE of supported thin films. We also employ two substrates, similar to the method of Retajczyk and Sinha [6], in order to generate two coupled equations with two unknowns. As with Retajczyk and Sinha, the modulus and CTE are deduced from the temperature induced curvature of two coated bimaterial circular plates; however, in this approach, the modulus and CTE can be temperature dependent. The method for obtaining the modulus and CTE is a modification of the classic flexural solution for a laminated plate. The film properties are derived from the direct relationship among the changes in curvature as a function of temperature, the mismatch in the thermal expansion strain, and the known properties of the substrates. The strain approach presented here does not require the differentiation of thermal stress of the film. In this paper, the accuracy of the solution was also examined by finite element analysis. In addition, uncertainty analyses were performed to assess the sensitivity of the solution to the uncertainties of experimental variables.

Theoretical Aspects of the Technique

Figure 1 shows the cross-sectional bending of a bimaterial circular plate built up of a thin film and a substrate, due to a change in temperature. A perfect bond is assumed between the film and the substrate. If the edges of the uniformly heated bimaterial plate are entirely free (no constraints), the plate will deform to a spherical shape when its temperature (T) differs from a reference temperature (T_o) at which the plate is stress-free (flat). The above argument holds if the deflections of the plate are small in comparison with the thickness of the bimaterial plate and the materials are homogenous and isotropic. Thus, the bending curvature, $(1/R)$, at temperature T, can be inferred from a classic solution for a bimaterial plate [12,13] as follows:

$$\frac{1}{R}=\frac{6(\varepsilon_s-\varepsilon_f)(h_s+h_f)}{h_s^2 K}. \tag{1}$$

with

$$K=\frac{\overline{E}_s}{\overline{E}_f}\left(\frac{h_s}{h_f}\right)+4+6\frac{h_f}{h_s}+4\left(\frac{h_f}{h_s}\right)^2+\frac{\overline{E}_f}{\overline{E}_s}\left(\frac{h_f}{h_s}\right)^3, \tag{2}$$

$$\varepsilon_s=\int_{T_o}^{T}\alpha_s dT, \quad \text{and} \quad \varepsilon_f=\int_{T_o}^{T}\alpha_f dT. \tag{3}$$

Contributed by the Materials Division for publication in the JOURNAL OF ENGINEERING MATERIALS AND TECHNOLOGY. Manuscript received by the Materials Division April 25, 2001; revised manuscript received November 26, 2001. Associate Editor: N. Chandra.

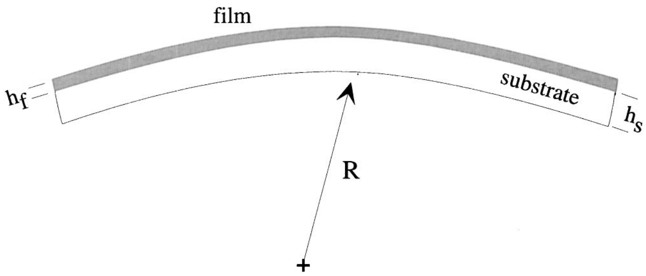

Fig. 1 A schematic of the cross-sectional bending of a bimaterial circular plate due to change in temperature

The subscript f represents the film and s represents the substrate. $\overline{E}_i = E_i/1-\nu_i$ $(i=s,f)$ is the bimodulus of material. E_i is the Young's modulus. ν_i is the Poisson's ratio. α_i is CTE. ε_i and h_i are stress-free thermal expansion strain and the thickness of the layers, respectively. R, the radius of the curvature at the temperature T, is determined experimentally. Although we assumed the reference temperature (T_o) as the temperature at which the bimaterial plate is stress free, T_o can be chosen at any temperature where experimental data are available, and $1/R$ in Eq. (1) should be changed to an incremental form $(\Delta 1/R)$.

For two different bimaterial plates (using the same film material but different substrate materials) with changes in curvature under an identical temperature history, in principle one can set up two coupled equations in form of Eq. (1) using the K expressed in Eq. (2). Thus, $\overline{E}_f$ can be obtained by solving a quadratic equation, and ε_f could be also derived afterward. However, in practice, the coefficient of the quadratic order term in the equation might endanger the stability of the solution in the case of a thin and soft film. Instead, if $\overline{E}_s/\overline{E}_f(h_s/h_f) \gg 1$ and $h_f/h_s < 1$, then K in Eq. (2) can be approximated as K^*:

$$K \simeq K^* \equiv \frac{\overline{E}_s}{\overline{E}_f}\left(\frac{h_s}{h_f}\right) \tag{4}$$

This approximation is valid for many organic film applications, since the film thickness is typically much smaller than that of the substrate, and the film stiffness is generally less than or comparable to that of the substrate. Then, for two different coated plates, one can rewrite Eq. (1), substituting K^* from Eq. (4), and get

$$R_i = \frac{1}{6(\varepsilon_{s_i}-\varepsilon_f)}\frac{\overline{E}_{s_i}}{\overline{E}_f}\frac{h_{s_i}^3}{h_{f_i}(h_{f_i}+h_{s_i})} \equiv S_i\overline{h_i} \tag{5}$$

with

$$S_i = \frac{1}{6(\varepsilon_{s_i}-\varepsilon_f)}\frac{\overline{E}_{s_i}}{\overline{E}_f} \tag{6}$$

and

$$\overline{h_i} = \frac{h_{s_i}^3}{h_{f_i}(h_{f_i}+h_{s_i})} \tag{7}$$

The index, i, ranges from 1 to 2, which corresponds to the two different bimaterial systems. S_i depends only on material properties and is a constant at any given temperature, while $\overline{h_i}$ is only a function of the geometry used in the experiments and is referred to as the effective thickness of a bimaterial plate. Therefore, for each bimaterial plate at temperature T, there is a linear relationship between these two physically measured variables, the curvature R and the effective thickness $\overline{h}$.

By solving two simultaneous equations of the form of Eq. (6), the stress-free strain (ε_f) and bimodulus $(\overline{E}_f)$ of a supported film at temperature T can be obtained from the following two formulas:

$$\varepsilon_f = \frac{\overline{E}_{s_2}S_1\varepsilon_{s_1} - \overline{E}_{s_1}S_2\varepsilon_{s_2}}{\overline{E}_{s_2}S_1 - \overline{E}_{s_1}S_2} \tag{8}$$

$$\overline{E}_f = \frac{\overline{E}_{s_2}S_1 - \overline{E}_{s_1}S_2}{6S_1S_2(\varepsilon_{s_2}-\varepsilon_{s_1})} \tag{9}$$

where S_i are determined through Eq. (5). By carrying out in situ curvature-temperature measurements on the film deposited on two different substrates subject to a thermal cycle, the temperature-dependent ε_f and $\overline{E}_f$ of a supported film can be deduced from a series of solutions to Eqs. (8) and (9). Also, Eqs. (8) and (9) can simply accommodate the effect of temperature dependence of the substrate modulus (if any). Once the temperature-dependent ε_f is determined, the CTE of film (α_f) can be obtained from the temperature derivative of ε_f as:

$$\alpha_f = \frac{d\varepsilon_f}{dT} \tag{10}$$

Theoretically, one measurement of R_i and $\overline{h_i}$ is needed at a given temperature for each bimaterial plate in order to obtain the corresponding S_i, and then the bimodulus as well as in-plane CTE can be determined through Eqs. (8)–(10). However, by using several specimens with different combinations of film and substrate thicknesses for each bimaterial plate, one can get additional measurements of curvature R with various $\overline{h}$ for each bimaterial plate and average these results (in the same way) to reduce the uncertainty in S_i. The uncertainty in S_i is readily related to the uncertainties of the calculated quantities (α_f and $\overline{E}_f$). This uncertainty issue will be addressed later.

The stress approach of Retajczyk and Sinha [6] gives the estimates of average values for the modulus and CTE over the temperature range studied. The assumption of temperature-independence of the modulus and CTE often might not be valid for polymers. One may argue that by using small temperature increments, a series of two coupled equations can be set up, and one can subsequently obtain the temperature-dependent elastic modulus and CTE by solving them. However, the insensitivity of stress differentiation to a small temperature range may affect the stability of the solution for the two algebraic equations created by the stress approach. Furthermore, from a statistical point of view, the uncertainty in the stress-temperature slope approach (differentiating step) can significantly affect the accuracy of solutions for the elastic modulus and CTE, since the scatter of curvature data with temperature usually is larger than the uncertainty of a single measurement of the curvature.

In the strain approach proposed in this work, the film properties are derived from the direct relationship among the changes in curvature as a function of temperature, the mismatch thermal expansion strain, and the known properties of the substrates. The technique presented here does not invoke the differentiating of thermal stress of a film and can provide the temperature-dependent in-plane modulus and CTE of a supported thin film at any temperature of interest. The temperature-dependent substrate properties (if any) can also be incorporated into the solution. Finally, although, Eq. (10) involves the differentiation of strain with respect to temperature to obtain the CTE of film, this differentiation will not affect the solution accuracy of the two algebraic equations in the form of Eqs. (8) and (9). However, in the stress approach, the accuracy of stress differentiation can significantly affect the solutions for both the elastic modulus and CTE of the film.

If the known properties of the substrates and the expected properties of film are not temperature dependent, then the thermal strain ε_i in Eq. (3) can be replaced in Eqs. (8) and (9) by $\alpha_i(T-T_o)$. Consequently, the CTE and bimodulus of a supported film can be obtained directly from the following two formulas:

$$\alpha_f=\frac{\bar{E}_{s_2}S_1\alpha_{s_1}-\bar{E}_{s_1}S_2\alpha_{s_2}}{\bar{E}_{s_2}S_1-\bar{E}_{s_1}S_2} \tag{11}$$

$$\bar{E}_f=\frac{\bar{E}_{s_2}S_1-\bar{E}_{s_1}S_2}{6S_1S_2(\alpha_{s_2}-\alpha_{s_1})(T-T_o)} \tag{12}$$

So far, the aforementioned derivation has demonstrated that the α_f and $\bar{E}_f$ of films are calculated from formulas using S_i obtained from two constituent measurements (R_i and h_i). The uncertainty in S_i is a function of the uncertainties in R_i and h_i. The number of specimens used and method of averaging the data will also affect the uncertainty in S_i (see [14], for example). Our concern in this study is not with the details of the experiment used to obtain S_i and the uncertainty in S_i, but instead we are interested in how the uncertainty in S_i propagates through the calculation of α_f and $\bar{E}_f$ in this approach. In some cases, because of the selection of the film/substrate systems, the calculations could produce large errors in the final results. For illustration purposes we consider the CTE and elastic modulus are not temperature dependent, and that other uncertainties are negligible compared to the uncertainties in S_i (denoted δS_i). Thus, the value for the relative uncertainty of the calculated elastic modulus and CTE ($\delta\alpha_f/\alpha_f$ and $\delta\bar{E}_f/\bar{E}_f$) can be formulated based on Eqs. (11) and (12) as:

$$\frac{\delta\alpha_f}{\alpha_f}=\left|\frac{\left(1-\frac{\alpha_{s_1}}{\alpha_f}\right)\left(1-\frac{\alpha_{s_2}}{\alpha_f}\right)}{\left(\frac{\alpha_{s_i}}{\alpha_f}-\frac{\alpha_{s_2}}{\alpha_f}\right)}\right|\sqrt{\left(\frac{\delta S_1}{S_1}\right)^2+\left(\frac{\delta S_2}{S_2}\right)^2}\equiv M_\alpha\frac{\delta S_2}{S_2} \tag{13}$$

and

$$\frac{\delta\bar{E}_f}{\bar{E}_f}=\frac{\sqrt{\left(\frac{\alpha_{s_1}}{\alpha_f}-1\right)^2\left(\frac{\delta S_1}{S_1}\right)^2+\left(\frac{\alpha_{s_2}}{\alpha_f}-1\right)^2\left(\frac{\delta S_2}{S_2}\right)^2}}{\left|\frac{\alpha_{s_1}}{\alpha_f}-\frac{\alpha_{s_2}}{\alpha_f}\right|}\equiv M_e\frac{\delta S_2}{S_2} \tag{14}$$

where

$$M_\alpha=\left|\frac{\left(1-\frac{\alpha_{s_1}}{\alpha_f}\right)\left(1-\frac{\alpha_{s_2}}{\alpha_f}\right)}{\left(\frac{\alpha_{s_1}}{\alpha_f}-\frac{\alpha_{s_2}}{\alpha_f}\right)}\right|\sqrt{\beta^2+1} \tag{15}$$

and

$$M_e=\frac{\sqrt{\left(\frac{\alpha_{s_1}}{\alpha_f}-1\right)^2\beta^2+\left(\frac{\alpha_{s_2}}{\alpha_f}-1\right)^2}}{\left|\frac{\alpha_{s_1}}{\alpha_f}-\frac{\alpha_{s_2}}{\alpha_f}\right|} \tag{16}$$

$$\text{with}\quad \beta\equiv\left(\frac{\delta S_1}{S_1}\right)\Big/\left(\frac{\delta S_2}{S_2}\right) \tag{17}$$

M_α is the relative uncertainty propagation factor for CTE. M_e is the relative uncertainty propagation factor for the elastic modulus.

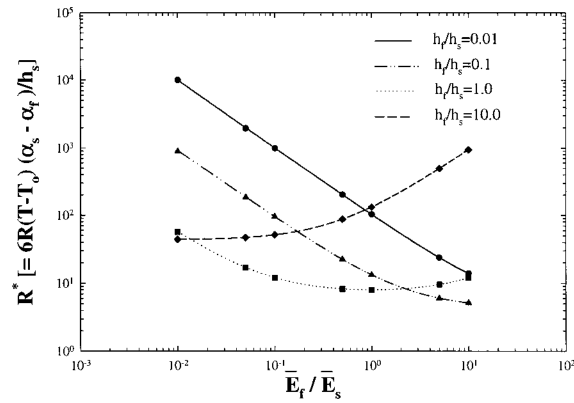

Fig. 2 The variation of the normalized radius of curvature as function of the film deformability and film/substrate stiffness ratio. The lines represent the curvature based on Eq. (1) while the symbols are the results of FEA.

Also, one notices that if the indexes, 1 and 2, in Eqs. (8)–(17) are interchanged, the uncertainty results still remain unchanged. This is expected since there is no preference in the sequence for selecting the substrate. Thus, once the relative uncertainties, $\delta S_1/S_1$ and $\delta S_2/S_2$, are determined from the measurements, and α_f and $\bar{E}_f$ of the film are obtained, the relative uncertainty of α_f and $\bar{E}_f$ can be readily obtained from Eqs. (13) and (14).

Results and Discussion

The Abaqus finite element program [15] was employed to validate the solution presented in Eq. (1) based on the specimen shown in Fig. 1. The elastic modulus and CTE of film and substrate were assumed to be temperature independent. Eight-node axially-symmetric elements and thermal stress analysis were used. The thickness of the substrate was 10 μm. The film thickness was set as a variable in the finite element analysis, ranging from 0.1 μm to 100μm for various combinations of film/substrate stiffness ratios. In order to minimize shear deformation, the aspect ratio of the plate was kept around 60. The aspect ratio of the finite elements were kept less than 10. Convergence of the finite element solutions, particularly the displacement at the center of the plate, was assessed by employing more refined meshes. The applicable range of Eq. (1) was evaluated by comparing the curvature results of Eq. (1) with that of the finite element analysis (FEA).

The variation of the normalized radius of curvature (R^*) as a function of the film deformability is shown in Fig. 2 as dependent on the film/substrate stiffness ratio. The lines in the figure represent the normalized curvature based on Eq. (1) with K expressed in Eq. (2) while the symbols are the results of FEA. One can see, from Fig. 2, that Eq. (1) does account for the change in curvature of a coated plate at a fixed temperature over the range of thickness and stiffness ratios considered in this study. A noteworthy feature of the results in Fig. 2 is the sensitivity of the change in curvature to the presence of the film. As shown in the figure, for realistic specimen dimensions in thin film applications ($h_f/h_s<1$), the change in curvature is extremely sensitive to the film/substrate modulus ratios of interest over a range of 4 decades.

Figure 3 presents the dependence of the ratio K^*/K on the ratios of the film to substrate stiffness and of the film to substrate thickness. From the results of FEA, the applicability of replacing K with K^* in Eq. (1) can be assessed by the deviation of the K^*/K curve from $K^*/K=1$. Accordingly, the appropriateness of the two formulas in Eqs. (6) and (7) for deducing the elastic modulus and CTE can be evaluated. For example, if $h_f/h_s \leqslant 0.001$, the expression in Eqs. (6) and (7) can be valid for any range of stiffness ratio considered in this study.

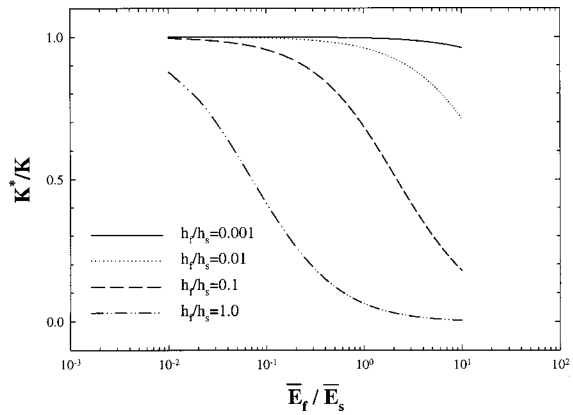

Fig. 3 The dependence of the K^*/K on the ratios of the film to substrate stiffness and of the film to substrate thickness

Both Eqs. (15) and (16) show that when $\alpha_{s_1} \approx \alpha_{s_2}$, the value of M_α and M_e becomes singular. This implies that the solutions for α_f and $\bar{E}_f$ in Eqs. (11) and (12) will be unstable and sensitive to any imperfection in the measurements. This is the basic reason that why the two substrates need to be two different materials with a gross difference in CTE. The intensity of the singularity for M_e or M_α depends on the value of α_{s_1}/α_f and α_{s_2}/α_f. Figure 4 indicates ranges of M_α and M_e for certain combinations of α_{s_1}/α_f and α_{s_2}/α_f corresponding to $\beta = 1$, where it is assumed that the relative uncertainties of S_1 and S_2 for the two bimaterial plates are the same. For example, when the CTE of the film is larger than that of both substrates (i.e., $\alpha_{s_1}/\alpha_f < 1$ and $\alpha_{s_2}/\alpha_f < 1$), the uncertainty propagation factor for the CTE is smaller than that of elastic modulus ($M_\alpha < M_e$). In other words, the calculated elastic modulus is more sensitive to imperfection in the measurements than the calculated CTE. One can also see from Fig. 4 that as long as one of the substrates has a larger CTE than the film and the other substrate has a smaller CTE than the film, the magnitude of M_α is less than $\sqrt{2}$ and the magnitude of M_e is less than 1. Such a combination of bimaterial plates will enhance the solution accuracy for the calculated elastic modulus and CTE. Although the above discussion is based on the assumption that $\beta = 1$, our analysis shows that the general information provided in Fig. 4 is applicable for β other than one. Thus, this uncertainty analysis can provide a guidance for optimal selection of film/substrate combinations for deducing the desired thin film modulus and CTE. Finally, in a separate study, preliminary testing with a polymeric film deposited on two different inorganic substrates has shown the feasibility of this technique in deducing the temperature-dependent, in-plane CTE and modulus of the film.

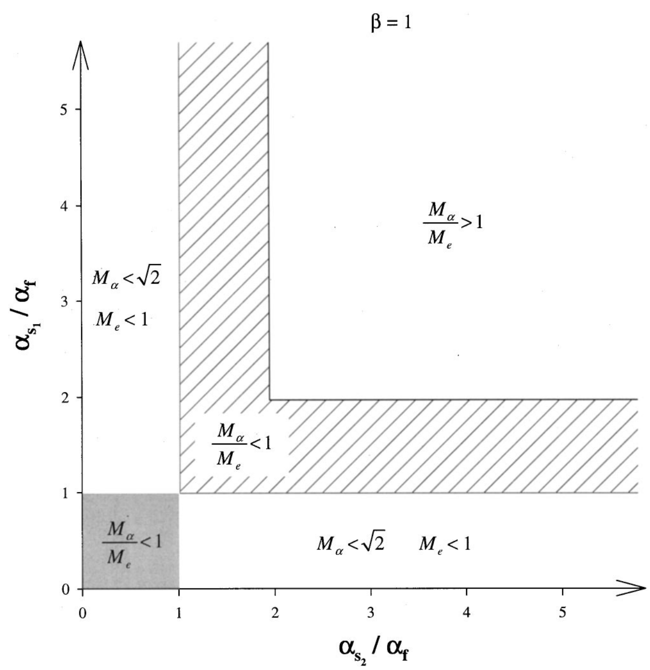

Fig. 4 Ranges of the relative uncertainty propagation factor for CTE (M_α) and the elastic modulus (M_e) for certain combinations of film/substrate

Conclusions

A simple and direct method has been developed for deducing the in-plane elastic modulus and CTE of a supported thin film. The modulus and CTE are calculated by solving two coupled equations that relate the thermally induced curvature of film samples deposited on two different substrates with the thermal and mechanical properties of the constituents. This strain approach does not require a temperature differentiation to calculate the modulus and in-plane CTE, and can provide the temperature-dependence of these properties for a supported thin film at any temperature of interest. The sensitivity analysis for the proposed solution method provides a guideline for choosing the appropriate substrate pair to improve the accuracy in the calculated elastic modulus and CTE.

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