Mechanism of Surface Wrinkle Modulation for a Stiff Film on Compliant Substrate

In this work, the surface wrinkle modulation of the film/substrate system caused by eigenstrain in the film is studied. A theoretical model is proposed which shows the change of the wrinkle amplitude is completely determined by four dimensionless parameters, i.e., the eigenstrain in the film, the plane strain modulus ratio between the film and the substrate, the film thickness to wrinkle wavelength ratio, and the initial wrinkle amplitude to wavelength ratio. The surface wrinkle amplitude becomes smaller (even almost flat) for the contraction eigenstrain in the film, while for the expansion eigenstrain it becomes larger. If the expansion eigenstrain exceeds a critical value, secondary wrinkling on top of the existing one is observed for some cases. In general, the deformation diagram of the wrinkled film/substrate system can be divided into three regions, i.e., the change of surface wrinkle amplitude, the irregular wrinkling, and the secondary wrinkling, governed by the four parameters above. Parallel finite element method (FEM) simulations are carried out which have good agreement with the theoretical predictions. The findings may be useful to guide the design and performance of stretchable electronics, cosmetic products, biomedical engineering, soft materials, and devices. [DOI: 10.1115/1.4036256]
system, examples such as post-buckling [12,15], creasing [16], folding [17,18], and film delamination [19,20].

Most previous studies focused on how wrinkles emerge from a thin film on a smooth substrate, i.e., the formation of wrinkles [21–26]. Often overlooked is the mechanical behavior of a surface that has already wrinkled. It is of great interest to explore whether one can remove, exemplify, or manipulate an existing surface morphology. For example, Xiao et al. found that in a wrinkled film/substrate system, the wrinkle amplitude decreases if external tension strain is applied to the wrinkled film/substrate system [27]. However, to the best of our knowledge, the surface modulation of a wrinkled film/substrate system has not been systematically studied.

The modulation of a wrinkled surface is important in many practical applications. Skin is a very important organ of human body that not only plays an important role to protect human body from environmental injury but also is an important part of cosmetology. As skin ages, its mechanical integrity is degraded, such as the loss of elastic recoil and baseline laxity. Therefore, wrinkles are generated in the aged skin [28]. Many wrinkle removal products are very popular in the cosmetic market. One effective way of wrinkle removal is to change the tension in the skin, through surgical or nonsurgical ways, this may be regarded as, from a mechanics point of view, changing the effective eigenstrain in the skin. Recently, an elastic second skin has been reported which can partly mimic the functionalities of human skin [29]. The elastic second skin consists of a wearable crosslinked polymer layer that can reshape the skin surface and erase skin wrinkles by shrinkage of the polymer layer due to the solvent loss. Again, the wrinkle erasing function of the elastic second skin can be modeled as the wrinkle modulation of the film/substrate system caused by the eigenstrain in the film.

The amplitude and the wavelength are also very important factors for flexible electronics, since they are closely related to maximum achievable stretchability, which is the critical factor in flexible electronics. Inspired by these applications, in this work, we focus on studying the wrinkling evolution of a wrinkled film/substrate system caused by eigenstrain in the film. Besides cosmetics and flexible electronics, the findings in this study may be extendable to a variety of applications in biomedical engineering, wearable devices, and micro/nanofabrication, to name a few.

The paper is organized as follows. In Sec. 2, a theoretical model is proposed to describe the deformation of the wrinkled film/substrate system under different eigenstrain values in the film. In Sec. 3, parallel FEM simulations are carried out to verify the theoretical model. The effects of eigenstrain in the film, film modulus, thickness, and morphology of the wrinkles on the surface wrinkle modulation are explored. The phase diagram of the deformation patterns of the wrinkled film/substrate system is given. Finally, Sec. 4 summarizes the main conclusions drawn from this study.

2 Theoretical Analysis

The wrinkled film/substrate system studied herein is schematically illustrated in Fig. 1. At the initial state, a film is adhered to the wrinkled substrate, as shown in Fig. 1(a). Here, the initial state is assumed as a stress free state which can be achieved by bonding a curved film with the same wrinkles to the substrate or directly coating a film on the wrinkled substrate [27,30]. Then, an eigenstrain $\varepsilon_{com}$ is induced in the film, which may be generated by the thermal loading (thermal expansion) [26,31,32], mechanical loading [1,33], solvent loss in the film, and other mechanisms [34,35]. Due to the eigenstrain in the film, the amplitudes of the surface wrinkles may be decreased (increased) depending on the contraction (expansion) of the film. In Sec. 2.1, a theoretical model is developed to describe the surface wrinkle modulation caused by eigenstrain in the film. The constitutive relations of the film and substrate are assumed as linear elasticity with Young’s modulus and Poisson’s ratio $E_f$, $v_f$, $E_s$, and $v_s$, respectively. The film is assumed to be perfectly bonded to the substrate during the deformation of the wrinkled film/substrate system.

2.1 Theoretical Model. Here, the wrinkles of the film/substrate system consist of one-dimensional sinusoidal wave for the simplification of theoretical analysis, which are described as $w = A \cos kx$, where $A$ is the initial wrinkle amplitude, $k = 2\pi/\lambda$ is the wave number, and $\lambda$ is the wavelength, as shown in Fig. 1(a). The thickness of the film is $h$, while the substrate is treated as a semi-infinite body due to its much larger thickness than that of the film. Besides, the deformation in $z$ direction is ignored, so that the deformation of the wrinkled film/substrate system is simplified as a plane strain problem.

In order to modulate the surface wrinkles, a uniform eigenstrain along the longitudinal direction of the film $\varepsilon_{com}$ is induced in the film. Indeed, the surface wrinkle modulation is dominated by the eigenstrain along the longitudinal direction of the film, and we have also tested the cases with biaxial and triaxial isotropic eigenstrains which have the similar results. Due to the constraint of the substrate, a tensile (compressive) stress is generated in the film for...
the contraction (expansion) eigenstrain, which results in the smoothing (roughening) of the wrinkles, as shown in Figs. 1(b) and 1(c).

When an eigenstrain is induced in the film, the wrinkles of the film/substrate change from \( w \) to \( w_1 \) and the membrane strain in the film can be expressed as

\[
e_1 = \frac{\partial u_1}{\partial x_1} + \frac{1}{2} \left[ \left( \frac{\partial w_1}{\partial x_1} \right)^2 - \left( \frac{\partial w}{\partial x_1} \right)^2 \right]
\]

(1)

where \( u_1 \) is the displacement of the film in \( x \) direction, and \( w_1 = A_1 \cos kx \). As the dimension of the wrinkled film/substrate in \( x \) direction is much larger than the wavelength of the wrinkles, we assume that the wrinkle wavelength is constant before and after applying eigenstrain in the film. Further FEM simulations have proved the validity of this assumption. Besides, the shear stress between the film and the substrate is ignored in consistent with the previous literatures of studying the buckling behaviors of a thin film on compliant substrate [26].

Based on Eq. (1), the membrane stress in the film is expressed as

\[
\sigma_1 = E_f (e_1 - e_{\text{com}})
\]

(2)

where \( E_f = E_f / (1 - \nu_f^2) \) is the plane strain modulus of the film, and \( e_{\text{com}} \) is the uniaxial eigenstrain in the film. By ignoring the shear stress between the film and the substrate, the membrane stress is constant, which gives

\[
d\sigma_1 / dx = E_f d\varepsilon_1 / dx = 0
\]

(3)

Then, the uniaxial displacement in the film \( u_1 \) is obtained by solving Eq. (3) with respect to \( x \)

\[
u_1 = ax + c + \frac{k}{8} \left[ A^2 - (A + \Delta A)^2 \right] \sin 2kx
\]

(4)

where \( \Delta A = A_1 - A \) is the wrinkle amplitude change caused by eigenstrain in the film. There are two undetermined parameters \( (a \) and \( c) \), which can be deduced by assuming that the rigid body displacement of the film is zero and the wavelength of the wrinkles is constant. According to the boundary conditions, the axial displacement in the film is then obtained

\[
u_1 = -\frac{k}{8} \left[ A^2 - (A + \Delta A)^2 \right] \sin 2kx
\]

(5)

Substituting Eq. (5) into Eq. (1), the membrane strain in the film is expressed as

\[
e_1 = -\frac{k^2}{4} \left[ A^2 - (A + \Delta A)^2 \right]
\]

(6)

With respect to the force equilibrium of the film, the normal traction force acting on the film is

\[
q = -\frac{E_f h^3}{12} d^2 \Delta \omega + \sigma_f h d^2 w_1 / dx^2
\]

(7)

Note that as the initial configuration of the film is wrinkled, we use the change of the wrinkles \( \Delta \omega \) to calculate the normal traction force caused by bending of the film, i.e., the first term in right hand of Eq. (7). Then, substituting Eqs. (2) and (6) into Eq. (7), the normal traction force is represented as

\[
q = -\frac{E_f h^3}{12} d^2 \Delta \omega + \sigma_f h \left( -\frac{k^2}{4} \left[ (A + \Delta A)^2 - A^2 \right] - e_{\text{com}} \right) \ast d^2 w_1 / dx^2
\]

(8)

For the semi-infinite substrate, as illustrated in the previous literatures [20,26], the relation between the normal loading \( q \) and the deflection of the surface \( \Delta \omega \) is

\[
q = \frac{k E_s \Delta w}{2}
\]

(9)

Substituting Eq. (8) into Eq. (9) to eliminate the normal traction force \( q \), we obtain the governing equation for the change of the wrinkle amplitude \( \Delta A / A \) induced by eigenstrain in the film

\[
\frac{\Delta A}{A} = \frac{2E_f}{E_s} \left[ -\frac{2k^3 h^3 \Delta A}{3} A \right. \left. - \frac{2\pi h}{k} \left( \frac{\pi A^2}{2A^2} \left( \left( 1 + \frac{\Delta A}{A} \right)^2 - 1 \right) - e_{\text{com}} \right) \ast \left( 1 + \frac{\Delta A}{A} \right) \right]
\]

(10)

It is a cubic equation for the variable of \( \Delta A / A \) and its solution is determined by four parameters, i.e., the eigenstrain \( e_{\text{com}} \) in the film, the plane strain modulus ratio \( E_f / E_s \), the film thickness to wavelength ratio \( h / \lambda \), and the initial wrinkle amplitude to wavelength ratio \( A / \lambda \). Solving Eq. (10) with respect to \( \Delta A / A \), the change of the wrinkle amplitude is expressed as

\[
\frac{\Delta A}{A} = \left[ \frac{1}{8} \frac{E_s \lambda^3}{E_f A^2 h} + \frac{h^2}{6A^2} \left( \frac{1}{8} \frac{E_s \lambda^3}{E_f A^2 h} + \frac{h^2}{6A^2} \right)^2 + \frac{1}{12} \frac{E_s \lambda^3}{E_f A^2 h} + \left( \frac{h^2}{9A^2} - \frac{1}{3} \frac{\pi \lambda^2}{3\pi A^2 e_{\text{com}} - 1/3} \right) \right]^{1/3}
\]

(11)

In order to verify the analytical solution of the wrinkle amplitude change \( \Delta A / A \), FEM simulations are carried out in Sec. 3, and the relations of \( \Delta A / A \) to the three dimensionless parameters \( E_f / E_s \), \( h / \lambda \), and \( A / \lambda \) under contraction and expansion eigenstrain are studied, respectively.

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2.2 Critical Eigenstrain for Secondary Wrinkling. For the expansion eigenstrain, the membrane stress in the film is compressive which will roughen the wrinkled surface. However, as the stiffness of the film is usually much larger than that of the substrate and the film thickness is small, the compressive stress may lead the buckling of the thin film if it is larger than a critical value.
The film buckling occurs on a wrinkled surface, so we call it secondary wrinkling. The critical buckling stress for a film bonded to a flat compliant substrate is \( \sigma^*_{\text{com}} = -\frac{1}{3} \frac{E_f}{E_s} (3E_s/E_f)^{2/3} \) corresponding to the critical buckling strain \( \varepsilon^* = -\frac{1}{3} (3E_s/E_f)^{2/3} \) and wavelength \( \lambda^* = 2\pi h (E_f/E_s)^{1/3} \) for the onset of buckling. If the ratio of the initial amplitude to wavelength approaches to zero, the buckling behaviors of the film will degenerate to that of the film bonded to a flat compliant surface.

According to Eqs. (6) and (10), the relation between the membrane strain and the eigenstrain for the critical buckling stress is

\[
\varepsilon^*_{\text{com}} = \frac{\left( \frac{\Delta A}{A} \right)^3 + 3 \left( \frac{\Delta A}{A} \right)^2 + \frac{3}{2} \left( \frac{E_s}{E_f} \right)^{2/3} \left( \frac{1}{4\pi^2} \frac{E_s}{E_f} \lambda^3 + \frac{\lambda^2}{3A} + 2 \right)}{1 + \frac{\Delta A}{A}} \frac{1}{1 - \left( \frac{\Delta A}{A} \right)^2 - 1^2} \varepsilon_1
\]

(12)

Substituting the solution of \( \Delta A/A \) (Eq. (11)) into Eq. (12), the relation between the membrane strain \( \varepsilon_1 \) and \( \varepsilon^*_{\text{com}} \) is plotted in Fig. 2. According to Eq. (2), the relation between the membrane stress \( \varepsilon_1 \) and \( \varepsilon^*_{\text{com}} \) is also plotted in Fig. 2. As the eigenstrain in the film increases, the membrane stress decreases, and the secondary buckling may occur when the membrane stress gets to the critical buckling stress \( \sigma^*_{\text{com}} \). The value of the wrinkle amplitude change \( \Delta A/A \) at the critical buckling stress is derived as

\[
\Delta A^*/A = \frac{\left( \frac{E_s}{E_f} \right)^{2/3}}{\pi h E_f} \frac{E_s}{3 \lambda^2} + \left( \frac{3E_s}{E_f} \right)^{2/3}
\]

(13)

and the membrane strain \( \varepsilon^*_1 \) in the film for the critical buckling stress is

\[
\varepsilon^*_1 = \frac{\pi^2 A^2}{2^2} \left[ \frac{1 + \frac{3E_s}{E_f}}{1 + \frac{E_s}{\pi h E_f} + \frac{4\pi^2 h^2}{3\lambda^2} + \left( \frac{3E_s}{E_f} \right)^{2/3}} - 1 \right] \left( \frac{3E_s}{E_f} \right)^{2/3}
\]

(14)

Then, substituting Eqs. (13) and (14) into Eq. (2), the critical eigenstrain for the critical buckling stress is

\[
\varepsilon^*_{\text{com}} = \frac{1}{4} \left( \frac{3E_s}{E_f} \right)^{2/3} \frac{\pi^2 A^2}{2^2} \left[ 1 + \frac{E_s}{\pi h E_f} + \frac{4\pi^2 h^2}{3\lambda^2} + \left( \frac{3E_s}{E_f} \right)^{2/3} \right] \left( \frac{3E_s}{E_f} \right)^{2/3} - 1
\]

(15)

The critical buckling stress and eigenstrain are marked in Fig. 2. If the applied eigenstrain in the film is larger than \( \varepsilon^*_{\text{com}} \), the secondary buckling may occur. In Sec. 3.3, FEM simulations are carried out to verify the secondary buckling behavior, and the effects of \( \varepsilon^*_{\text{com}} \) and initial wrinkle wavelength \( \lambda \) on the secondary buckling will be systematically explored.

### 3 Finite Element Method Simulations

In order to verify the theoretical results in Sec. 2, FEM simulations are carried out via commercial software, **ABAQUS**. The film and the substrate are described by plane strain element in consistent with the theoretical analysis, and the film is perfectly bonded to the substrate via the tie procedure in **ABAQUS**. The displacement at the bottom and lateral surfaces of the substrate is fixed as shown in Fig. 1. Different eigenstrains in the film are induced by applying different thermal expansion strains in the film. Here, the length of the wrinkled film/substrate system (dimension in \( x \) direction) is much larger than the wavelength of the surface wrinkles, so the deformation in every wrinkle is identical. Besides, the thickness of the substrate (dimension in \( y \) direction) is also much larger than the initial amplitude of the surface wrinkles, so that the substrate can be treated as a semi-infinite body. The constitutive relations of the film and substrate are linear elasticity with Poisson’s ratio of \( v_f = v_s = 0.3 \). The mesh convergence is checked to ensure the accuracy of the numerical results.

#### 3.1 Evolution of the Wrinkle Amplitude Caused by Eigenstrain in the Film

The evolution of the surface wrinkles for different contraction and expansion eigenstrains in the film is shown in Fig. 3, respectively. Here, the ratio of the plane strain modulus between the film and the substrate is \( E_f/E_s = 300 \), the film thickness is \( h = 0.5 \text{ mm} \), the wavelength of the surface wrinkles is \( \lambda = 10 \text{ mm} \), and the initial amplitude is \( A = 0.5 \text{ mm} \). For the contraction eigenstrain in the film, the wrinkle amplitude decreases as the eigenstrain increases, and it becomes almost flat as the contraction eigenstrain exceeds \(-0.04 \), as shown in Fig. 3(a). In contrast, for the expansion eigenstrain in the film, the wrinkle amplitude gradually increases as the eigenstrain increases, as shown in Fig. 3(b). Besides, the membrane strain \( \varepsilon_1 \) in the film is almost uniform, and the wavelength is constant during the contraction and expansion of the film, both of which are consistent with the assumptions in the theoretical analysis of Sec. 2. The change of the surface wrinkle amplitude during FEM simulation is tracked for further quantitative comparison to the theoretical predictions in Sec. 2.

The relations between the relative change of the wrinkle amplitude \( \Delta A/A \) and the applied eigenstrain \( \varepsilon^*_{\text{com}} \) in the film are given in

![Fig. 2 The relations of the membrane strain (solid line) and membrane stress (dashed line) to the applied eigenstrain in the film. The marked point corresponds to the critical buckling stress \( \sigma^*_1 \) and eigenstrain \( \varepsilon^*_{\text{com}} \). Secondary buckling may occur for \( \varepsilon^*_{\text{com}} \) greater than \( \varepsilon^*_{\text{com}} \). Here, the ratio of the plane strain modulus between the film and the substrate is \( E_f/E_s = 50 \), film thickness to wavelength ratio \( h/\lambda = 0.025 \), and initial wrinkle amplitude to wavelength ratio \( A/\lambda = 0.05 \).](image-url)
Fig. 3 The evolution of the surface wrinkles as the eigenstrain induced in the film. (a) The deformed configurations of the wrinkled film/substrate system for the contraction eigenstrain from $\varepsilon_{\text{com}} = 0$ to $\varepsilon_{\text{com}} = -0.1$ and (b) for the expansion eigenstrain from $\varepsilon_{\text{com}} = 0$ to $\varepsilon_{\text{com}} = 0.1$, where $E_f/E_s = 300$, $h = 0.5\text{ mm}$, $\lambda = 10\text{ mm}$, and $A = 0.5\text{ mm}$. The color contour represents the distribution of $\varepsilon_1$.

3.2 Effects of Film Modulus, Thickness, and Wrinkle Amplitude on the Wrinkle Modulation. Based on the theoretical analysis in Sec. 2, the change of the surface amplitude $\Delta A/A$ is completely determined by $E_f/E_s$, $h/\lambda$, $A/\lambda$, and eigenstrain $\varepsilon_{\text{com}}$ in the film. In this part, the effects of the film modulus, thickness, and initial wrinkle amplitude on the wrinkle modulation are studied, and the results are shown in Fig. 5. Here, the eigenstrain in the film is $-10\%$ for contraction and $10\%$ for expansion. Besides, the mechanical properties of the substrate are the same for all of the cases.

In general, the magnitude of $\Delta A/A$ quickly increases for the plane strain modulus ratio $E_f/E_s < 50$, and then it approaches an asymptotic value, as shown in Figs. 5(a) and 5(b). For the contraction eigenstrain in the film, the asymptotic values of $\Delta A/A$ approach $-1$ for different initial wrinkle amplitudes $A/\lambda = 0.05$ and $A/\lambda = 0.025$, which means that the wrinkled surface becomes almost flat. Meanwhile, for the expansion eigenstrain in the film, the asymptotic values are different, and the smaller initial wrinkle amplitude has larger $\Delta A/A$. The film thickness is another important parameter to determine the wrinkle modulation. As shown in Figs. 5(c) and 5(d), the magnitude of $\Delta A/A$ first increases, and then it decreases with the increasing of $h/\lambda$. The optimum value of $h/\lambda$ for the wrinkle modulation is about 0.05. Therefore, appropriate wrinkle modulation can be achieved by properly selecting the modulus and thickness of the film (e.g., $E_f/E_s > 50$ and $h/\lambda = 0.05$). The mechanism of the wrinkle modulation is the competition between the membrane strain energy in the film and the summation of the bending energy in the film and strain energy in the substrate. For the film with relatively small thickness, the small membrane strain energy will generate small wrinkle amplitude change, while for relatively large thickness, the larger bending energy will also prevent the variation of wrinkle amplitude. For the film with larger modulus, the membrane strain energy is also larger compared to the strain energy in the substrate which can generate a larger change in wrinkle amplitude. The asymptotic wrinkle amplitude is determined by the minimum strain energy of the film/substrate system.

Besides the film modulus and thickness, the wrinkle modulation is also influenced by the initial wrinkle morphology, such as the initial wrinkle amplitude $A$ and the wavelength $\lambda$. The absolute value of $\Delta A/A$ decreases with the increasing of $A/\lambda$. For the contraction eigenstrain, the reduction of $\Delta A/A$ is very small if $A/\lambda$ is smaller than 0.05, as shown in Figs. 5(e) and 5(f). Note that the
effect of wrinkle wavelength on the wrinkle modulation is explicitly included in the parameters $h/\lambda$ and $A/\lambda$. The theoretical predictions agree well with the FEM simulations for the ranges of parameters considered in Fig. 5, which further verify the validity of the theoretical model in Sec. 2.

3.3 Deformation Patterns of Wrinkled Film/Substrate System. In the aforementioned FEM simulations, the expansion eigenstrain is smaller than the critical eigenstrain for the secondary buckling (Eq. (15)). If we further increase the expansion eigenstrain to exceed a critical value, the secondary wrinkling may be observed in the FEM simulation depending on the ratio between the wrinkle wavelength $\lambda$ and the critical buckling wavelength of the film on flat substrate $\lambda^*$. In this subsection, the deformation patterns of the wrinkled film/substrate system will be explored.

As the expansion eigenstrain in the film raises, the surface wrinkle amplitude first increases, and then the secondary wrinkling is observed, as shown in Fig. 6(a). The relation of the strain energy in the film to the eigenstrain is plotted in Fig. 6(b). Here, a bifurcation point is found, i.e., when the second derivative of the strain energy–eigenstrain relationship reaches zero, as shown in the inset of Fig. 6(b). Therefore, the critical eigenstrain at the onset of secondary wrinkling is identified. Indeed, the membrane strain in the film predicted by the theoretical model (Eq. (12)) deviates from the FEM simulation results after the bifurcation point, as shown in Fig. 6(c). This is because the current theoretical model does not include the secondary wrinkling.

Fig. 5 Comparison of the relations of the wrinkle amplitude change to plane strain modulus ratio between theoretical predictions (Eq. (11)) and FEM simulations with $h/\lambda = 0.05$, $h/\lambda = 0.05$ (a) and $h/\lambda = 0.05$, $A/\lambda = 0.05$ (b), respectively. (c) and (d) Comparison of the relations of the wrinkle amplitude change to the film thickness $h/\lambda$ with $E_f/E_s = 50$, $A/\lambda = 0.05$ and $E_f/E_s = 50$, $A/\lambda = 0.05$, respectively. (e) and (f) Comparison of the relations of the wrinkle amplitude change to the initial wrinkle amplitude $A/\lambda$ with $E_f/E_s = 300$, $h/\lambda = 0.025$ and $E_f/E_s = 100$, $h/\lambda = 0.025$, respectively. Note that the negative value of $\Delta A/A$ corresponds to the contraction eigenstrain in the film, while the positive value of $\Delta A/A$ corresponds to the expansion eigenstrain.
According to the theoretical analysis in Sec. 2.2, the critical eigenstrain in the film at the onset of secondary wrinkling is determined by Eq. (15). Similarly, in the FEM simulation, the critical eigenstrain is defined as the eigenstrain at the bifurcation point of the strain energy–eigenstrain curve. The comparison between the theoretical prediction and the FEM simulation is given in Fig. 7, which shows good agreement for relatively small plane strain modulus ratio \( E_f/E_s \). However, the secondary wrinkling is not observed during FEM simulations for large plane strain modulus ratio \( E_f/E_s \), i.e., the dashed line in Fig. 7. This is because the initial buckling wavelength for a stiff film on compliant substrate is 
\[
\lambda^* = 2\pi h(3E_f/3E_s)^{1/3}
\]
which increases with \( E_f/E_s \). If \( \lambda^* \) is comparable to the wavelength of surface wrinkles, the secondary wrinkling is not easy to occur. In the following content, we will discuss the deformation diagram of the wrinkled film/substrate system caused by the eigenstrain in the film.

The wrinkle morphology (e.g., the wrinkle wavelength and the wrinkle amplitude) is another important parameter to influence the evolution of the surface wrinkles. In this work, we only
consider the effect of wrinkle wavelength on the surface wrinkle evolution and keep $A/\lambda = 0.025$ and $h/\lambda = 0.025$. If $A/\lambda$ approaches zero, the deformation of the wrinkled film/substrate should degenerate to the buckling behaviors of the film on a flat substrate. In general, the phase diagram of the deformation patterns of the wrinkled film/substrate system can be divided into three regions, as shown in Fig. 8. In the lower left region (region I), when $A/\lambda$ and $e_{com}/\varepsilon_{com}$ are relative small, it is only the wrinkle amplitude changes (with pure roughening), and no secondary buckling is observed even for the eigenstrain $\varepsilon_{com}/\varepsilon_{com} > 8$. Whereas, in the upper right region (region III), the deformation pattern dominated by the secondary wrinkling. Between region I and region III, there is a small irregular buckling region (region II), i.e., the initiation of the buckling irregularly locates at the film. The primary reason is that for relative small $\lambda/\lambda'$ the superposition of the buckling morphology of the film on the wrinkled configuration is energetically unfavorable even for large $e_{com}/\varepsilon_{com}$ and for large $\lambda/\lambda'$ the buckling of the film can behave like that on a flat substrate. For the intermediate $\lambda/\lambda'$, the superposition of the film buckling on the wrinkled configuration may cause irregular buckling pattern.

4 Conclusions

A theoretical model is developed to study the surface evolution of a wrinkled film/substrate system modulated by eigenstrain in the film. The surface profile evolution is governed by four dimensionless parameters, i.e., the eigenstrain in the film, the plane strain modulus ratio $E_f/E_s$, the film thickness to wrinkle wavelength ratio $h/\lambda$, and the initial wrinkle amplitude to wavelength ratio $A/\lambda$. The surface wrinkle amplitude becomes smaller (approaching flat) for the contraction eigenstrain in the film, while for the expansion eigenstrain it becomes larger. For a given eigenstrain in the film, the absolute change of the wrinkle amplitude $\Delta A/A$ first quickly increases with the increasing of $E_f/E_s$, then it gradually approaches an asymptotic value for $E_f/E_s > 50$. On the other hand, $\Delta A/A$ first increases and then decreases with the increasing of $h/\lambda$. Therefore, the preferred parameters for the surface wrinkle modulation are $E_f/E_s > 50$ and $h/\lambda = 0.05$. Besides, the surface wrinkle modulation effect is also influenced by the initial wrinkle amplitude $(A/\lambda)$, and the value of $\Delta A/A$ is larger for smaller $A/\lambda$. Parallel FEM simulations are carried out which show good agreement with the theoretical predictions. When the expansion eigenstrain in the film exceeds a critical value, the secondary wrinkling is observed. The phase diagram of the deformation patterns of the wrinkled film/substrate system can be divided into three regions, i.e., the “pure roughening,” the irregular wrinkling, and the secondary wrinkling, determined by $\lambda/\lambda'$ and $e_{com}/\varepsilon_{com}$. The results presented in this work shed some useful insights for the design and modulation of surface wrinkle morphology, which may have potential applications in stretchable electronics, cosmetic industry, soft materials, and engineering structures. Note that in some practical applications, such as skin, the structure is of multilayer, and the “film” and its corresponding properties should be regarded as effective ones. A more direct, quantitative application of the theory to skin and cosmetics will be pursued in future.

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Nomenclature

$A = $ initial amplitude of the surface wrinkles
$E_f = $ plane strain modulus of the film
$E_s = $ plane strain modulus of the compliant substrate

$h = $ thickness of the film
$\Delta A = $ the change of the wrinkle amplitude
$e_{com} = $ eigenstrain in the film
$\varepsilon_{com} = $ critical eigenstrain for the secondary buckling
$\lambda = $ wavelength of the surface wrinkles
$\lambda' = $ the initial buckling wavelength for a stiff film on flat compliant substrate

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