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Effects of pre-buckling on the bending of organic electronic structures

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This paper explores the extent to which pre-buckling of layers (in thin film multilayered structures) can be used to increase the flexibility of organic electronic devices. The deformation of wavy/buckle profiles, with a range of nano- and micro-scale wavelengths, is modeled using finite element simulations. The predictions from the models are then validated using experiments that involve the bending of layered structures that are relevant to flexible organic electronics. The introduction of pre-buckled profiles is shown to increase the range of deformation that is applied to model structures, prior to onset of significant stresses and strains. The implications of the work are discussed for the design of robust flexible organic solar cells. [http://dx.doi.org/10.1063/1.4975396]

I. INTRODUCTION

In recent years, there has been considerable interest in the development of low cost flexible organic solar cells with the potential to replace conventional silicon cells that are fabricated typically on glass substrates.[1–3] However, the rigidity of the glass substrates limits the extent to which flexible electronics structures can be deformed without inducing cracks in the rigid and brittle glass substrates.[4] There is, therefore, a need for approaches that can improve the deformation of flexible organic solar cells, without inducing cracks in the underlying substrates.[5]

Two approaches have been used in literature to improve the flexibility of organic solar cells structures.[6–13] One involves the replacement of the glass substrates with bendable polymers, such as polyethylene terephthalate (PET)[14–19] or polydimethylsiloxane (PDMS),[10,20–23] while the second

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approach involves the introduction of pre-buckles that stretch and flatten out during deformation. The pre-buckles also increase the range of deformation that can be applied to flexible organic electronics prior to the onset of cracking and other stress/strain-induced failure mechanisms.[24]

Prior work on the deformation of flexible and stretchable organic electronics structures has been reported by Bao et al.,[4,25] Volinski et al.,[26] Groenwold,[27] Huang,[28–30] Stafford et al.[31] and Sariciftci and co-workers.[6,32,33] Their work shows that pre-buckles with different wavelengths can improve the flexibility of layered organic solar cell structures with different substrates. Similar reports of improved deformability have also been presented by Huang and co-workers,[30,34–37] while Rogers et al.[38] and Akogwu et al.[20] have used pre-buckles to improve the "stretch-ability" of flexible organic electronic structures.

Bao et al.[4] have presented two methods that can be used to influence the elasticity of rigid materials. The first involves the dispersion of conductive materials in an elastic matrix (as exploited by Wagner,[39] Suo,[40] Rogers,[41] Sariciftci[33]), while the second involves the pre-buckling of electronic structures deposited on elastomeric substrates. The latter have been investigated by Stafford,[42] Sariciftci,[43] Tarasovs and Andersons.[44] The results show clearly that the potential application of pre-buckled films (with controlled wavelengths and amplitudes) could have a significant effect on the deformability and reliability of electronic devices.

Rogers et al.[38] have also suggested that the buckle wavelength increases with increasing buckle film thickness. However, there have been only limited prior efforts to model the deformation of flexible organics cell structures under bending loads and deformation conditions[45] that are relevant to flexible organic electronics.[41]

Furthermore, the flexibility of transparent electrodes has been achieved by structurally configuring devices to accommodate most of the strain under mechanical deformation that minimizes the strains in the conducting materials. These structural configurations have been classified into out-of-plane and in-plane structures.[47–50] The most common out-of-plane structure is the 'wavy' structure, which is generally obtained by depositing conductive materials on pre-strained elastomeric substrates. When subsequently released, these conductive films spontaneously form periodic wavy structures in which most of the induced strains are absorbed by the structural changes.[47] These have been likened to accordion bellows, where increased buckling wavelengths and decreased buckling amplitudes can be achieved.[47,51,52]

The fundamental advantage of the presented pre-buckled structure is its ability to permit large elastic deformation protecting the active layers with limited strains. Thin films fabricated this way are more compliant when compared to the bending strain which is less than that of pre-stretch.[47–53]

However, although the effects of pre-buckling have been studied,[53] there is only a limited understanding of pre-buckling on the deformation of flexible organic structures. Hence, this paper, therefore, presents the results of an experimental and computational study of the effects of this most common out-of-plane structure (pre-buckling) on the deformation and failure of flexible organic solar cell structures on polydimethylsiloxane (PDMS) substrates. The effects of pre-buckle wavelengths and amplitudes are modeled using finite element method. The stress/strain distributions and the deformation profiles associated with the bending of the pre-buckled profiles are computed for the model flexible multilayer structures deposited experimentally. The increased deformation associated with the flattening of the pre-buckles is determined for pre-buckles with different wavelengths and amplitudes. The implications of the results are then discussed for the design of flexible organic electronic structures.[54,55]

II. EXPERIMENTAL PROCEDURE

This section utilizes un-stretched, pre-stretched and post-stretched structures in a study of the bending of model organic semiconductor layers on flexible PDMS substrates. It is important to note that the un-stretched condition corresponds to the un-stretched PDMS substrate, while the pre-stretched condition corresponds to the stretched condition prior to the deposition of poly 3, 4-ethylenedioxythiophene polystyrene sulfonate (PEDOT:PSS) thin film layer. The post-stretched condition corresponds to the subsequent deformation of the pre-buckled structure that is formed after the release of the PEDOT:PSS/PDMS structure.

A. PDMS substrate preparation

The PDMS substrate was prepared from a Sylgard 184 elastomeric base and a silicone curing agent with 1:10 weight ratio, as described in our prior work.[56] Observations show that increasing the weight ratio of the silicon curing agent makes the resulting PDMS less stretchable, as expected, since silicon acts as the cross-link (hardener) agent between the polymer chains.[20] In any case, PDMS substrates with dimensions of 1.5mm and 4.5 cm × 1.25 cm were prepared by casting and curing them in an aluminum mold.

B. PEDOT:PSS coating deposition

PEDOT: PSS was procured from H.C. Starck Inc., Newton, MA, USA. It was filtered with a 0.2 μm filter paper before depositing 0.4 ml of the filtrate with the Model WS 650 Laurel spin-coater at an initial rate of 500 revolutions per minute (rpm) for 5 s. The PEDOT:PSS was then spin coated at 1500 rpm for 3 s and 3000 rpm for 60 s.[18] The principal challenge in depositing PEDOT:PSS on PDMS was its high water contact angle on PDMS. It was observed that when PDMS was stretched to a high degree before deposition, PEDOT:PSS deposition was possible. The pre-stretching experiments were carried out on 3D printed fixtures that were fabricated from polylactide (PLA). These were used for the deformation of the PEDOT:PSS/PDMS structures to different levels of pre-stretch. The fixtures were used to apply pre-stretch levels of 35% and 50% by varying the PLA stands (Figure 1a).

PDMS was pre-stretched by clamping to the PLA stands. PEDOT:PSS was spin coated while the PDMS is pre-stretched. Afterwards, the clamps were carefully removed to introduce the buckling. Since PDMS is not rigid enough to compensate the force of the water-air-surface tension perpendicular to the surface, a ridge is pulled up around the edge of the water drop, locally increasing the surface roughness and thus hysteresis via pre-stretching enhancing the PEDOT:PSS deposition.[57,58]

C. Surface characterization and bending experiments

Microscopic observations of the surfaces were made with atomic force microscopy (AFM) and optical microscopy (OM). The latter was performed under different stretching conditions. After pre-stretching, the PDMS substrates were spin-coated with PEDOT:PSS. They were then mounted on 3D rollers with different diameters (Figure 1b). Rollers with different diameters were used to apply different bending strains to the pre-buckled/wrinkled PEDOT:PSS on PDMS substrates.

Deformation was applied until the thin film flattened out on the substrates (Figure 1b). Atomic force microscopy (AFM) images of the coated and uncoated surfaces were then obtained in the contact mode, using a Bruker Instruments Nanoscope IIIa atomic force microscope (Bruker Instruments, Plainview, NY, USA). The three dimensional (3D) printed rollers with diameters of 20, 18, 16, 15, and 12 mm used to apply controlled bending strains of 0.0375, 0.0417, 0.0469, 0.0500 and 0.0625,

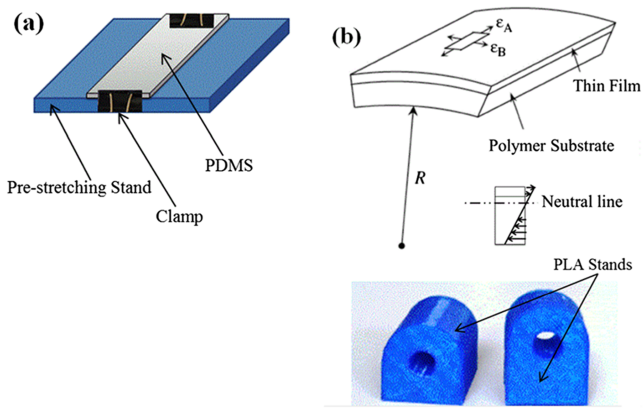

FIG. 1. (a) PDMS clamped to a pre-stretching stand made of PLA. (b) PEDOT:PSS thin film on a polymer substrate (PDMS) undergoing bending showing its acquired radius of curvature, R when placed on PLA stands with different diameters. (Ref. 62,87).

respectively. The axial strains were determined using the following expression:[59–63]

$$Strain = \frac{\left(\frac{Thickness - of - PDMS}{2}\right)}{Diametter - of - Roller} \quad (1)$$

AFM was then used to characterize the wavelengths of the surfaces of the wrinkled/buckled PEDOT:PSS layers on PDMS. This was done after pre-stretching and bending to different strain levels. AFM imaging was carried out in the contact mode using a Bruker Instruments Dimension 3000 atomic force microscope (Bruker Instruments, Plainview, NY, USA).

III. MODELING

Finite element simulations where used to model the effects of bending on the pre-buckled films. The structures were modeled undergoing three-point bending, after introducing pre-buckled profiles with different wavelengths. Finite element modeling was carried out using the ABAQUS software package (ABAQUS CAE 6.12-1, Dassault Systèmes, Pawtuckett, Rhode Island, USA). A 2D (two dimensional) plane stress model was built (Figures 2a and 2b).

The thickness of the PDMS was 1.5 mm in the model. This corresponded to the thickness of the PDMS substrate that was used in the experiments. It should be noted that the pre-buckles were simulated using profiles of PEDOT:PSS that were in partial contact with the PDMS substrates prior to the application of bending (Figure 2a). The deformation of the pre-buckled structures then resulted in the flattening of the layers as the bending strains were increased (Figure 2b).

The mechanical properties of the individual layers (Young moduli and Poisson's ratios) that were used in the simulation are presented in Table I.[63–66] Linear elastic deformation was also assumed in each of the layers. Structured quadrilateral meshes were used in the finite element model, along with standard bi-linear plane stress elements with incompatible modes. The two aluminum stoppers at both ends were fixed in the X, Y and Z directions (i.e. U1, U2 and U3, respectively). The middle roller was constrained to displace upward (i.e. move only in the U2 direction) upon the application of pressure.

The above procedure was repeated for the other layered structures in which the anode layer (ITO), the active layer (P3HT: PCBM), and the cathode (Aluminum) were pre-buckled and deformed on the relevant layers in model organic photovoltaic structures.[16,18,19,67–71] The layer mechanical properties that were used in the simulations are summarized in Table I.

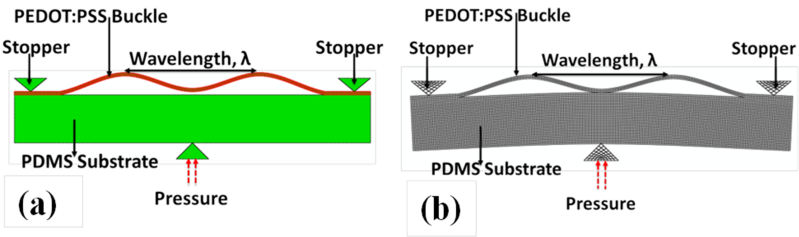

FIG. 2. (a) Schematics of multilayer parts designed for the finite element analysis taking into consideration the experimental parameters and (b) elemental view of buckle on PDMS.

TABLE I. Material Properties.

Materials	Young's Modulus, E/GPa	Poisson's ratio	References
PDMS	0.003	0.48	64,86,87
PEDOT: PSS	1.42	0.3	64
Al	70	0.3	63

IV. RESULTS AND DISCUSSION

A. Effects of pre-stretching on surface Topography

The AFM images revealed the presence of buckles, following release from small pre-stretches of 18%, 25% and larger pre-stretches of 35% and 50% (Figure 3a–3d). These show clearly that the wavelengths of the resulting buckles decreased with increasing pre-stretch. Optical Microscopy images of the surfaces of the PDMS and PEDOT:PSS/PDMS structures also revealed similar trends in the pre- and post-stretched conditions (Figures 4a–4c)). These images also showed that the surface roughness (wavelengths) decrease with increasing pre-stretch.

Plots of the wavelengths of the buckled profiles (obtained from the contact mode AFM images) are presented in Figure 5. These were obtained for surfaces that were subjected to different levels of pre-stretch. These show clearly that the wavelengths of the pre-buckled profiles decrease with increasing pre-stretch. Similar results have been reported by Oyewole et. al.[23] for the formation of pre-buckles on the surfaces of pre-stretched and released Au films on PDMS substrates.

B. Effects of deformation on surface topography

In general, increased pre-stretch resulted in a reduction in the wavelengths of the pre-buckled profiles (Figures 3 and 5). However, upon subsequent bending of the pre-buckled structures, the surfaces of the pre-buckled structures flattened out (Figures 6a and 6b). In the case of the samples produced after 50% pre-stretch, the initial pre-buckled surfaces had wavelengths of about 0.13 microns. The surface wavelengths increased to 0.4545 microns, after applying a bending strain 0.0417. This is consistent with a tendency to flatten out the buckles with increasing applied strain. In the case of the samples that were subjected to a pre-stretch of 70%, the initial PEDOT:PSS/PDMS structures had pre-buckles with a wavelength of about 16.7 nanometers. However, upon applying a bending strain of 0.0417, the surface to about 1.67 microns (See Table II).

C. Finite element simulations of three-point bending

The finite element simulations of the deformation of the pre-buckled PEDOT:PSS profiles on a PDMS substrate are presented in Figures 7a–7c. These show the progression of stress and deformation profiles from the initial pre-buckled states to the conditions at which the buckles become flattened out due to the application of tensile stresses through bending. The results clearly show that the stretching of the surface layers flattens out the initially sinusoidal profiles.

The finite element predictions of the initial pre-buckle flattening conditions confirmed results presented in Table II, along with the experimental measurements of the forces per unit width required for the flattening of the pre-buckles. These forces were observed to increase with decreasing pre-buckle wavelength. Hence, increased pre-stretch of the films, which gives rise to decreased pre-buckled wavelengths, is likely to increase the apparent film "deformability".

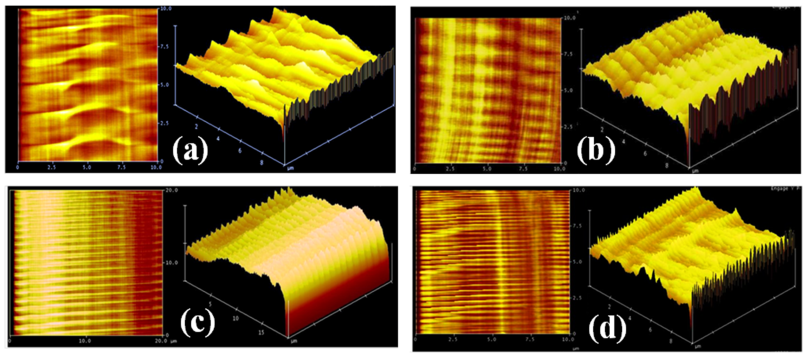

FIG. 3. AFM images of micro-wrinkles observed with pre-stretched PDMS substrates after PEDOT:PSS deposition. (a): 18% pre-stretch. (b): 25% pre-stretch. (c): 35% pre-stretch. (d): 50% pre-stretch. [Note that the wavelength decreases in this order: (a) > (b) > (c) > (d)].

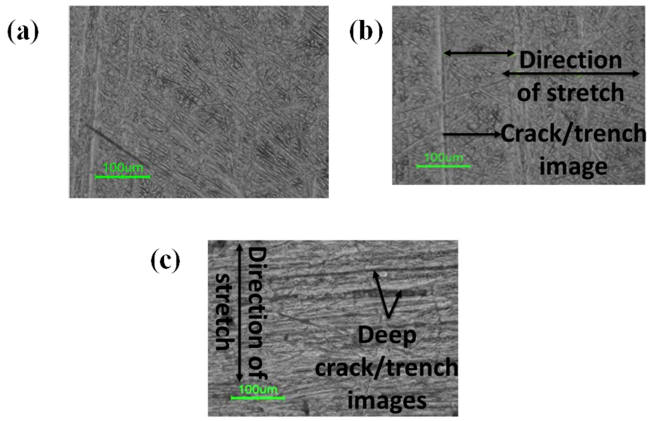

FIG. 4. Optical Microscopic images of the surface topology of PDMS and PEDOT:PSS/PDMS structures; (a) is an optical image of un-stretched PDMS substrate; (b) 35% post-stretched PDMS substrate with PEDOT:PSS deposition, showing less wrinkles, and (c) 50% post-stretched PDMS substrate with PEDOT:PSS deposition showing deep crack/trench images observed as surface features indicating more wrinkles.

Furthermore, the computed von Mises stress distributions obtained for the deformed pre-buckled films were used to characterize the deformation in multilayered structures (Al/P3HT:PCBM/PEDOT:PSS/ITO/PDMS and Al/P3HT:PCBM/PEDOT:PSS/PDMS) that are relevant to organic solar cells. The results are presented in Figures 8a and 8b. These show that the bending of the films (for different initial pre-buckle wavelengths) results in increased von Mises stresses that can lead ultimately to the onset of plastic deformation in the Al cathode layers and the polymeric layers within the model organic solar cell structures.

Hence, the deformation that occurs, prior to flattening, is likely to extend the deformability of the flexible multilayer structures, while the deformation that occurs, after the flattening can result in the build-up of stresses until the onset pf plasticity or fracture. Interfacial failure can also occur between the layers, depending on the adhesion between the layers.[12,72,73]

Finally, it is important to discuss the potential effects of transparent indium tin oxide (ITO) layers that are often used as the anode of organic solar cells. Since these layers have relatively high moduli (Table I), they can result in higher stress distributions and elastic strain energies in flexible model solar cell structures (Figures 8a and 8b). These can lead ultimately to the cracking of the ITO layer, as observed in earlier work.[17,70,74,75] There is, therefore, a need to avoid the use of ITO layers in the development of flexible organic solar cells.

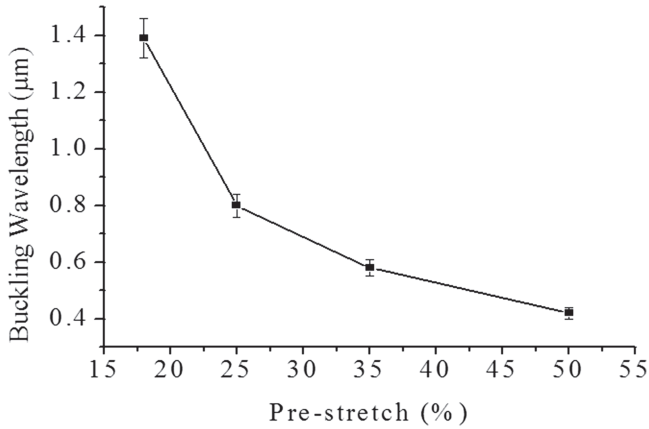

FIG. 5. Buckling/wrinkling wavelengths changes as a function of pre-stretching percentage.

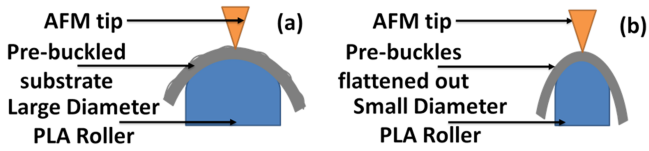

FIG. 6. Schematics of an AFM tip measuring the wavelength of a substrate with wrinkles/buckles undergoing bending on different PLA rollers; (a) Buckles exhibit small wavelengths due to less bending strains applied by courtesy of larger diameter PLA roller used, (b) Buckles flattened implying very high wavelength values due to large bending strain achieved on a small diameter PLA roller.

TABLE II. Pre-buckled wavelength before and after bending.

Pre-buckled Wavelength before Bending/μm	Pre-buckled Wavelength after Bending/μm
0.1300	0.4545
0.0167	1.6700

D. Implications

The implications of the results are very significant. First, they show that pre-buckling can be used to increase the deformability of flexible organic solar cells (see Figures 8a and 8b), prior to the onset of failure by the plasticity or fracture of the film constituents. The improvements in flexibility can also be enhanced by the control of initial buckle wavelengths and amplitudes, which can be achieved by the use of pre-stretching methods (as done in the experimental section of this paper).[50,76]

However, increased pre-buckle wavelengths can also result in higher film stresses that can induce failure within the layers or between them. A balanced approach is, therefore, needed to determine the pre-buckled configurations that improve flexibility/deformability, without compromising the conditions for final film failure.

Furthermore, from the literature,[6,46,48,77,78] it is clear that further flexibility can be achieved by fabricating the devices on ultra-thin polymer substrates and laminating them unto a pre-strained elastomers such as PDMS. The application of such ultra-thin substrates could decrease the bending

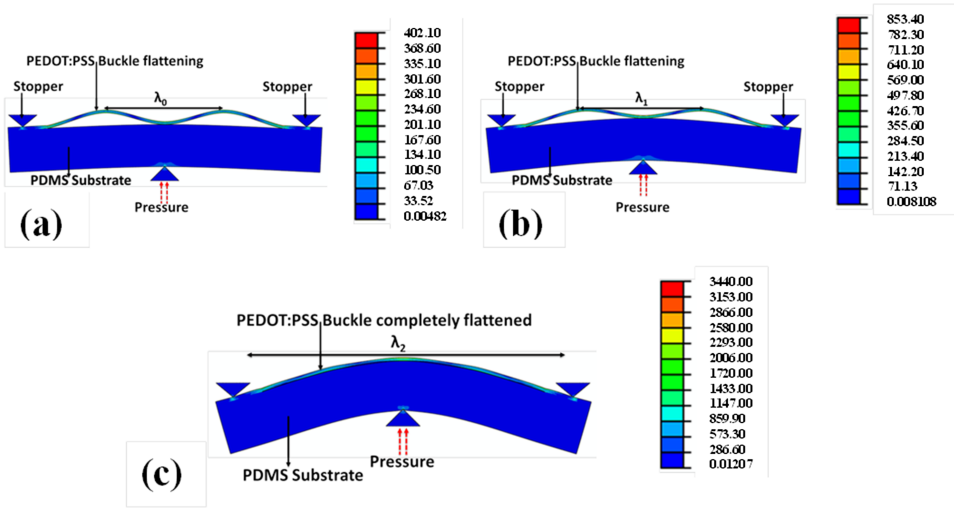

FIG. 7. Pre-buckled structure of PEDOT: PSS on PDMS with von Misses stress distribution at the onset of flattening for various wavelengths; (a) buckle with wavelength, λ_0, at 15 N/mm^2 pressure (b) buckle wavelength gradual increase on further application of bending moment to λ_1 due to the stretching of the buckle at 37 N/mm^2 pressure, and (c) complete flattening of buckle on PDMS substrate on applying 50 N/mm^2 pressure.

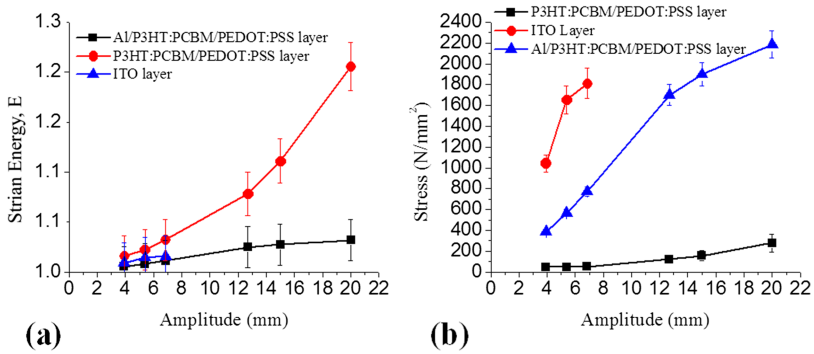

FIG. 8. Maximum strain and von Misses stresses in the different films for different amplitudes at constant wavelength with effects of indium tin oxide layers in the pre-buckle structures; (a) strain energy variations and (b) stress distribution in the different layers.

strains. However, the compliance of these structures would also be increased significantly. In any case, the model PEDOT:PSS transparent electrode, explored in the current work is consistent with the work of Drack et al.[48] and Kim et al.[77] who have produced flexible devices in which resistance increases to ~30% after 10,000 cycles of stretching to strains of 50%.[48,77]

Furthermore, the current work suggests that improvements in the flexibility of pre-buckled organic electronic structures can be estimated by considering the ratio of the strains to flattening of a pre-buckled structure to the strain to failure of a non-pre-buckled structure. This means that the surface area of the device (if it is a solar cell) would have more wavy characteristics that could improve the harnessing of sunlight for photo-conversion to electricity.

The fabrication of the above organic solar cell structures could be achieved by the use of roll-to-roll printing[16,79,80] and lamination processes.[64,81] The optimization of such manufacturing processes could lead to the future scale up of emerging approaches for the design and fabrication of flexible solar cells.[82] The flexible organic solar cells could also be integrated into roofing tiles[83] and electronic textiles[84–86] in which significant bending strains[87] can be applied during fabrication and service.

V. SUMMARY AND CONCLUDING REMARKS

This paper presents the results of an experimental and computational study of the deformation behavior of pre-buckled thin films structures that are relevant to the deformation of flexible organic electronics. The results show that the additional strain to flattening (of the pre buckles) enhances the deformability/flexibility of the films. The strains to flattening also increase with increased pre-buckle wavelength. However, such increased pre-buckle amplitudes and wavelengths may also induce failure by film plasticity, fracture or delamination. A balanced approach is, therefore, needed for the design of robust pre-buckled layers for potential applications in flexible organic electronic structures.

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