

Prestress Driven Improvement in Fracture Behavior of in Situ Sputtered Zinc Oxide Thin Films on Stretched Polymer Substrates

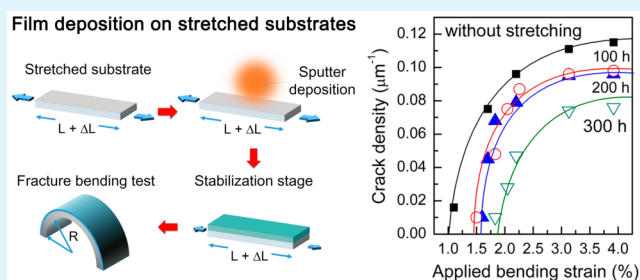
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S Supporting Information

ABSTRACT: Flexible electronic devices need to survive bending or stretching operation without mechanical failure. If inorganic thin films are involved in the device structure, the evolution of cracks is a major challenge to overcome. Here, we report a novel way to substantially improve the fracture behavior of films that are based on intentional utilization of residual stress on the films by in situ sputtering on a stretched polymer substrate. The in situ sputtering combined with a stabilization stage yielded ZnO:Al thin films with a nearly 2-fold improvement in crack initiation strain, which indicates greater resistance to bending. The critical strain of the optimal ZnO:Al films was $\sim 1.83\%$, which is a significant improvement compared to the current tolerance value of $\sim 1\%$. This was accompanied by a $\sim 300\%$ improvement in fracture energy. We attributed the improved fracture behavior to the presence of residual compressive stresses, which creates a barrier for crack formation by acting opposite to the applied bending strain.

KEYWORDS: flexible electronics, fracture behavior, transparent conducting oxide, zinc oxide



INTRODUCTION

Despite the recent progress in the field of flexible electronics, the inferior fracture behavior of thin films grown on polymer substrates remains a key challenge.¹ In particular, successful fabrication of inorganic oxide-based devices on polymer substrates is highly challenging due to their fragility and limited mechanical integrity during bending, stretching, and folding.^{1–3} The allowable tensile strain of flexible inorganic thin films such as ZnO, Sn:In₂O₃ (ITO), and Al₂O₃ before the occurrence of fracture has been reported to be $\leq \sim 1\%$.^{1,4,5} Various transparent conducting oxide (TCO) thin films have been used extensively in intriguing applications such as flat panel displays, touch-screen devices, and solar cells.⁶ Recent developments in the fabrication of TCO thin films on various polymer substrates have paved the way for their future applications in potential flexible and stretchable devices.^{7–9} Hence, there is a need to improve the fracture behavior of TCO thin films to realize competitive flexible devices that can operate at strain levels substantially above the current tolerance value. In this work, we study the fracture behavior of inorganic thin films on polymer substrates using ZnO:Al thin films. Several studies reported the fracture behavior of flexible ZnO thin films, providing baseline data. The fracture energy and crack-initiating critical strain of ZnO:Al thin films grown on PES substrates are known to be in the range of 50–70 J cm⁻² and 0.95–1.1%, respectively.^{5,7}

Earlier examples for improving fracture behavior, such as inkjet-printed silver electrodes,¹⁰ e-beam evaporated metallic films,¹¹ and transferred Si ribbons,¹² focused mainly on the use of elastomeric poly dimethylsiloxane (PDMS) substrates by the

concept of wavy buckled surfaces. These “wavy” thin films can be stretched to larger strain levels than flat films.^{11,12} The use of wrinkled Al₂O₃ film in graphene–carbon nanotube transistors dramatically improved stretchability from 0.475 to 30%.¹ In contrast, few methods to improve the fracture energy and critical strain of inorganic thin films grown on polymer substrates have been reported. Our group recently reported a 70% improvement in critical strain of ZnO:Al thin films using an AlN passivation layer.⁷ It is known in the integrated circuit field that utilization of a passivation layer enhances the robustness and mechanical stability of devices.¹³ It may also be possible to take advantage of the relation between the fracture formation and the existence of residual compressive or tensile stress in inorganic thin films to improve the fracture behavior. For instance, during stretching or bending, residual compressive stresses are known to prevent the formation of cracks, whereas tensile residual stresses facilitate crack opening and propagation.^{14–16} In this regard, there have been a few reports of intentional induction of compressive stress in films by using the curved surface of the polymer (PET) substrate upon sputtering of ZnO, ITO, and TiO₂ thin films.^{17,18} Likewise, Watanabe has reported in situ sputter deposition of gold thin films on stretched PDMS and silicone substrates to produce ordered wavy wrinkles upon releasing.^{19,20} However, these studies did not investigate the mechanical behavior of the flexible films.

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Here, for the first time, we present a new in situ growth method to improve the fracture behavior of flexible ZnO:Al thin films on a polymer substrate by enabling the deliberate formation of compressive stress on the dense film. The key processing steps include the stretching of the polymer substrate prior to deposition of ZnO:Al thin films, and the assurance of a sufficient time for relaxation of the stress without causing mechanical failure. The stretching operation provides pretensile stress upon film growth, while the relaxing period, referred to as the stabilization process, builds a residual compressive stress in the thin film before releasing the stretched substrate. This process dramatically reduces crack density. We confirmed that the residual compressive stress in the thin films improved the fracture energy by $\sim 300\%$, with near double enhancement of the crack-initiating critical strain. The in situ formation of residual stresses in thin films during sputter deposition must be controlled in such a way that they can be utilized as an independent degree of freedom to control fracture behavior.

EXPERIMENTAL SECTION

ZnO:Al thin films were deposited on linearly stretched 200 μm thick polyethersulfone (PES) substrates using RF magnetron sputtering at room temperature. The size of the PES substrate was 50×20 mm. Prior to sputter-deposition, stretching strain was intentionally applied to the PES substrate using a fixture designed to control the level of stretching. A schematic illustration of the process of in situ sputtering on stretched substrates is shown in Figure 1. Figure 1a,b represent the

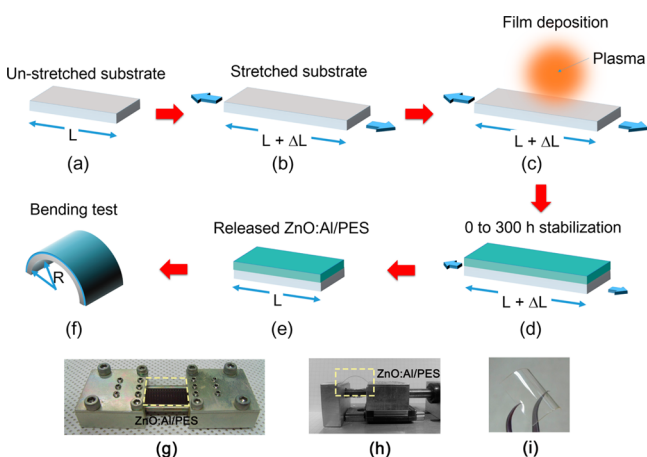


Figure 1. Schematics depicting the processing steps involved in ZnO:Al thin film growth on a stretched polymer substrate and the procedure used to evaluate fracture behavior. (a and b) Stretching of the PES substrate from the unstretched state, (c) in situ sputter deposition process of ZnO:Al film on the stretched substrate, (d) stabilization process, (e) released film with the original length, and (f) bent film with a given radius, R , for evaluation of mechanical flexibility. Photographs of (g) the setup for the stretching process, (h) the bending test, and (i) an actual flexible film sample.

PES substrate in the normal and stretched condition prior to film deposition, respectively. An actual photograph of the custom substrate holder that was used to clamp the two ends of the substrate, is shown in Figure 1g. Using a rotating knob, one end of the PES substrate was stretched, and the increased distance was measured using a connected micrometer. The level of the stretching strain applied to the PES substrate, ϵ , was quantitatively expressed via the following simple equation, $\epsilon = \ln(L/(L + \Delta L))$, where L is the original length of the substrate and ΔL is the increase in length with applied stretching stress. To prevent misinterpretation as compressive strain, we used the absolute value of ϵ even though this definition creates a negative sign.

The fixture was ultimately located inside the sputtering chamber, and deposition was carried out on the linearly stretched substrate as shown in Figure 1c. Details of the film deposition and sputtering system are given in our previous studies.^{7,21} A 2 in. disk-typed target (99.995% purity, CERAC Inc., Milwaukee, WI) of 2 wt % Al_2O_3 -doped ZnO was used. The distance between the target and substrate was ~ 10 cm. Films of ~ 200 nm in thickness were deposited at an RF power of 100 W in pure Ar atmosphere (2 mTorr). Another critical parameter, the stabilization time, which is the holding time before releasing the as-deposited film from the substrate holder in the stretched condition, was adjusted up to 300 h to investigate the fracture behavior of ZnO:Al thin films (Figures 1d). It should be mentioned that the original length of the sample was maintained when released after stretching and stabilization processes according to measurements using a Vernier caliper with an accuracy of ± 0.02 mm. The structural properties of the ZnO:Al thin films were investigated via X-ray diffraction (XRD) using a PANalytical model X'Pert PRO XRD unit with Cu-K α radiation in Bragg-Bretano ($\theta/2\theta$) geometry.

The cracking behavior of the ZnO:Al thin films was investigated using an optical microscope and expressed in terms of crack density, ρ , the number of cracks per unit length generated on the surface of the thin films. Details of the bending test (Figure 1f) can be found in our previous publications.^{5,7} The Young's modulus of the ZnO:Al thin films ($E_f = 150$ GPa) is significantly larger than that of the PES substrates ($E_s = 2.6$ GPa).⁷ Therefore, the neutral surface in the ZnO:Al/PES structure shifted upward toward the film, and the applied bending strain, ϵ_a , on the ZnO:Al film was calculated as^{22,23}

$$\epsilon_a = \left(\frac{t_f + t_s}{2r} \right) \frac{(1 + 2\eta + \chi\eta^2)}{(1 + \eta)(1 + \chi\eta)} \quad (1)$$

where t_f and t_s are the thickness of the film and the substrate, respectively, $\eta = t_f/t_s$, and $\chi = E_f/E_s$. The radius of the curvature, r , at the center of the bent sample was calculated as reported earlier.²⁴

RESULTS AND DISCUSSION

As described in the Experimental Section, ZnO:Al films were sputter deposited on linearly stretched PES substrates with stretching strain values ranging from 0.49 to 4.87%. After each deposition, the ZnO:Al film/PES was subjected to stabilization process before releasing the substrate. This stabilization process was performed to avoid the sudden release of stress. The stabilization time was varied from 0 to 300 h. Figure 1e shows the released ZnO:Al film/PES after the stabilization process. The fracture behavior of the released film was evaluated under bending. Figure 1h shows the measurement setup used to apply bending strain to the ZnO:Al film. A photograph of an actual sample of the transparent flexible ZnO:Al/PES structure is shown in Figure 1i.

The effects of applied stretching strain (or pretensile strain), ϵ , and stabilization time on the fracture behavior of the ZnO:Al thin films were investigated by observing the evolution of cracks on the film surface using an optical microscope. Several examples of the surface of ZnO:Al thin films deposited under applied stretching strains of 0.49 and 4.87%, each stabilized for 0 and 300 h, are provided in Figure 2a–d; the micrographs were photoedited to have a blue background for better contrast visualization. Straight cracks perpendicular to the stretching and releasing direction were observed. Irrespective of the level of applied stretching strain, all samples that did not undergo the stabilization process exhibited sporadic cracks on the surface. The number of cracks increased as the stretching strain increased from 0.49% to 4.87%, while the space between two adjacent cracks, the crack spacing, λ , decreased. No cracks were observed in the sample with a stretching strain of 0.49% after stabilization for 300 h (Figure 2c). In contrast, even after

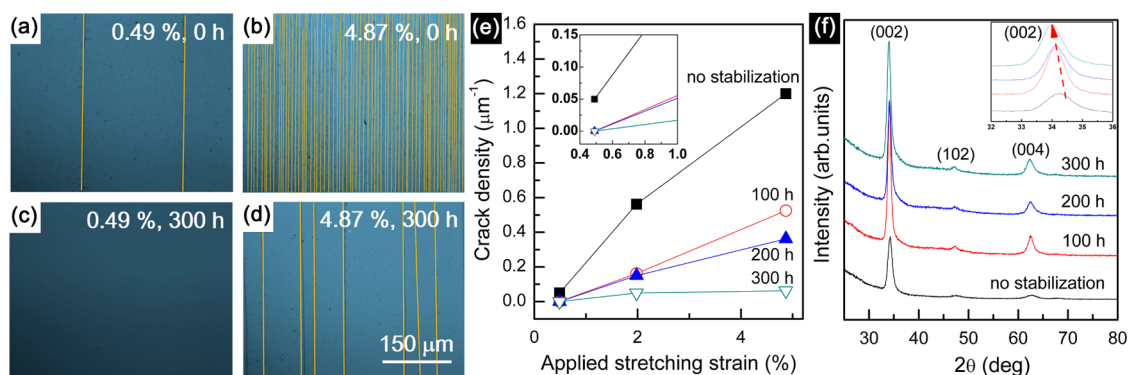


Figure 2. (a–d) Representative optical micrographs showing the fracture behaviors of ZnO:Al thin films grown with a stretching strain of 0.49 or 4.87% and stabilized for 0 or 300 h. (e) Dependence of crack density on the applied stretching strain and stabilization time, with an inset highlighting the lowest region of stretching strain. (f) XRD patterns of ZnO:Al films grown with a stretching strain of 0.49% and stabilized for 0–300 h. The shift of the (002) peak with increase in stabilization time is shown as an inset.

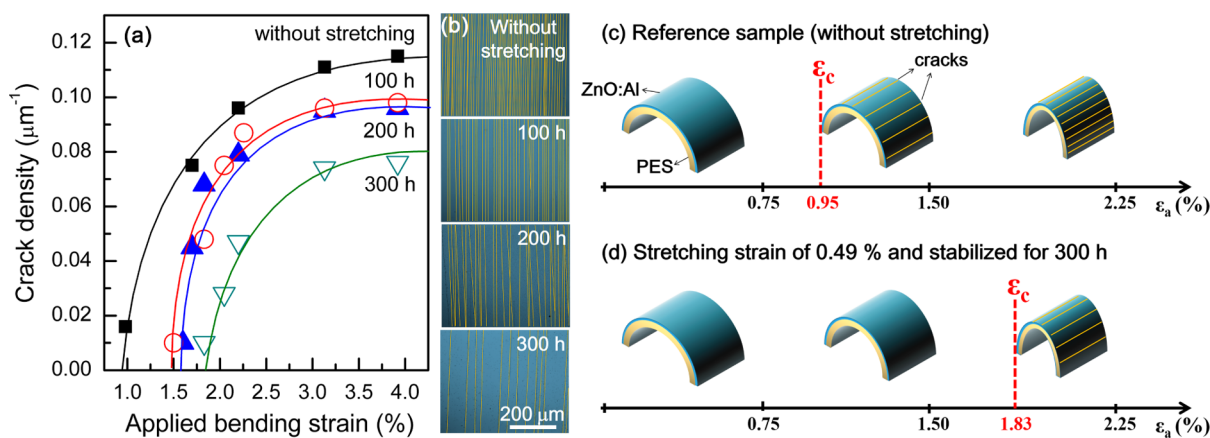


Figure 3. (a) Plots depicting variations in crack density, ρ , as a function of applied bending strain for samples grown with a stretching strain of 0.49% and stabilized for 100, 200, and 300 h (a plot for the reference sample prepared without stretching is shown for comparison). (b) Examples of optical micrographs showing the linear cracks that developed on the film surfaces when the highest applied bending strain of $\sim 3.92\%$ was applied. (c and d) Schematic illustrations of the occurrence of fracture in the sample grown with 0.49% strain and stabilized for 300 h, shown with the reference sample.

stabilization for 300 h, the sample grown with the high stretching strain of 4.87% exhibited cracks (Figure 2d), suggesting a strong dependence on the degree of stretching.

The level of fracture of ZnO:Al thin films can be expressed in terms of crack density, ρ , which we defined as the number of cracks per unit micrometer length. The plots shown in Figure 2e reflect the effect of applied stretching strain and stabilization time on the crack density of ZnO:Al thin films. The crack density tended to increase with increasing applied stretching strain, regardless of the stabilization time, but the tendency for crack density to increase slowed significantly with stabilization time. Without the stabilization process, severe cracking was evident at higher stretching strains. It is worth noting that no cracks were observed at a stretching strain of 0.49%, with a stabilization time of 100 h or longer, as highlighted in the inset of Figure 2e. Crack density could not be completely removed at stretching strains of 1.98 and 4.87% up to 400 h stabilization. From the viewpoint of fracture behavior, excessive stretching strain must be avoided. Stabilization time combined with a proper stretching strain plays a crucial role in preventing the initiation of cracks; the optimized conditions for a crack-free ZnO:Al film are a stretching strain of 0.49% and stabilization time of ≥ 100 h.

Figure 2f shows the effect of stabilization time on the XRD pattern of ZnO:Al thin films deposited at room temperature

under an applied stretching strain of 0.49% and then released after stabilization. The deposited ZnO:Al films were predominantly oriented along the c -axis of the wurtzite hexagonal structure.²¹ The XRD patterns revealed that the peak positions shifted toward lower 2θ value when the stabilization time increased from 0 to 300 h, as represented by the (002) peak shift from 34.25 to 34.01° (Figure 2f, inset). This indicated that the films were subjected primarily to residual stress, as estimated using the biaxial strain model.²⁵ Details of the residual stress calculation from the XRD pattern can be found in the Supporting Information. With an increase in stabilization time, the residual stress, σ_{res} , increased from -2.25×10^9 N/m² (0 h) to -3.80×10^9 N/m² (100 h) and then to -5.36×10^9 N/m² (300 h) (Figure S1, Supporting Information). The negative sign of σ_{res} suggested that the residual stress was compressive in nature. During release of the applied stretching strain, the PES substrate returned to its original length, L , and the applied stretching strain from the substrate promoted the formation of compressive stress in the films. Excessive stress beyond the allowable limit accompanies the occurrence and propagation of cracks, which worsens when stress is released instantaneously.²⁶ The stabilization process appeared to allow the slow incorporation of more residual stress into the films by hindering the formation of cracks. Despite the clear positive

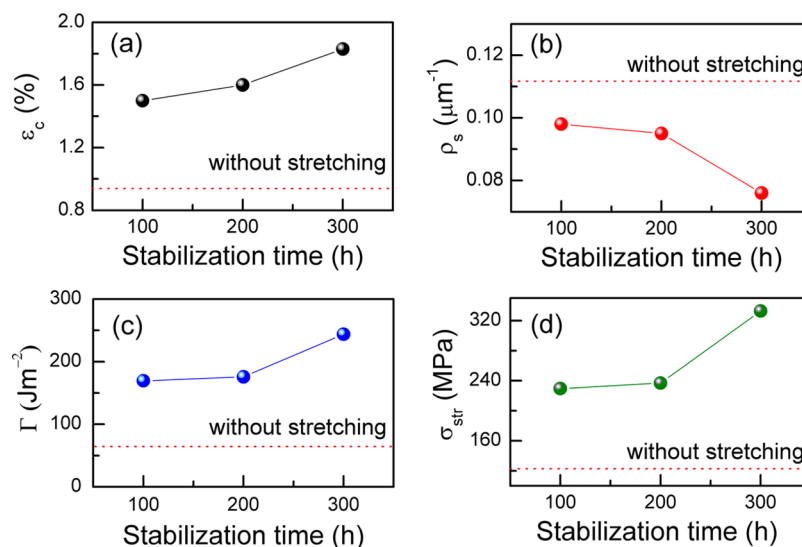


Figure 4. Effect of stabilization time on (a) critical strain, ϵ_c , (b) saturated crack density, ρ_s , (c) fracture energy, Γ , and (d) the film strength, σ_{str} of the ZnO:Al films. Horizontal dotted lines represent the corresponding value for the reference sample grown without any stretching strain.

effect of the stabilization process, the underlying mechanism is not clear.

Optimized conditions of a stretching strain of 0.49% and stabilization time ≥ 100 h to obtain crack free ZnO:Al films were used to further investigate fracture behavior under various applied bending strains, ϵ_a , using an optical microscope. A reference sample, which was grown under the same growth conditions but without any stretching strain, was also included for comparison. Figure 3a shows the effect of applied bending strain and stabilization time on the crack density, ρ , of ZnO:Al thin films grown with a stretching strain of 0.49%. The minimum applied bending strain required to initiate cracks in the ZnO:Al thin films, referred to as the critical strain, ϵ_c , was obtained by extrapolating the corresponding ρ versus ϵ_a curve to $\rho = 0$. The critical strain for the reference sample was only 0.95%; in contrast, the samples grown with a stretching strain of 0.49% and stabilized for ≥ 100 h showed higher critical strains of 1.5% (100 h) and 1.83% (300 h). This near double improvement in ϵ_c from 0.95 to 1.83% implied that these films could withstand greater bending strain without any crack formation.

When the applied bending strain is less than the critical strain, $\epsilon_a < \epsilon_c$, all films are capable of withstanding the applied bending strain without any crack openings. However, when ϵ_a exceeds ϵ_c ($\epsilon_a > \epsilon_c$), nucleation of parallel cracks perpendicular to the bending direction begins and then propagates over the films.^{5,7} Initially, the number of parallel cracks increased rapidly as the applied bending strain increased beyond the critical strain, leading to increased crack density and decreased crack spacing. However, the crack density remained nearly constant with further increases in the applied bending strain, indicating a saturated state. Figure 3b shows optical micrographs of the samples subjected to the highest applied bending strain of $\sim 3.92\%$, at which crack formation was nearly saturated, as an example of crack formation after bending fracture evaluation. It is evident that, even at the highest applied bending strain, the number of cracks was greatly reduced for the sample stabilized for 300 h, relative to the reference sample.

Figure 3c,d shows the effect of the applied bending strain on the reference and 300 h stabilized samples, respectively. During bending evaluation, the direction of the applied tensile bending

stress, σ_a , was opposite to the residual compressive stress, σ_{res} , in the film. Initially, for the 300 h stabilized sample, which possessed the highest residual compressive stress, the applied bending stress was completely canceled out by the residual compressive stress, and hence, the ZnO:Al films were unaffected. The residual compressive stress induced by the stretching and stabilization treatment appeared to create a higher energy barrier for crack initiation than for the reference sample; this energy barrier depends on the level of locking of the residual stress. Hence, more applied bending strain was required to initiate cracks relative to the reference sample. This is the reason the stabilized samples (≥ 100 h) showed higher critical strains than the reference sample.

The effect of stabilization time on the critical strain and saturated crack density are plotted in Figure 4a,b. Critical strain increased with an increase in stabilization time, revealing the importance of the stabilization process on fracture behavior. The ZnO:Al film grown with an optimized stretching strain of 0.49% and stabilization time of 300 h exhibited a critical strain of 1.83%, which was significantly larger than that of the reference sample of 0.95%. The saturated crack density decreased from $0.112 \mu\text{m}^{-1}$ for the reference sample to $0.076 \mu\text{m}^{-1}$ for the 300 h stabilized sample. In a recent study,⁷ we found that AlN-passivated ZnO:Al films grown under similar conditions exhibited a critical strain of 1.4%. Hence, the critical strain of 1.83% is the highest value obtained thus far for the thin films grown on polymer substrates. This is highly relevant for inorganic film-involved flexible devices that can benefit from higher resistance to mechanical failure in bendable environments.

Further investigation of the fracture behavior of the ZnO films was performed by evaluating fracture energy, Γ , and film strength, σ_{str} , which are depicted in Figure 4c,d as a function of stabilization time. As reported earlier,^{7,27–30} the Γ value for ZnO:Al films was calculated using the energy criterion proposed for the multiple film cracking phenomenon. Cracks are assumed to develop in the films if the change in strain energy due to cracking of the film is in equilibrium with the energy required for film cracking. Fracture energy of the ZnO:Al film is given by^{27–29}

$$\Gamma = \frac{3(f_1 \varepsilon_c + f_2 \Delta \varepsilon)^2 f_3 f_4 + 2\Delta \varepsilon (f_1 \varepsilon_c + f_2 \Delta \varepsilon) f_5}{4\kappa} E'_f \quad (2)$$

where ε_c is the critical strain and $\Delta \varepsilon$ is the mismatch strain. Values of $\Delta \varepsilon$ were estimated from the compressive stresses obtained from XRD measurements. The parameters κ , E'_f , f_1 , f_2 , f_3 , f_4 , and f_5 were estimated using Young's modulus, Poisson's ratio, the biaxial modulus ratio, and the thickness ratio of the ZnO:Al film and PES substrate.²⁷ Detailed mathematical expressions and values for these parameters are provided in the Supporting Information. Similarly, film strength, σ_{str} , can be estimated using the strength criterion, which assumes that cracks occur when the film center stress reaches the film strength.²⁷ Film strength can be expressed in terms of critical strain and mismatch strain as²⁷

$$\sigma_{str} = E'_f (f_1 \varepsilon_c + f_2 \Delta \varepsilon) \quad (3)$$

Figure 4c,d reveals that both fracture energy and film strength increased with an increase in the stabilization time. Horizontal dotted line represents the corresponding value for the reference sample. All values in Figure 4 are given in Table S1 (Supporting Information). Because the residual compressive stress increased with an increase in stabilization time, we can safely state that the higher the residual compressive stress, the higher the fracture energy and film strength. The highest fracture energy and film strength values obtained here were 244 J cm⁻² and 333 MPa, respectively, corresponding to the improvements of ~300 and ~170%. The reported fracture energy of ZnO thin films ranges from 11.7 to 70 J cm⁻² depending on the film thickness (200 nm to 1 μ m).^{5,7,22} The use of an AlN passivation layer on ZnO:Al films is associated with a fracture energy of 236 J cm⁻².⁷ Computational estimation of a SiO_x/PET structure demonstrated that the square root of fracture energy and film strength increased linearly with an increase in residual compressive stress.²⁷ It should be mentioned that the Γ and σ_{str} values obtained here are apparent ones that can be influenced by material parameters such as mismatch strain, critical strain, and thickness of the film.

CONCLUSIONS

We deposited ZnO:Al thin films on linearly stretched polymer substrates using an in situ sputter deposition technique followed by a stabilization process. The ZnO:Al film grown with a stretching strain of 0.49% and stabilized for 300 h showed the highest critical strain of 1.83% and a fracture energy of 244 J cm⁻², in contrast to the corresponding values of 0.95% and 61 J cm⁻² for the reference sample. We attributed the improved fracture behavior of the ZnO:Al thin films to the formation of residual compressive stress during the stabilization process, which acted against the bending strain. The near double improvement in fracture behavior of the ZnO:Al thin films may be extended to other flexible inorganic thin films that require more bendable environments to realize competitive flexible devices.

ASSOCIATED CONTENT

Supporting Information

Residual stress calculation from XRD, calculation of fracture energy, and actual values of the bending evaluation parameters of the ZnO/PES structures. The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.5b01836.

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Notes

The authors declare no competing financial interest.

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