

Supporting Information

Enhanced fracture resistance in flexible ZnO:Al thin films *in situ* sputtered on bent polymer substrates

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Calculation of fracture energy and film strength

When evaluating fracture energy Γ and film strength σ_{str} , the mismatch strain $\Delta\varepsilon$ was used as one of the parameters that control the fracture behavior in the pre-bending condition. In the energy criterion, it is assumed that the cracks begin to develop if the change in strain energy due to cracking of the film is in equilibrium with the energy required for film cracking. The fracture energy of the film can be written as,¹⁻³

$$\Gamma = \frac{3(f_1\varepsilon_c + f_2\Delta\varepsilon)^2 f_3 f_4 + 2\Delta\varepsilon(f_1\varepsilon_c + f_2\Delta\varepsilon)f_5}{4\kappa} E'_f \quad (S1)$$

where f_1, f_2, f_3, f_4, f_5 and κ are the parameters considering the Young's modulus, Poisson's ratio, biaxial modulus ratio and thickness ratio of the ZnO:Al film and PES substrate.^{1,4} The parameters are given by the following specific equations:

$$f_1 = \frac{(1 + \Sigma\eta^3) \left[(1 - \nu_s)(1 + \nu_f) + \Sigma\eta(1 - \nu_f\nu_s) \right]}{(1 + 3\Sigma\eta^2 + 4\Sigma\eta^3 + \Sigma\eta + \Sigma^2\eta^4)(1 + \nu_f)} \quad (S2)$$

$$f_2 = -\frac{(1 + \Sigma\eta^3)}{(1 + 3\Sigma\eta^2 + 4\Sigma\eta^3 + \Sigma\eta + \Sigma^2\eta^4)} \quad (S3)$$

$$f_3 = \frac{(1 + \nu_f)(1 - 2\nu_f\nu_s + \nu_s^2)}{(1 - \nu_f\nu_s)^2} \quad (S4)$$

$$f_4 = 1 + 3 \left[\frac{\Sigma\eta^2(1 + \eta)}{(1 + \Sigma\eta^3)} \right]^2 \quad (S5)$$

and

$$f_5 = \frac{2\nu_s(1 + \nu_s)(1 - \nu_f^2)}{(1 - \nu_f\nu_s)^2} \quad (S6)$$

The parameters f_1, f_2, f_3, f_4, f_5 and κ are obtained as being 0.79, -0.85, 1.44, 1.00, 0.49 and 1.46, respectively. The parameter E'_f is given by the relation of $E'_f = E_f/(1 - \nu_f^2)$ where E_f and ν_f are the Young's modulus and Poisson's ratio (0.35) of ZnO film, respectively.⁵⁻⁷ The values

of $\Delta\varepsilon$ were estimated from the compressive stresses obtained from XRD measurements as 0.014, 0.016 and 0.019 for the films grown with a pre-bending strain of 0.70 %, 0.99 % and 1.21 %, respectively.

Similarly, film strength, σ_{str} , was quantitatively expressed using the several parameters given in Eq. (S1) with the assumption in which the cracks occur when the film center stress reaches the film strength.¹ The σ_{str} is given by¹

$$\sigma_{str} = E'_f (f_1 \varepsilon_c + f_2 \Delta\varepsilon) \quad (S7)$$

The plots of all the resultant values of fracture energy Γ and film strength σ_{str} as a function of pre-bending strain are given in Fig. 3(c,d) of the main text.

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