Supplementary Information for

Graphene meta-interface for enhancing the stretchability of brittle oxide layers

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S1. Derivation of the shear-lag model

Strain transfer between the PET substrate and the ITO layer via the graphene layer can be described using the 2D shear lag model of McGuigan *et al.*[32] The governing equation is derived from the force equilibrium relating the interfacial shear stress $\tau(x)$ of the graphene layer to the tensile stress σ transferred to the ITO layer; i.e.,

$$q\frac{d\sigma(x)}{dx} = -\tau(x) \quad \text{(S1)}$$

The shear stress in the multilayer graphene is given by

$$\tau(x) = \frac{G(\varepsilon x - \emptyset(x))}{p} \quad \text{(S2)}$$

when $x \leq x_p$ and

$$\tau(x) = G\gamma_p = \tau_p \quad \text{(S3)}$$

when $x \geq x_p$, where $\Phi(x)$ is the displacement of the ITO layer, ε is the applied strain and γ_p is the plastic shear strain in the multilayer graphene at which the shear stress reaches the critical value τ_p at $x = x_p$. Because the strain is the gradient of the displacement, we have

$$\sigma(x) = E\frac{d\emptyset(x)}{dx}, \quad \text{(S4)}$$

Combining Eqs. (S1) with (S2) yields the following differential equation:

$$\frac{d^2\emptyset}{dx^2} - \beta^2\emptyset(x) = -\beta^2\varepsilon x, \quad \text{(S5)}$$

where $\beta = \sqrt{G/pqE}$. The following two boundary conditions are required to solve Eq. (S5):

$$\emptyset(x=0) = 0 \quad \text{(S6)}$$

and

$$\sigma(a) = E\frac{d\emptyset(x)}{dx}_{(x=a)} = 0. \tag{S7}$$

The solution to Eq. (S5) is given by

$$\emptyset(x) = \varepsilon\left(x - \frac{sinh(\beta x)}{\beta cosh(\beta a)}\right). \tag{S8}$$

The maximum stress occurs in the center of each segment, and we look for a solution where this stress is equal to the tensile strength σ^*, which corresponds to the maximum length of the segment before fracture at that strain. The tensile stress in the center of a segment is given by

$$\sigma(0) = \frac{1}{q}\int_0^a \tau(x)dx. \tag{S9}$$

Substitution of Eqs. (S2) and (S3) into Eq. (S9) leads to the following expressions for the crack density n as a function of strain.

① The fully elastic case ($x_p > a$)

When maximum stress reaches the tensile strength, a is equal to half the maximum crack length; i.e.,

$$\sigma(0) = \frac{1}{q}\int_0^a \tau(x)dx = \sigma^*, \tag{S10}$$

and we have

$$a = \frac{1}{\beta}cosh^{-1}\left(\frac{1}{1 - \frac{\sigma^*}{\varepsilon E}}\right). \tag{S11}$$

Because the crack density is now given by $n = 1/2a$, we have

$$n = \frac{1}{2a} = \sqrt{\frac{G}{E}}\left[2\sqrt{pq}cosh^{-1}\left(\frac{1}{1 - \frac{\sigma^*}{\varepsilon E}}\right)\right]^{-1}. \tag{S12}$$

② The elastic–plastic case ($x_p < a$)

When $x < x_p$, Eq. (S11) can be used for a; i.e.,

$$\tau(x) = \frac{G}{p\beta}\left(\varepsilon - \frac{\sigma^*}{E}\right) sinh(\beta x) \tag{S13}$$

and when $x \geq x_p$ we have

$$\tau(x_p) = G\gamma_p. \tag{S14}$$

Because we require that the shear stress is continuous at x=x_p, we obtain

$$x_p = \frac{1}{\beta} sinh^{-1}\left(\frac{\gamma_p\sqrt{\frac{Gp}{qE}}}{\left(\varepsilon - \frac{\sigma^*}{E}\right)}\right), \tag{S15}$$

and a is equal to half of the maximum crack length when the maximum stress becomes equal to the tensile strength. We therefore have

$$\sigma(0) = \frac{1}{q}\int_0^a \tau(x)dx = \frac{1}{q}\left(\int_0^{x_p} \tau(x)dx + \int_{x_p}^a G\gamma_p dx\right) = \sigma^*, \tag{S16}$$

which leads to

$$a = \frac{Eq}{G\gamma_p}\left[\left(\frac{\sigma^*}{E} - \varepsilon\right) cosh\left(sinh^{-1}\left(\frac{\gamma_p\sqrt{\frac{Gp}{Eq}}}{\left(\varepsilon - \frac{\sigma^*}{E}\right)}\right)\right) + \varepsilon + \gamma_p\sqrt{\frac{GP}{Eq}} sinh^{-1}\left(\frac{\gamma_p\sqrt{\frac{Gp}{Eq}}}{\left(\varepsilon - \frac{\sigma^*}{E}\right)}\right)\right]. \tag{S17}$$

Because the crack density is $n = 1/2a$, this can be expressed as follows:

$$n = \frac{G\gamma_p}{2Eq}\left[\left(\frac{\sigma^*}{E} - \varepsilon\right) cosh\left(sinh^{-1}\left(\frac{\gamma_p\sqrt{\frac{Gp}{Eq}}}{\left(\varepsilon - \frac{\sigma^*}{E}\right)}\right)\right) + \varepsilon + \gamma_p\sqrt{\frac{GP}{Eq}} sinh^{-1}\left(\frac{\gamma_p\sqrt{\frac{Gp}{Eq}}}{\left(\varepsilon - \frac{\sigma^*}{E}\right)}\right)\right]^{-1}. \tag{S18}$$

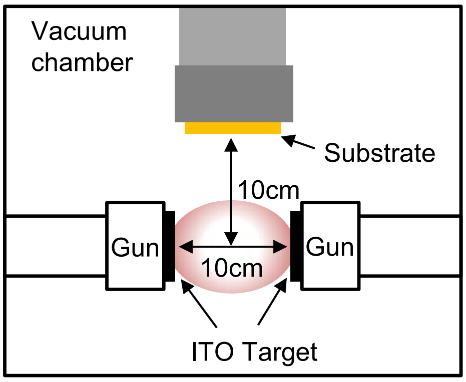

Figure S1. Schematic of an off-axis RF magnetron sputtering.

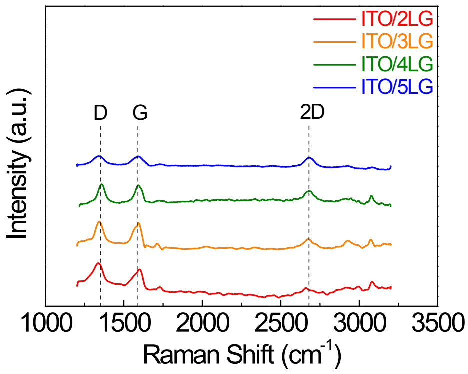

Figure S2. Raman spectra (excitation wavelength λ = 514nm) of the ITO/MLG after subtracting the peaks of PET substrates. The existence of the D peak indicates the damage on the graphene layers during ITO sputtering, but even with this damage, the graphene meta-interface can reduce the strain transfer by the interlayer sliding. By decreasing the sputtering damage of graphene, the performance of the graphene meta-interface can be improved further.

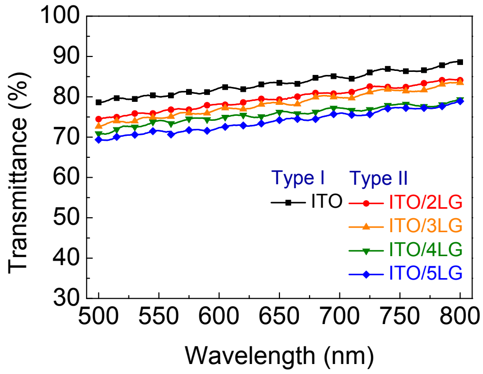

Figure S3. Transmittances of type-I and type-II structures over a range of wavelength.

Samples		Transmittance (%)	
		Mean	Standard deviation
Type-I	ITO	80.7	0.86
Type-II	ITO/2LG	76.6	0.57
	ITO/3LG	74.5	0.18
	ITO/4LG	74.1	0.64
	ITO/5LG	72.1	0.69

Table S1. Transmittances of type-I and type-II structures at a wavelength of 550 nm.

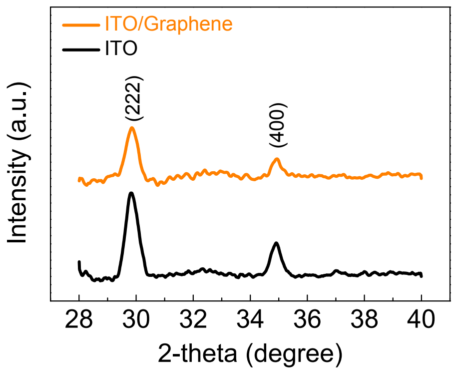

Figure S4. XRD diffraction patterns of ITO and ITO/graphene on the PET substrates.

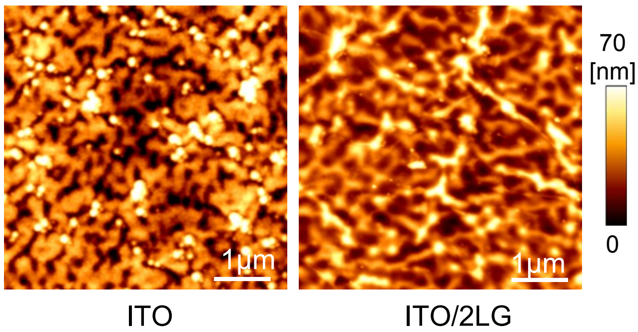

Figure S5. Topographies of ITO and ITO/2LG on PET substrates.

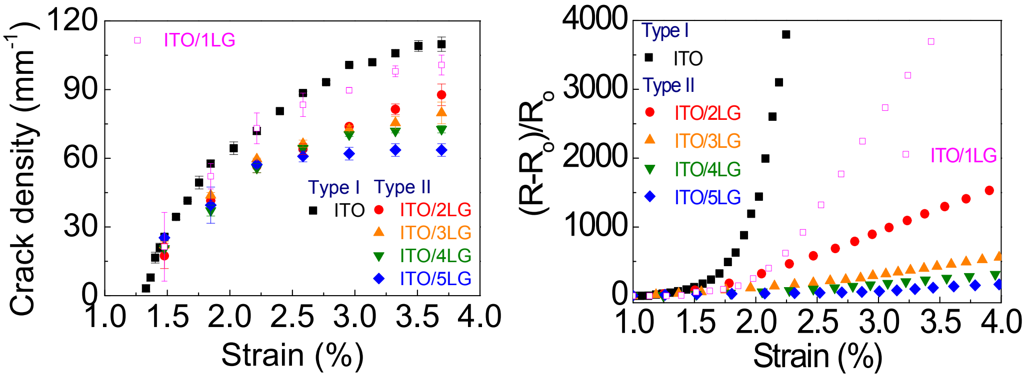

Figure S6. Crack densities and the normalized electrical resistance for the ITO/1LG. a) Crack densities for the ITO/1LG, type-I and type-II structures as a function of the strain. b) The normalized electrical resistance of the ITO/1LG, type-I and type-II structures as a function of the strain.