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. The governing equation is derived from the force equilibrium relating the interfacial shear stress \( \tau(x) \) of the graphene layer to the tensile stress \( \sigma \) transferred to the ITO layer; i.e.,

\[
q \frac{d\sigma(x)}{dx} = -\tau(x)
\]  
(S1)

The shear stress in the multilayer graphene is given by

\[
\tau(x) = \frac{G(\varepsilon x - \Phi(x))}{p}
\]  
(S2)

when \( x \leq x_p \) and

\[
\tau(x) = G\gamma_p = \tau_p
\]  
(S3)

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

\[
\sigma(x) = E \frac{d\Phi(x)}{dx}
\]  
(S4)

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

\[
\frac{d^2\Phi}{dx^2} - \beta^2 \Phi(x) = -\beta^2 \varepsilon x
\]  
(S5)

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

\[
\Phi(x = 0) = 0
\]  
(S6)

and
\[ \sigma(a) = E \frac{d\phi(x)}{dx} \bigg|_{x=a} = 0. \]  \hspace{1cm} (S7)

The solution to Eq. (S5) is given by

\[ \phi(x) = \varepsilon \left( x - \frac{\sinh(\beta x)}{\beta \cosh(\beta a)} \right). \]  \hspace{1cm} (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 \( \sigma^* \), 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

\[ \sigma(0) = \frac{1}{a} \int_{0}^{a} \tau(x) dx. \]  \hspace{1cm} (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.,

\[ \sigma(0) = \frac{1}{a} \int_{0}^{a} \tau(x) dx = \sigma^*, \]  \hspace{1cm} (S10)

and we have

\[ a = \beta \cosh^{-1} \left( \frac{1}{1 - \frac{\sigma^*}{\varepsilon E}} \right). \]  \hspace{1cm} (S11)

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

\[ n = \frac{1}{2a} = \sqrt{\frac{G}{E}} \left[ 2 \sqrt{\beta q \cosh^{-1} \left( \frac{1}{1 - \frac{\sigma^*}{\varepsilon E}} \right)} \right]^{-1}. \]  \hspace{1cm} (S12)
② The elastic–plastic case \((x_p < a)\)

When \(x < x_p\), Eq. \((S11)\) can be used for \(a\); i.e.,
\[
\tau(x) = \frac{G}{p\beta} \left(\frac{\epsilon - \sigma^*}{E}\right) \sinh(\beta x) \tag{S13}
\]
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
\[
x_p = \frac{1}{\beta \sinh^{-1} \left(\frac{G\gamma_p}{\sqrt{qE}} \left(\epsilon - \frac{\sigma^*}{E}\right)\right)} \tag{S15}
\]
and \(a\) is equal to half of the maximum crack length when the maximum stress becomes equal to the tensile strength. We therefore have
\[
\sigma(0) = \frac{1}{q} \int_0^a \tau(x) dx = \frac{1}{q} \int_0^{x_p} \tau(x) dx + \int_{x_p}^a G\gamma_p dx = \sigma^* \tag{S16}
\]
which leads to
\[
a = \frac{Eq}{G\gamma_p} \left[\frac{\sigma^*}{E - \epsilon} \cosh \left(\sinh^{-1} \left(\frac{G\gamma_p}{\sqrt{qE}} \left(\epsilon - \frac{\sigma^*}{E}\right)\right)\right) + \epsilon + G\gamma_p \frac{G\gamma_p}{\sqrt{qE}} \sinh^{-1} \left(\frac{G\gamma_p}{\sqrt{qE}} \left(\epsilon - \frac{\sigma^*}{E}\right)\right)\right] \tag{S17}
\]
Because the crack density is \(n = \frac{1}{2a}\), this can be expressed as follows:
\[
n = \frac{G\gamma_p}{2Eq} \left[\frac{\sigma^*}{E - \epsilon} \cosh \left(\sinh^{-1} \left(\frac{G\gamma_p}{\sqrt{qE}} \left(\epsilon - \frac{\sigma^*}{E}\right)\right)\right) + \epsilon + G\gamma_p \frac{G\gamma_p}{\sqrt{qE}} \sinh^{-1} \left(\frac{G\gamma_p}{\sqrt{qE}} \left(\epsilon - \frac{\sigma^*}{E}\right)\right)\right]^{-1} \tag{S18}
\]
Figure S1. Schematic of an off-axis RF magnetron sputtering.

Figure S2. Raman spectra (excitation wavelength $\lambda = 514\text{nm}$) 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.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Transmittance (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean</td>
</tr>
<tr>
<td>Type-I</td>
<td>ITO</td>
</tr>
<tr>
<td>Type-II</td>
<td>ITO/2LG</td>
</tr>
<tr>
<td></td>
<td>ITO/3LG</td>
</tr>
<tr>
<td></td>
<td>ITO/4LG</td>
</tr>
<tr>
<td></td>
<td>ITO/5LG</td>
</tr>
</tbody>
</table>

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.