

# Supporting Information

## Investigating Multiaxial Mullins Effect of Carbon Black Reinforced Elastomers using Electrical Resistivity Measurements

Yuga Taniguchi,<sup>1,†</sup> Thanh-Tam Mai,<sup>1,†</sup> Masashi Yamaguchi,<sup>2</sup> Katsuhiko Tsunoda,<sup>2</sup> Kenji Urayama<sup>1,\*</sup>

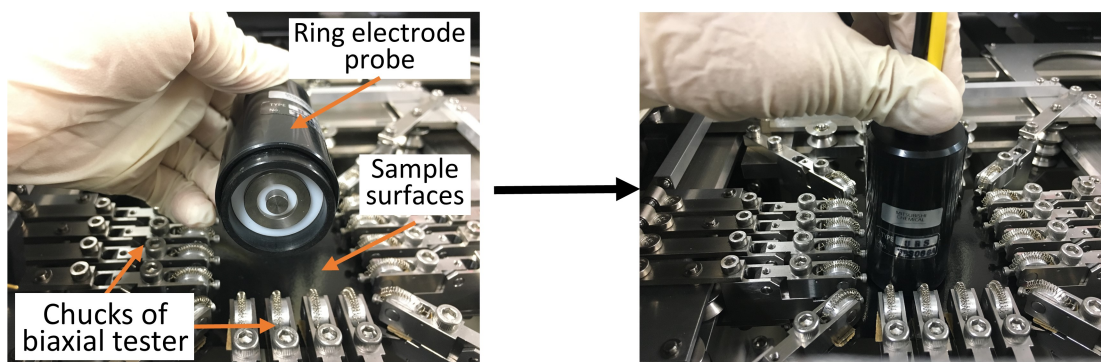
<sup>1</sup> Department of Macromolecular Science & Engineering, Kyoto Institute of Technology,  
Sakyo-ku, Kyoto 606-8585, Japan

<sup>2</sup> Research Department I, Central Research, Bridgestone Corporation,  
Tokyo 187-8531, Japan

<sup>†</sup>These authors contributed equally to this work.

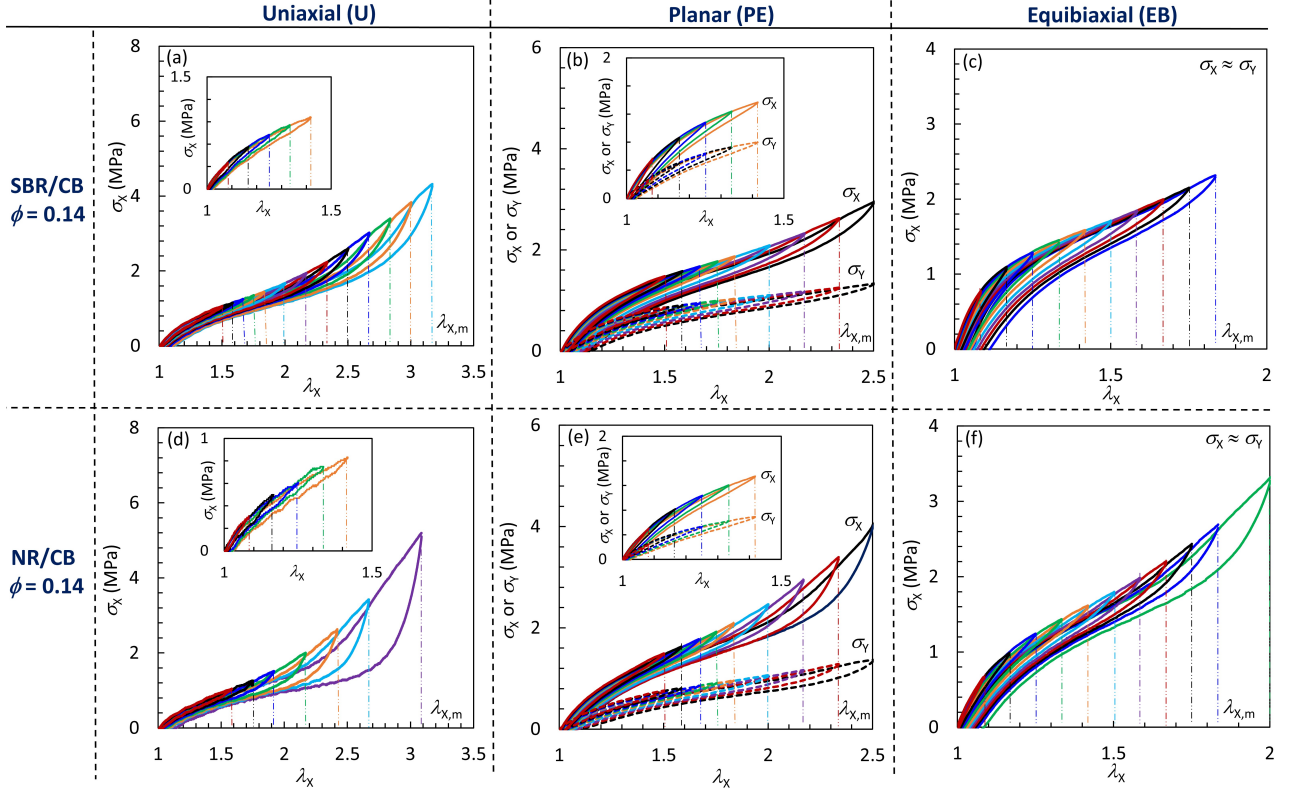
\*Corresponding author: urayama@kit.ac.jp

### 1. The resistivity measurement under biaxial deformation



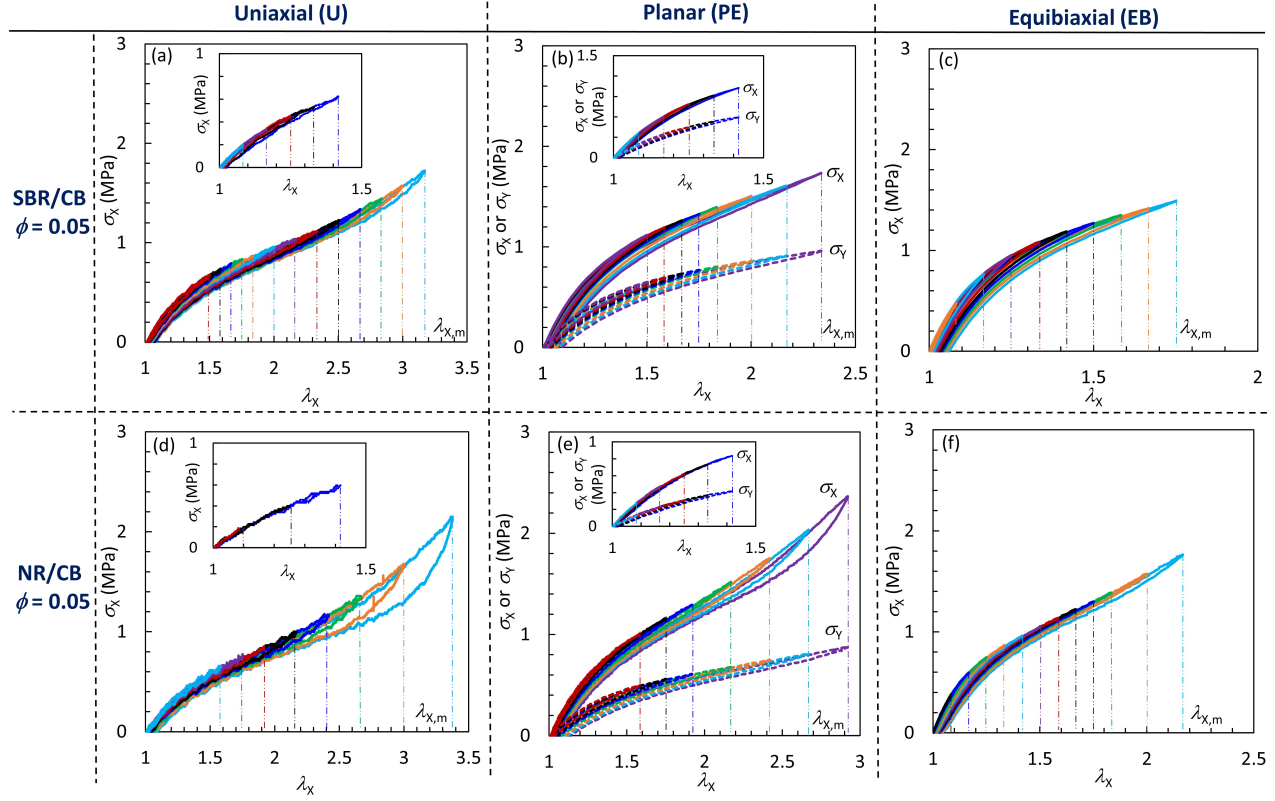
**Figure S1.** The illustration of resistivity measurement under biaxial deformation by using the ring electrode probe.

**2. Loading-unloading cycles under various deformation types for SBR/CB and NR/CB with  $\phi = 0.14$**



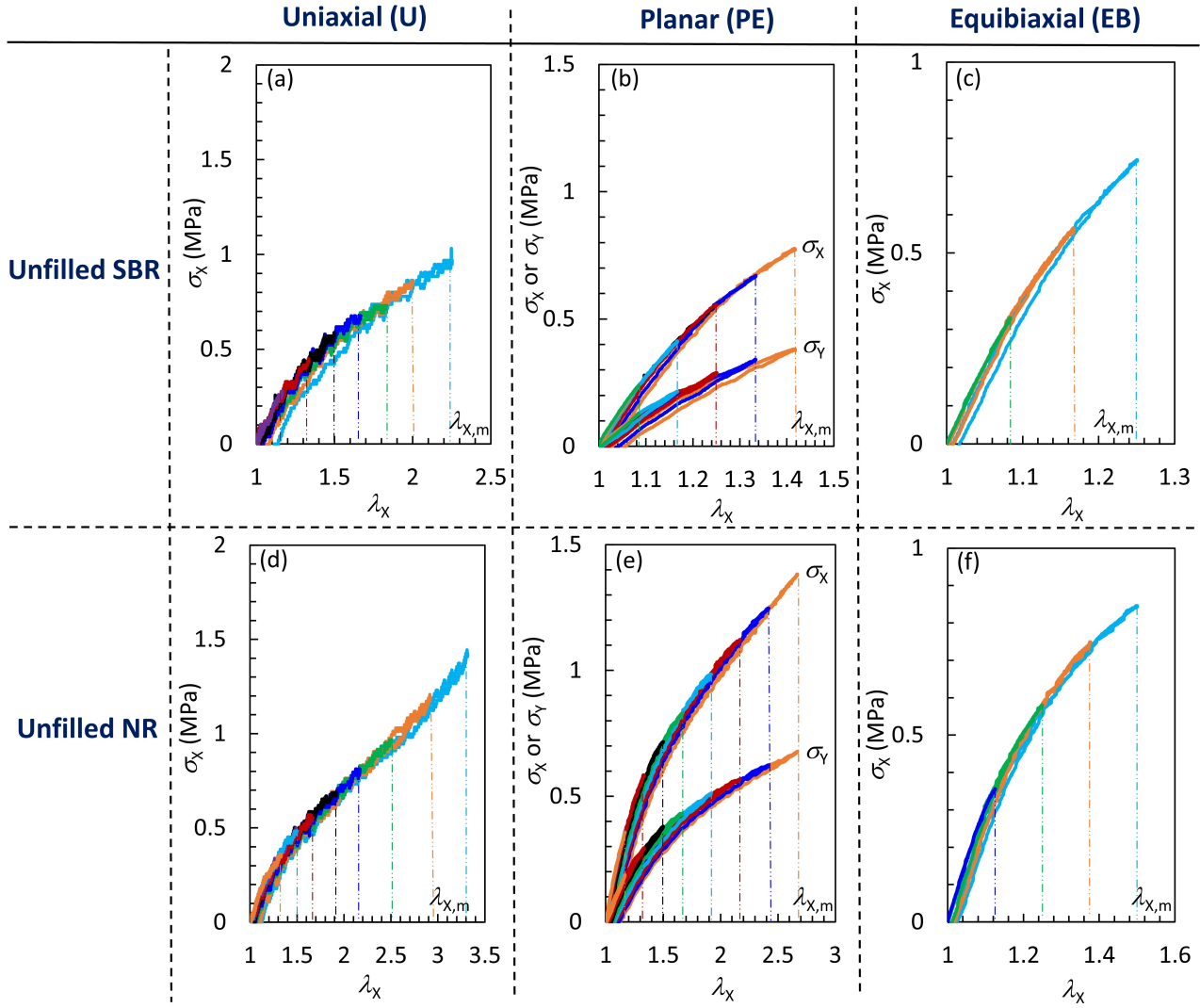
**Figure S2.** Nominal  $\sigma$ - $\lambda_x$  relation in loading-unloading cycles with uniaxial, planar, equibiaxial extensions in the x- and y-directions for (a-c) SBR/CB and (d-f) NR/CB with  $\phi = 0.14$ . The vertical dashed lines point out the  $\lambda_{x,m}$  values in each cycle. The insets in figures are the magnifications of the small  $\lambda_x$  region.

### 3. Loading-unloading cycles under various deformation types for SBR/CB and NR/CB with $\phi = 0.05$



**Figure S3.** Nominal  $\sigma$ - $\lambda_x$  relation in loading-unloading cycles with uniaxial, planar, equibiaxial extensions in the x- and y-directions for (a-c) SBR/CB and (d-f) NR/CB with  $\phi = 0.05$ . The vertical dashed lines point out the  $\lambda_{x,m}$  values in each cycle. The insets in figures are the magnifications of the small  $\lambda_x$  region.

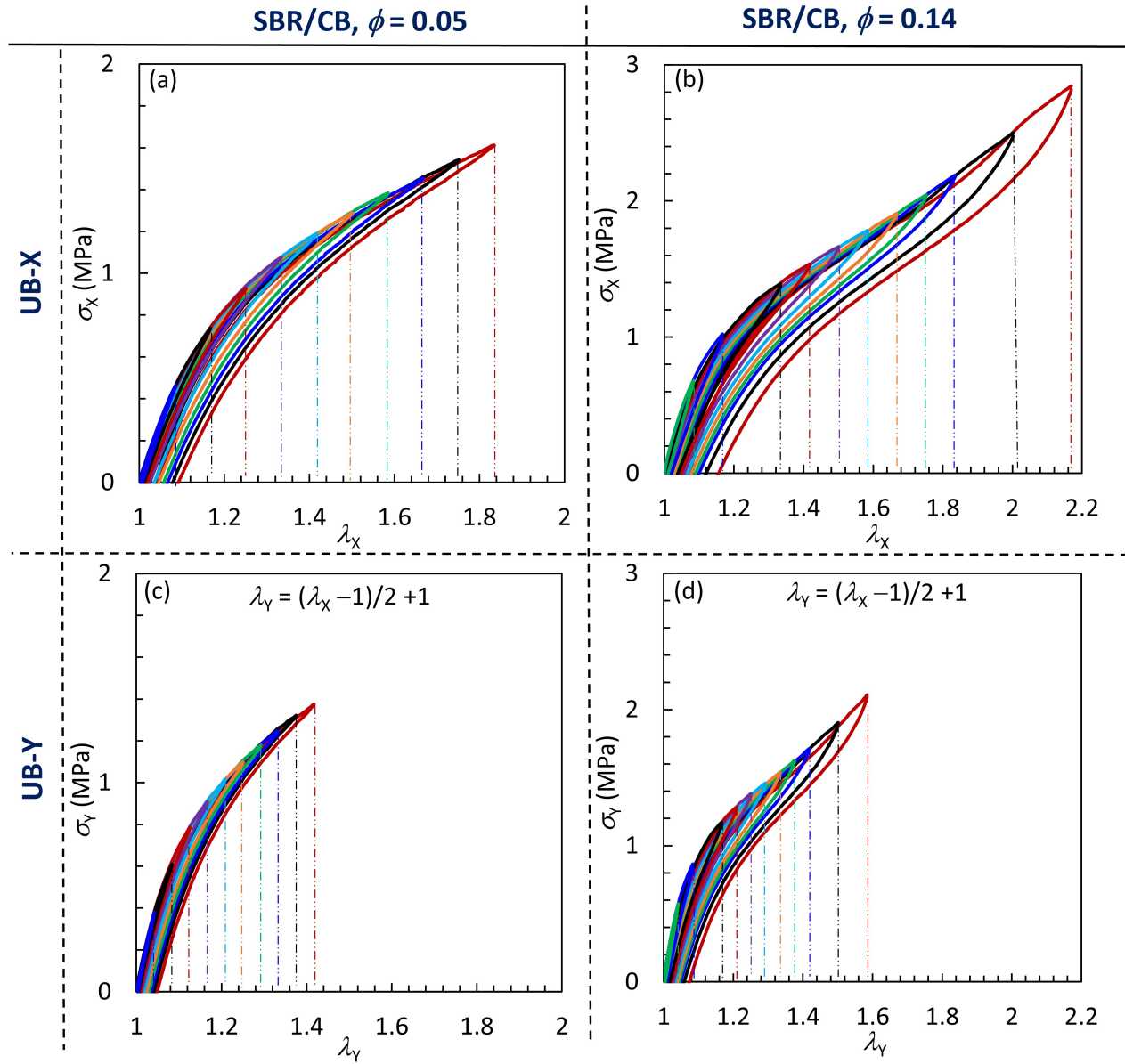
#### 4. Loading-unloading cycles under various deformation types for unfilled SBR and NR



**Figure S4.** Nominal  $\sigma$ - $\lambda_x$  relation in loading-unloading cycles with uniaxial, planar, equibiaxial extensions in the x- and y-directions for unfilled (a-c) SBR and (d-f) NR. The vertical dashed lines point out the  $\lambda_{x,m}$  values in each cycle.

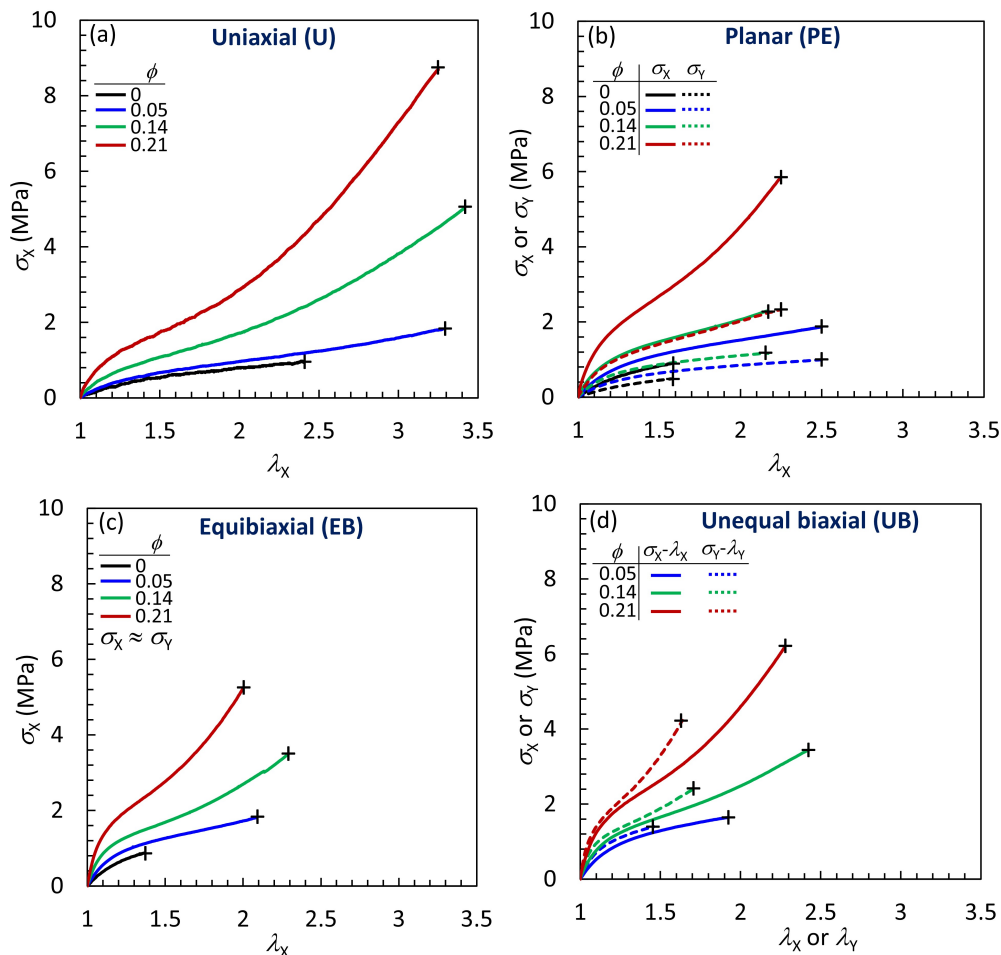


5. Loading-unloading cycles under unequal biaxial extension for SBR/CB with  $\phi = 0.14$  and 0.05.



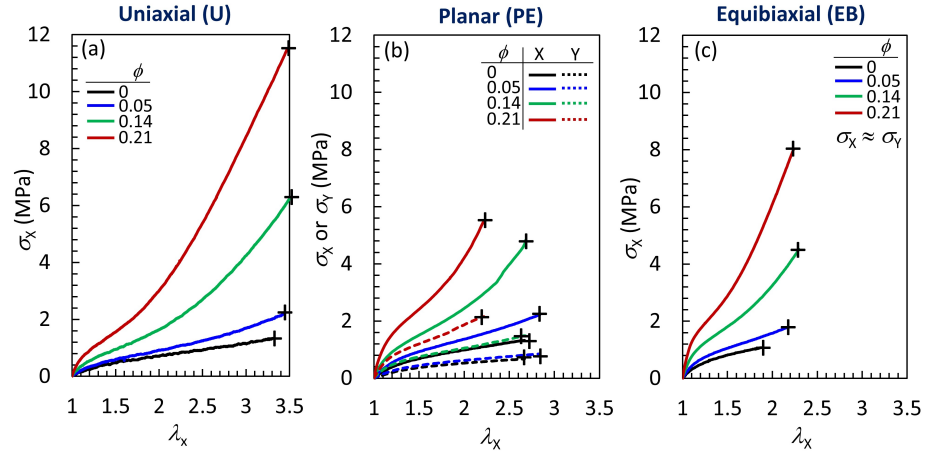
**Figure S5.** Nominal  $\sigma_x$ - $\lambda_x$  and  $\sigma_y$ - $\lambda_y$  relations in loading-unloading cycles with unequal biaxial extensions in the (a-b) x- and (c-d) y-directions for SBR/CB with  $\phi = 0.05$  and 0.14. The vertical dashed lines point out the  $\lambda_{x,m}$  values in each cycle.

## 6. Pristine loading under various deformation types for SBR/CB with various $\phi$



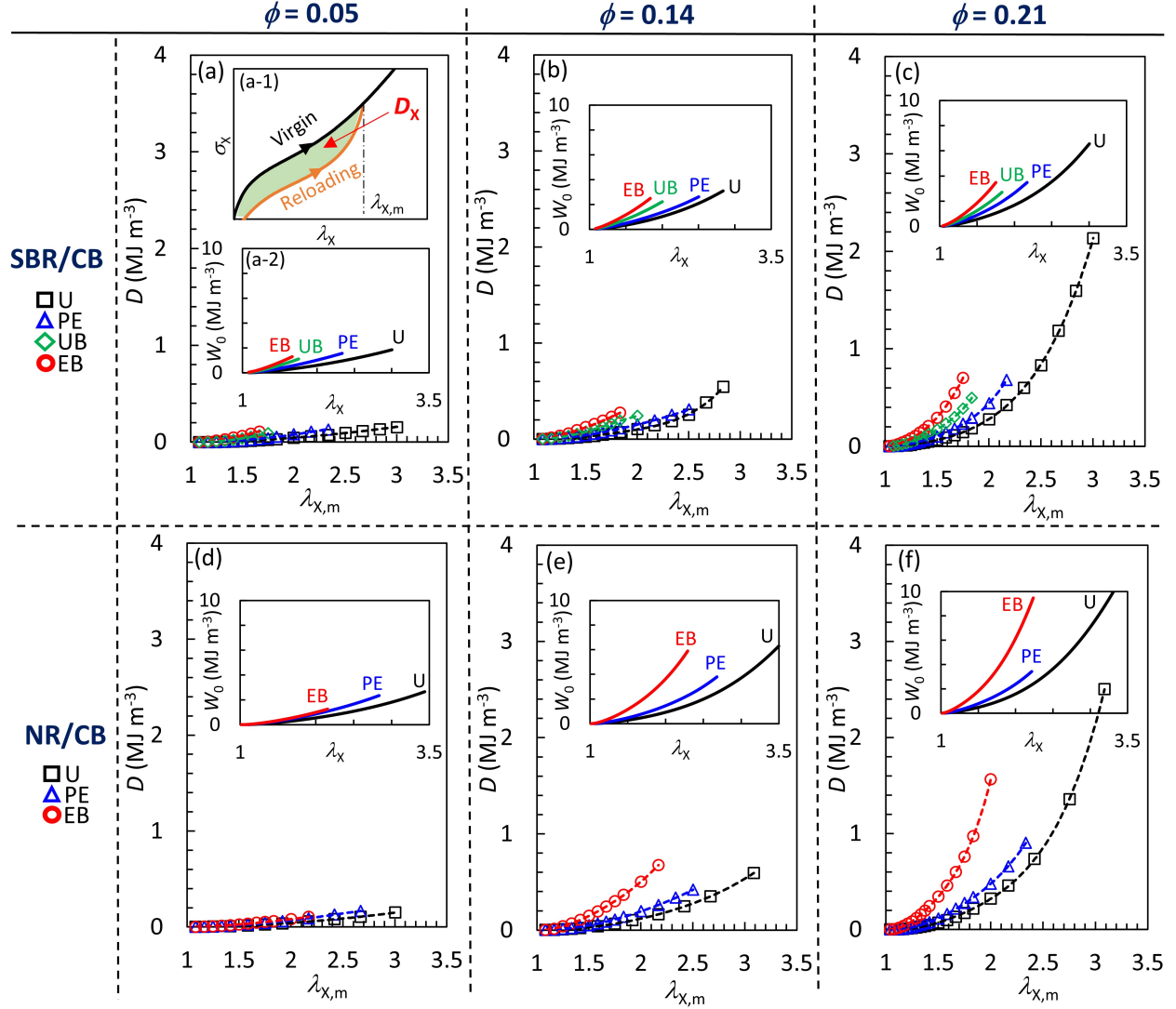
**Figure S6.** Nominal stress ( $\sigma_x$  or  $\sigma_y$ ) elongation ( $\lambda_x$  or  $\lambda_y$ ) curves under pristine loading with (a) uniaxial, (b) planar, (c) equibiaxial, and (d) unequal biaxial extensions for SBR/CB with various volume fractions of CB ( $\phi$ ). The cross symbols denote the rupture points.

## 7. Pristine loading under various deformation types for NR/CB with various $\phi$



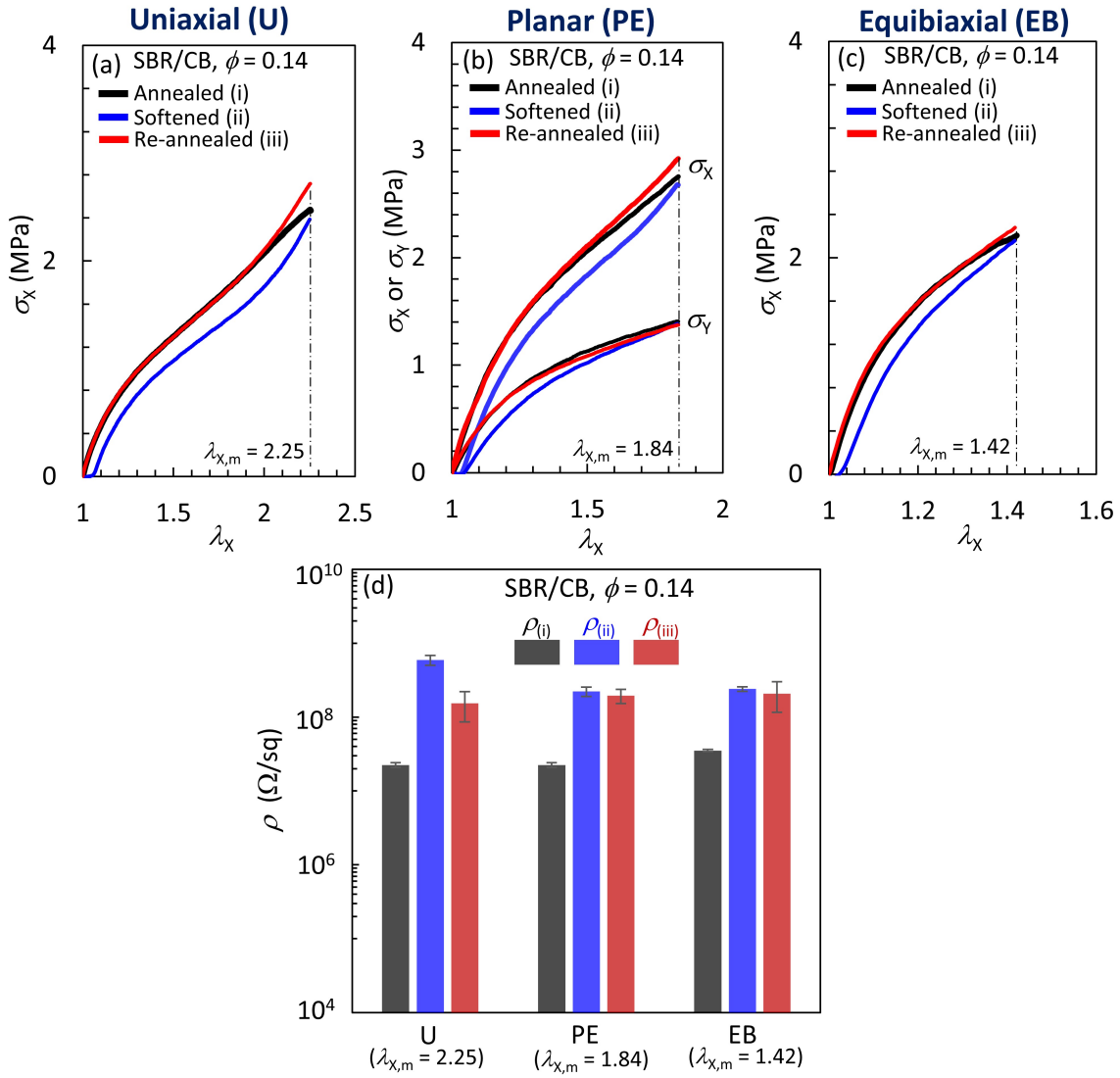
**Figure S7.** Nominal  $\sigma_x$ – $\lambda_x$  curves under pristine loading with (a) uniaxial, (b) planar, and (c) equibiaxial extensions for NR/CB with various values of  $\phi$ . The cross symbols denote the rupture points.

**8. Energy dissipation per unit volume ( $D$ ) under various deformation types for SBR/CB and NR/CB with various  $\phi$**



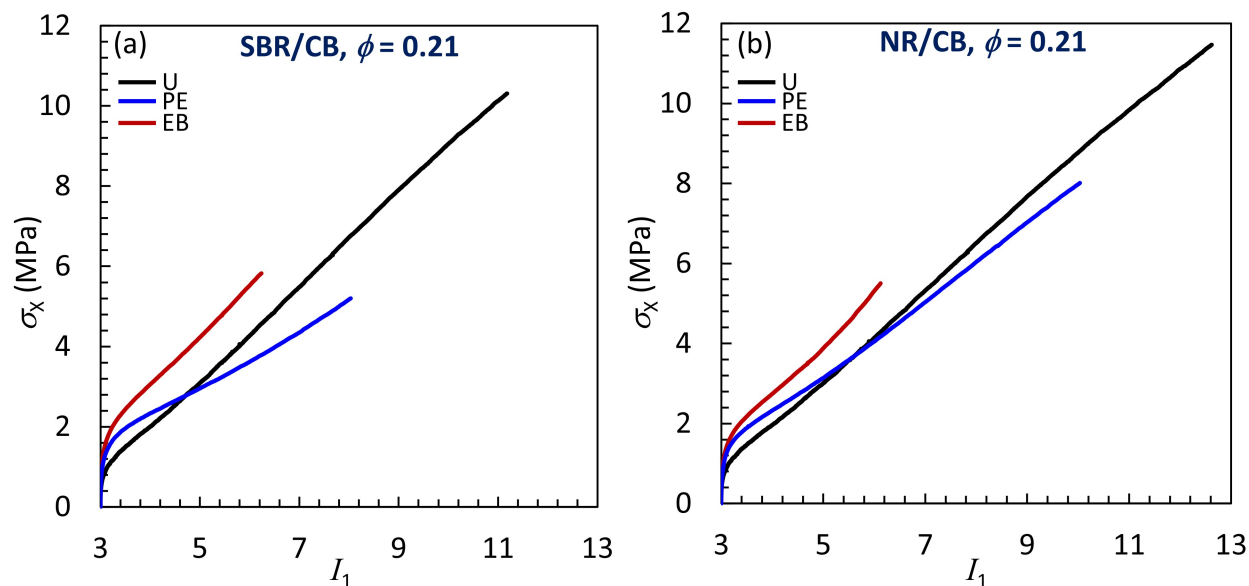
**Figure S8.** Energy dissipation ( $D$ ) as a function of  $\lambda_{x,m}$  under various deformation types for (a–c) SBR/CB and (d–f) NR/CB with  $\phi = 0.05, 0.14$ , and  $0.21$ . The inset in (a) illustrates the estimation of  $D(\lambda_{x,m})$  from the pristine loading and reloading curves. The insets in (a)–(f) show the mechanical work per unit volume in pristine loading ( $W_0$ ) as a function of  $\lambda_x$ . The dashed lines in each figure represent guides for the eyes.

## 9. Mechanical and electrical recoveries by thermal annealing of SBR/CB with $\phi = 0.14$



**Figure S9.** Comparison of (a-c) nominal  $\sigma$ - $\lambda_x$  relation under a single loading-unloading cycle and (d) surface electrical resistivity ( $\rho$ ) among the three specimens (i), (ii), and (iii) for SBR/CB with  $\phi = 0.14$  in each of uniaxial (U), planar (PE), and equibiaxial (EB) extensions. The error bar in each column chart indicates the standard deviation.

# 10. Stresses as a function of the first invariant ( $I_1$ ) under various deformation types



**Figure S10.** Nominal  $\sigma_x$ – $I_1$  relation under various deformation types for (a) SBR/CB and (b) NR/CB with  $\phi = 0.21$ . The  $\sigma_x$  values were reproduced from Figures S2-S3.