Supporting Information for "Structural Evolution of OBC/Carbon Nanotube Bundle Nanocomposites under Uniaxial Deformation"

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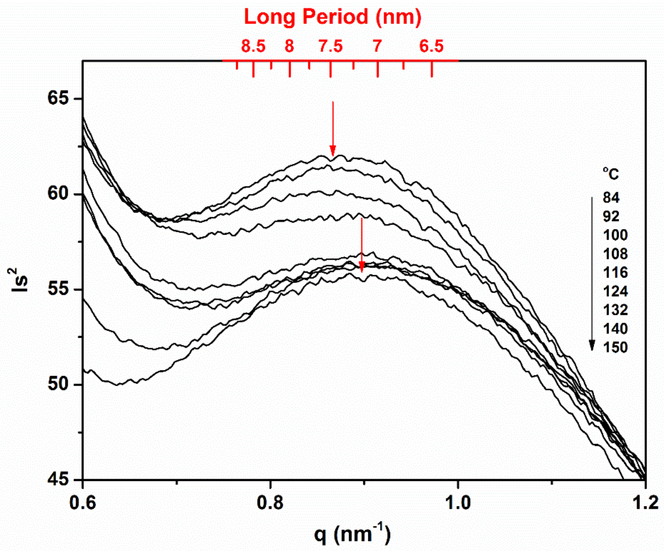
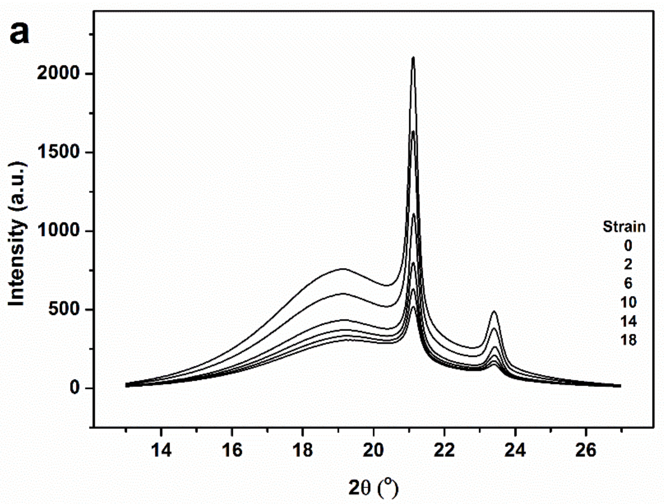


Figure S1. Selected Lorentz-corrected integrated SAXS profiles as a function of temperature for CNTB2 at a heating rate of 5 ℃/min.



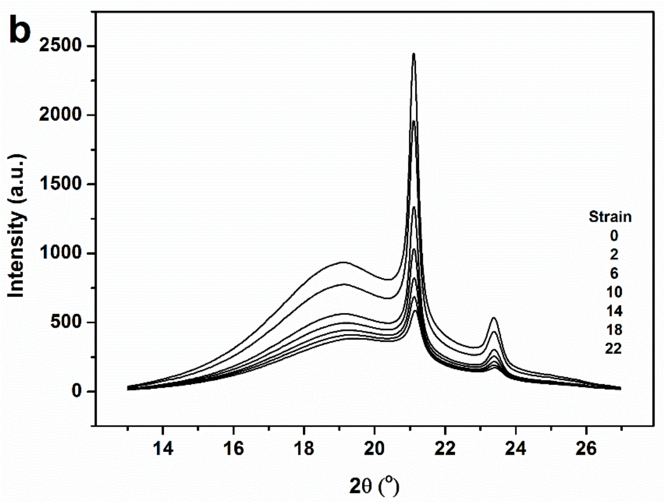


Figure S2. Selected integrated WAXD profiles of (a) NeatOBC and (b) CNTB2.

The (010) monoclinic reflection is highly oriented by relatively diffuse. The (110) and (200) orthorhombic crystal reflections as well as amorphous reflection are all effected by the contribution of (010) monoclinic reflection. And the azimuthal intensity distribution exhibit six peaks after multiple Gauss peak fit. The peak contributed by (010) monoclinic reflection can be perfectly subtracted, as is illustrated in Figure S3.

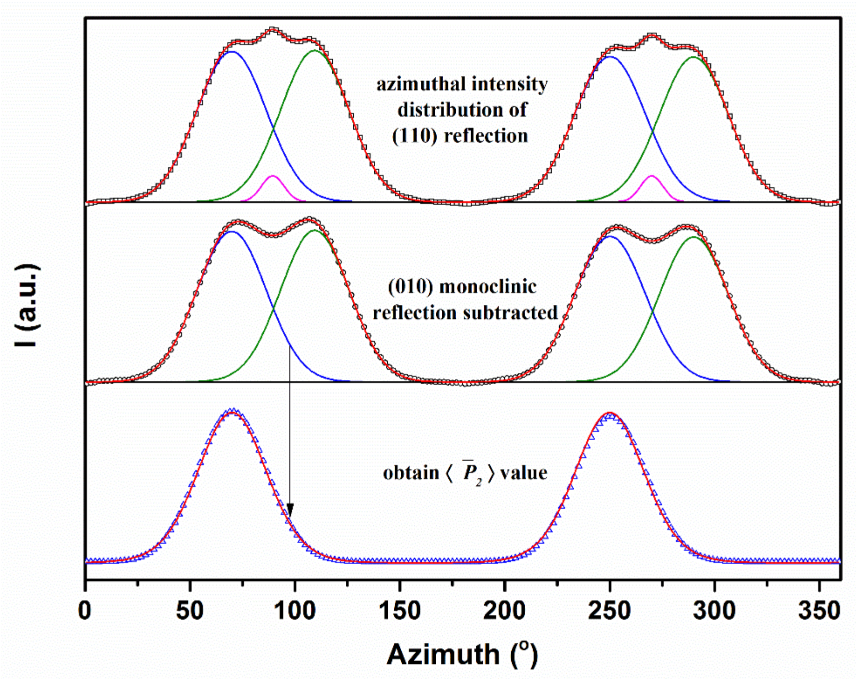
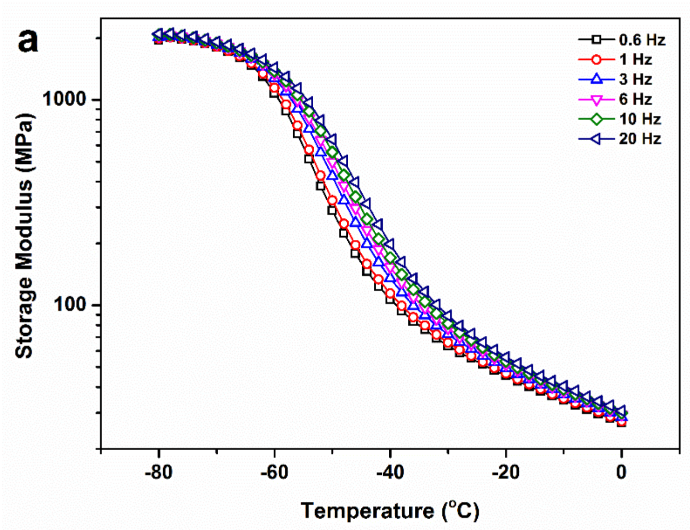


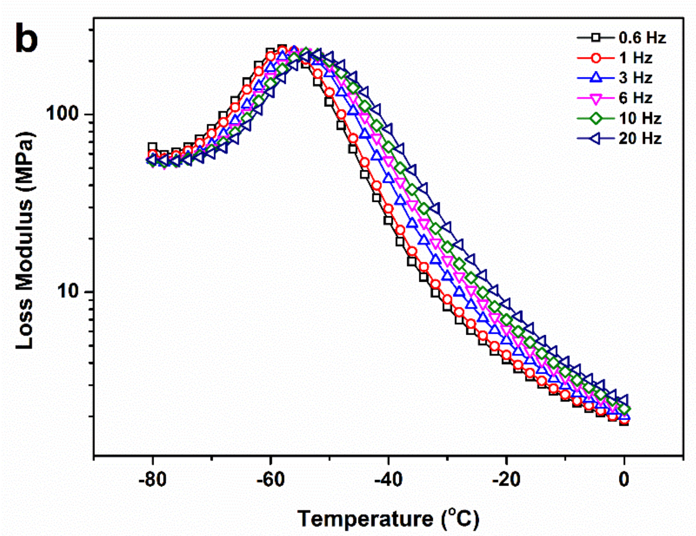
Figure S3. Representative azimuthal intensity distribution of the (110) reflection of NeatOBC at the strain of 1200%. The pink curve is the contribution of (010) monoclinic reflection.

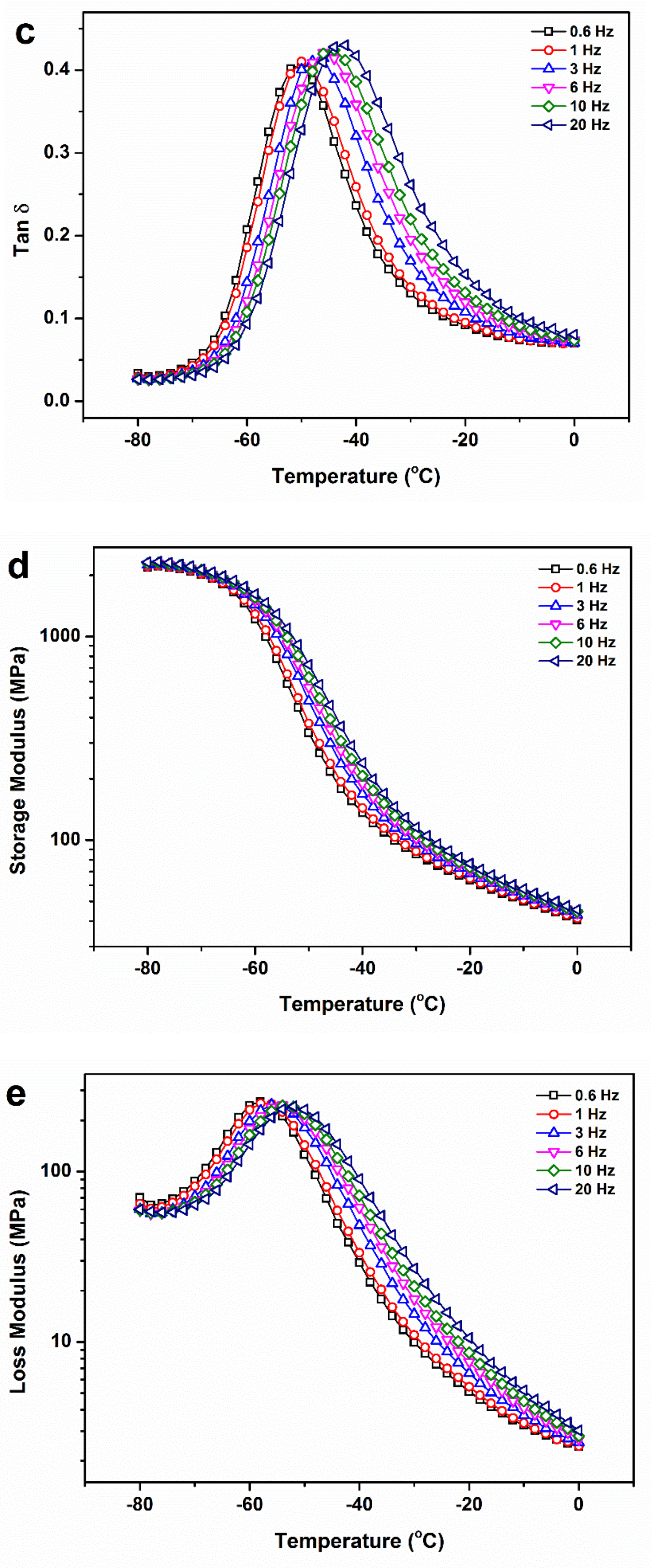
The storage modulus, loss modulus and tan δ in dynamic mechanical multifrequency tests in temperature scanning mode in the range of -80 to 0 ℃ is presented in Figure S4a-f. Compare to the peak position and value of tan δ, the activation energy of glass transition is more convincing to describe the chain mobility. Activation energy of transition can be evaluated from the frequency-dependent spectrum. And the relaxation frequency can be represented by the Arrhenius equation:

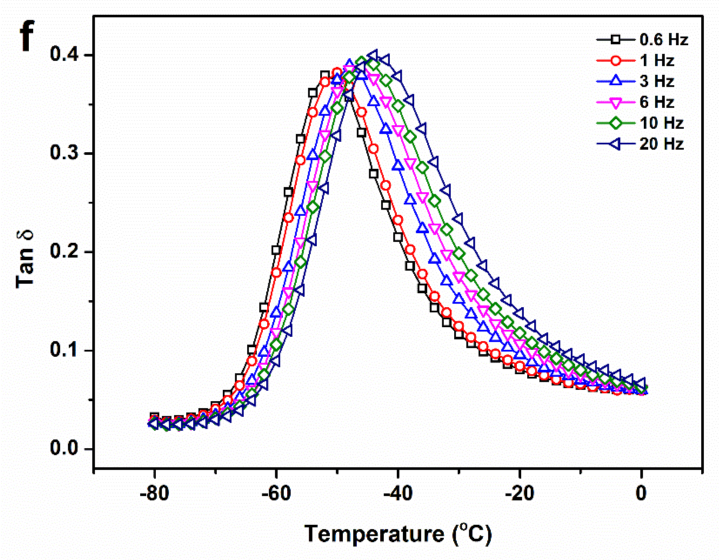
$$\ln f = A - \frac{E_a}{RT}$$

where f is the peak frequency, A is the logarithm form of preexponential factor, E_a is the activation energy, R is the gas constant and T is temperature. The Arrhenius plot is shown in Figure S2g, from which we can see the Arrhenius plots of NeatOBC and CNTB2 showed almost no difference. The calculated E_a is 236.11 kJ for NeatOBC, and 236.50 kJ for CNTB2.






c
0.6 Hz
1 Hz
3 Hz
6 Hz
10 Hz
20 Hz
Tan δ
0.0
0.1
0.2
0.3
0.4
-80
-60
-40
-20
0
Temperature (°C)
d
0.6 Hz
1 Hz
3 Hz
6 Hz
10 Hz
20 Hz
Storage Modulus (MPa)
100
1000
-80
-60
-40
-20
0
Temperature (°C)
e
0.6 Hz
1 Hz
3 Hz
6 Hz
10 Hz
20 Hz
Loss Modulus (MPa)
10
100
-80
-60
-40
-20
0
Temperature (°C)



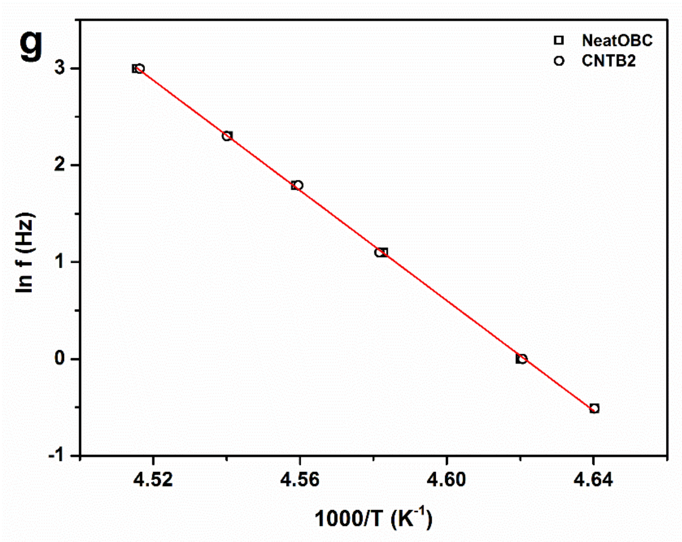


Figure S4. Storage modulus, loss modulus and tan δ of NeatOBC (a, b, c) and CNTB2 (d, e, f). (g) Arrhenius plot of β transition computed in the frequency range of 0.6 to 20 Hz for NeatOBC and CNTB2 (The red line is the linear fitting curve of the data points).

As is reported, the four-arc (110) reflection of OBC changes to two-arc-like at the strain of 500%. In our observation, the four-arc pattern is not very obvious, but the splitting of (110) reflection can be clearly seen in azimuthal intensity distribution curves in Figure S5, and the separated peaks approaches the stretching axis with increasing strain. We deduce the change of (110) reflection rings from four arcs to two arcs is contributed by both the contribution from (010) monoclinic reflection as well as the approaching of separated peaks of (110) reflection toward stretching axis.

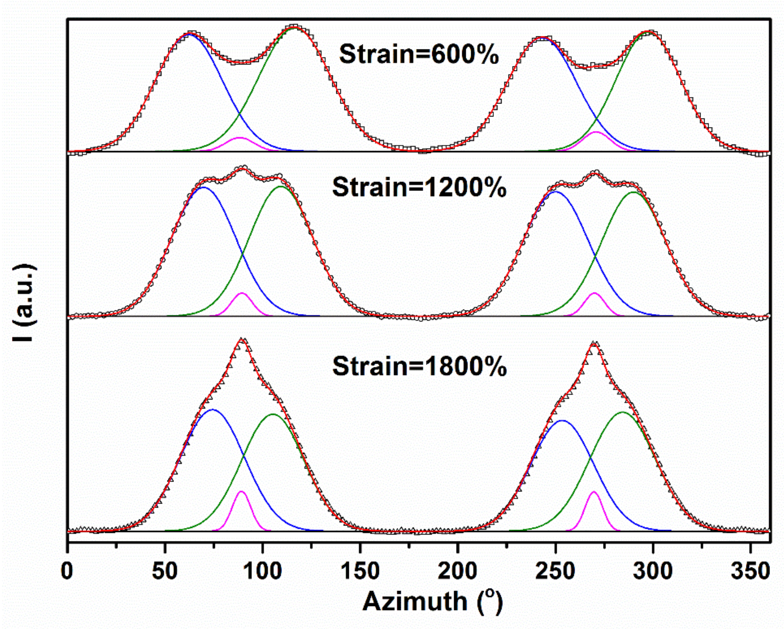


Figure S5. Representative azimuthal intensity distribution of (110) reflection of NeatOBC at the strain of 600%, 1200% and 1800%.

The contribution of (010) monoclinic reflection can also affect the azimuthal intensity distribution of (002) CNTB reflection of CNTB15. And the contribution of (010) reflection can be subtracted in the Gaussian peak fitting process.

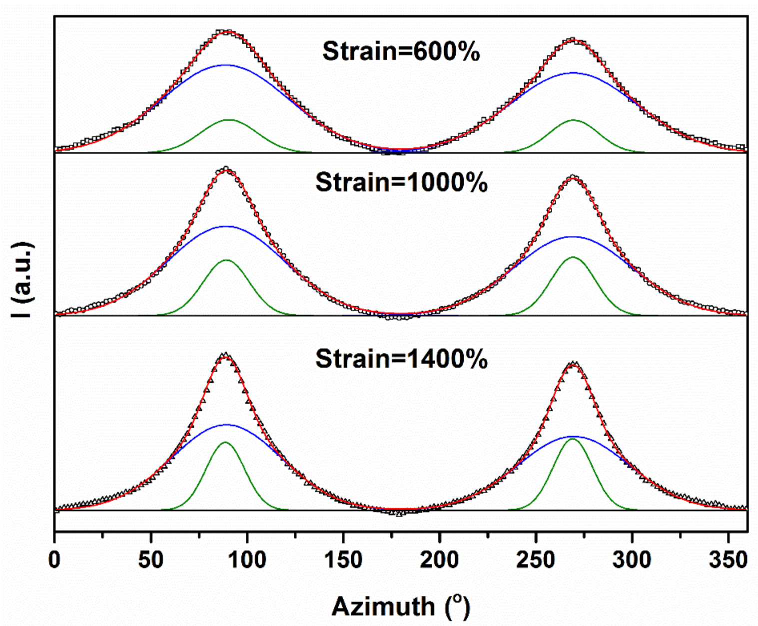


Figure S6. Representative azimuthal intensity distribution of (002) CNTB reflection of CNTB15 at the strain of 600%, 1000% and 1400%.