

Supplementary Information

Details of Raman and XRD spectra for the tested samples:

1. Experimental details

1.1 Raman characterization of the samples

Like most of the graphite materials, pyrocarbon presents two main peaks under laser excitation: G peak at $\sim 1575\text{ cm}^{-1}$ and D peak at $\sim 1330\text{ cm}^{-1}$. The G peak of pyrocarbon is related to the in plane E_{2g} symmetry of the grapheme lattice and the D peak is related to disorders in the grapheme lattice. The effective in-plane crystallite size (L_a) in each single graphene layer can be calculated using the following equation ²:

$$L_a = 4.4 \times \frac{1}{I_D/I_G}$$

where I_D is the intensity of D peak and I_G is the intensity of the G peak in Raman spectrums.

1.2 XRD characterization of the samples

XRD patterns were further obtained to analyze the out-of-plane crystallite parameters. XRD patterns for pyrocarbon matrix in the tested samples is carried out under the step-scanning mode in the range of $23^\circ \sim 27^\circ$, which corresponding to the (002) diffractions of graphite-related materials due to the stacking structure of graphene layers. The mean out-of-plane crystallite size L_c along the stacking orientation of pyrocarbon matrix can be estimated using the Scherrer-Warren equation:

$$L_c = \frac{K\lambda}{B\cos\theta}$$

where K is a geometry dependent constant (0.9 for graphite-related materials), λ is the wave length of the applied X-ray (Cu K α , 0.154nm), B is the full-width-half-max (FWHM) of the peak and θ is the Bragg angle.

2. Testing results

2.1 Raman spectra for the prepared samples

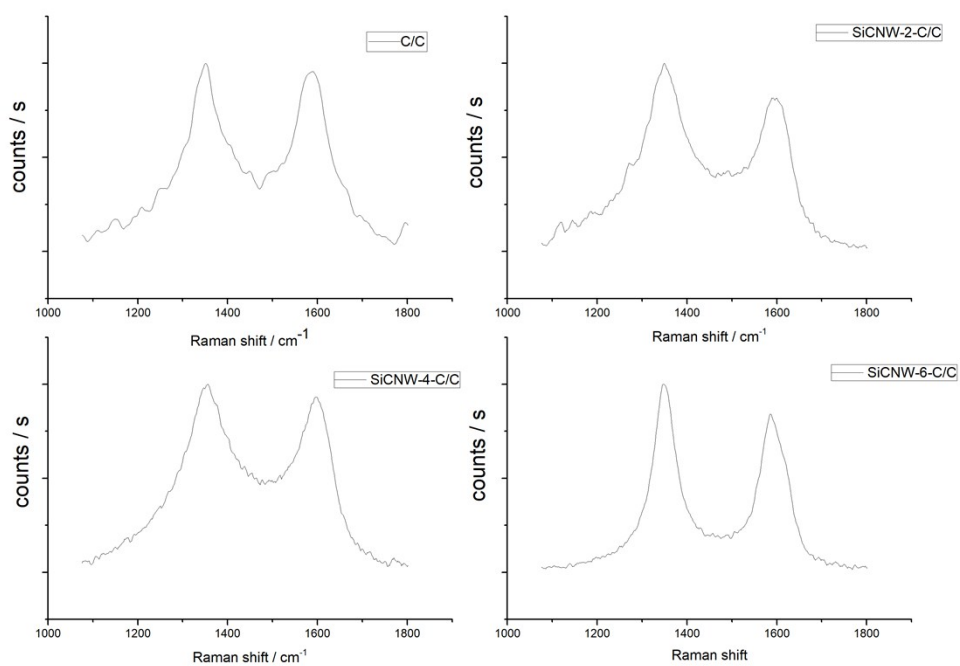


Fig.1 Raman spectra for the tested samples

2.2 XRD spectra for the prepared samples

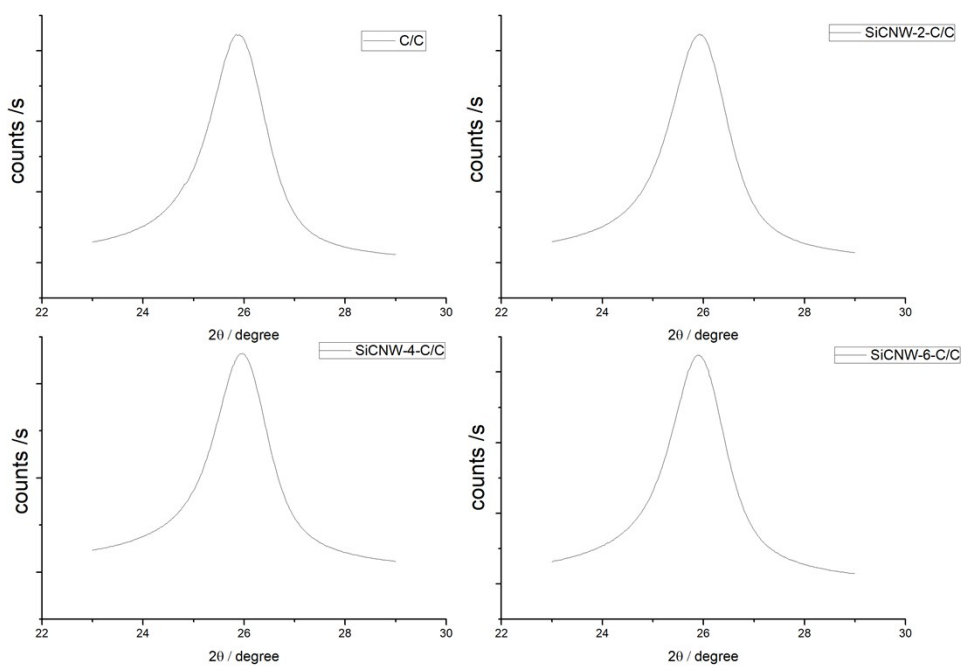


Fig.2 XRD spectra for the tested samples

References:

1. F. Tuinstra and J. L. Koenig, *Journal of Composite Materials*, 1970, **4**, 492-&.
2. F. Tuinstra and J. L. Koenig, *Journal of Chemical Physics*, 1970, **53**, 1126-&.