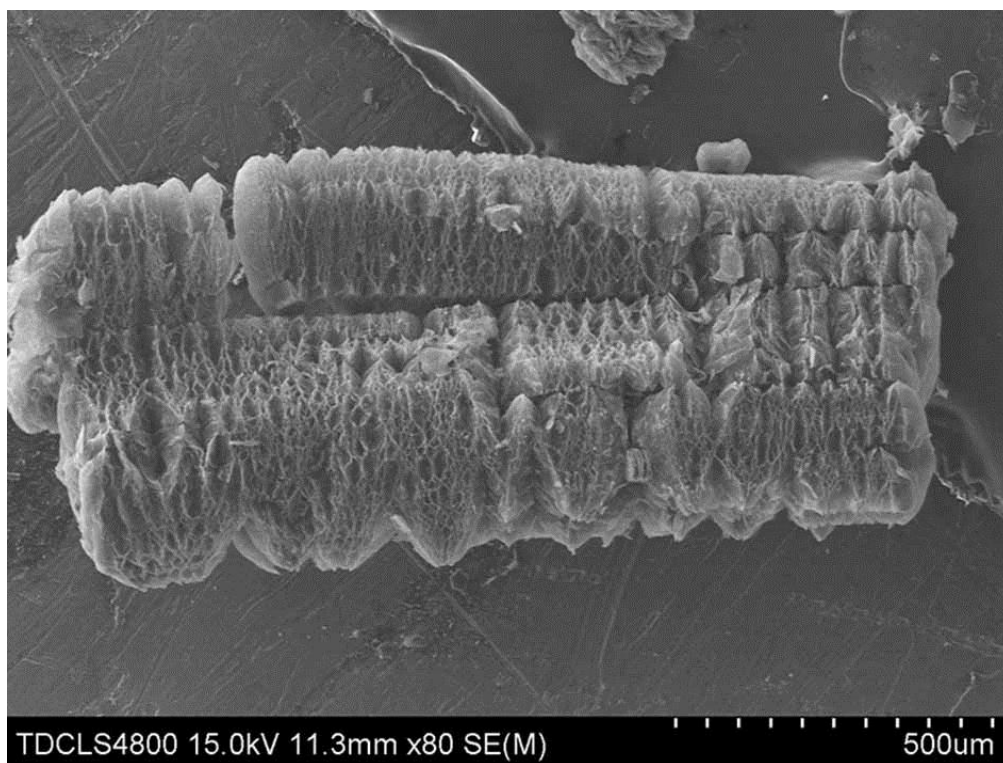


## Electronic Supplementary Information

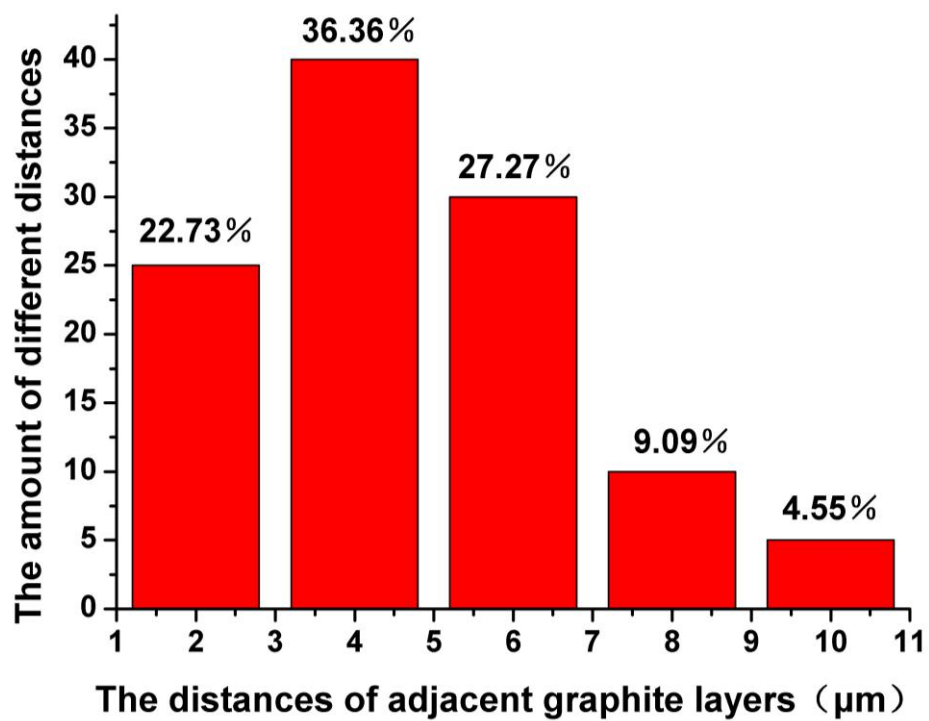
### **Enhanced cross-plane thermal conductivity and high resilience of three-dimensional hierarchical carbon nanocoil-graphite nanocomposites**

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**Fig. S1** SEM image of EG.



**Fig.S2** The distances distribution histograms of adjacent graphite layers.

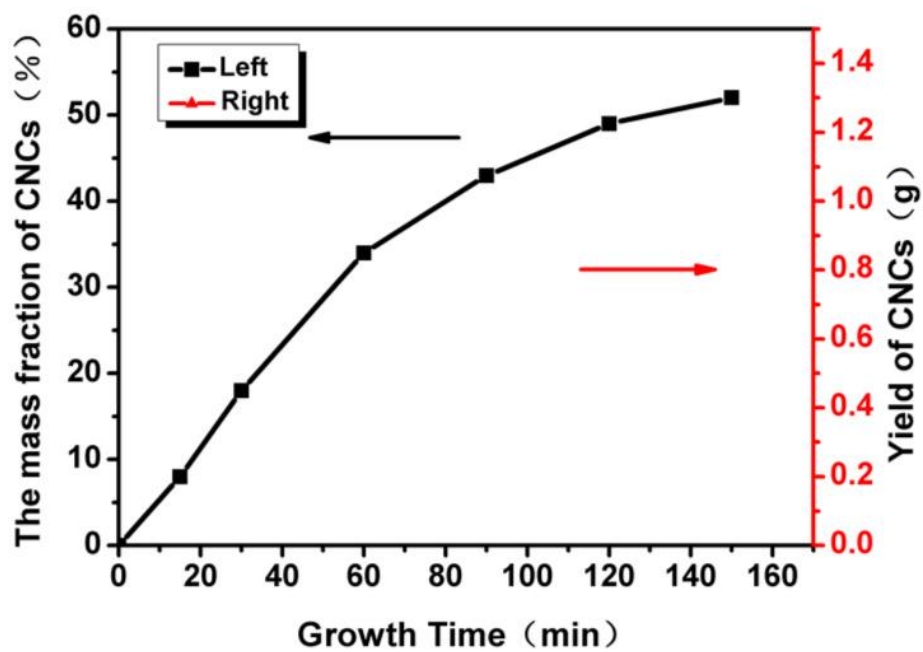
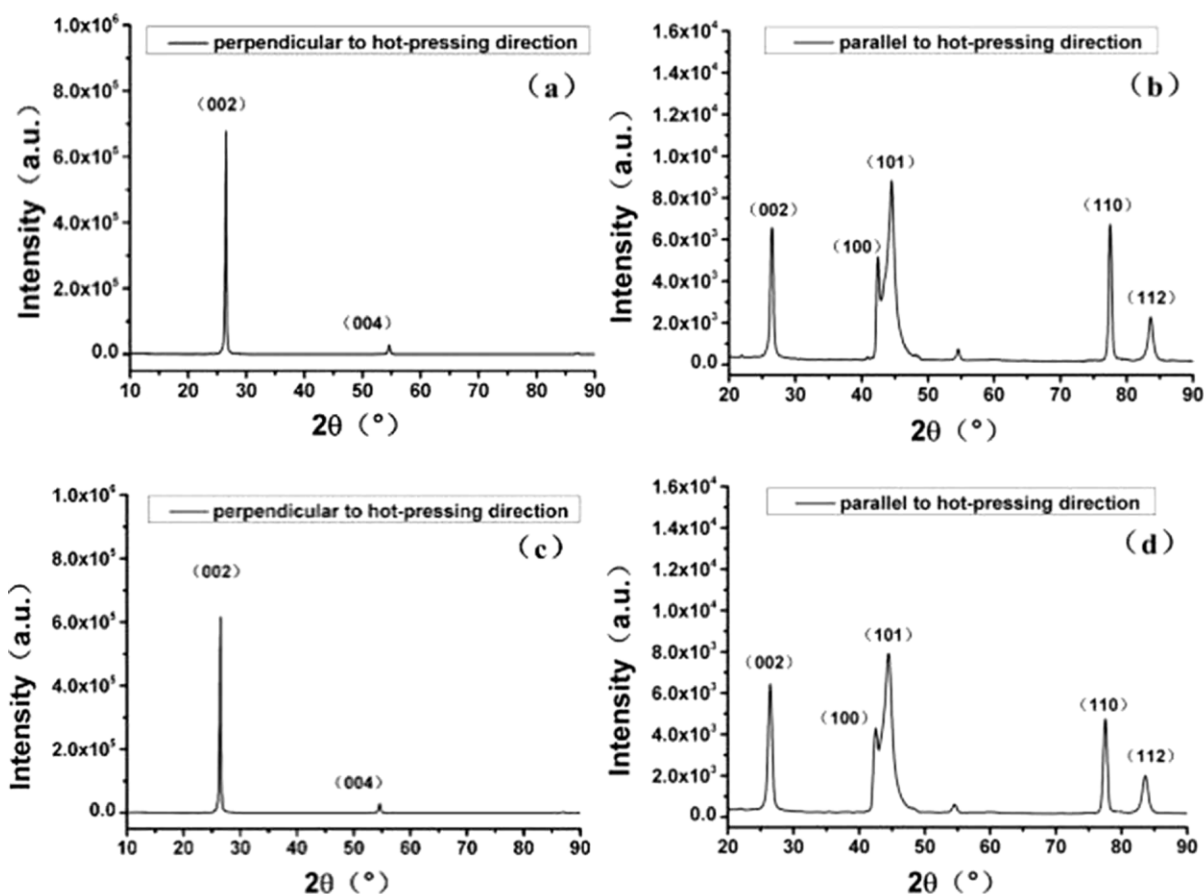


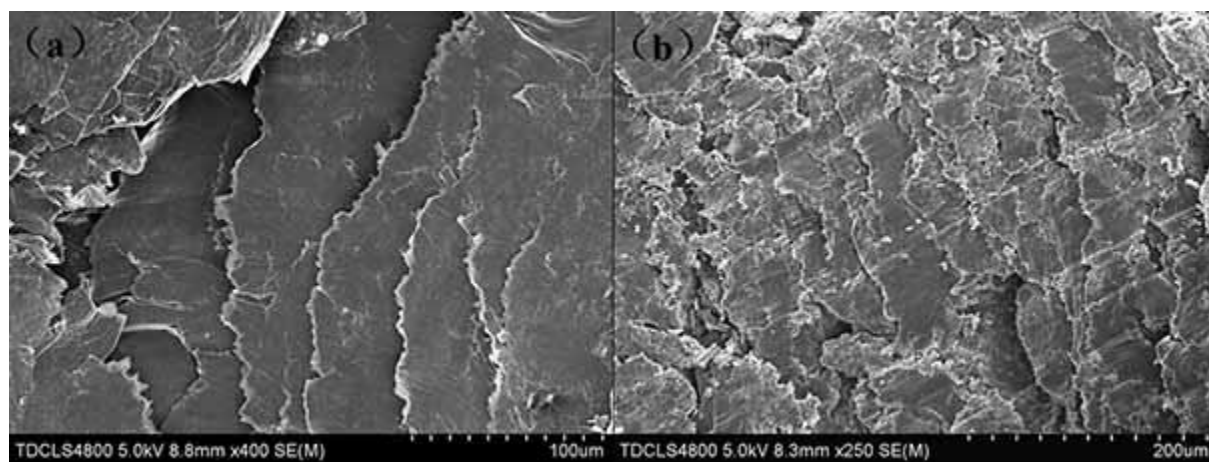
Fig. S3 The mass fraction of CNC in CNC-EG composite as a function of growth time



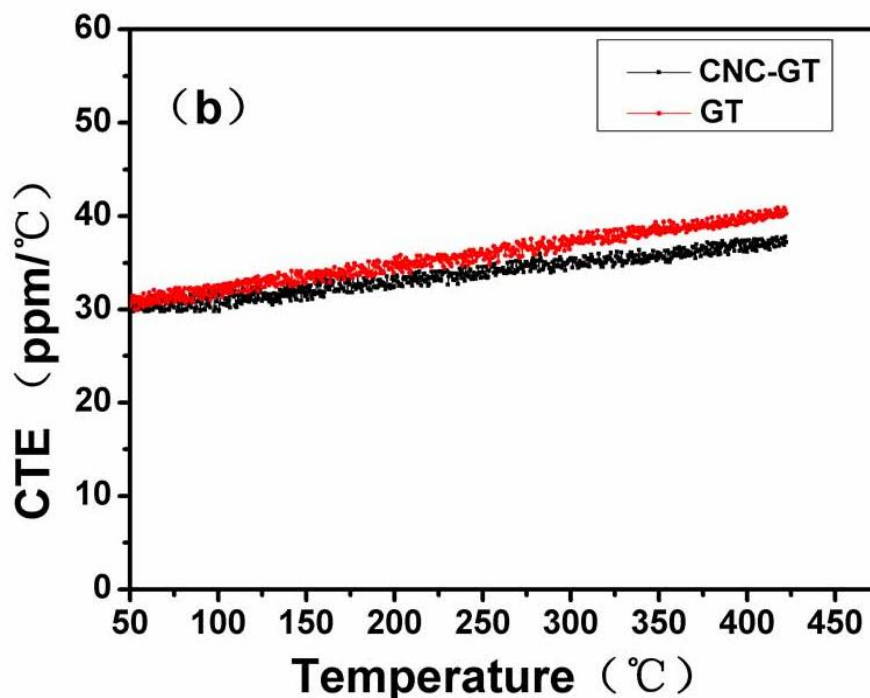
**Fig. S4** XRD patterns in the (a, c) perpendicular and (b, d) parallel to hot-pressing direction of (a) and (b) GT block (1873 K, 40MPa), (c) and (d) CNC-GT block (growth time of 15 min, 1873 K, 40MPa).

Fig. S4a and 4c show that only two sharp (002) and weak (004) diffraction peaks are clearly observed in the perpendicular to hot-pressing direction (in-plane direction). Fig. S4b and 4d display that both CNC-GT and GT blocks exhibit a small (0 0 2) peak and four diffraction peaks corresponding to (1 0 0), (1 0 1), (1 1 0) and (1 1 2). It can be seen that the intensities of (002) peaks of CNC-GT and GT blocks in the parallel to hot-pressing direction decreased compared with that in the perpendicular to hot-pressing direction. This feature indicates the preferred orientation of graphite layers in the in-plane direction by hot-pressing.<sup>1</sup> Compared with GT, the

intensities of (001) and (002) peaks in CNC-GT in both perpendicular and parallel to hot-pressing direction decrease greatly. Moreover, the intensity ratios of (002) to (001) peaks of CNC-GT blocks are much lower than that of GT blocks. This result indicates that the intercalation of CNC at the interface of GT results in the reduced orientation and size of crystalline structure.<sup>2</sup>



**Fig. S5** SEM images of the in-plane surface (a) GT and (b) CNC-GT blocks prepared by hot-pressing.



**Fig. S6** Coefficient of thermal expansion (CTE) in cross-plane direction of CNC-GT (the growth time of 120 min, 1873 K, 40MPa) and GT blocks (1873 K, 40MPa) with the increasing temperature.

As shown in Fig. S6, GT and CNC-GT blocks show an increasing CTE in cross-plane direction with the increase in the temperature. CET values of CNC-GT and GT blocks are slightly higher than that of fine graphite lattice at 400 °C (~28 ppm/°C)<sup>3</sup> due to a relatively loose packing of graphite layers.<sup>4</sup> Compared with GT, a low CTE of CNC-GT block is attributed to the intercalation of CNC with negative or approximate zero CTE, which produces the back-tension and decrease thermal expansion.<sup>5,6</sup> In addition, CNC-GT block is more compact with the strong interaction between GT layers, which also leads to the decrease in thermal vibration and expansion. Results suggest that CNC-GT block with a low CTE maintains a good thermal-stability at a wide range of working temperature.



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