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Interfacial construction and multi-scale structural evolution in nanocomposites

Siduo Wu
Peking University Shenzhen Graduate School, China

We first report the construction of hybrid shish-kebab (HSK) superstructure in low-crystallinity elastomer nanocomposites with incorporation of carbon nanotube bundles (CNTBs). With strong interfacial adhesion, the tensile strength of olefin block copolymer (OBC)/CNTBs nanocomposites was tripled from 7.34 MPa to 24.11 MPa. With innsitu synchrotron small angle X-ray scattering (SAXS) and wide-angle X-ray diffraction (WAXD) as well as in situ Raman spectra, the multi-scale structural evolution was thoroughly studied. The formation of HSK leads to lower lamellar density, exhibiting remarkably increased long period. Unlike that in neat OBC, the lower density of crystal lamellaes in nanocomposites does not dominate the evolution of long range orderness at low strain regions, the long period of nanocomposites exhibit an inversely increasing trend. With addition of CNTBs, the orientation behavior still follows the slip-link theory. With HSK acting as larger but fewer physical junctions, the chain connectivity of the soft and hard segments in OBC chains in nanocomposites is lower than that in neat OBC, it is less necessary for HSK to adjust their orientation status along the stretching direction. Thus the orientation factor of orthorhombic crystals at low strain regions is lower than that of neat OBC. The mesoscopic structural evolution of CNTBs can be directly revealed by the downshift trend of the Raman G-band of CNTBs in nanocomposites, which reveals the axial deformation of CNTBs. The downshift can reach a maximum of 10.2 cm^{-1} and the downshift under axial deformation also confirm to the slip-link theory and is consistent with the orientation status of HSK superstructure.

373970592@qq.com