## Nanoscale patterning of carbon nanotubes in polyacrylonitrile fibers

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Polymer nanocomposites (PNCs) have been a research focus for more than a few decades due to their unique physical and chemical properties. In terms of their processing, polymer and nanoparticles are often mixed for synergistic and hybrid properties, yet, the spatial geometry and nanoscale structural features within such composite are often overlooked. In this presentation, a scalable fabrication method, combining the forced assembly process and dry-jet-wet fiber spinning technique is demonstrated for location-regulated deposition of carbon nanotubes (CNTs) within polyacrylonitrile (PAN) matrix for high mechanical performance fiber. Two polymer solutions (i.e., PAN and CNT/PAN) first entered a 3D-printed multiplier die side-by-side and then were physically separated along the horizontal direction (top-down) and repositioned along the vertical direction (right-left). In this way, a 512 layered fiber with hierarchical structures, including highly aligned nanotubes (hierarchy level 1, ~20 nm), alternatively packed nanolayers (hierarchy level 2, ~170 nm), microscale fibers (hierarchy level 3, ~80 µm) is constructed for macroscale fiber reinforcement in traditional fabrics or laminates (hierarchy level 4, ~ 1 m) (Figure 1a). The introduction of alternating layers facilitated the quality of CNT dispersion due to nanoscale confinement and, at the same time, enhanced their orientation due to shear stress generated at each layer interface (Figure 1b). We demonstrated an example with 0.5 wt%CNTs loading and the inclusion of 170 nm layers in composite fibers showed a 27.4% increase in modulus and a 22.2% increase in strength compared to the traditional CNT/PAN mixing method without any hierarchical structures (Figure 1c).



Figure 1. (a) Hierarchical fiber structure from macro to nanometer scale. (b) 512 layered fiber with a layer thickness of 170nm. (c) Enhanced mechanical behaviors with increasing layer numbers