

# Supporting information

## Hydrogen Bonding and Phase Separation Cooperatively Guide the Self-assembly of U60 and Polymer to Fabricate Multiscale Nanostructures

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### Materials and Methods

DLS measures the intensity–intensity time correlation function by means of a BI-9000AT multichannel digital correlator. The field correlation function  $|g^{(1)}(\tau)|$  was analyzed by the constrained regularized CONTIN method to yield information on the distribution of the characteristic line width  $\Gamma$  from  $|g^{(1)}(\tau)| = \int G(\Gamma) e^{-\Gamma\tau} d(\Gamma)$ . The normalized distribution function of the characteristic line width,  $G(\Gamma)$ , so obtained, can be used to determine an average apparent translational diffusion coefficient,  $D_{app} = \Gamma/q^2$ . The hydrodynamic radius  $R_h$  is related to  $D$  via the Stokes–Einstein equation:  $R_h = kT/(6\pi\eta D)$  where  $k$  is the Boltzmann constant and  $\eta$  the viscosity of the solvent at temperature  $T$ . From DLS measurements, we can obtain the particle-size distribution in solution from a plot of  $\Gamma G(\Gamma)$  vs  $R_h$ . The  $R_h$  of the particles is obtained by extrapolating  $R_{h,app}$  to zero scattering angle.

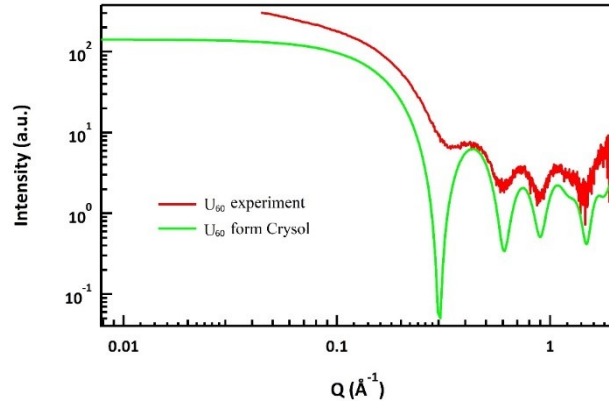


Fig. S1 SAXS data of U60.

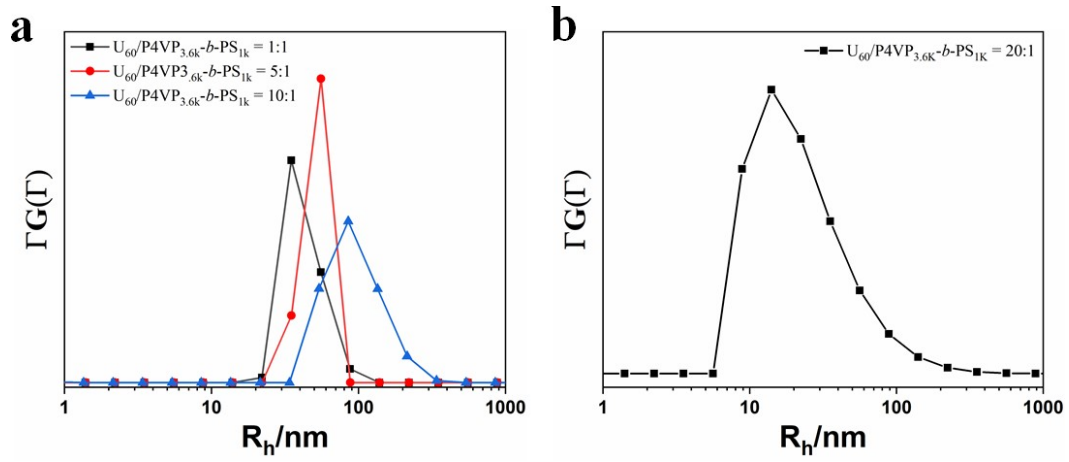


Fig. S2 (a) and (b)  $U_{60}$  and  $P4VP_{3.6k}\text{-}b\text{-}PS_{1k}$  form a complex aqueous solution DLS analysis diagram (the molar ratio of  $U_{60}$  and  $P4VP_{3.6k}\text{-}b\text{-}PS_{1k}$  are 1:1, 5:1, 10:1 and 20:1).

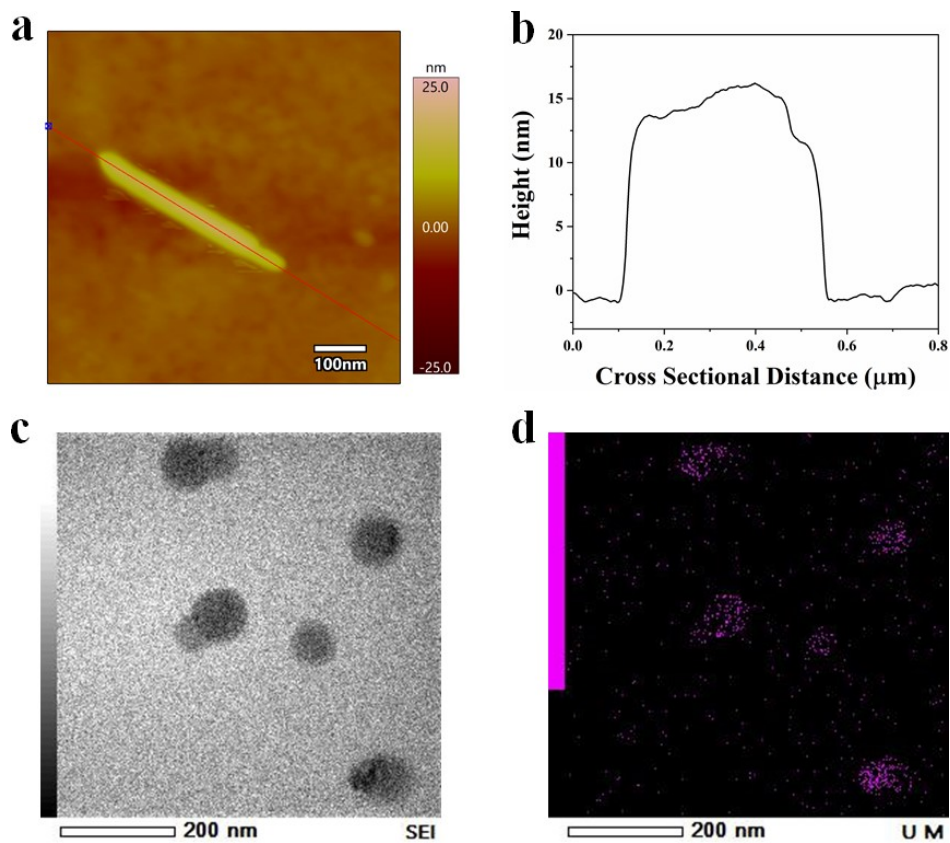


Fig. S3 (a) AFM image of  $U_{60}/P4VP_{3.6k}\text{-}b\text{-}PS_{1k}$  wormlike composite. (b) Corresponds to the height of the compound in (a). (c) TEM image of  $U_{60}/P4VP_{3.6k}\text{-}b\text{-}PS_{1k}$  composite (the molar ratio of  $U_{60}$  and  $P4VP_{3.6k}\text{-}b\text{-}PS_{1k}$  is 1:1). (d) Corresponds to the distribution of U elements in (c).

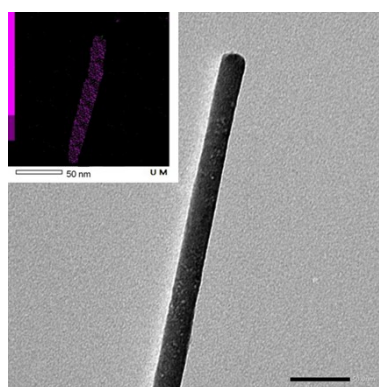


Fig. S4 TEM images of  $U_{60}/P4VP_{3.6k}\text{-}b\text{-}PS_{1k}$  wormlike composite, the inset is the distribution diagram of U.

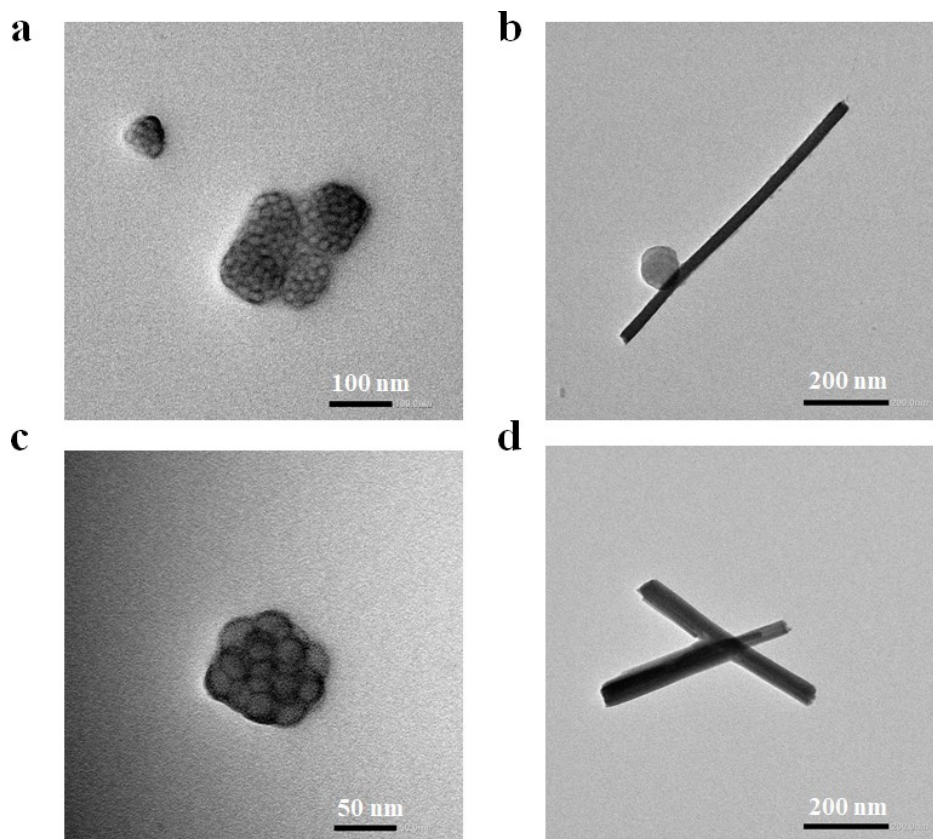


Fig. S5 (a) and (b) TEM image of  $U_{60}/P4VP_{3.6k}-b-PS_{3.1k}$  composite (the molar ratio of  $U_{60}$  and  $P4VP_{3.6k}-b-PS_{3.1k}$  is 1:1 and 10:1 respectively). (c) and (d) TEM image of  $U_{60}/P4VP_{3.6k}-b-PS_{5.1k}$  composite (the molar ratio of  $U_{60}$  and  $P4VP_{3.6k}-b-PS_{5.1k}$  is 1:1 and 10:1 respectively).

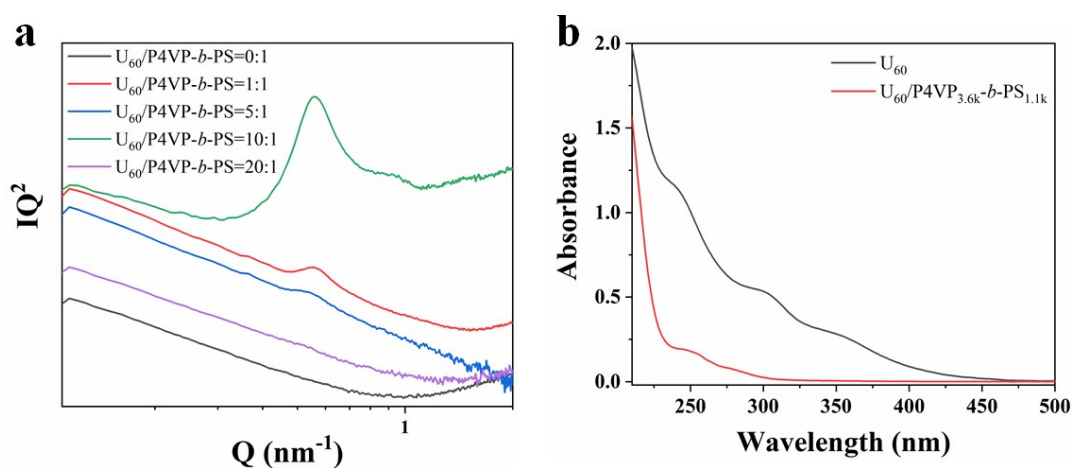


Fig. S6 (a)  $U_{60}/P4VP_{3.6k}-b-PS_{1k}$  SAXS images with different molar ratios. (b) The UV spectrum of the sample supernatant after  $P4VP_{3.6k}-b-PS_{1.1k}$  and  $U_{60}$  were assembled and adsorbed one month later.