

# Supporting Information

## **Hierarchical Polymer-Carbon Nanotube Hybrid Mesostructures by Crystallization Driven Self-Assembly**

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## SUPPLEMENTARY TABLES AND FIGURES

**Table S1** Structural information of PFS crystal-coated MWCNT structures (**NHSK-H**) and PFS-based micelle-decorated MWCNT structures (**NHSK-M**).

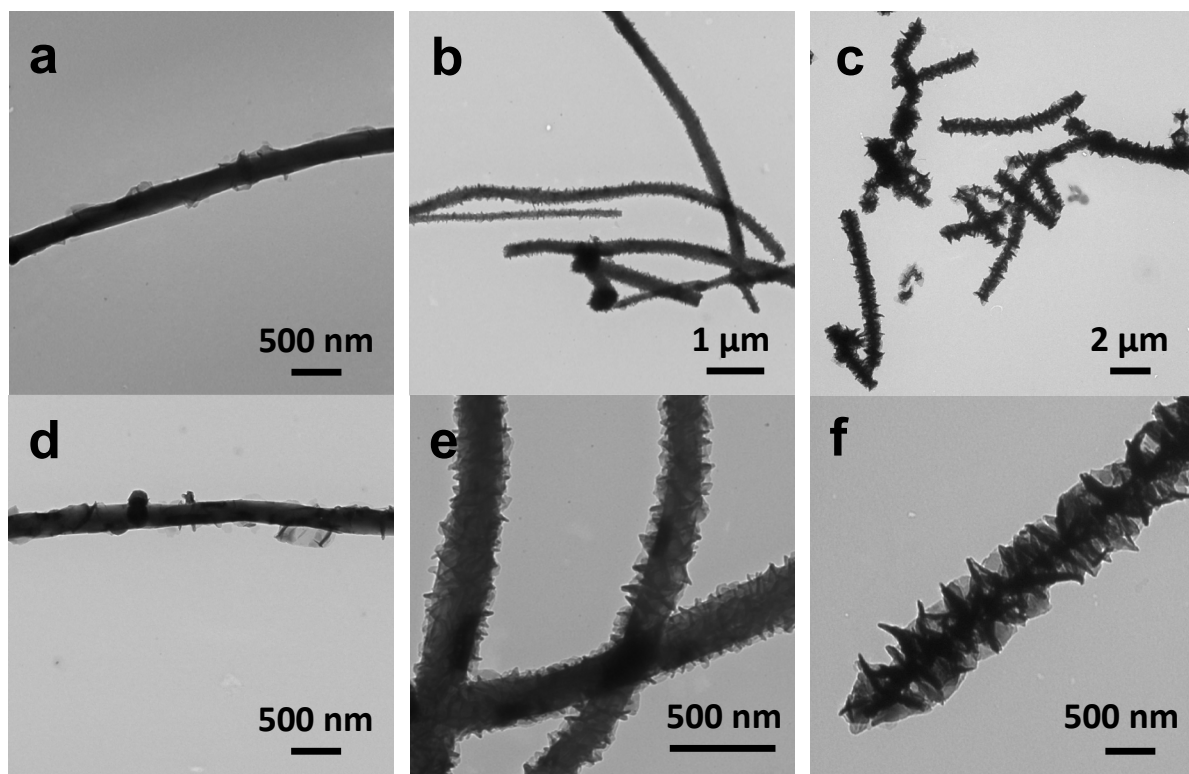
Samples	Diameter (nm) <sup>a</sup>	Size of PFS <sub>31</sub> crystals (nm) <sup>b</sup>	Length of the micelles (PFS <sub>53</sub> - <i>b</i> -PI <sub>637</sub> ) <sup>c</sup>	Length of the micelle (PFS <sub>17</sub> - <i>b</i> -P2VP <sub>170</sub> ) <sup>c</sup>
<b>SK0.1</b>		90		
<b>SK1</b>	560			
<b>SK2</b>	870			
<b>SK0.1-M<sub>PFS-PI(4)</sub></b>		90	1340	
<b>SK1-M<sub>PFS-PI(4)</sub></b>	560		780	
<b>SK2-M<sub>PFS-PI(4)</sub></b>	870		470	
<b>SK2-M<sub>PFS-PI(8)</sub></b>	870		1010	
<b>SK1-M<sub>PFS-PI(8)</sub></b>	560		2000	
<b>SK1*-M<sub>dPFS-P2VP(8)</sub></b>	560			570

<sup>a</sup> Diameter refers to the number average diameter of the cylindrical **SK**.

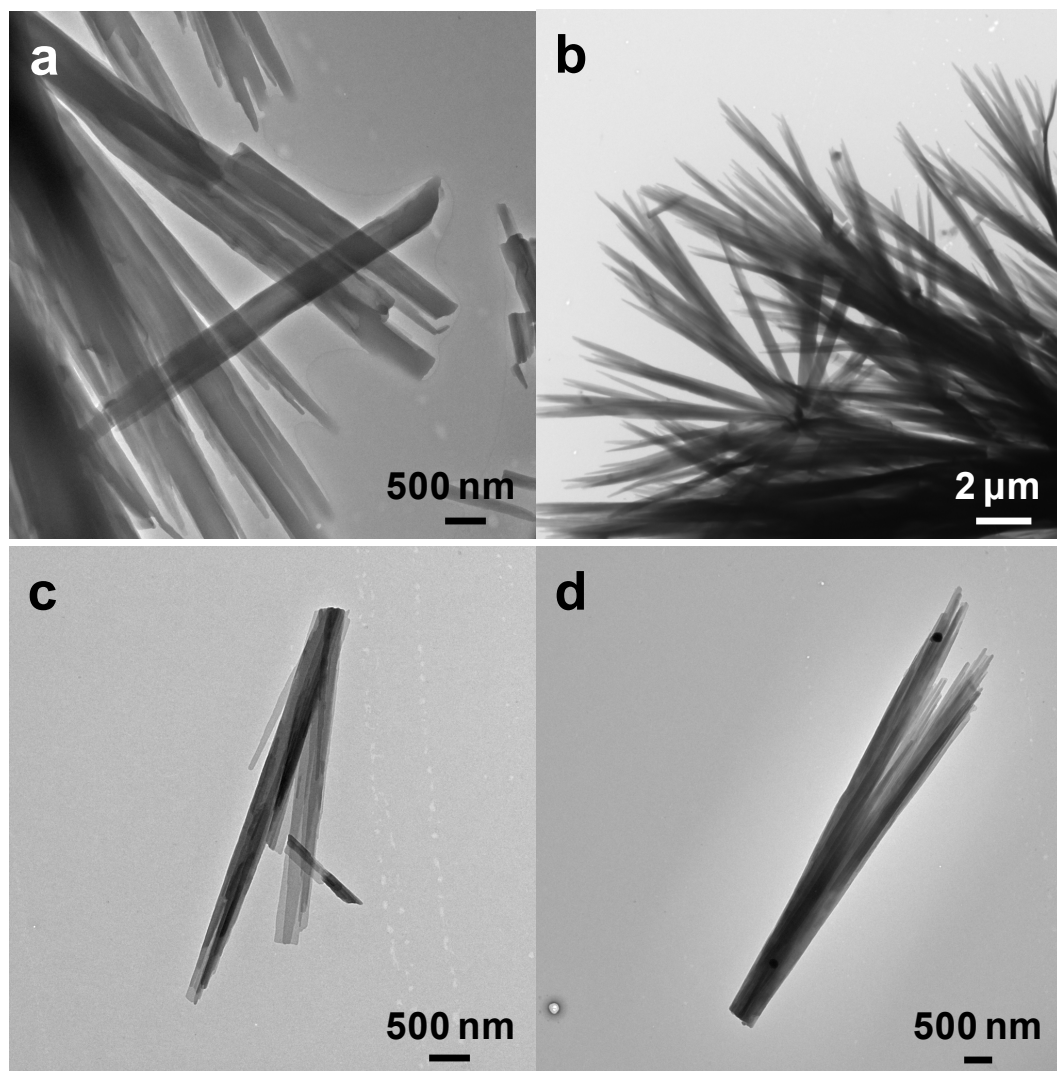
<sup>b</sup> Size of PFS<sub>31</sub> crystals refers to the average distance from the surface of MWCNT to the edge of PFS<sub>31</sub> crystals in **SK0.1**.

<sup>c</sup> Length of micelle refers to the number average distance from the surface of **SK** to the end of the PFS micelle. All of the values were calculated from TEM images by tracing by hand more than 100 individual crystals or micelles using the software ImageJ (NIH, US).

<sup>d</sup> **SK1\*** refers to a sample of **SK1** in decane that was sedimented and then redispersed in 2-propanol. **SK1\*-M<sub>PFS-P2VP(8)</sub>** refers to the structure formed after seeded growth of PFS<sub>17</sub>-*b*-P2VP<sub>170</sub> micelles from the PFS crystals of **SK1\***.

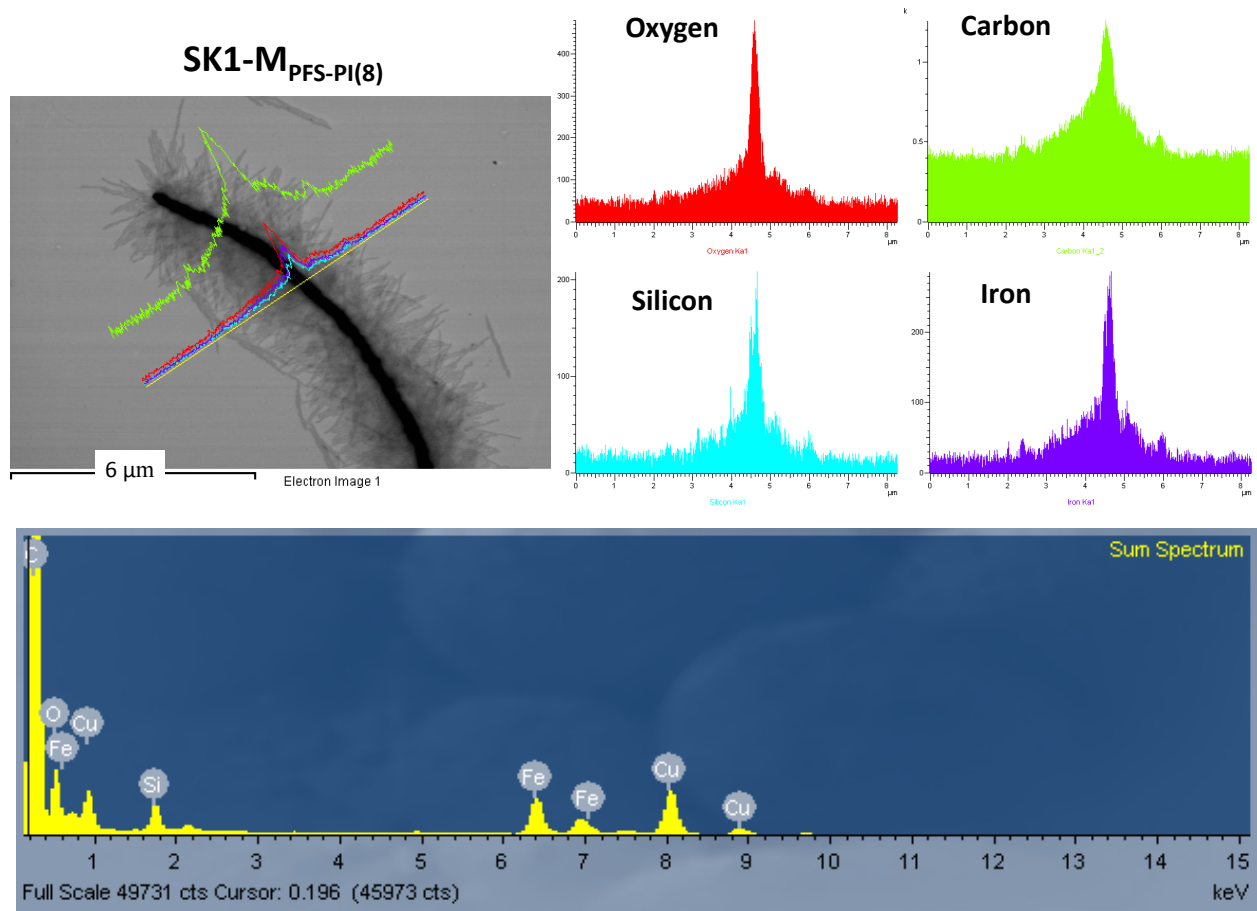


**Figure S1.** Additional TEM images of (a), (d) **SK0.1**, (b), (e) **SK1**, (c), (f) **SK2**.

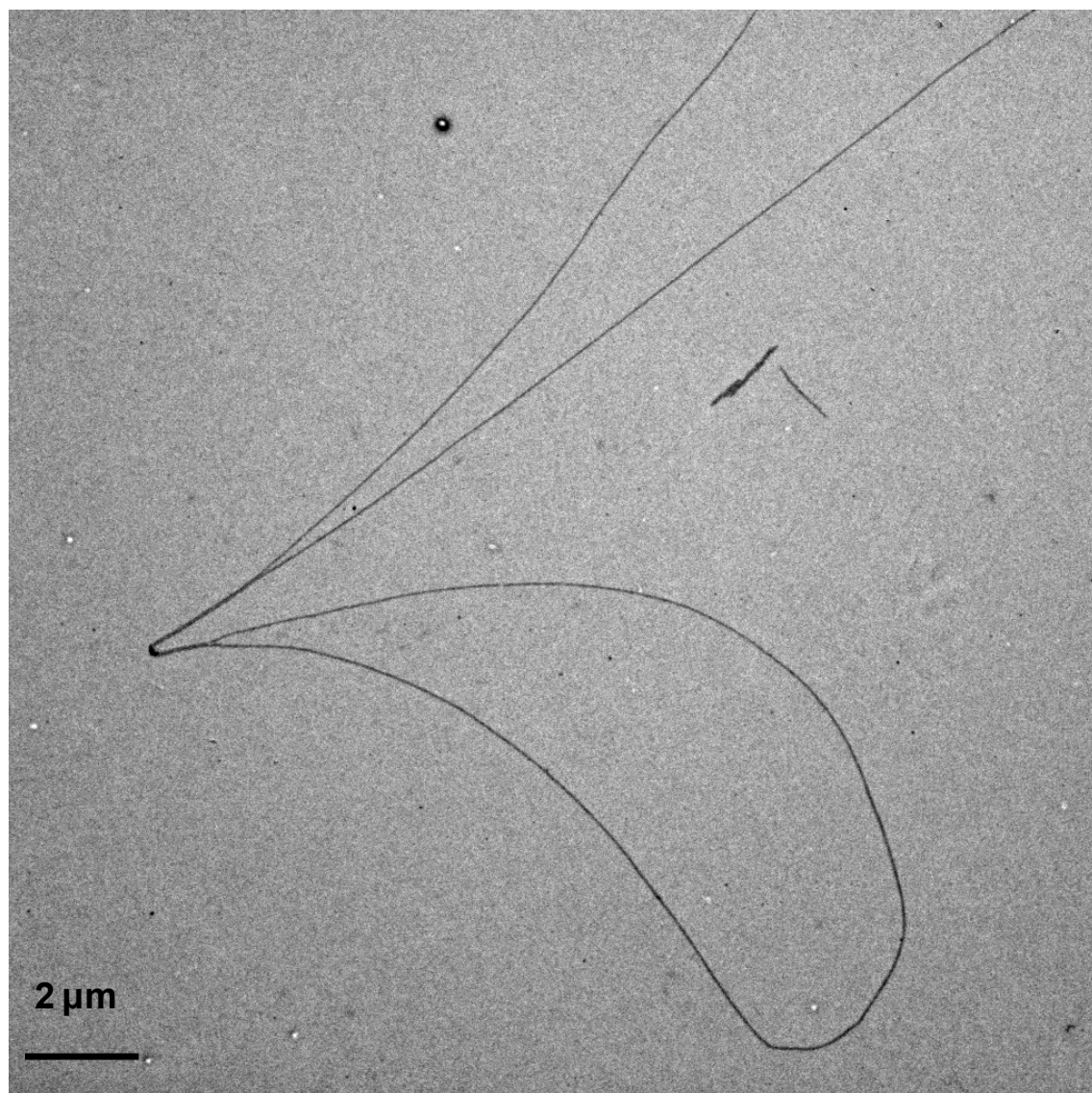


**Figure S2.** (a), (b), (c) and (d) Additional TEM images of homonucleated PFS<sub>31</sub> crystals.

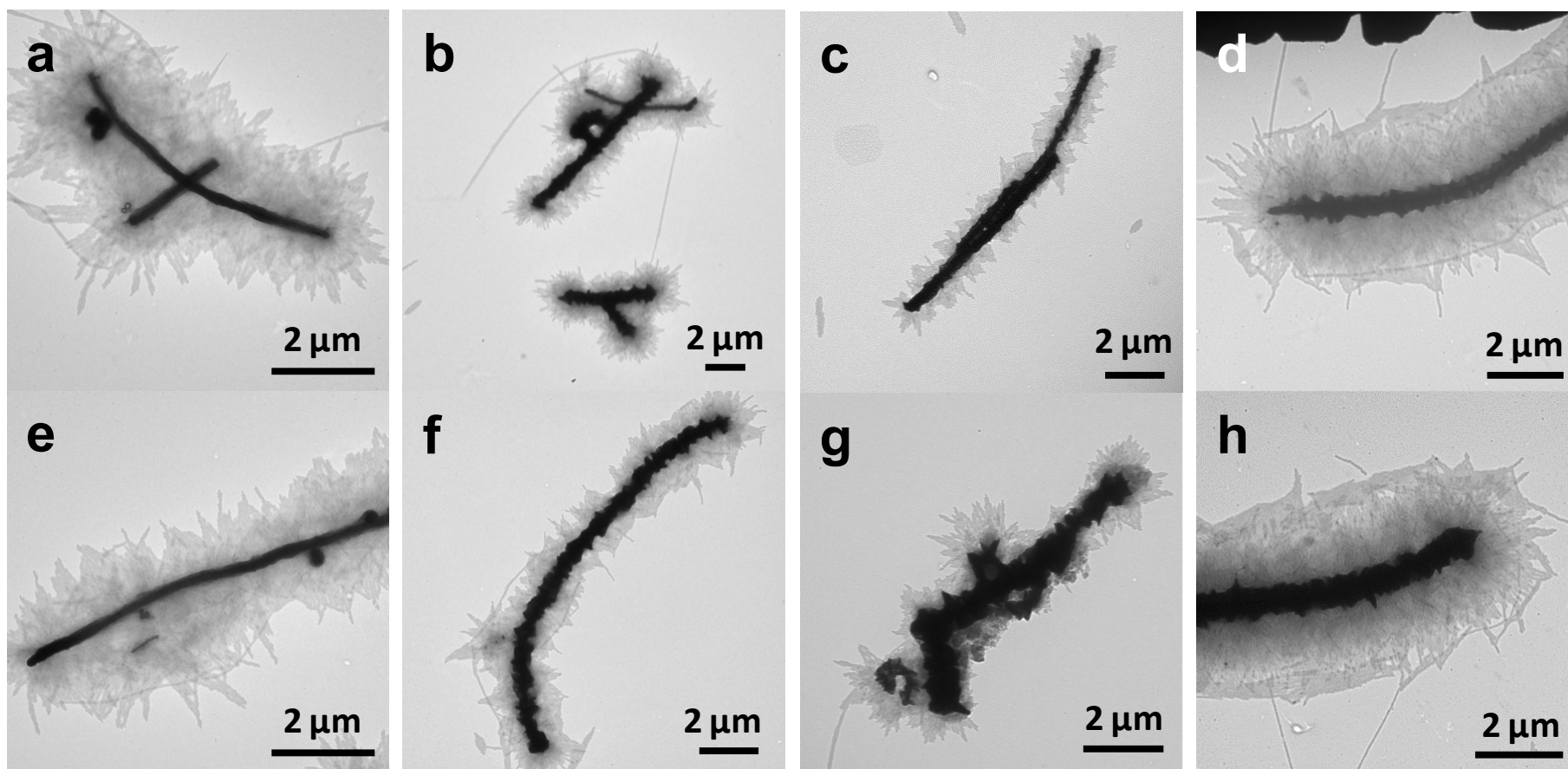




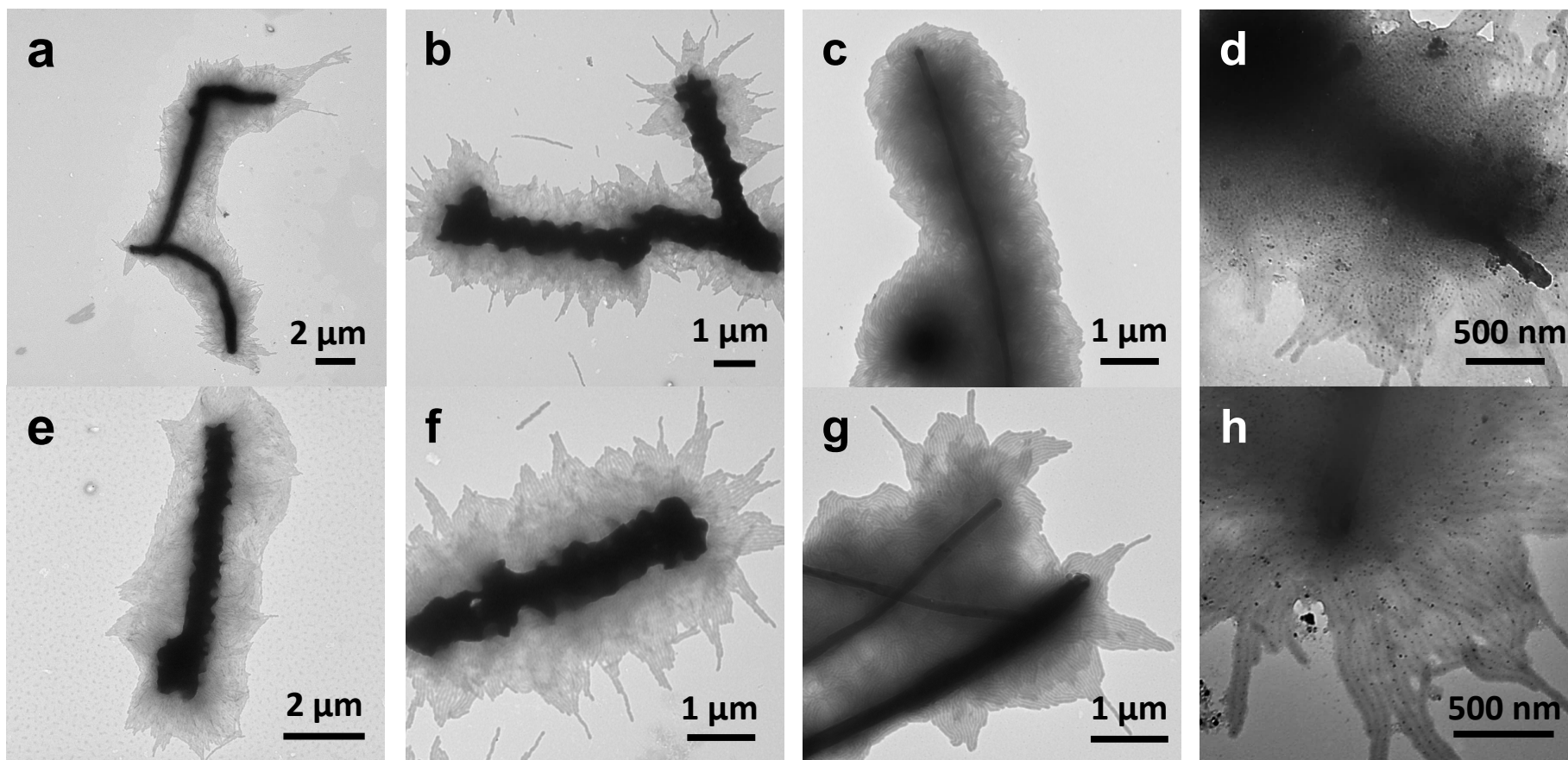
**Figure S3.** EDX Line scan analysis of **SK1-M<sub>PFS-PI(8)</sub>**.



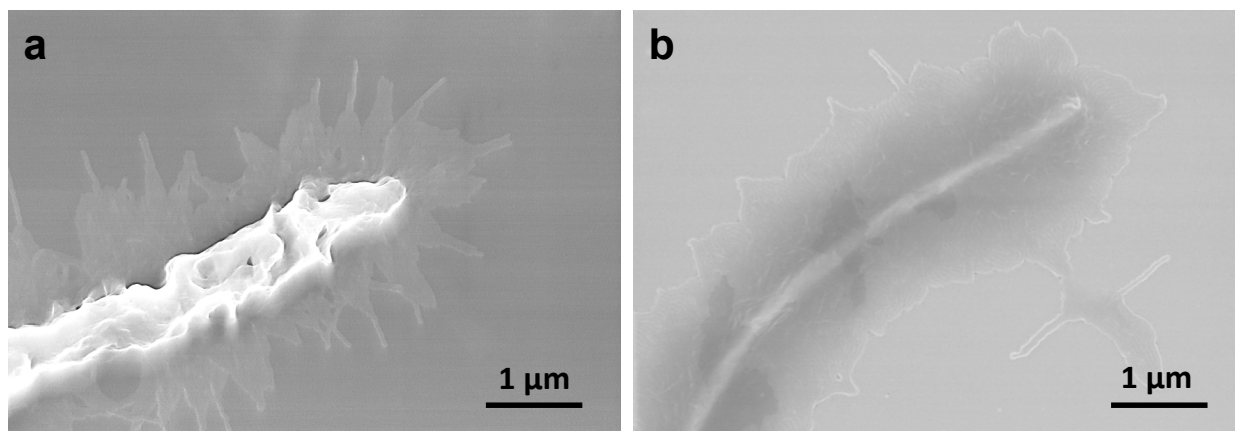
**Figure S4.** Low magnification of the TEM image shown in Figure 3b of the main text. The  $\text{PFS}_{53}\text{-}b\text{-PI}_{637}$  micelle seen in this image is at least 40  $\mu\text{m}$  long, as measured by Image J. It strongly resembles homonucleated  $\text{PFS}_{53}\text{-}b\text{-PI}_{637}$  micelles formed in decane. As indicated in the main text, this micelle was formed during the preparation of sample **SK1- $\text{M}_{\text{PFS-PI}(8)}$** .



**Figure S5.** Additional TEM images of (a), (e) **SK0.1-M<sub>PFS-PI(4)</sub>**, (b), (f) **SK1-M<sub>PFS-PI(4)</sub>**, (c), (g) **SK2-M<sub>PFS-PI(4)</sub>**, (d), (h) **SK1-M<sub>PFS-PI(8)</sub>**.

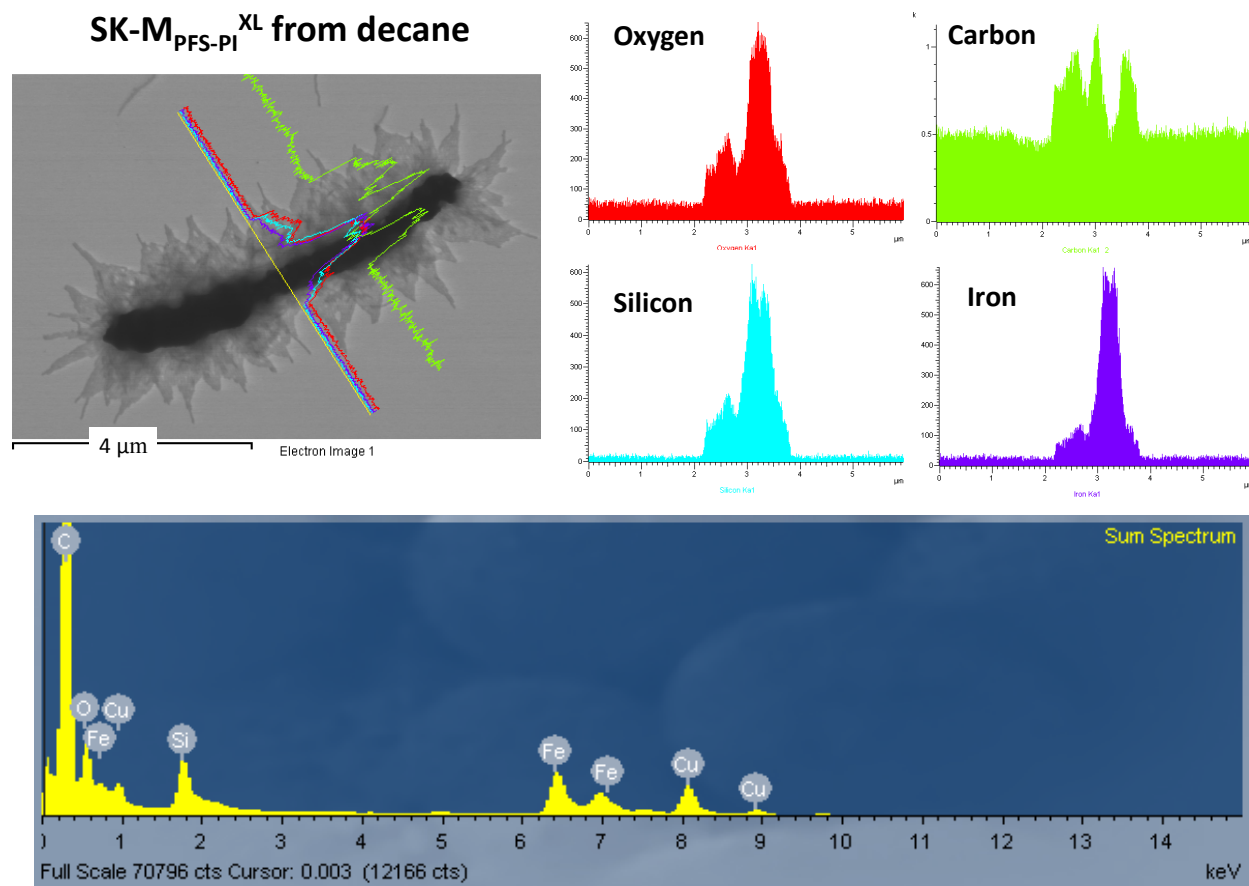


**Figure S6.** Additional TEM images of (a), (e)  $\text{SK2-M}_{\text{PFS-PI}(8)}$ , (b), (f)  $\text{SK-M}_{\text{PFS-PI}}^{\text{XL}}$ , (c), (g)  $\text{SK-M}_{\text{PFS-PI}}^{\text{XL}}$  after treatment with DCM ( $(\text{SK-M}_{\text{PFS-PI}}^{\text{XL}})_{\text{DCM}}$ ), (d), (h) Ag NPs embedded in  $(\text{SK-M}_{\text{PFS-PI}}^{\text{XL}})_{\text{DCM}}$ .

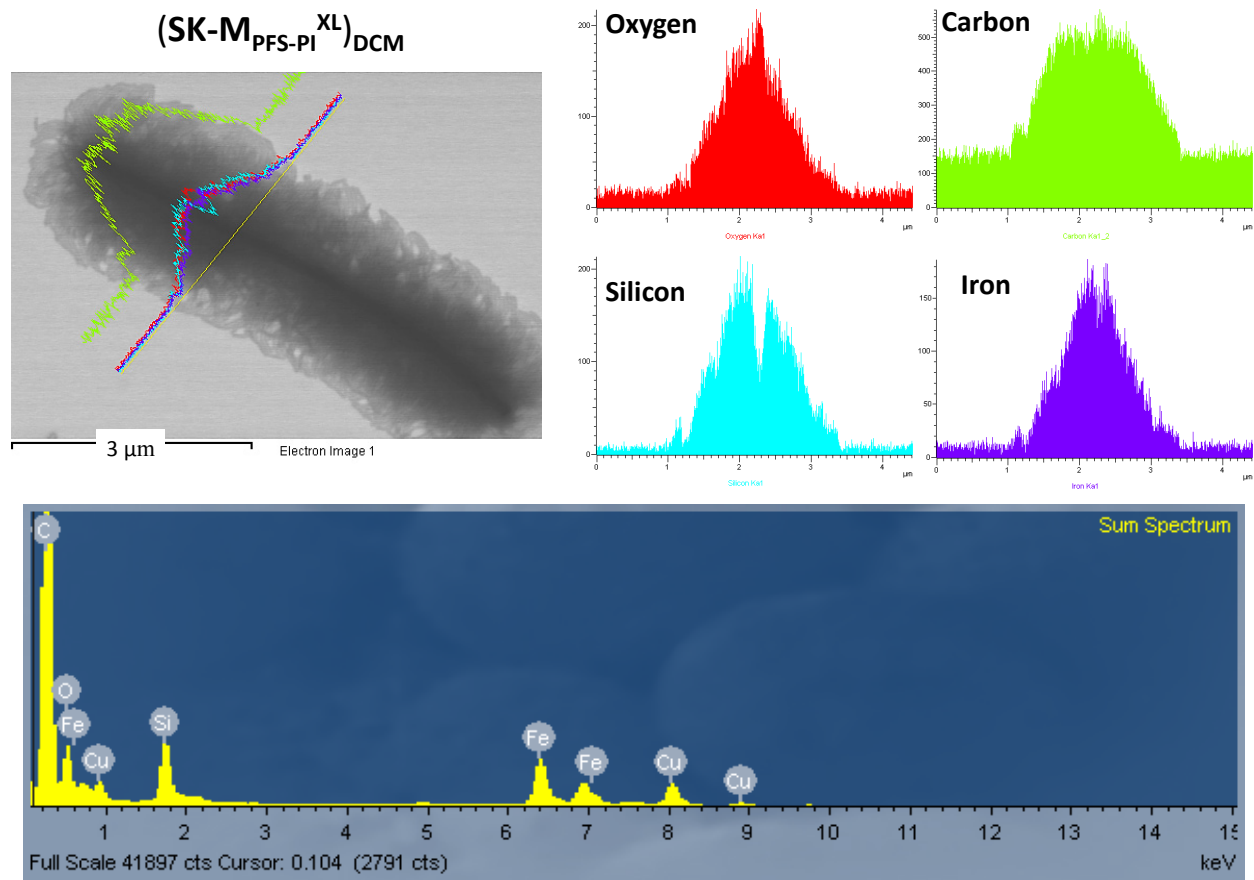


**Figure S7.** SEM images of (a)  $\text{SK-M}_{\text{PFS-PI}}^{\text{XL}}$  from decane, (b)  $\text{SK-M}_{\text{PFS-PI}}^{\text{XL}}$  after treatment with DCM ( $(\text{SK-M}_{\text{PFS-PI}}^{\text{XL}})_{\text{DCM}}$ ).

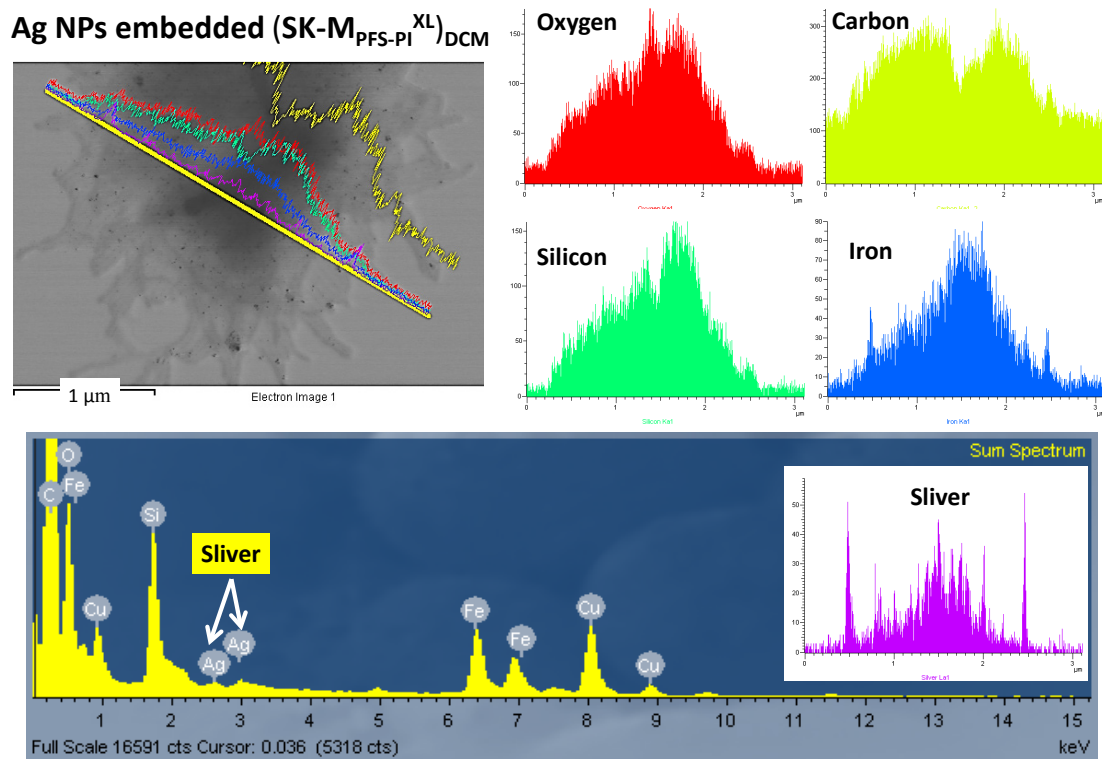




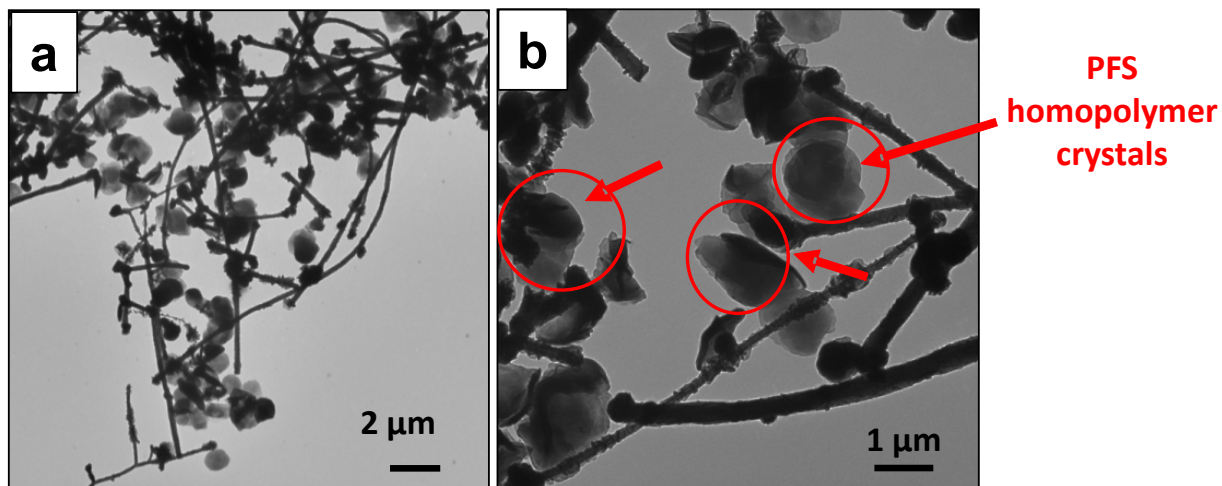
**Figure S8.** EDX Line scan analysis of **SK2-M<sub>PFS-PI(8)</sub>** after cross-linking the PI corona (**SK-M<sub>PFS-PI</sub><sup>XL</sup>**) from decane solution.



**Figure S9.** EDX Line scan analysis of (SK-M<sub>PFS-PI</sub><sup>XL</sup>)<sub>DCM</sub>.

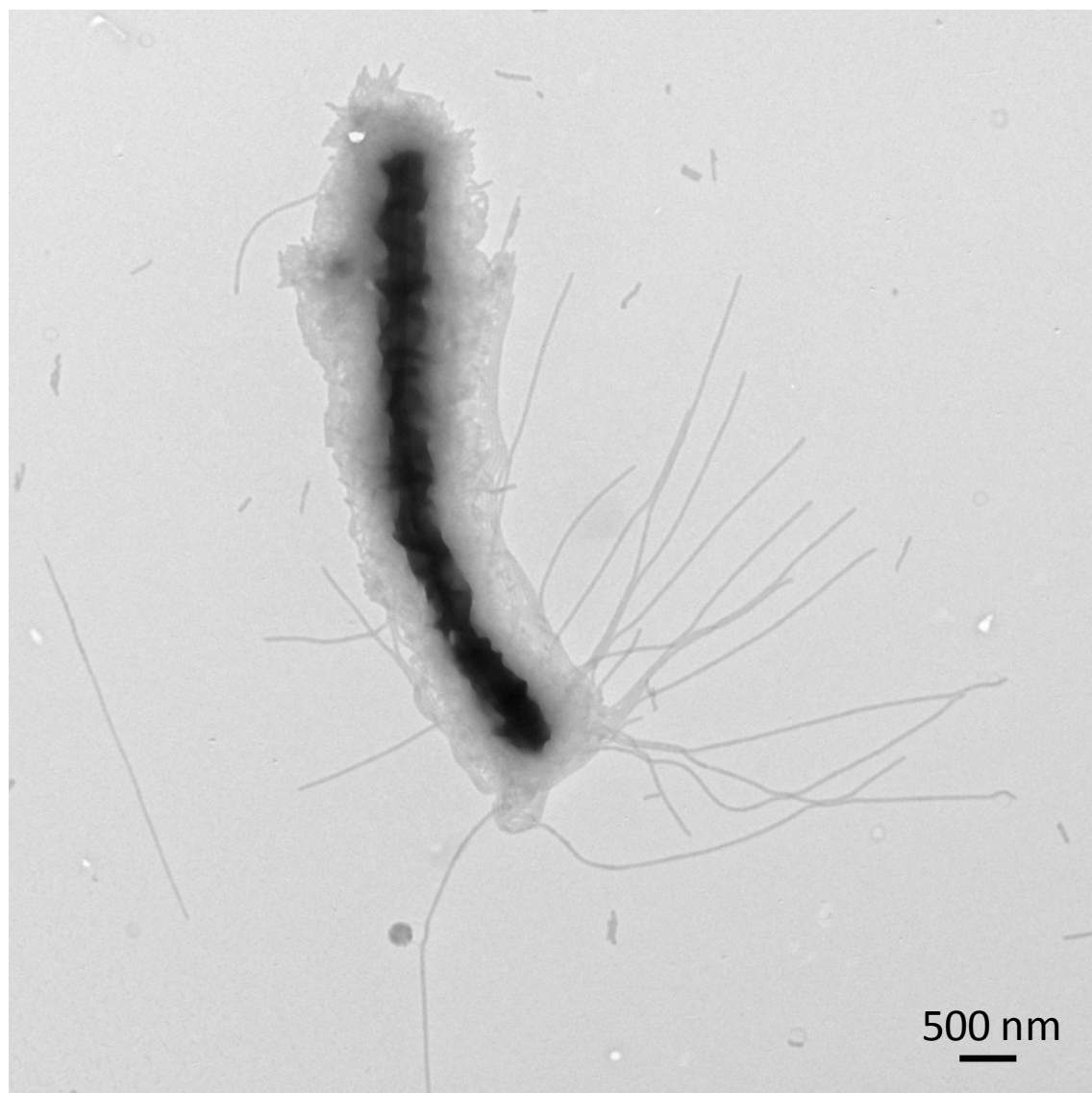


**Figure S10.** EDX Line scan analysis of  $(SK-M_{PFS-PI}^{XL})_{DCM}$  with Ag NPs embedded in the structure.



**Figure S11.** TEM images of a sample containing MWCNTs and  $PFS_{31}$ . This sample was prepared by addition of a THF solution (0.25 mL) containing a mixture of  $PFS_{31}$  (0.25 mg) and MWCNT (0.25 mg) to a pre-heated 2-propanol (10 mL, 80 °C), cooled slowly to room temperature and aged for 24 h. The sample prepared in this way appears to be a mixture of  $PFS_{31}$ -coated CNTs plus  $PFS_{31}$  homopolymer crystals.





**Figure S12.** Low magnification of the TEM image of sample **SK1\*-M<sub>PFS-P2VP(8)</sub>** shown in Figure 6d of the main text. This sample of PFS-*b*-P2VP micelles grown from the PFS homopolymer crystals of **SK1\*** in 2-PrOH was obtained by adding a solution in THF (200  $\mu$ L) containing PFS<sub>17</sub>-*b*-P2VP<sub>170</sub> (2 mg) to a 2-PrOH solution (10 mL) of **SK1\***. After brief swirling, the solution was allowed to age for 24 h. this image shows that both long smooth micelles and shorter micelles (seen to be kinked in Figure 6d, main text) are formed when PFS<sub>17</sub>-*b*-P2VP<sub>170</sub> was added to a suspension of **SK1\*** in 2-PrOH.