Supporting Information

Small-Angle Neutron Scattering from Aqueous Dispersions of Single-Walled Carbon Nanotubes with Pluronic® F127 and poly(vinylpyrrolidone)

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UV-visible absorbance measurements for evaluation of the relative CNT content

The production process of CNTs the raw material consists yields different types of nanotubes and many impurities. In order to achieve high quality and stable dispersions, a light centrifugation process has to take place during the CNTs/polymer sample preparation. This is needed in order to reduce the residual catalyst and amorphous carbon particles. This, however, reduces the CNTs content in dispersion by an unknown amount. Thus, a specific procedure is used in order to verify that the final dispersions contain the same amount of CNTs. UV-visible light absorptions is used to verify that the dispersions in a given set of samples have the same degree of "blackness" which indicates the same CNT content, and thus differ only in contrast due to their D_2O/H_2O content. Unfortunately the absorbance measurements cannot indicate directly the CNT content. The relative amount of CNTs in dispersion was evaluated using a UV-visible spectrophotometer (Evolution-201 Computer Control UV-Vis, Thermo Scientific) with 10-mm quartz cuvettes, at the wavelength range 200-400 nm.

Figures S1 and S2 present the absorbance data for the two of CNT/polymer systems. Within each group of sample, both CNT/Pluronic and CNT/PVP dispersions, the absorbance measurements indicated the same CNT content, within experimental error.

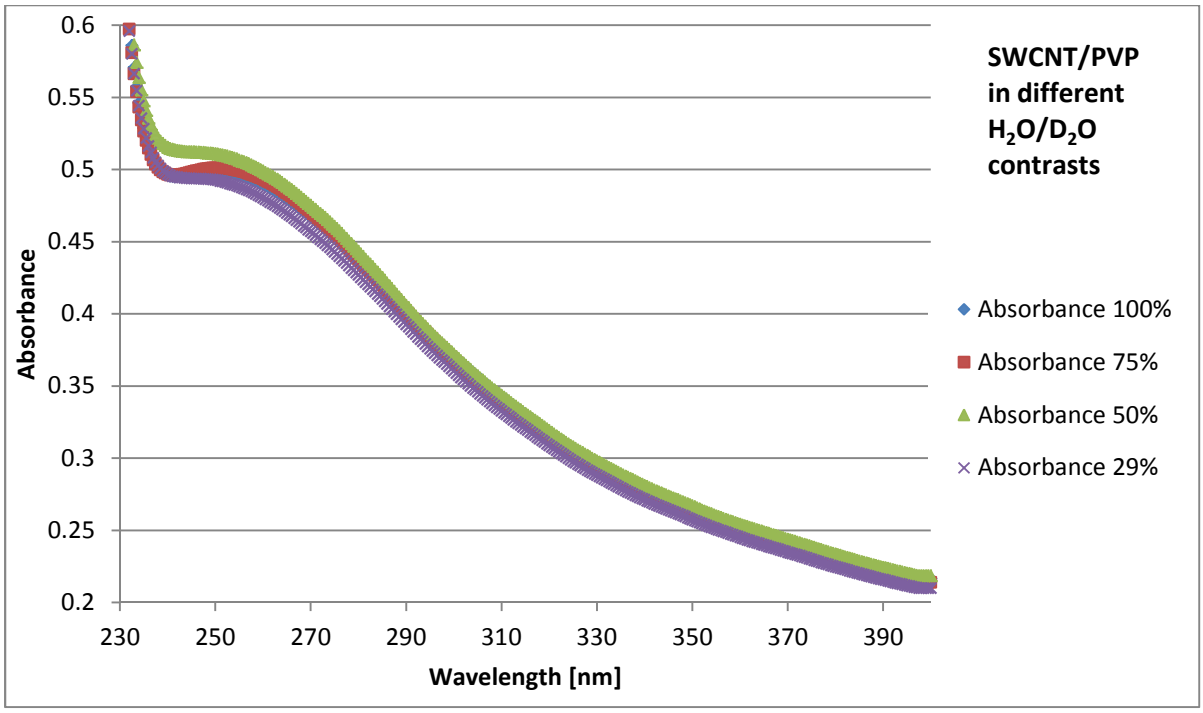

Figure S1. Absorbance patterns from SWCNT/PVP dispersions at solvent compositions (wt. % D_2O) of: 100, 75, 50 and 29.

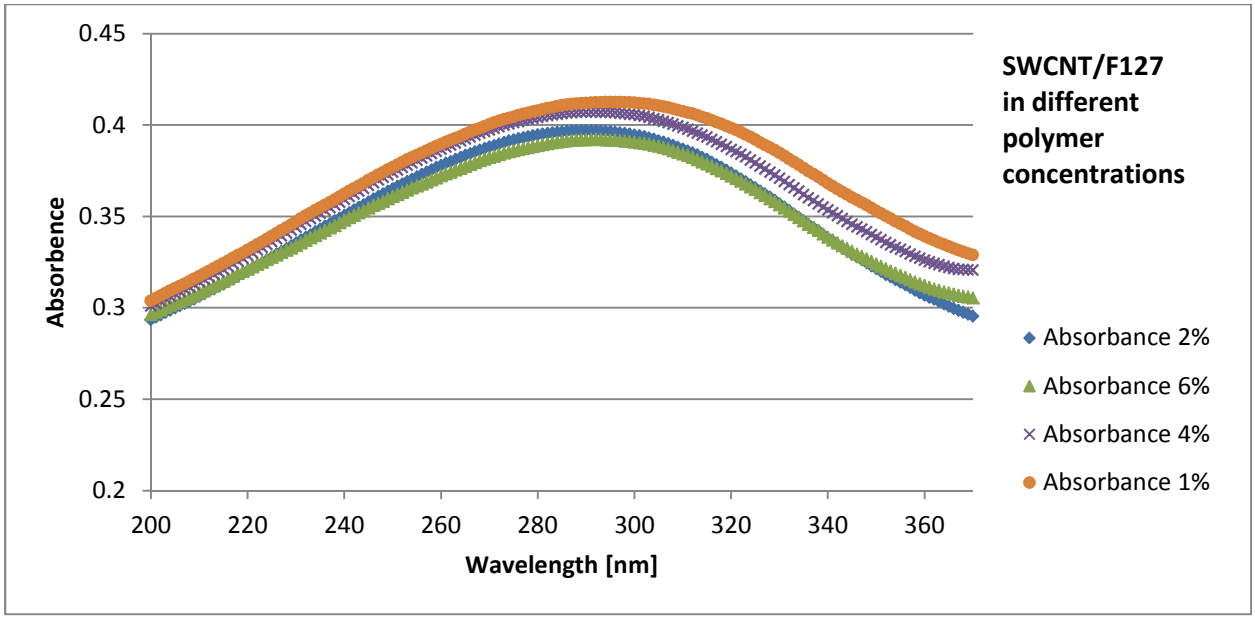

Figure S2. Absorbance patterns from SWCNT/F127 dispersions in 100% D_2O, at four different concentrations (% w/w) of: 6%, 4%, 2% and 1%.

SANS scattering patterns from the F127 polymer solutions

The scattering patterns from F127 polymer solutions, which are below the critical micelle temperature (CMT), were best fitted to the Debye[1] function of random coils and are presented in Figure S3.

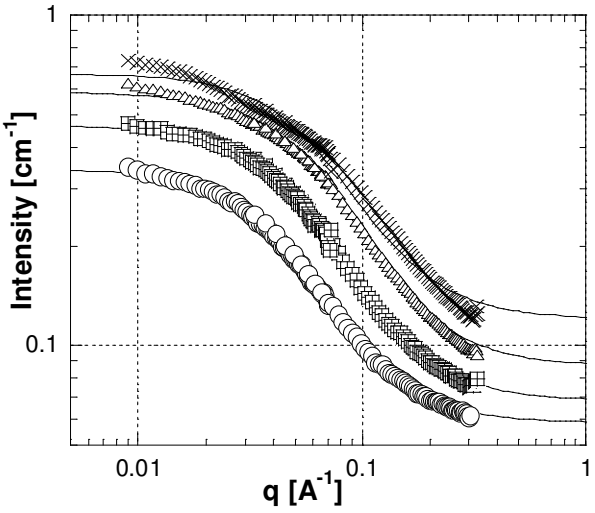

Figure S3. F127 dispersions in 100% D_2O at four different concentrations (% w/w) of: 1% (○), 2% (⊞), 4% (△), 6% (×). The solid curves through the symbols are Debye function fittings.

Normalization of the intensity from the SWCNT/F127 dispersions with $(\Delta\rho)^2$

SANS patterns from SWCNT/F127 dispersions in 100% D_2O are shown in Figure S4,

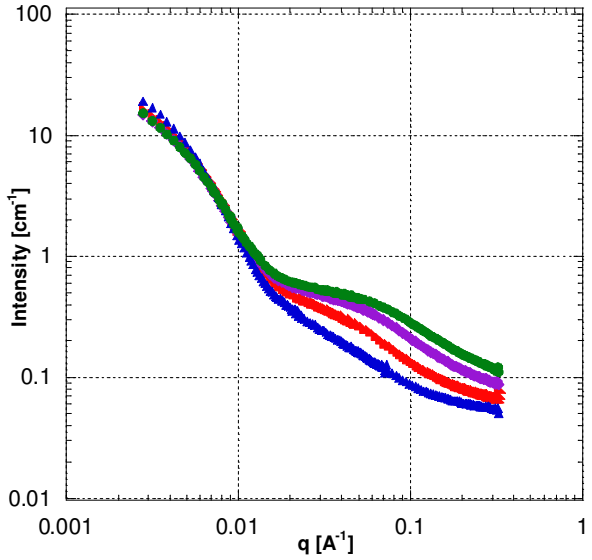

Figure S4. SANS patterns (before normalization of the intensity by contrast, $(\Delta\rho)^2$), from SWCNT/F127 dispersions in 100% D_2O, at four different concentrations (% w/w) of: 6% (green), 4% (purple), 2% (red) and 1% (blue).

The normalization factors, the squared differences in scattering length densities of the pure polymer and the polymer solution, were calculated according to equations 1, 2 and 3 below:

(1) $$(\Delta\rho)^2 = (\rho_{F127} - \rho_{Solution})^2$$

(2) $$\rho_{Solution} = \phi_{F127} \cdot \rho_{F127} + \phi_{solvent} \cdot \rho_{solvent}$$

(3) $$\phi_{F127} = \frac{w_{F127} / d_{F127}}{w_{F127} / d_{F127} + w_{solvent} / d_{solvent}}$$

where: $\rho_{F127} = 5.04 \cdot 10^{-7}$ Å^{-2}, $\rho_{solvent} = 6.34 \cdot 10^{-6}$ Å^{-2}and $\rho_{Solution}$ are the scattering length densities (SLDs) (Å^{-2}) of the polymer F127, the D_2O solvent and the solution (the solvent and the dissolved polymer according to its concentration), respectively; ϕ_{F127} and $\phi_{solvent}$ are the volume fractions of the polymer F127 and the D_2O solvent in the solution, respectively; w_{F127} and $w_{solvent}$ are the mass fractions of the polymer F127 and the D_2O solvent in the solution, respectively; d_{F127}=1.01 gram/cm^3 (using bulk densities of 1.01 gram/cm^3 for both EO and PO, following Mortensen[2]) and $d_{solvent}$ =1.107 gram/cm^3 , are the mass densities of the polymer F127 and the D_2O solvent, respectively; Finally the normalization factors and the squared differences in scattering length densities of the pure polymer and the polymer solution are presented in the Table below:

Table S1. Normalization factors for SWCNT/Pluronic®F127 dispersions

SWCNT/F127 in different polymer concentration (% w/w)	$\rho_{Solution}$ [Å^{-2}]	$(\Delta\rho)^2$ [cm^{-4}]
SWCNT/F127 1%	$6.28 \cdot 10^{-6}$	$3.33 \cdot 10^{21}$
SWCNT/F127 2%	$6.21 \cdot 10^{-6}$	$3.26 \cdot 10^{21}$
SWCNT/F127 4%	$6.09 \cdot 10^{-6}$	$3.12 \cdot 10^{21}$
SWCNT/F127 6%	$5.96 \cdot 10^{-6}$	$2.98 \cdot 10^{21}$

Cylindrical core-adsorbed chains model for the analysis of SANS patterns form SWCNT/polymer hybrids

The specific equations of the form factors and the cross terms of the cylindrical core-adsorbed chains model, which were reported elsewhere[3], are provided below:

(4) **(a)** $$\mathcal{F}_{core}(q) = Amp_{core}{}^2 \cdot \frac{1}{L^2} \cdot F_L(q)$$

(b) $$Amp_{core}(q) = \frac{2J_1(qR_{core})}{qR_{core}}$$

(c) $$F_L(q) = \frac{\pi L}{q}$$

(5) $$F_{chain}(q) = \frac{2\left(\exp(-q^2R_g^2) - 1 + q^2R_g^2\right)}{\left(q^2R_g^2\right)^2}$$ (Glatter and Kratky[1])

(6) $$S_{core-chain}(q) = \psi(qR_g) \cdot J_0[q(R_{core} + dR_g)] \cdot \frac{J_1(qR_{core})}{qR_{core}} \cdot \frac{1}{L^2} \cdot F_L(q)$$

(7) $$S_{chain-chain}(q) = \psi^2 \cdot (qR_g) \cdot J_0{}^2[q(R_{core} + dR_g)] \cdot \frac{1}{L^2} \cdot F_L(q)$$

where: L (=5000 Å) is an arbitrarily chosen unit length; d presents the penetration level of the chains into the core structure (it has values between 0 and 1); R_{core} and R_g are the nanotube core radius and the gyration radius of the polymer chains, respectively; N_{agg} is the number of adsorbed polymer chains in the hybrid (of length L); J_0, J_1 are the zero and first order of the Bessel functions, respectively; Thus equation S1 describes $\mathcal{F}_{core}(q)$ which is the form factor of the SWCNT thin bundle core. F_L, shown in equation S1(c), is the form factor of an infinitely thin

rod of length L. F_{chain} is the form factor of the polymer chain behaves as a random coil with radius of gyration R_g, is given by the Debye function shown in equation 5.

$\psi(qR_g) = \left(1 - \exp(-q^2 R_g{}^2)\right) / q^2 R_g{}^2$ is the scattering amplitude of the polymer chain.[4,5] The cross term between the nanotube core cylinders and the polymer chains, $S_{core\text{-}chain}$, is elaborated in equation 6, and the cross term between every two chains, $S_{chain\text{-}chain}$, is given in equation 7.

PVP Dispersions

SANS patterns of PVP 4% (w/w) solutions in water at four different contrasts (100%, 75%, 50% and 29% D_2O in the solvent) are shown in Figure S5. The solutions, measured at ambient conditions, present the typical characteristics of a semidilute solution, best fit to a Lorentzian function:[6]

(8) $$I(q) = (I(0) / \left(1 + \xi^2 q^2\right)) + B$$

where ξ is a mesh-size ($\xi \approx 22$ Å is fit to the data at100% and the 75% D_2O), $I(0)$ is the extrapolated zero-angle intensity and B is the background level incorporating the incoherent scattering and small-scale solvent density fluctuations. The latter is so dominant at 50 and 29% D_2O so as to obscure the polymer scattering signal.

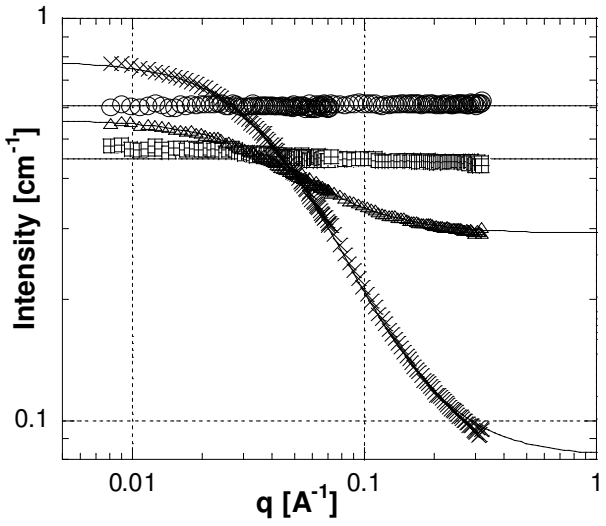

Figure S5. Scattering patterns from PVP 4% (w/w) aqueous solutions at four different contrasts (% D_2O in the solvent): 100% (×), 75% (△), 50% (田) and 29% (○). The solid curves through the symbols are the fit of Eq. 8.

References

[1] O. Glatter, O. Kratky, Small angle x-ray scattering. *Academic Press, New York,* **1982***, Ch. 12,* p. 402.

[2] Mortensen, K. Structural properties of self-assembled polymeric aggregates in aqueous solutions. *Poly. Adv. Tech*. **2001**, *12*, 2-22.

[3] Granite, M.; Radulescu, A.; Pyckhout-Hintzen, W.; Cohen, Y. Interactions between block copolymers and single-walled carbon nanotubes in aqueous solutions: a small-angle neutron scattering study. *Langmuir* **2011**, *27*, 751.

[4] Hammouda, B. Structure factor for dendrimer polymer gels. *J. Polym. Sci., Part B: Polym. Phys.* **1992**, *30*, 1387-1390.

[5] Hammouda, B. SANS from homogeneous polymer mixtures: a unified overview. *Adv. Polym. Sci.* **1993**, *106*, 87.

[6] Daoud, M.; Cotton, J. P.; Farnoux, B.; Jannink, G.; Sarma, G.; Benoit, H.; Duplessix, R.; Picot, C.; de Gennes, P. G. Solutions of flexible polymers - neutron experiments and interpretation. *Macromolecules* **1975**, *8*, 804-818.