## Supporting Information for: Tailoring Organic-Organic Poly(vinylpyrrolidone) Microparticles and Fibers with Multi-Walled Carbon Nanotubes for Reinforced Composites

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## XRD

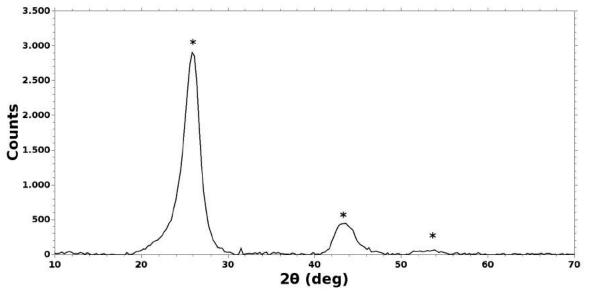
X-ray Diffraction analyses were performed in an XRD Bruker diffractometer D8I-90 by standard powder method (Cu-K $\alpha$  radiation,  $\lambda\alpha 1=0.154$  nm), from 10.00° to 70.00° ( $\Delta(2\theta) = 0.015^{\circ}$ , counting time = 80 s)

## SEM

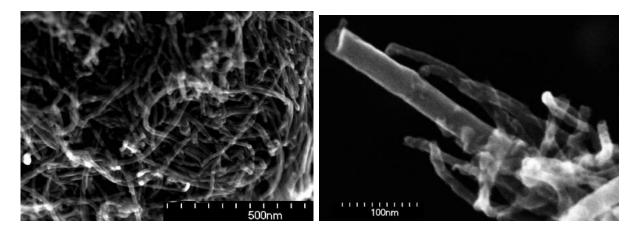
Scanning Electron Microscopy images were taken in a HITACHI S5200 using an acceleration voltage of 5 kV. No metallic coating was used. Nanotube widths were measured using ImageJ software: nanotube diameter =  $18 \pm 7$  nm. Typically, MWNT powder were poured onto a double face and conductive scotch, and imaged without further metallization.

## TEM

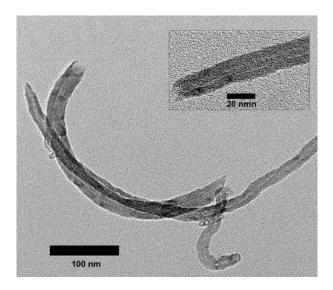
Transmission electron microscopy was performed in a Philips CM200 working at an acceleration voltage of 200 kV. In a typical procedure, MWNT powder was dispersed in ethanol, and subsequently a drop of the MWNT-ethanol dispersion was casted onto a TEM grids.



**Figure S1**. XRD pattern of as-received MWCNT where typical graphite pattern is remarked by the stars (Reference pattern: 0-001-0646)



**Figure S2**. Typical SEM images of the as produced MWCNT powder, where the width of the tubes can be easily assessed. Besides, the open ends of the tubes can be also confirmed.



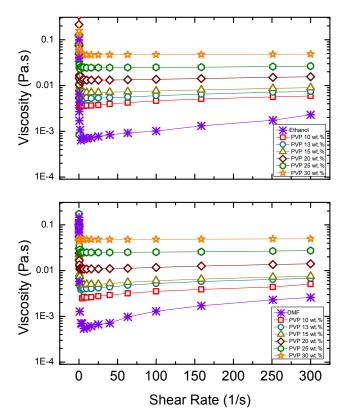
**Figure S3**. Typical TEM images of the as produced MWCNT reveal the width of the tubes, the open ends, and the thickness of the layered walls of the tubes.



**Figure S4**. Macroscopic pictures of 7 ml vials: a) PVP/Ethanol 30 wt.%. (b-c) Functionalized Multi Walled Carbon Nanotubes (NanoAmor) suspensions in PVP/Ethanol at different content of nanotubes, b) 0.02 wt.%, c) 0.2 wt.% and d) 0.4 wt.%. PVP content was maintained constant at 30 wt.% in all samples.



**Figure S5**. Show a photograph of this experimental set up for mechanical measurements. The specimens were tested as yarns of approximate diameter df = 900  $\mu$ m and length L = 3 cm by rolling flat fiber mats.



**Figure S6**. Viscosity vs Shear rate for PVP solutions for different content of PVP for shear rate from 0 to 300 (1/s)



**Figure S7.** Macroscopic pictures of 7 ml vials: a) PVP/Ethanol 30 wt.%. (b-c) pristine Single Walled Carbon Nanotubes (Elicarb) suspension in PVP/Ethanol at different content of nanotubes, b) 0.02 wt.%, c) 0.1 wt.% and d) 0.2 wt.%. PVP content was maintained constant at 30 wt.% in all samples. Single walled Carbon nanotubes (SWNT) form Elicarb "Thomas Swan" (UK) with purity > 95%, average diameter = 2 nm, average BET specific surface area > 800 m2/g, metal oxide content: maximum of 5 wt.% and moisture content: maximum of 1 wt.% are presently used.

DMF							
Sample	Flow Rate (mL/hr)	Distance (cm)	Voltage (kV)	Humidity (%)	Temperature (°C)		
PVP 10 %	0.1	12	8.5	56	21.3		
PVP 13 %	0.1	12	8.8	53	22.3		
PVP 15 %	0.1	12	8.4	56	21.1		
PVP 20 %	0.1	12	9.4	45	22.3		
PVP 25 %	0.1	12	8.5	45	22.5		
PVP 30 %	0.1	12	7.2	45	22.7		

**Table S1.** Experimental parameters for microparticle and fiber preparation using<br/>dimethylformamide (DMF) as solvent

**Table S2.** Experimental parameters for microparticle and fiber preparation using ethanol (EtOH) as solvent

EtOH								
Sample	Flow Rate	Distance	Voltage	Humidity	Temperature			
	(mL/hr)	(cm)	(kV)	(%)	(°C)			
PVP 10 %	0.1	12	13.8	53	22.5			
PVP 13 %	0.1	12	13.9	53	22.3			
PVP 15 %	0.1	12	14.1	49	22.5			
PVP 20 %	0.1	12	14.2	45	22.3			
PVP 25 %	0.1	12	14.5	42	21.3			
PVP 30 %	0.1	12	14.7	46	20.5			