## 1. Determination of water in DMF by NMR

DMF and NMP are hygroscopic solvents. As long as absorbed water in the solvent is kept below 2000 ppm we do not observe a significant effect on SWCNT aggregation kinetics. Water determination in a solvent is using NMR is a quantitative method. First the NMR is tuned and shimmed with deuterated DMF-D7 (D 99.5%). These setting are saved and used when analyzing the un-deuterated dispersions. Depending on the concentration of water present in the processed samples, the number of scans was adjusted to acquire good signal to noise for quantitative integration of the proton NMR signals. The methyl protons peak of the DMF at 8 ppm chemical shift is integrated and the integrated area is set to 123,000. Then the integrated signal of the water protons at 3.5 ppm chemical shift directly yields the concentration of water in the DMF in units of ppm (m/m) where the integrating factor is determined from:

The integrating factor was calculated as follows:

$$M_{H2O} = \left(\frac{mol}{g}DMF * \frac{g}{mol}H_2O\right) * 10^6 \left(\frac{S_{3.5}}{S_8}\right)$$
$$H_2O = \frac{1mol}{73.09g} * \frac{18g}{1mol} * 10^6 * \frac{1prot}{2prot} \frac{DMF}{H_2O} \left(\frac{S_{3.5}}{S_8}\right)$$
$$1(S_8) = 123,000 S_{3.5}$$

where  $S_{3.5}$  is the measure area at 3.5 ppm chemical shift and  $S_8$  is the measured area at 8 ppm chemical shift.

Typical water concentrations during the processing of a dispersion are listed in Table S.1. Results are average of 5 trials.

Table	<b>S.1</b>
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Process step	Average Concentration of Water (ppm) +/- standard error			
Open Bottle of DMF kept under N2	266 +/- 19			
Sample After Sonication				
(open to air 30 min)	361 +/- 30			
Sample After Centrifugation				
(open to air 45 min, 20 °C)	790 +/- 130			



**S. 1**. SEM of hybrid supraparticle assembly (Au-SWCNT-Au)



**S. 2.** SEM of hybrid supraparticle assembly (Au-SWCNT-Au)

A decanuclear ruthenium coordination complex<sup>21</sup> is mechanically wrapped about the ends of a dispersed single walled carbon nanotube (SWCNT).<sup>22</sup> The synthesis of the coordination complex is fully described in MacDonnell et al. The characterization of the mechanically wrapped complex around SWCNTs is described in Chaturvedi et al. 10  $\mu$ L of 10  $\mu$ M complex is added to 1000  $\mu$ l of 10 mg/L SWCNT in DMF and incubated for several hours. The complex-SWCNT particles are removed by centrifugation and washed with DMF to remove any unbound ruthenium complex. After removal of unbound complex, citrate stabilized 15 nm colloidal Au nanoparticles (NPs) were added into the dispersion and allowed to incubate for a few days. 10  $\mu$ L of the Au-complex-SWCNT dispersion was spun cast onto a Si chip and prepared for SEM.

From main text:

21. F. M. MacDonnell, M. J. Kim and S. Bodige, Coord. Chem. Rev., 1999, 186, 535-549.

22. H. Chaturvedi, A. N. Giordano, M.-J. Kim, F. M. MacDonnell, S. S. Subaran and J. C. Poler, *J. Phys. Chem. C*, 2009, **113**, 11254-11261.



**S. 3.** Molar conductivity  $\Lambda$  versus concentration<sup>1/2</sup> corresponds to Kohlrausch's Law for strong electrolytes:  $\Lambda = \Lambda^{\circ} - K[+yRu_x]^{1/2}$ 



**S. 4:** Zeta potential and electrophoretic mobility (EPM) of SWCNTs in dry (<1000 ppm  $H_2O$ ) DMF T = 25.0 °C. EPN measured by phase analysis light scattering, calculated zeta using Smoluchowski model. Zeta drops as +2**Ru1** coagulant is added.



**S. 5:** Looking down the length of two parallel (7,7) SWCNTs in DMF at 273.15 K. COMPASS force field, NVT, 1fs step, Anderson thermostat, periodic box. System was energy minimized, then annealed at 900 K for 50 ps, then run at 273.15 for 1.0 ns. The tubes were then moved together by 0.05 nm and the process repeated. Inter-tube separation is measured along surface normal from opposing carbon nuclei. Three solvent layers (b) are clearly formed between the tubes. As the tubes are compressed further two solvent layers (c) collapse into one tube-mediated solvent structure between the tubes when the tubes are  $\sim$ 0.90 nm apart (d.) and collapses as the tubes are moved closer together (e., f.)



**Fig. S. 6** Schulze-Hardy plot of dispersion stability versus charge on coagulant's cation. Inorganic salts in black with a slope =  $-4.4 \pm 0.4$  and mononuclear ruthenium complexes in red with a slope =  $-4.0 \pm 0.7$ .



Fig. S. 7 Molecular Dynamics calculation (COMPASS force field, NVT, 1fs step, Anderson thermostat, periodic box. The system was energy minimized, then run at 273.15 for 50 ps) of +1Ru1 cation solvated by DMF.  $1^{st}$  solvation sphere was estimated by including all solvent molecules within 0.35 nm of the cation's atoms. Average calculated coordination number is 20 with a ~11 nm solvated ion radius.



**Fig. S. 8** SWCNT RLCA kinetics for various coagulants. Data fit by both zeroth (left) and second order (right) aggregation models. Residual Sum of Squares listed for NaBr (A,B), +2Ru1 (C,D), and +2Ru2 (E,F).

Coagulant	Conc. (uM)	2 <sup>nd</sup> order kinetics			0 <sup>th</sup> order kinetics		
		[SWCNT] <sub>o</sub> (mg/L)	k (mg/L) <sup>-1</sup> h <sup>-1</sup>	RSS	[SWCNT] <sub>o</sub> (mg/L)	$k$ (mg/L) $h^{-1}$	RSS
NaBr	50	$9.17 \pm 0.007$	$(11.8, 2)^{-1}$ $(2.54 \pm 0.2)10^{-6}$	5.5E-5	$9.17 \pm 0.007$	$(1.0 \pm 0.18)10^{-4}$	6.1E-5
	120	$7.58\pm0.16$	$(1.70 \pm 0.11)10^{-4}$	1.7E-2	$7.19 \pm 0.46$	$(4.8 \pm 1)10^{-3}$	0.223
	150	$6.15 \pm 0.35$	$(3.14 \pm 0.48)10^{-4}$	6.8E-2	5.74 ± 0.19	$(4.9 \pm 0.4)10^{-3}$	3.9E-2
+2 Ru1	0.80	$9.12 \pm 0.09$	$(1.51 \pm 0.29)10^{-5}$	2.0E-2	9.11 ± 0.08	$(1.15 \pm 0.19)10^{-4}$	1.7E-2
	1.44	$8.59\pm0.46$	$(2.11 \pm 0.32)10^{-4}$	27E-2	$7.94 \pm 0.18$	$(6.61 \pm 0.45)10^{-3}$	9.1E-2
	1.50	$8.28\pm0.28$	$(2.95 \pm 0.24)10^{-4}$	7.9E-2	$7.31 \pm 0.33$	$(6.81 \pm 0.85)10^{-3}$	0.320
	1.64	$7.95 \pm 0.53$	$(6.28 \pm 0.84)10^{-4}$	7.2E-2	$6.38 \pm 0.84$	$(7.6 \pm 2)10^{-3}$	0.733
	1.70	$1.00 \pm 0.25$	$(4.8 \pm 2)10^{-3}$	3.4E-2	$0.63 \pm 0.07$	$(6.6 \pm 1.8)10^{-4}$	1.5E-2
+2 Ru2	0.95	$7.66 \pm 0.07$	$(2.01 \pm 0.09)10^{-4}$	8.9E-3	$7.44 \pm 0.12$	$(7.26 \pm 0.63)10^{-3}$	4.4E-2
	1.00	$8.40 \pm 0.42$	$(5.28 \pm 0.62)10^{-4}$	0.269	$6.92 \pm 0.37$	$(8.3 \pm 1.1)10^{-3}$	0.959
	1.20	$7.5 \pm 1$	$(1.30 \pm 0.27)10^{-3}$	0.433	$4.93 \pm 54$	$(7.0 \pm 1.6)10^{-3}$	2.042
	1.65	2.87 ± 0.16	$(6.32 \pm 0.33)10^{-3}$	2.2E-3	$1.42 \pm 0.23$	$(2.12 \pm 0.66)10^{-3}$	0.352
+3 Ru2	0.253	8.37 ± 0.16	$(5\pm 6)10^{-6}$	8.9E-2	8.37 ± 0.15	$(4 \pm 4)10^{-4}$	8.9E-2
	0.303	$7.24 \pm 0.12$	$(5 \pm 1)10^{-5}$	1.8E-2	$7.22 \pm 0.11$	$(2.33 \pm 0.57)10^{-4}$	1.8E-2
	0.320	$6.81 \pm 0.10$	$(4.71 \pm 0.67)10^{-5}$	3.1E-2	$6.79\pm0.07$	$(1.81 \pm 0.19)10^{-3}$	1.9E-2
	0.354	$6.33 \pm 0.32$	$(2.11 \pm 0.43)10^{-5}$	0.252	$6.12 \pm 0.23$	$(4.41 \pm 0.61)10^{-3}$	0.200
+4 Ru2	0.100	$7.87 \pm 0.27$	$(0.3 \pm 1)10^{-5}$	0.209	$7.86 \pm 0.27$	$(2 \pm 7)10^{-4}$	0.211
	0.120	$7.06 \pm 0.05$	$(3.73 \pm 0.29)10^{-5}$	6.0E-3	$7.03 \pm 0.05$	$(1.56 \pm 0.12)10^{-3}$	6.2E-3
	0.150	$3.48\pm0.4$	$(0.4 \pm 1)10^{-4}$	0.599	$3.47 \pm 0.45$	$(0.4 \pm 1)10^{-3}$	0.600
	0.180	$6.07\pm44$	$(2\pm 2)10^{-2}$	0.137	$0.52 \pm 0.3$	$(4 \pm 8)10^{-4}$	0.331

Table S2. RCLA kinetics data for SWCNTs by various coagulants in DMF at 25  $^{\circ}\mathrm{C}$