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Stability of Aqueous Suspensions of COOH-Decorated Carbon Nanotubes to Organic Solvents, Esterification, and Decarboxylation

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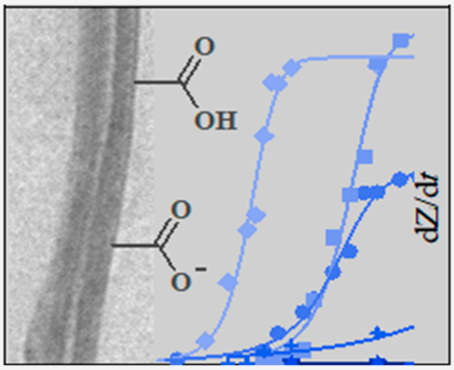
ABSTRACT: Carbon nanotubes are among the most widely used nanosystems, and stability of carbon nanotube suspensions is critical for nanotechnology and environmental science. Remaining in aqueous environment alone misses important factors that regulate colloidal stability in the presence of electrolytes. Indeed, introduction of (80−95) vol % organic solvents leads to sharp changes in suspension properties depending on the solvent. For example, the critical coagulation concentrations for a given inorganic or organic coagulating ion can change by 2 orders of magnitude when going from dimethyl sulfoxide to acetonitrile. We establish and explain these trends by Lewis acid−base interactions and show that a strong interaction extending beyond the standard theory of aggregation plays an important role.



Carbon nanotubes (CNT) are among the most widely utilized nanodispersed systems.[1−3] A huge number of studies are devoted to aqueous suspensions of both single-walled and multiwalled carbon nanotubes (SWCNT and MWCNT). The colloidal particles of the suspensions are usually negatively charged. Such systems are of interest both for nanotechnologies and environmental impact.[1,4−6] Colloidal properties of oxidized SWCNTs, decorated with COOH groups, in aqueous environments are of particular interest.[7] In addition to the generally accepted methods for determining the critical concentrations of coagulation (CCC) with various electrolytes, insights into the properties of suspensions of the same SWCNTs after either esterification of carboxyl groups or thermal decarboxylation carry significant importance.[7]

The present Communication establishes the factors that determine colloidal stability of SWCNT in aquatic environments. Experiments have shown that changes in CCCs caused by introduction of water-miscible organic solvents provide valuable insights. Therefore, we furthered our investigations using the CCC values as an important criterion for determining the SWCNT stability to aggregation in various solvents. Remaining in aqueous systems alone, one can miss important factors that regulate colloidal stability in the presence of electrolytes. Using the data obtained with CNTs in water−organic solvents, we demonstrate the presence of an additional strong stabilizing factor in aqueous suspensions, besides the electrostatic repulsion.

The previous work[7] clearly shows that various details of CNT oxidation, suspension preparation, etc., have a strong influence on the CCCs in water. On the one hand, this is characteristic of lyophobic systems, and on the other hand, it is due to the fact that CNTs are not a chemical compound (unlike fullerenes) but a material. This situation creates a difficulty in identifying important patterns, even though they are definitely present. In particular, the dependence of the CCC on the degree of oxidation is established.[1] In addition to observing the Schulze−Hardy rule, an unusual mechanism of the influence of two- and three-charged cations manifests itself owing to the so-called bridging effect.[1,7]

With regard to organic solvents, there are two trends in the literature. First, the dispersion and exfoliation of CNTs have been quantitatively studied in solvents of different chemical nature. Second, the properties of thus obtained colloidal solutions have been investigated in detail.

Already, 10−15 years ago, detailed studies of the dependence of the limiting concentration of CNTs in organic solvents on the descriptors of these solvents were performed.[8−12] Ausman et al.[8] demonstrated that stable dispersions of SWCNT are formed in solvents with a high Kamlet−Taft electron pair donation parameter, β, and a low hydrogen bond parameter, α. Bergin et al.[9] examined nitrogen-containing solvents and revealed that the level of dispersibility is more sensitive to the dispersive parameter δ_D than to the the polar δ_P

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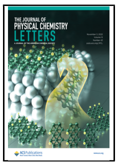

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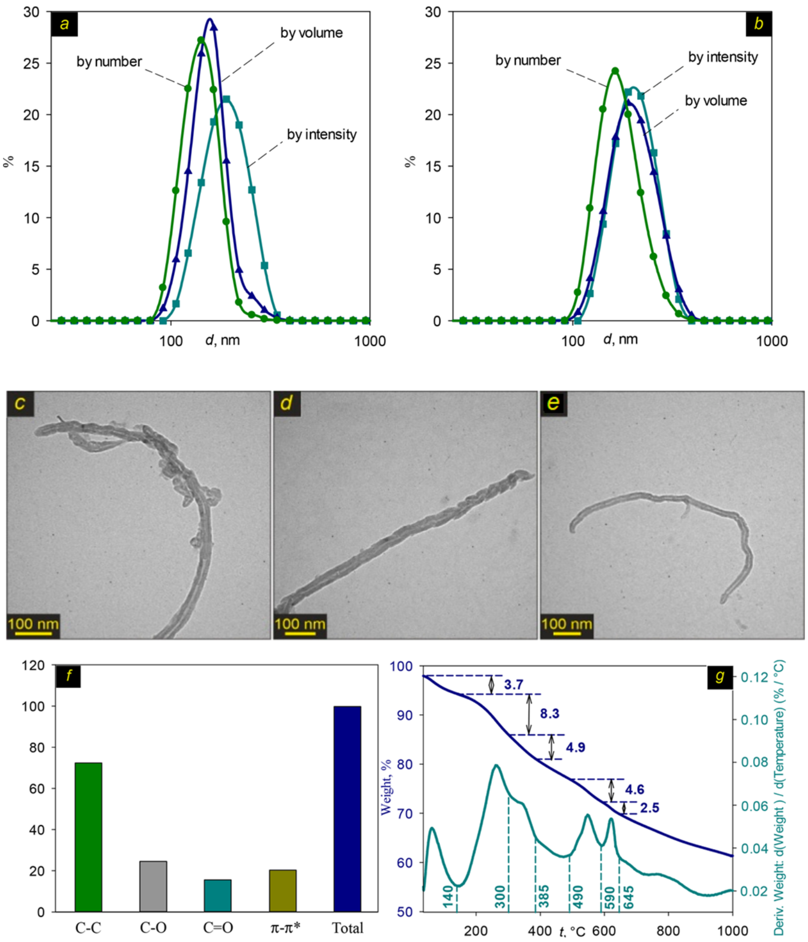

Figure 1. Characterization of the initial salt-free sample: size distribution of the 0.001 g L^{-1} SWCNT aqueous solution, at pH 5.7 (a); the same in 95 vol % DMSO (b); typical TEM images of the dried solution (c−e); relationships between different carbon bonds (XPS spectrum) after filtration and drying of the solution (f); TGA after filtering of the solution and drying (g).

or H-bonding δ_H Hansen parameter of the solvent. At the same time, Cheng et al.[10] reported that SWCNTs are easily dispersed in solvents with a Hildebrand solubility parameter of ≈22−24 $MPa^{1/2}$ and a Hansen δ_P parameter of ≈12−14 $MPa^{1/2}$. Coleman considered the solute−solvent interaction among other factors governing the exfoliation of CNTs and their maximum attainable concentration.[11] Premkumar et al.[12] investigated the dispersion of CNTs in liquids, focusing on the role of different types of interactions with solvents.

Our recent studies on C_{60} and C_{70} fullerenes revealed that comparison of the CCCs determined in water and in solvents of different nature allows one to shed light on the intrinsic properties of colloids.[13,14] Considering similar features of CNTs and fullerenes, we expect that corresponding study of SWCNTs can provide important insights.

Coagulation of CNT in organic solvents has been studied.[15−17] However, analysis of the data obtained with different CNTs[15−17] is hindered by variations in sample sources of the samples, lack of electrokinetic potential values, and unknown oxidation degree.

The strategy used here is as follows. As initial solutions for preparation of the suspensions, the stock aqueous solutions of the earlier used sample[7] are prepared via sonication as described previously.[7] The aliquots of these stock solutions are diluted by different organic solvents. This allows one to observe the solvent effects as such and avoid differences in the properties of samples prepared in water and directly in organic solvents.

The organic solvents fractions are 80 or 95 vol %. The need to use a range of solvent concentrations is caused, on the one hand, by the low stability of suspensions at high content of some organics and, on the other hand, by the high viscosity of DMSO at concentrations below 95 vol %, which hinders the utilization of the dynamic light scattering in accordance with the principle of operation of the device. However, the differences in the influence of the solvents are so striking that some variations of the content of organic cosolvents cannot change the general conclusions.

The stock suspensions are prepared by sonication in water as described previously; their concentrations are determined using optical absorbance.[7] Characterization of the system is presented in Figure 1. Importantly, whereas the XPS analysis of the initial sample results in an C/O atomic ratio of 5.34,[7] after dispersion in water via sonication, filtration, and drying, the ratio decreases to 1.88. The decrease indicates that the most hydrophobic (less oxidized) fraction of the SWCNTs and amorphous carbon are retained at the filter.

At working concentrations, the SWCNT species are mainly isolated.

Sodium and barium chlorides, cetyltrimethylammonium bromide (CTAB), and a cationic dye, Neutral Red (NR), are used as electrolytes. The size of colloidal particles in water,

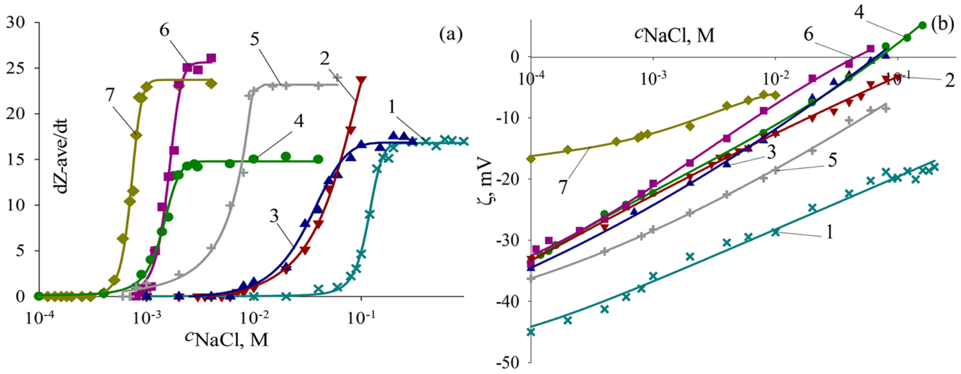

Figure 2. d*Z*-average/d*t*, nm/min (a) and the zeta potential (b) vs the concentration of NaCl, logarithmic scale, for SWCNT dispersions in water (1), 95 vol % DMF (2), 95 vol % DMSO (3), 80 vol % ethanol (4), 80 vol % methanol (5), 80 vol % acetonitrile (6), and 80 vol % acetone (7). The data are obtained using a Zetasizer Nano ZS from Malvern Instruments. The SWCNT concentration is 0.001 g L^{-1}; 25 °C, pH = 5.7 ± 0.1. See the Supporting Information for details. The data for water are from ref 7.

Table 1. Critical Coagulation Concentrations of Different Electrolytes for a SWCNT Suspension in Water-Organic Solvents[a]

solvent (x_2)[b]	critical coagulation concentrations of electrolytes, mM			
	NaCl	$BaCl_2$	Neutral Red	CTAB
water[c]	150 ± 50	1.0 ± 0.1	0.011 ± 0.001	0.050 ± 0.002
95 vol % DMF (0.807)	100 ± 50	–	–	–
95 vol % DMSO (0.812)	96 ± 4	1.5 ± 0.5	–	–
80 vol % methanol (0.608)	9.5 ± 0.5	–	–	–
80 vol % ethanol (0.527)	1.8 ± 0.2	0.0057 ± 0.0009	0.042 ± 0.001	11 ± 1
80 vol % acetonitrile (0.563)	2.0 ± 0.2	0.0085 ± 0.0004	0.044 ± 0.003	7.6 ± 0.1
80 vol % acetone (0.600)	0.85 ± 0.05	–	–	–

[a] SWCNT concentration is 0.001 g L^{-1}; *t* = 25 °C, pH = 5.7 ± 0.1. [b] Molar fraction of the organic cosolvent. [c] The data for water are from ref 7.

80 vol % methanol, ethanol, acetonitrile, and in 95 vol % DMF and DMSO is within the range of 185 to 200 nm, while in 80 vol % acetone it is around 260 nm, substantially higher. All sizes correspond to the diameter of an equivalent sphere and are expressed as *Z*-average values. All the ζ values are substantially negative and vary from −(53−54) mV in 80 vol % methanol, ethanol, and acetonitrile to −(44−48) mV in 95 vol % DMSO and water and to −(29−33) mV in 80 vol % acetone and 95 vol % DMF. More detailed characterization of the salt-free suspensions, the CCC estimation procedure, and other experimental data can be found in the Supporting Information.

Liu et al.[18] demonstrated the dependence of the sign of the ζ values of unfunctionalized graphene on the nature of the solvent depending on the electron transfer, while Damasceno and Zarbin[17] received such results with MWCNTs. Sonication was performed in both cases, and positive ζ values were observed only in the case of Cl-containing solvents.[17,18] We decided not to use the sonication in entire organic solvents because of possible unexpected side effects. For example, acidic admixtures appear in DMSO during sonication.[14] Therefore, only previously prepared initial aqueous suspensions are diluted with purified organic solvents. The above-mentioned ζ values obtained in our study may reflect various processes, including the dissociation of the carboxylic group. Therefore, we focused our efforts on interpreting of the CCC values.

The determination of the CCC values for NaCl in different media using a modification of the Fuchs function is exemplified in Figure 2a. The CCC corresponds to the beginning of the plateau. The results are presented in Table 1. In Figure 2b, the corresponding alterations of the electrokinetic potential are shown.

With regard to inorganic salts, a sharp drop in the CCC values with the introduction of alcohols, acetonitrile, and acetone and a slight variability in the case of DMF and DMSO are immediately striking. The drop of the CCC for NaCl on going from water to aqueous alcohols, acetonitrile, and acetone is 16−176-fold. In the case of DMF and DMSO, it is only 1.5−1.6. In the case of $BaCl_2$, the difference between DMSO and ethanol or acetonitrile is even more pronounced than that for NaCl. The absolute CCC values of the two salts are in line with the Schulze−Hardy rule.

There is a distinct connection with the basicity of the solvents. Indeed, for methanol, ethanol, acetonitrile, the values of the Gibbs energy of proton transfer from water to the given solvent are as follows:[19] $\Delta G_{tr}(H^+)$/kJ mol^{-1} = +8.7, + 11.1, and +44.8, while for DMF and DMSO they are −14.4 and −19.4; for water, $\Delta G_{tr}(H^+) = 0$ by definition. The normalized Gutmann's Donor Numbers are DN^N = 0.37 (acetonitrile), 0.44 (acetone), 0.49 (methanol, by convention), 0.63 (water, by convention), 0.68 (DMF), and 0.77 (DMSO).[17] Hence, DMSO and DMF are typical protophilic and cationophilic solvents,[20] whereas acetonitrile, acetone, and alcohols are protophobic solvents.[21] Some other solvent descriptors are given in the Supporting Information.

Better solvation of CNTs by protophilic solvents is also illustrated by Cheng et al.[10] These authors determined the maximal concentrations of their nanotube samples in DMF and DMSO as 0.022 and 0.006 mg mL^{-1}, respectively, while in acetonitrile and nitromethane they are below 0.001 mg mL^{-1}.

The results obtained now (Table 1) are in line with the solvent influence on the CCCs for electrolytes in C_{60} and C_{70} colloids in acetonitrile and methanol, on the one hand, and in DMSO and DMF, on the other.[13,14] Fullerenes are Lewis acids, and their interaction with proto- and cationophilic solvents can be expected. The CCCs in these solvents are even higher than in water. Note that the surfaces of CNT and fullerenes are of similar nature.

Even in the case of the colloids of detonated nanodiamond with positively charged particles, the CCC for NaCl in 95 vol % DMSO is 5.9 times higher than in 80 vol % CH_3CN, despite the sp^3-hybridization of the carbon atom in this case.[19] For $NaClO_4$, the difference is 10-fold.[22]

Hence, it can be expected that in water, like in solvents with high Gutmann's Donor Numbers, DMF and DMSO, specific stabilizing solvation factors are present, which can be considered as a kind of non-DLVO (Derjaguin, Landau, Verwey, Overbeek) contribution. By considering only aqueous systems and only the electrostatic repulsion and molecular attraction, one can miss important factors that regulate colloidal stability in the presence of electrolytes. It is the comparison of CCCs in water with those in protophobic solvents that reveals the hidden non-DLVO factors, manifested by peculiarities of solvation.

At this place, it is worth to recall the concept of the structural effect, introduced by Churaev and Derjaguin.[23,24] An additional solvation (hydration) contribution, V_s, to the total energy of interaction between two plates in an electrolyte solution is expressed in J m^{-2}, in eq 1.

$$V = V_{el} + V_{attr} + V_s \tag{1}$$

Here V_{el} is the energy of electrostatic repulsion, V_{attr} is the attraction energy that is caused by dispersion forces. Similar to the first two contributions, the V_s value is a function of the interparticle distance, h: $V_s = Kl\exp(-h/l)$. Here K and l are constants; for hydrophilic and hydrophobic surfaces, $K > 0$ (structural repulsion) and $K < 0$ (structural attraction), respectively.[23,24]

$$V = f(\Psi, I, \varepsilon_r)\exp\left(-\frac{h}{\lambda}\right) - \frac{({A_{CC}}^{1/2} - A_{SS}^{1/2})^2}{12\pi h^2} + Kl\exp\left(-\frac{h}{l}\right) \tag{2}$$

(We use here the DLVO equation in a general form, without specifying the shape of the colloidal particles.) The electrostatic contribution is dependent not only on h but also on the surface potential Ψ, ionic strength I, and relative permittivity of the solvent ε_r. A_{CC} and A_{SS} stand for the Hamaker constants of CNT−CNT and solvent−solvent interactions. Such equation can be used only qualitatively, because most SWCNTs at the working concentration are rather in the monomeric state.[7] The A_{CC} values published in the literature are within the range of $(4-60) \times 10^{-20}$ J (see the Supporting Information). Validation of the A_{CC} values based on the CCCs in water without accounting for the last item in eqs 1 and 2 seems to be erroneous.

It is reasonable to expect that for aqueous alcohols, acetonitrile, and acetone, the K values are negative, or at least much lower compared with those in water and aqueous DMSO and DMF. If, for instance, aqueous methanol is conditionally taken as the reference point ($K = 0$), then for water, aqueous DMSO and DMF, $K > 0$, whereas for acetonitrile and other solvents, $K < 0$.

The origin of the structural, or solvation, V_{el} value can be the acid−base interaction of both the SWCNT aromatic surface and the COOH groups with DMF and DMSO, in view of the van Oss concept,[25] whereas it should be ascribed to the hydration in pure water.

Contrary to the inorganic electrolytes, the CCCs of salts with organic cations even increase on going from water to water-organic mixtures, a 4-fold increase is observed for the cationic dye NR, whereas for CTAB it reaches ≈150−200, which is connected with the decisive role of adsorption of the amphiphilic cation. In water, the adsorption results in neutralization of the surface charge and hydrophobization of the surface of the colloidal particles. In the presence of 80 vol % ethanol or acetonitrile, the adsorption decreases owing to good solvation of the organic ions and hence stability of the suspension increases.

Overcharging, i.e., the change of the sign of the surface charge of colloidal particles, provides a clear evidence of adsorption of cations. Adsorption of counterions results in decelerating the coagulation and even formation of narrow stability zones at medium electrolyte concentration.

In aqueous CNT suspensions, this takes place for many organic cations and polyvalent inorganic cations. However, at high content of an organic solvent, solvation of the mentioned cations can change the picture. Interestingly, in the case of the NR dye, the overcharging and decelerating of the coagulation takes place in aqueous ethanol and acetonitrile (Figure 3a,c), whereas for CTAB the solvation of hydrophobic $C_{16}H_{33}$ tail weakens the adsorption (Figure 3b,d).

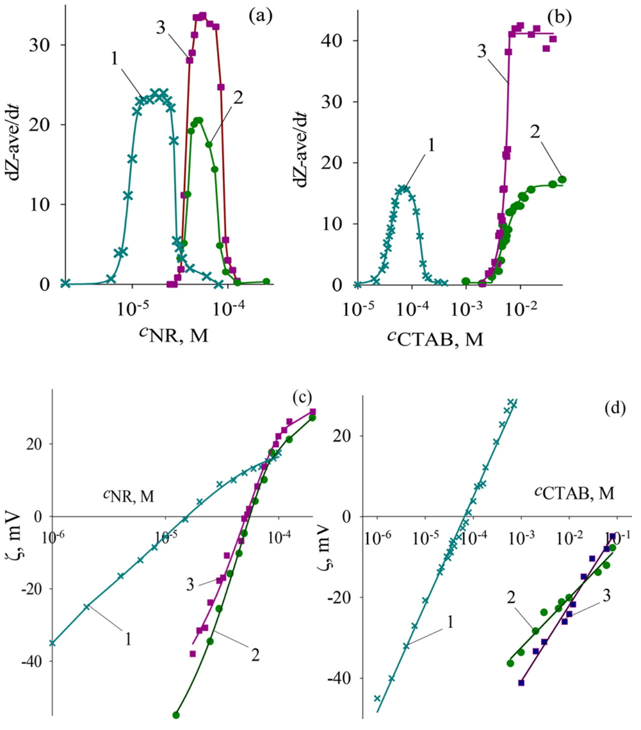

Figure 3. Coagulation of the SWCNT by NR (a) and CTAB (b) in water (1), in 80 vol % ethanol (2), and in 80 vol % acetonitrile (3); corresponding alterations of the ζ values for NR (c) and CTAB (d).

Overcharging of the colloid particles in NaCl solutions (Figure 2b) is more pronounced in solvents that solvate the Na^+ ions poorly, while in water, aqueous DMSO and DMF screening of the surface charge and compression of the electrical double layer plays an important role. In the case of $BaCl_2$, the overcharging of SWCNT particles is much more pronounced in the ethanol−water and acetonitrile−water

Table 2. Critical Coagulation Concentrations of NaCl for Suspensions of Modified SWCNT in Water−Organic Solvents[a]

solvent	CCC of NaCl, mM				
	initial	methylated		heated	
		for 240 h	for 370 h	at 350 °C	at 600 °C
water[b]	150 ± 50	110 ± 10	50 ± 2	43 ± 4	20 ± 1
95 vol % DMSO	96 ± 4	59 ± 5	50 ± 5	30 ± 4	52 ± 5
80 vol % ethanol	1.8 ± 0.2	7.1 ± 0.5	–	1.8 ± 0.2	10.3 ± 0.2
80 vol % acetonitrile	2.0 ± 0.2	6.0 ± 0.6	2.0 ± 0.5	1.9 ± 0.2	–

[a]SWCNT concentration: 0.001 g L^{-1}; 25 °C. [b]The data for water are from ref 7.

systems than in water and its mixtures with DMSO (see the Supporting Information).

Overcharging takes place also in water in the presence of 0.4 mg mL^{-1} lysozyme: $\zeta = +(30 \pm 1)$ mV at particle size of 250 ± 10 nm. This is in line with the data reported by Nepal and Geckeler.[26]

Comparison with the CCC values of C_{60} and C_{70} fullerenes in water, methanol, acetonitrile, DMF, and DMSO[13,14] reveals a striking similarity with the trends described in the present report. However, the current case is more complicated because of the presence of COOH groups on SWCNT surface. These groups can be hydrated to various extent, solvated, and ionized, and COO^- can associate with metal cations, influencing the ζ and CCC values. In order to clarify the role of the COOH groups, we examined suspensions of both esterified and decarboxylated SWCNT in the presence of organic solvents (Table 2).

The characterization of the SWCNTs modified by transformation of some of the COOH groups into $COOCH_3$[7] and (partly) decarboxylation via heating (Figure 1) is given in the Supporting Information. Importantly, XPS spectroscopy of the 240 h-esterified sample revealed that after dispersion in water and drying, the C/O ratio on the surface of the species is 2.68, whereas before the contact with water it equals 12.04.[7] Hence, again, most hydrophobic species are retained on the filter. After dispersion and drying of a 1000 °C heated sample, this ratio decreases even further to 2.29. Removal of oxygen-containing groups is seen in a typical TGA (Figure 1); see also ref 27.

In the case of DMSO, the decrease in stability with respect to the electrolyte is usually reduced. This is not surprising considering the esterification or removal of the COOH groups, which is a result of a decrease in the $|\zeta|$ values (see Table S4). However, in the case of ethanol and acetonitrile, the stability mainly increases. The reason is better solvation of $COOCH_3$ and H as compared with COOH. For instance, the CCC ratio in water and in 80 vol % ethanol changes from 83 for the initial sample to 15.5 (methylated for 240 h); 24 (heated at 350 °C); and 1.9 (heated at 600 °C). Also, in the case of hydrophobic surfaces, an alternative charging mechanism connected with the HO^- adsorption should be taken into account.[28]

Summarizing, the colloidal stability of an aqueous suspension of COOH-decorated SWCNT with respect to inorganic electrolytes drops sharply on substantial addition of methanol, ethanol, acetonitrile, and acetone. Contrary to it, in the case of cationophilic (protophilic) solvents, such as DMF and DMSO, the critical concentrations of coagulation are much closer to those in water. This is in concert with other details of coagulation of esterified and decarboxylated SWCNT and points to a substantial non-DLVO contribution to the stability of suspensions of SWCNT in water.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.jpclett.2c02902.

Characterization of the SWCNT sample, materials, apparatus, and details of coagulation studies, solvent parameters, and a summary of literature data on Hamaker constant of CNT (PDF)

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Notes

The authors declare no competing financial interest.

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