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Aqueous nanostructures in water induced by electromagnetic fields emitted by EDS

A conductometric study of fullerene and carbon nanotube EDS

V. Elia · L. A. Marrari · E. Napoli

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Abstract This article reports the experimental results of a conductometric study on the time evolution, over a 541-day period, of 450 samples of Extremely Diluted Solutions (EDS) of fullerene and carbon nanotube and 450 samples of twice-distilled water, stored in alternate rows of EDS and water spaced 0.5 cm apart. The purpose was to establish whether these two aqueous systems are able to transmit, via electromagnetic fields, their variations in the supramolecular structure of the solvent water which has not undergone any previous perturbation. The chemical-physical method employed was conductometry, which proved to be the simplest and most efficient means for quickly and accurately monitoring the structural variations. In addition, since it has been demonstrated that there is a clear linear correlation between specific conductivity and heat of mixing with alkaline solutions, the conductometric result can also be extended to the calorimetric result. These findings, though doubtless unexpected and intriguing, are highly significant. The alterations over time of the pure water samples follow those of the EDS surprisingly closely.

Keywords Water · EDS Extremely Diluted Solutions · Electromagnetics fields · Dissipative structures · Conductivity measures

Introduction

“No one really understands water. It is embarrassing to admit it, but the stuff that covers two-thirds of our planet is still a mystery. Worse, the more we look, the more the problems accumulate: new techniques probing deeper into the molecular architecture of liquid water are throwing up more puzzles” said Ball in 2008 [1]. Many biological phenomena lead to this fascinating mystery, either in vegetal models [2], animal models [3], cellular models [4, 5] as well as in geochemistry [6]. Over the past 20 years, chemical-physical studies on extremely diluted aqueous solutions (EDS), obtained through a repeated process of successive centesimal dilutions and succussion (vigorous vertical shaking), have detected numerous “anomalies” in the chemical-physical behaviour of such systems [7–17, 21]. Amongst these [9, 11–13], the most striking are the evolution over time (“arrow of time”) marked by a succession of maxima and minima, and the dependence on volume (“volume effect”) of the sample’s evolution over time. Small volumes in fact exhibit a faster and greater time evolution of specific conductivity [10, 12]. The measure of specific conductivity, χ ($\mu\text{S cm}^{-1}$), was found to be extremely useful for tracking the structural alterations of the solvent water due to the combined arrow of time–volume effect. The versatility of this method is tied to the working hypothesis that water is a complex system capable of self-organising in response to perturbations of various kinds (mechanical and electromagnetic), even when these are of small magnitude. These elicit the formation in the water of “dissipative structures” [18, 19], that is to say ordered systems of water molecules (clusters, molecular aggregates, nanostructures). It is worth noting that the physical meaning of specific conductivity is the electricity flow through a cube of unit side per unit time,

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under a unit electrical potential gradient. Naturally, the electricity flow is dependent on the speed at which the charge carriers (ions) move. For what concerns H^+ and OH^- ions, which are always present in water due to the phenomenon of autoprotolysis, and their anomalous property of having a much greater mobility than other ions of similar ionic radius, in 1806 Grotthuss [20] proposed the hopping mechanism to account for the anomalous diffusion of these ions. When an H^+ ion encounters a water molecule in a favourable geometrical position, it forms a bond with the water molecule which in its turn releases an H^+ ion. It is thus, effectively, as if ions which meet a water molecule that enables this “hop” diffuse faster than ions which do not meet a water molecule in this favourable geometrical position. The proton hopping hypothesis, which entails an increased specific conductivity as a result of the faster ion diffusion, would account for the greater conductivity of these systems, without need to posit an increased concentration of electrical charge carriers, i.e. of ions. It would in fact be the “dissipative structures” which, through their dimensions (variation in the supramolecular structure of the water), augment the contribution of the hopping mechanism and hence the resultant diffusion of ions, and so also the specific conductivity. In reality, there are also a certain number of electrolytes released by the glass of the containers (sodium bicarbonate, silicic acid, boric acid), and these contributions are duly taken into account by performing a chemical analysis of these aqueous systems and subtracting their contribution to the parameter under study (conductivity, mixing heat, pH). During the course of the study in this delicate field, it became clear that the water itself is capable of self-organising in response to even slight perturbations by dissipating, as an open system, environmental energies of an electromagnetic nature. This implies that pure water is not a stable system, but instead subject to alterations of its chemical-physical parameters by effect of the formation of dissipative structures, especially if confined to “small” volumes [10, 12]. For this reason, the study of the EDS required the adoption of a new type of control (blank), consisting of an aqueous solution containing the same amounts of impurities released by the glassware containers. By so doing, the differences between the experimental parameters and those arising from the chemistry of the system (excess magnitudes) are attributed to the presence of “dissipative structures”. During the past 3 years of this research, it has been possible to characterise the nature of these structures in more detail. In particular, it has been possible to determine their concentration and the thermodynamic parameters of the formation of binding complexes with the OH^- and H^+ ions, through methodologies of conductometric, pH-metric and calorimetric titration [14–17]. The results obtained are highly self-consistent, and tend to support the working hypothesis that

the hopping mechanism is augmented by the presence of molecular aggregates of water molecules. The purpose of this work is to systematically verify, using a large number of samples, the possibility that EDS transmit to pure (twice-distilled) water, via electromagnetic fields, perturbations that induce alterations in the supramolecular structure of the water which are not generically ascribable to the environment, but specifically to the presence of the EDS. A similar assertion, but not identical, was put forward by Montagnier [4] using very different methodologies from those employed by us. In order to minimise the effects of ambient energies, it was decided to place the containers of the EDS and those of the twice-distilled water closely spaced, at a distance of 0.5 cm. To reduce the duration of the experiments and magnify the alterations in conductivity, it was decided to work with very small volumes, in the order of 0.25 mL. In this way, the combined time–volume effect enabled us to obtain reliable results over the course of only 2 years of testing. The close spacing of the vials probably made it possible to reduce the effects of ambient energies, allowing those of the adjoining EDS to prevail. It should be underlined that the untreated pure water is affected by external energy fields, in the same way as the EDS. However, for the EDS the perturbation which triggers the phenomenon has already occurred due to the preparation protocol, so that the system probably exploits the ambient energies, dissipating a part of them, to remain far from equilibrium, but with the direction of evolution already underway. For pure water, the perturbation which imparts the direction is instead probably ascribable to proximity with the EDS, whose evolutions over time are closely followed. In any case, irrespective of the possible interpretations of the phenomenon, the experimental data reveal a sufficiently certain phenomenon.

Experimental section

Procedures

An EDS is prepared by iterating two steps: dilution (centesimal or decimal) and “dynamization”. The process of “dynamization” (succussion step), consists in 100 violent vertical strokes in 12 s, given by a mechanical apparatus (DYNA HV 1 by Debofar N.V.S.A. Belgium). For a centesimal dilution, the starting point is a 1% in mass solution: to prepare, for example, a Magnesium Muriaticum ($MgCl_2$, symbol MM) solution, 1 g of $MgCl_2$ is added to 99 g of solvent. After the mechanical treatment, plain successions, the solution so obtained is called “1 centesimal hahnemannian” or 1CH in short, and the solution is labelled with the name or formula of the solute, or a simple abbreviation of it. So in the previous example, MM 1CH is obtained. In

order to prepare the 2 centesimal hahnemannian (MM 2CH), 1 g of the 1CH solution is added to 99 g of solvent, and the resulting solution is “dynamized”.

All the EDS studied in this work are therefore composed of pure water containing dissolved components of the glassware containers, at impurity level concentrations. The study was performed on 900 vials of 2 mL volume filled with 0.25 mL of EDS or twice-distilled water. The vials were arranged in 10 × 10 arrays for a total of 10 arrays. In each 100-vial array, the odd-numbered rows held the samples of EDS whilst the even-numbered rows held the samples of twice-distilled water. Each row or column was spaced 0.5 cm from the adjoining one. Over a time period from 0 to 541 days, at intervals of approximately 4 months, the EDS samples in each array (odd-numbered rows) were taken and placed in a single container. The specific conductivity and the sodium bicarbonate concentration were then measured. The same procedure was then repeated for the water samples (even-numbered rows). It must be underlined that the arithmetic mean of measured conductivity of each vial is in a very good agreement with the result obtained mixing the content of all the vials and then measuring the conductivity.

Conductivity measurements

Systematic measurements of the specific conductivity were performed on the EDS, using a conductometer, YSI model 3200, employing a conductivity cell with constant equal to 1.0 cm⁻¹. Before measuring the conductivity of the sample, the cell has to be calibrated by determining the cell constant *K* (cm⁻¹). The specific conductivity χ ($\mu\text{S cm}^{-1}$), is then given by the product of the cell constant and the conductivity of the solution. For a given cell, the cell constant is determined by measuring the conductivity of a KCl solution with a specific conductivity that is known with great accuracy for several concentrations and temperatures. All the values of conductivity were temperature corrected to 25 °C, using a pre-stored temperature compensation for pure water and performed in a room at controlled temperature of 25 ± 1 °C. In programmed experiments to obtain the standard deviation on a single experiment we repeated ten times the measurement of conductivity for a sample of about 42 $\mu\text{S cm}^{-1}$. The mean was 42.0 $\mu\text{S cm}^{-1}$ and the standard deviation 0.1 $\mu\text{S cm}^{-1}$, i.e. 0.23% of the mean.

Analytical determination of impurities

The sodium concentration was determined by the spectroscopic method of atomic absorption, using a Spectra A Varian instrument. Before measuring the samples, the necessary calibration curve was obtained using standard

solutions. To prepare the standard solutions, NaCl has been dissolved in water and diluted to 1 L to obtain 1,000 $\mu\text{g/mL}$ Na. The working conditions were: Lamp current: 5 mA; Fuel: acetylene; Support: air; Flame stoichiometry: oxidizing. The wavelength used, chosen on the basis of the concentration range of sodium, was of 589.6 nm. Sodium is partially ionised in the air-acetylene flame. To suppress the ionisation, some potassium chloride solution was added, to produce a final concentration of 2,000 $\mu\text{g/mL}$ in every solution, including the blank.

Results and discussion

Table 1 reports: the volume of the samples in the individual vials, their age (days), the average values of the

Table 1 Fullerene (active principle)

CH ^a	V/mL ^b	t/days ^c	$\chi_{\text{EDS}}^{\text{d}}$	$\chi_{\text{H}_2\text{O}}^{\text{d}}$	$\chi_{\text{EDS}}^{\text{Na+e}}$	$\chi_{\text{H}_2\text{O}}^{\text{Na+e}}$	$\chi_{\text{EDS}}^{\text{E f}}$	$\chi_{\text{H}_2\text{O}}^{\text{E f}}$
5CH	0.25	0	24.0	15.0	24.0	14.9	0	0.1
		49	39.0	34.0	35.2	30.1	3.8	3.9
		127	65.0	62.0	54.5	53.5	10.5	8.5
		322	75.0	68.0	59.5	53.1	15.5	15.0
		541	88.0	77.0	59.9	51.0	28.1	26.0
7CH	0.25	0	9.0	8.0	9.2	8.0	-0.2	0
		3	22.0	16.0	10.7	10.3	11.3	5.7
		7	22.0	23.0	12.5	13.3	9.5	9.7
		56	29.0	31.0	20.5	20.6	8.5	10.4
		135	37.0	36.0	27.8	21.8	9.2	14.2
		329	44.0	41.0	32.5	32.6	11.5	8.4
9CH	0.25	0	6.0	4.9	6.3	4.9	-0.3	0
		1	21.0	17.0	10.3	8.1	10.7	8.9
		55	43.0	36.0	24.7	22.7	18.3	13.3
		135	45.0	45.0	34.2	35.4	10.8	9.6
		329	55.0	55.0	40.4	40.7	14.6	14.3
12CH	0.25	0	20.0	18.0	20.2	18.2	-0.2	-0.2
		48	57.0	55.0	29.0	26.1	28.0	28.9
		128	69.0	63.0	53.4	50.1	15.6	12.9
		322	77.0	69.0	54.9	55.0	22.1	14.0
		541	82.0	87.0	55.6	58.6	26.4	28.4
30CH	0.25	0	23.0	11.0	23.2	11.2	-0.2	-0.2
		49	45.0	43.0	24.2	19.4	20.8	23.6
		128	59.0	52.0	44.0	40.3	15.0	11.7
		322	67.0	58.0	53.0	47.1	14.0	10.9
		541	78.0	75.0	54.2	48.7	23.8	26.3

^a Dilution, ^b volume/mL, ^c time/days, ^d specific electrical conductivity of EDS and twice-distilled H₂O, ^e specific electrical conductivity of NaHCO₃, ^f excess specific electrical conductivity of EDS and twice-distilled H₂O

specific conductivity χ ($\mu\text{S cm}^{-1}$) of the EDS and of the water, the conductivity of the sodium bicarbonate and the excess conductivity, meaning the difference between the experimental value and that of the bicarbonate solution, for the 500 samples of EDS (5, 7, 9, 12 and 30CH) of fullerene. Table 2 reports the degree of dilution (5CH) the volume of the samples, the average conductivity of EDS and of water for the 400 samples di EDS and twice-distilled water, the specific electrical conductivity of sodium bicarbonate and the excess conductivity of the 400 samples of carbon nanotube EDS (5CH) and water. It should be noted that the experimentally measured conductivities of the 50 EDS and water samples in the individual arrays yield an arithmetic mean that agrees well (within 2%) with the experimentally measured conductivity of the mixture of 50 samples. Finally, the individual vials were refilled with a volume of 0.25 mL taken from the mixture of the 50 samples.

Table 3 reports the degree of dilution, the values, after 541 days in the arrays, of the average of the specific conductivity χ ($\mu\text{S cm}^{-1}$) of EDS (5, 7, 9, 12 and 30CH), the specific conductivity of the sodium bicarbonate for the 500 samples of EDS (5, 7, 9, 12 and 30CH) and water. Table 4 reports the sequential number (1–4) of the arrays consisting

Table 2 Nanotubes (active principle)

CH ^a	V/mL ^b	t/days ^c	$\chi_{\text{EDS}}^{\text{d}}$	$\chi_{\text{H}_2\text{O}}^{\text{d}}$	$\chi_{\text{EDS}}^{\text{Na+e}}$	$\chi_{\text{H}_2\text{O}}^{\text{Na+e}}$	$\chi_{\text{EDS}}^{\text{E f}}$	$\chi_{\text{H}_2\text{O}}^{\text{E f}}$	
5CH	0.25	0	10.8	1.3	11.2	1.6	-0.4	-0.3	
		89	18.0	10.0	17.4	9.2	0.6	0.8	
		[1]	126	44.0	39.0	24.8	15.0	19.3	24.0
		301	66.0	52.0	61.0	43.2	5.0	8.8	
		511	68.0	65.0	58.0	45.0	10.3	20.0	
5CH	0.25	0	10.8	1.3	10.7	1.7	0.1	-0.4	
		89	20.0	10.0	18.3	7.6	1.7	2.4	
		[2]	126	53.0	47.0	24.3	15.1	28.7	31.9
		301	60.0	61.0	53.4	51.6	6.6	9.4	
		511	85.0	77.0	61.0	60.0	24.0	16.7	
5CH	0.25	0	10.8	1.3	10.7	1.7	0.1	-0.4	
		89	19.0	10.0	15.9	8.4	3.1	1.6	
		[3]	126	49.0	46.0	21.2	15.8	27.8	30.2
		301	58.0	60.0	46.9	51.6	11.1	8.4	
		511	79.0	75.0	61.0	48.0	18.3	27.3	
5CH	0.25	0	10.8	1.3	10.7	1.7	0.1	-0.4	
		89	20.0	8.0	17.0	7.1	3.0	0.9	
		[4]	126	49.0	43.0	23.5	13.8	25.5	29.2
		301	65.0	56.0	55.3	46.9	9.7	9.1	
		511	99.0	79.0	71.4	59.0	27.6	20.5	

^a Dilution, ^b volume/mL, ^c time/days, ^d specific electrical conductivity of EDS and twice-distilled H₂O, ^e specific electrical conductivity of NaHCO₃, ^f excess specific electrical conductivity of EDS and twice-distilled H₂O

Table 3 Specific conductivity of EDS and H₂O at 541 days

CH ^a	$\chi_{\text{EDS}}^{\text{b}}$	$\chi_{\text{H}_2\text{O}}^{\text{b}}$	$\chi_{\text{EDS}}^{\text{Na+c}}$	$\chi_{\text{H}_2\text{O}}^{\text{Na+c}}$
5	88.0	77.0	59.9	51.0
7	56.0	54.0	36.9	34.0
9	63.0	62.0	40.7	39.0
12	82.0	87.0	55.6	58.6
30	78.0	75.0	54.2	48.7

^a Dilution, ^b specific electrical conductivity of EDS (Fullerene) and twice-distilled H₂O, ^c specific conductivity of NaHCO₃ of EDS and H₂O

Table 4 Specific conductivity of EDS and H₂O at 511 days

N ^a	$\chi_{\text{EDS}}^{\text{b}}$	$\chi_{\text{H}_2\text{O}}^{\text{b}}$	$\chi_{\text{EDS}}^{\text{Na+c}}$	$\chi_{\text{H}_2\text{O}}^{\text{Na+c}}$
1	68.0	65.0	58.0	45.0
2	85.0	77.0	61.0	60.0
3	79.0	75.0	61.0	48.0
4	99.0	79.0	71.4	59.0

^a Set consisting of 100 samples, ^b specific electrical conductivity of EDS (nanotube) and twice-distilled H₂O, ^c specific conductivity of NaHCO₃ of EDS and H₂O

of 100 samples, the specific conductivity of the 5CH EDS of carbon nanotube and twice-distilled water, the conductivity of sodium bicarbonate for EDS and water samples.

Figures 1, 2, 3, 4, 5 illustrate the experimental and excess conductivities for the EDS and for water as a function of the time spent in the arrays. It should be noted that at time zero the EDS samples have a greater conductivity than the water samples, because they were prepared in advance. As can be easily seen from the figures, each EDS (5, 7, 9, 12, 30CH) exhibits a characteristic trend as a function of time. We observe a small peak in excess conductivity at around 2 months at least for fullerene EDS followed by a tendency to increase again. The excess conductance values are likewise characteristic for each EDS, and mimicked to a surprising extent by the water samples held in the same array. Figure 6 shows a histogram of the specific conductivity values for the EDS and water samples belonging to the same array after a period of 541 days in the individual array, for the 5 studied arrays containing respectively the 5, 7, 9, 12 and 30CH EDS. From Figs. 1, 2, 3, 4, 5 that illustrate the experimental values of χ for the EDS as a function of time, we note that the χ curves of the water samples follow those of the adjoining EDS, and that the values reached by the EDS differ depending on the number of centesimal dilutions (CH) in the iterative process. Figures 7, 8, 9, 10 illustrate the experimental and excess conductivity for nanotube EDS 5CH aged in four different arrays. Also in this case we observe a small peak in excess conductivity at about

Fig. 1 Specific electrical conductivity of EDS (Fullerene 5CH) and distilled H₂O versus *t* (days) and excess specific electrical conductivity of EDS versus *t* (days)

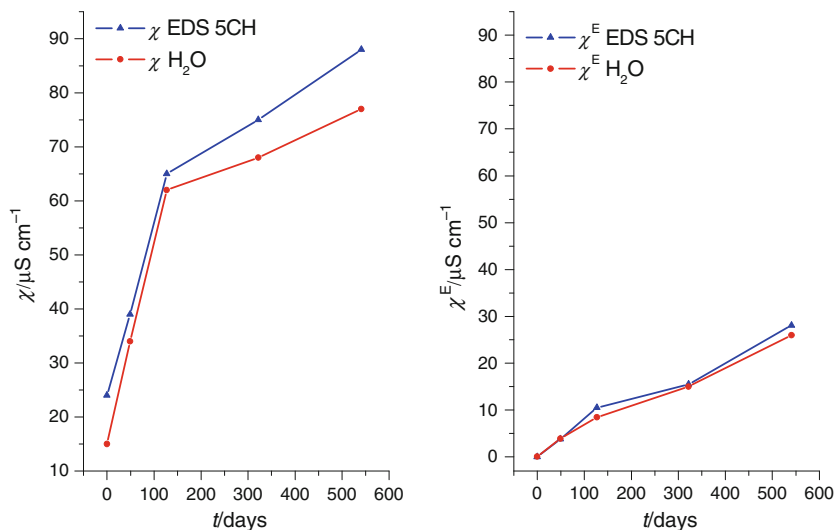


Fig. 2 Specific electrical conductivity of EDS (Fullerene 7CH) and distilled H₂O versus *t* (days) and excess specific electrical conductivity of EDS versus *t* (days)

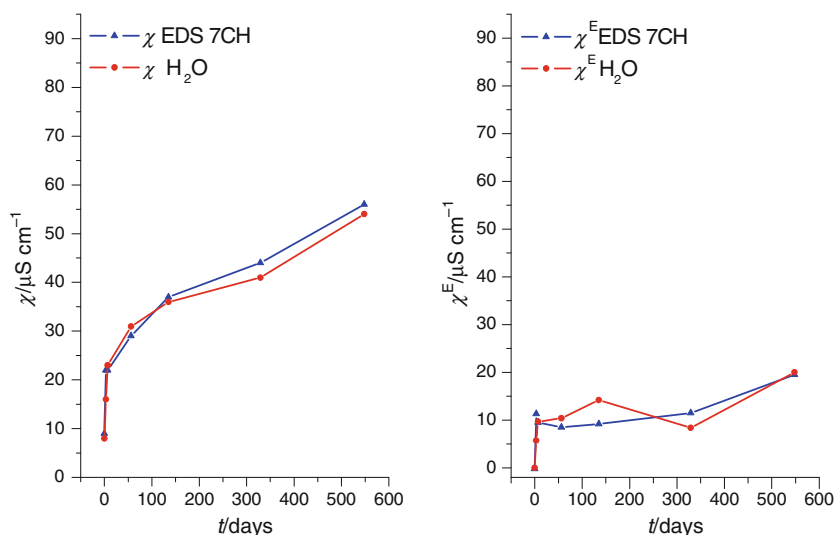


Fig. 3 Specific electrical conductivity of EDS (Fullerene 9CH) and distilled H₂O versus *t* (days) and excess specific electrical conductivity of EDS versus *t* (days)

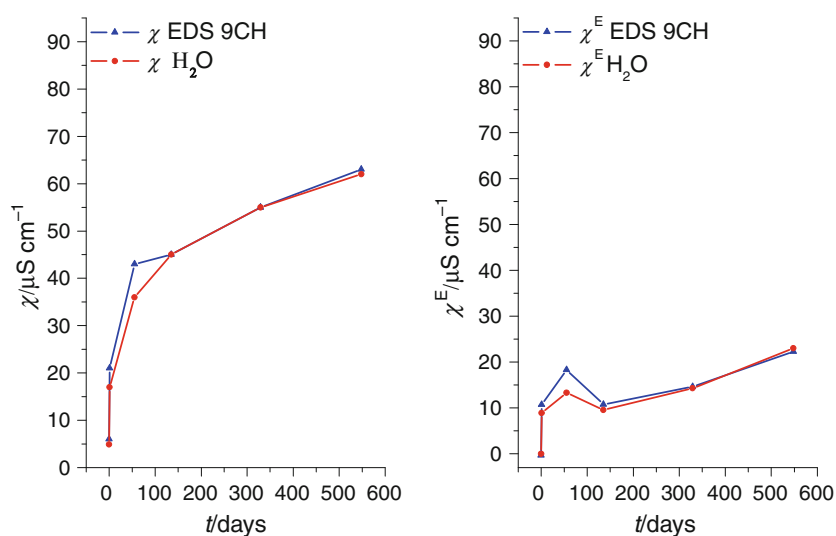


Fig. 4 Specific electrical conductivity of EDS (Fullerene 12CH) and distilled H₂O versus t (days) and excess specific electrical conductivity of EDS versus t (days)

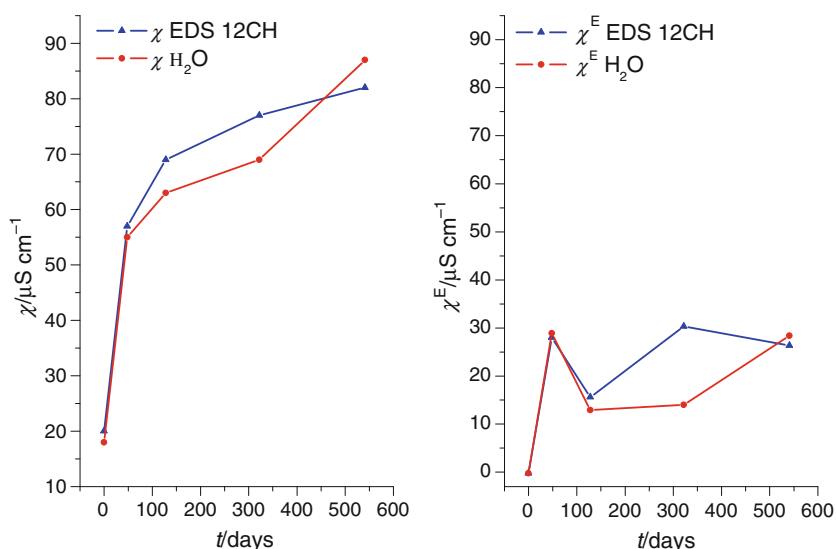


Fig. 5 Specific electrical conductivity of EDS (Fullerene 30CH) and distilled H₂O versus t (days) and excess specific electrical conductivity of EDS versus t (days)

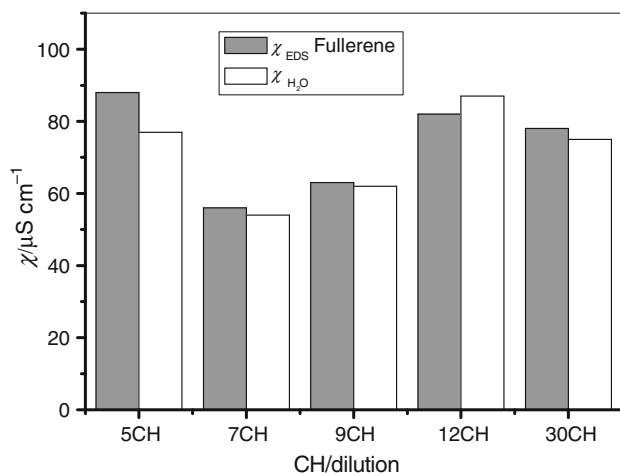
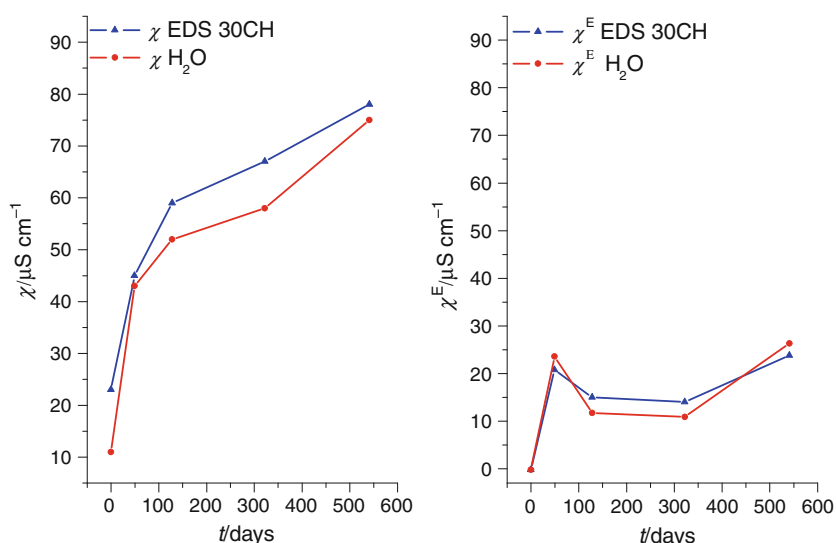


Fig. 6 Specific electrical conductivity of EDS (Fullerene) and distilled H₂O at 541 days

4 months followed by a tendency to increase. Figure 11 shows a histogram of the specific conductivity values for the EDS of carbon nanotube and water samples belonging to the four different arrays after a period of 541 days in the individual array, for the 5CH EDS. Though unexpected, these experimental results are nevertheless significant, and setting aside possible interpretations concerning the formation of dissipative structures and effects of electromagnetic fields, it is apparent that the proximity to the EDS causes the water to evolve with a time trend and numerical values that closely follow the behaviour of the EDS. Furthermore, the shape of the curve, with maxima and minima over time, strongly indicates that these systems cannot be described by classical thermodynamics. Rather, they are systems (including the pure water) operating far from equilibrium, where they remain or move further away

Fig. 7 Specific electrical conductivity of EDS (nanotubes set 1) 5CH and twice-distilled H₂O versus *t* and excess specific electrical conductivity of EDS (set 1) and twice-distilled H₂O versus *t*

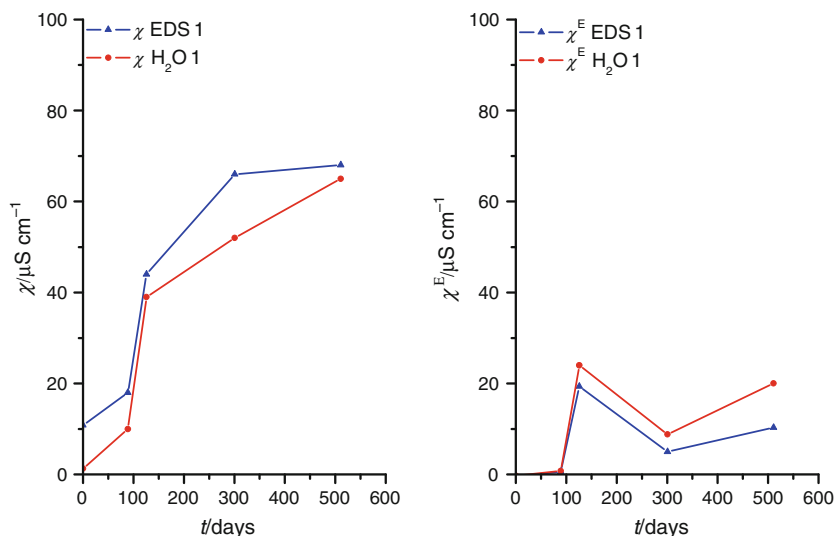


Fig. 8 Specific electrical conductivity of EDS (nanotubes set 2) 5CH and twice-distilled H₂O versus *t* and excess specific electrical conductivity of EDS (set 2) and twice-distilled H₂

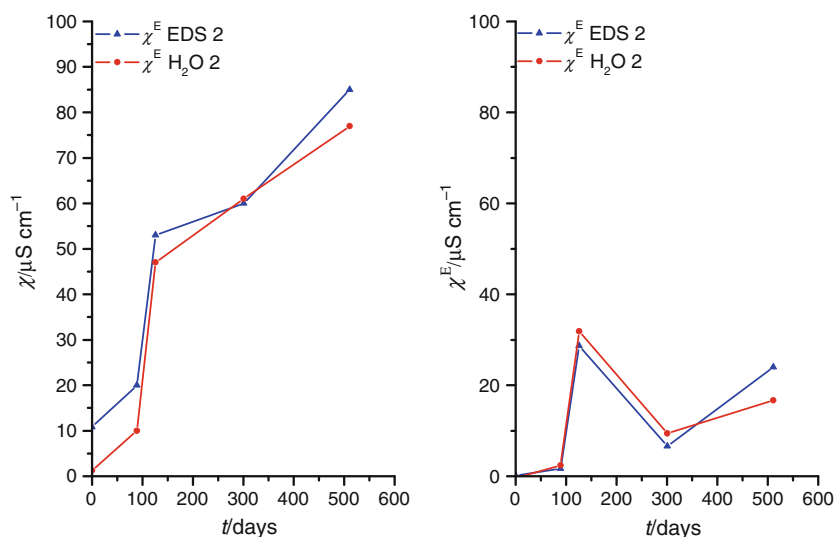


Fig. 9 Specific electrical conductivity of EDS (nanotubes set 3) 5CH and twice-distilled H₂O versus *t* and excess specific electrical conductivity of EDS (set 3) and twice-distilled H₂O versus *t*

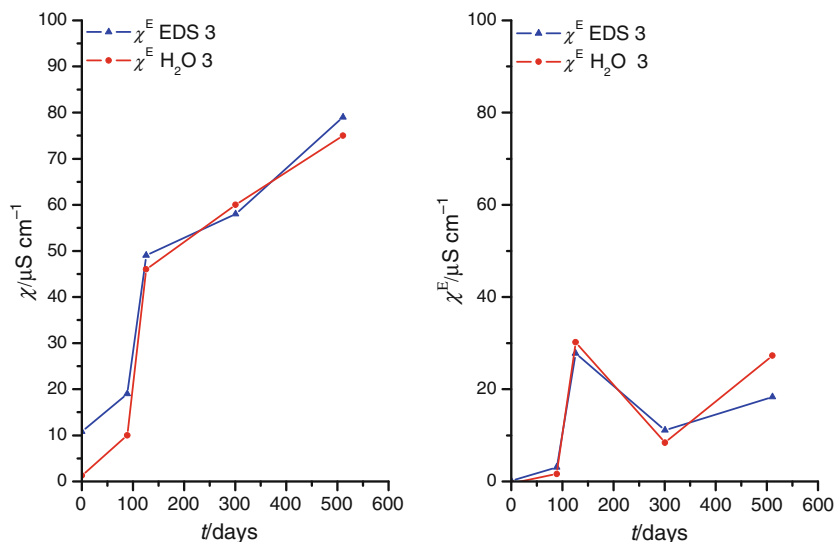


Fig. 10 Specific electrical conductivity of EDS (nanotubes set 4) 5CH and twice-distilled H₂O versus t and excess specific electrical conductivity of EDS (set 4) and twice-distilled H₂O versus t

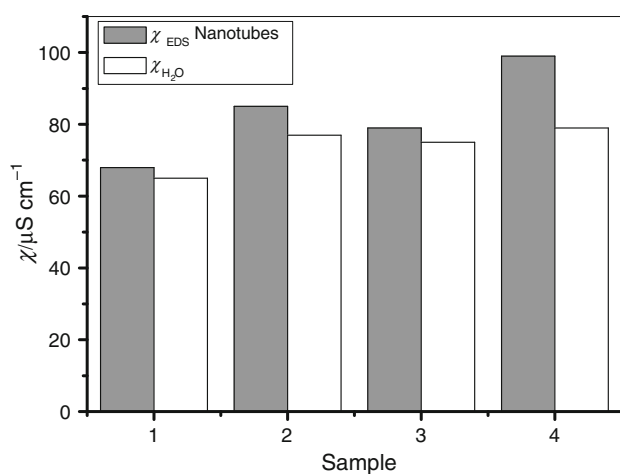
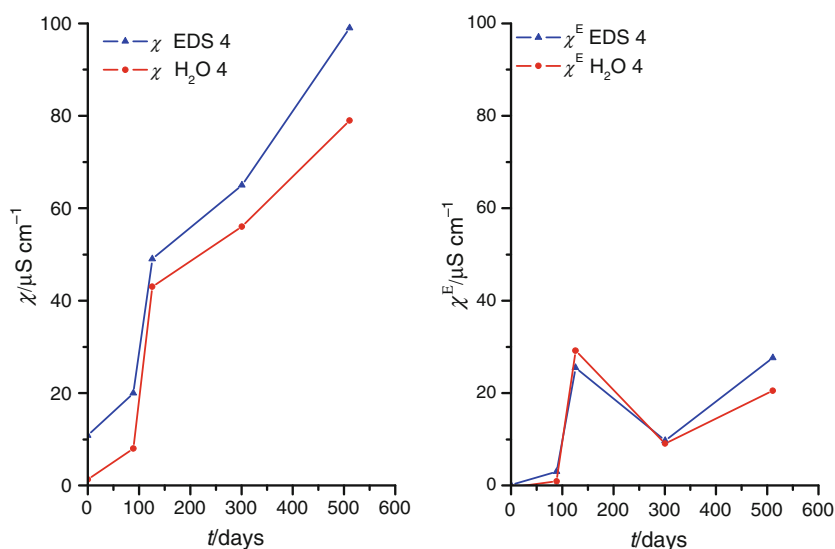


Fig. 11 Specific electrical conductivity of EDS (nanotubes) and distilled H₂O at 511 days

from, or re-approach the equilibrium condition, dissipating ambient energies of an electromagnetic nature. We are therefore dealing with systems capable of self-organising in response to even slight perturbations, by forming “dissipative structures”. It should further be emphasised that the described phenomena were rendered observable through a simple and non-destructive methodology, electrical conductivity, which is able to detect variations in the supramolecular structure of the water connected with the possible formation of aqueous nanostructures. This is a peculiarity of pure water, of which EDS represent a special case due to the protocol by which they are obtained. Finally, the identification of a time and volume dependency made it possible to obtain experimental measurements with values well above experimental error. A comparable study to this one, but performed over a shorter period of time and

using volumes in the order of tens of mL, would not have found any significant results.

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