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The 'Memory of Water': an almost deciphered enigma. Dissipative structures in extremely dilute aqueous solutions

V Elia^{1,*}, E Napoli¹ and R Germano²

¹*Dipto. di Chimica, Università 'Federico II' di Napoli, Complesso Universitario di Monte S. Angelo, via Cintia, 80126 Napoli, Italy;*

²*PROMETE Srl – INFN Spin off Company, Via Buongiovanni 49, San Giorgio a Cremano, 80046 Napoli, Italy*

In the last decade, we have investigated from the physicochemical point of view, whether water prepared by the procedures of homeopathic medicine (leading inexorably to systems without any molecule different from the solvent) results in water different from the initial water?

The answer, unexpectedly, but strongly supported by many experimental results is positive. We used well-established physicochemical techniques: flux calorimetry, conductometry, pHmetry and galvanic cell electrodes potential. Unexpectedly the physicochemical parameters evolve in time.

The water solvent exhibits large changes in measurable physicochemical properties as a function of its history, the solute previously dissolved, and time. In particular we found evidence of two new phenomena, both totally unpredicted, in homeopathic dilutions: the presence of a maximum in the measured physicochemical parameters vs sample age, and their dependence on the volume in which the dilution is stored. These new experimental results strongly suggest the presence of an extended and 'ordered' dynamics involving liquid water molecules. *Homeopathy* (2007) 96, 163–169.

Keywords: homeopathy; calorimetry; conductivity; pH; dissipative structures

Introduction

The 'Memory of Water' is a journalistic expression, first used in the French newspaper *Le Monde*, after the publication in 1988 of Jacques Benveniste's famous paper in the international scientific journal *Nature*.¹ In this paper he claimed, with biological experimental data, that 'homeopathic dilutions' of substances (ie so much diluted as to not contain any molecules of the substance initially diluted in it) are able to induce biological effects typical of the substance initially dissolved in it. The 'memory of water' is a synthesis of a still unexplained phenomenon. Recent scientific publications suggest

some possible ways to experimentally validate the reality of a whole new class of physicochemical new phenomena concerning liquid water.²

It seems that it really is possible to obtain physicochemical information depending on the recent or remote 'history' of a water sample (in Prigogine's terminology: breaking of the temporal symmetry), almost as in the better known case of magnetic materials (Prigogine would say: breaking of the spatial symmetry).³ The so-called memory of water, is connected to the capacity of this kind of solvent, a multi-variable complex system, to be influenced by very tiny perturbations, such as mechanical or electromagnetic actions, in such a way to move away from the initial equilibrium conditions, and this is increasingly established. The 'memory of water', in this sense, is comprehensible in the framework of the theory of Irreversible Processes Thermodynamics due to the Nobel Laureate for Chemistry (1977), Ilya Prigogine.³

*Correspondence: Vittorio Elia, Dipto. di Chimica, Università 'Federico II' di Napoli, Complesso Universitario di Monte S. Angelo, via Cintia, 80126 Napoli, Italy.

E-mail: elia@chemistry.unina.it

Received 2 April 2007; revised 22 May 2007; accepted 29 May 2007

In the last 10 years,⁴⁻¹³ our research group has investigated this problem from the point of view of the physicochemical properties of water when prepared following the procedures of homeopathic medicine preparation: iterative dilutions (of specific solutes of medical interest) followed by agitation (succussion). This method leads inexorably to systems without any molecule different from the solvent, in our case pure water.

Can the 'new water' thus obtained really be 'different' from the initial one? Answering this question was our challenge. The answer, unexpected but strongly supported by the experimental results, is affirmative. In the meantime, other research groups came to similar conclusions using different experimental models and other methodologies.¹⁴⁻²⁰ We also want to note here Giorgio Piccardi, the founder of the Italian physical-chemistry, and his pioneering work concerning fluctuating chemical reactions.²¹⁻²⁴ A critical mass of experimental data² necessary to evidence a new class of physicochemical phenomena of the water has now been reached.

Methods

The experimental methodologies used for our investigations were chosen as the most efficient among the many tested. We list them, without entering into the technical details, but emphasising that they are well-established physicochemical methodologies: flux calorimetry, conductometry, pHmetry and galvanic cell electrode potential. The greatest difficulty of the preliminary work, which lasted for many years, was the selection of the most enlightening experimental methodologies and the establishment of optimal experimental conditions. It was also difficult to evaluate the contribution of the impurities released by the glassware, to the measured experimental values. In fact, the problem of impurities has been the principal objection probably due to the strong prejudice against the possibility that the procedures followed might really change the physicochemical nature of water.

Figures 1-3 show that the presence of impurities released by the glassware makes a significant contribution to the physicochemical state of the dilutions, but it is not relevant in comparison with the unexpected contribution (much higher than the range of the experimental errors) of the auto-organisation process of the water molecules—the water is far from the thermodynamic equilibrium (see below) —and this auto-organization is triggered by external perturbations (such as iterative dilution and succussion).

Figures 1-3 show: (i) how the contributions of impurity were taken into account; (ii) the major contribution of 'something' different from any possible substance of chemical origin. This is a preliminary result but it cleared misunderstandings from the experimental methodologies and allowed us to proceed

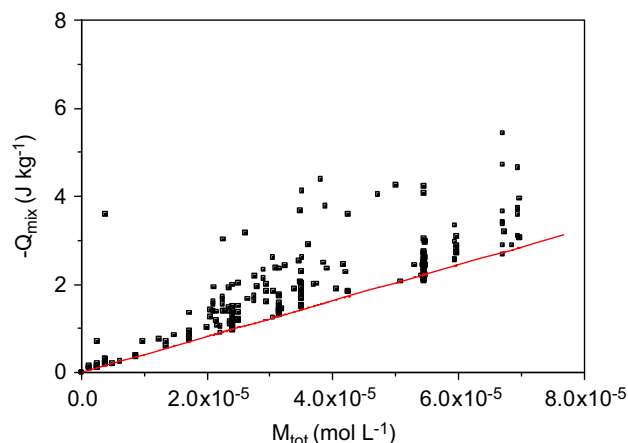


Figure 1 Heat of mixing, Q_{mix} , vs concentration (determined by analytic methods) of impurities, M_{tot} , released by the glass vessels. Black symbols: heat of mixing of homeopathic solutions with sodium hydroxide, NaOH, 0.01 M (mol kg^{-1}); red line: heat of mixing of aqueous solutions containing the same amount of impurities determined in the homeopathic solutions. The absolute values of the heat of mixing with sodium hydroxide using homeopathic solutions are always higher than the corresponding heat of mixing determined only by the 'chemical' contribution originating from the glassware.

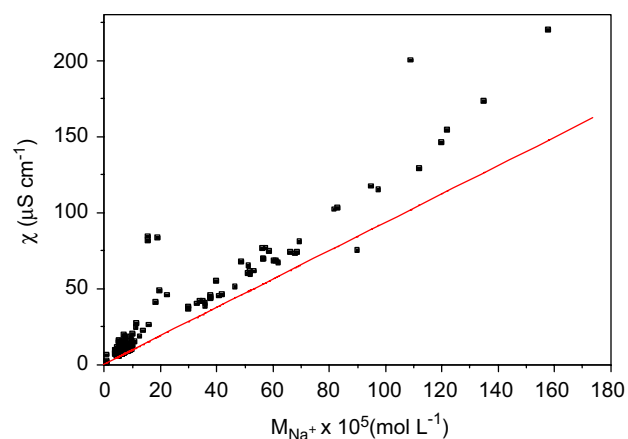


Figure 2 Specific conductivity, χ , vs concentration (determined by analytic methods) of impurities, M_{Na^+} , released by the glass vessels. Black symbols: specific conductivity of the homeopathic dilutions; red line: specific conductivity of aqueous solution containing only the same amount of impurities determined in the homeopathic solutions.

to collect further information and insights on the nature of the 'homeopathic dilutions'.⁴⁻¹³

It is important to emphasise that, from the studies so far conducted, we cannot derive reproducible information concerning the influence of the different degrees of homeopathic dilution or the nature of the active principle (solute) on the measured physicochemical parameters. For this reason the experimental data reported in the figures are not given in terms of homeopathic dilutions or name of homeopathic medicine.

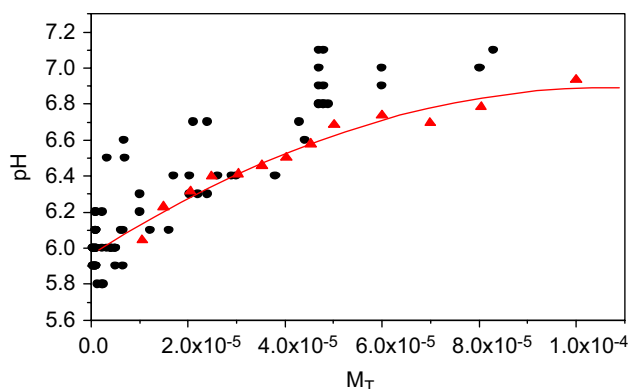


Figure 3 pH values vs concentration of impurities M_T (determined by analytic methods), released by the glassware. Circle symbols: pH of homeopathic dilutions; triangle symbols: pH of aqueous solution containing the same amount of impurities determined by analytic methods in the homeopathic solutions.

Ageing effects

A stimulating, and somewhat serendipitous, result very important in understanding the complex system under study, was that the physicochemical properties of the homeopathic solutions depend on time. The fact that the numerous experiences were performed over many years, naturally introduced the time parameter. The analysis of the experimental results vs the 'arrow of time' was of unexpected relevance,⁷⁻¹⁰ and led to the idea that the system under observation (homeopathic solution) is a closed system (able to exchange only energy with the external environment), far from thermodynamic equilibrium, which allows structures with a local order higher than the water around them to emerge from chaos ('dissipative structures').

Figures 4 and 5 show that, unexpectedly, the investigated physicochemical parameter increases with time. In other words, ageing modifies the physicochemical nature of homeopathic solutions.

What is the interpretation of this newly-observed characteristic of the homeopathic solutions? Are we simply observing a system seeking an energetic minimum and a new equilibrium in a slow kinetic process or this is something totally different? From the data in Figures 4 and 5 we deduce that the temporal variations of the reported parameters are very slow, because it takes many months to evidence them unambiguously. However, this temporal behaviour does not match the idea of a simple slow kinetic. In fact, following the *reductio ad absurdum*, if there exists an energetic minimum towards which the system could move, it would be impossible that in the time that water has existed, it has not reached this hypothetical minimum. Moreover for both parameters (specific conductivity and heat of mixing with alkaline solutions) an increase with time is observed. The correlation between the specific conductivity and the heat of mixing with alkaline solutions, shown in Figure 6 is linear; in other words, these two parameters have the

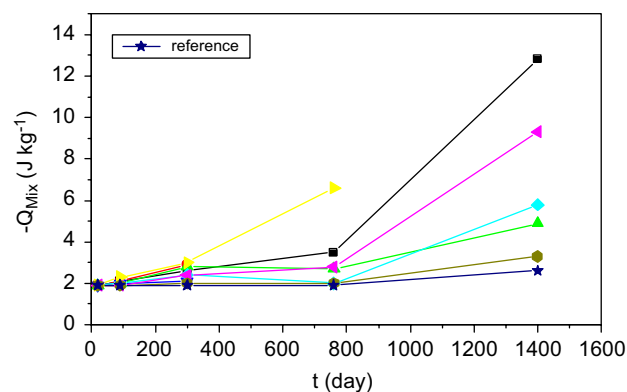


Figure 4 Heat of mixing, Q_{mix} , vs the samples age, t , for six homeopathic solutions.

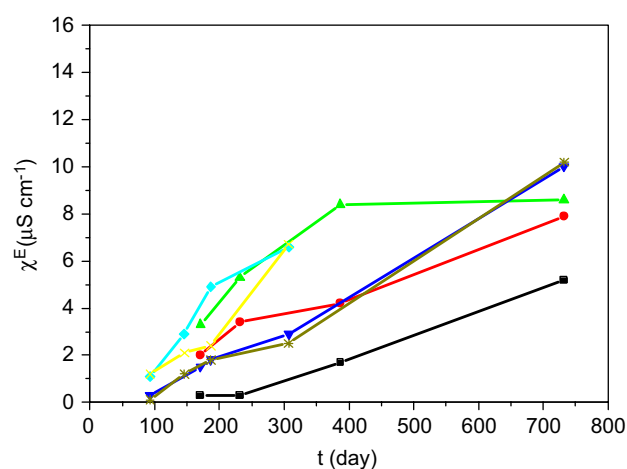


Figure 5 Excess specific conductivity, χ^E ($\mu\text{S cm}^{-1}$) (defined as the experimental difference between the experimental χ value and the contribution to this parameter by the presence of impurities (χ^{chem})) vs the samples age, t , for six homeopathic solutions. Each studied sample has its own peculiar χ^E vs time evolution but with overall similar behaviour: an increment of χ^E in time.

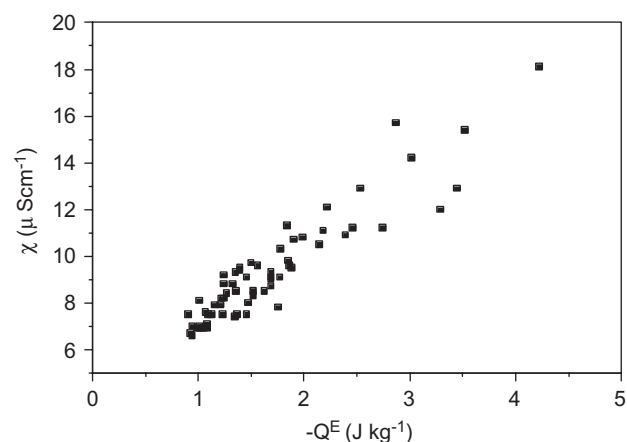


Figure 6 Specific conductivity, χ , vs excess heat of mixing, Q_{mix}^E (the difference between the experimental Q_{mix} value and the contribution to this parameter due to the presence of impurities), for a given volume of homeopathic solution.

same underlying cause. This result provoked us to investigate the nature of the mechanism able to increase simultaneously the electrical conductivity and the heat of mixing with alkaline solutions, after repeated dilutions and succussions. The descriptive model proposed below, although simple, is in agreement with the experimentally observed ageing effect.

An explanation of the electrical conductivity increasing after the preparation procedure and ageing may be based on the so-called 'hopping mechanism', proposed by C.J.T. Grotthuss (1806)²⁵ to explain the much higher mobility (about 5 times) of H^+ and OH^- ions (always present in liquid water) in comparison with other ions of comparable ionic radius. If H_2O molecular clusters are present in the solution, bonded by hydrogen bonds, the hydrogen ions H^+ colliding them experience the 'hopping' phenomenon (Figure 7): the water molecules catch an H^+ ion at one end of the cluster (for the sake of simplicity considered linear) and release instantaneously another H^+ ion at the other end of the cluster. The drift velocity under an electrical potential gradient (a measure of the conductivity) is much increased in comparison with that of ions which do not encounter H_2O molecular clusters. The greater the number of the clusters and/or their length, the higher the conductivity value. The correlation between the electrical conductivity and the heat of mixing with alkaline solutions is a consequence of H_2O clusters breaking, due to the pH variation (see Figure 8).

The greater the number of the clusters and the larger their dimensions, the more is the measured thermal effect (Figure 8). These two experimental phenomena witness the same thing, both are sensitive to the number and/or dimensions of the clusters.

Let us return to the question: Are we measuring the presence of stable clusters seeking an energetic minimum? Or of unstable clusters consisting of dissipative auto-organised structures that are far from equilibrium

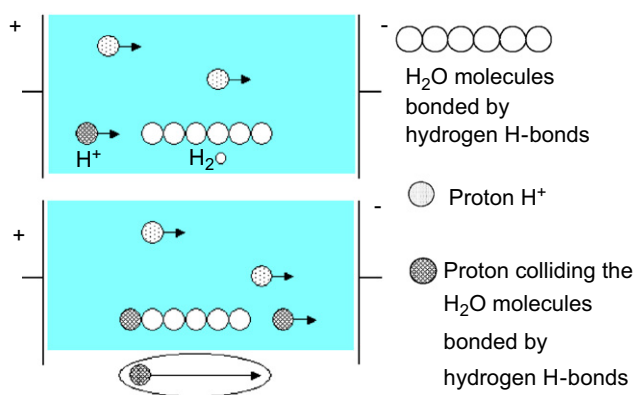


Figure 7 Schematic representation of the Grotthuss hypothesis of the proton (H^+) hopping mechanism to explain the much higher mobility (defined as the ionic drift velocity under a unitary gradient of electrical potential Vcm^{-1}) of H^+ and OH^- ions in water ($H_2O = H^+ + OH^-$) in comparison with other ions of comparable ionic radius.

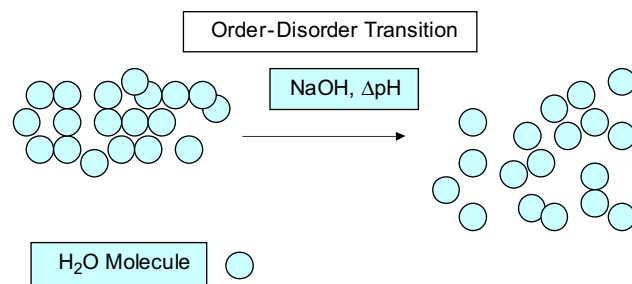


Figure 8 Schematic representation of the phenomenon of molecular clusters breaking, due to pH variation during the experimental procedure of determining the heat of mixing with hydroxide solutions ($NaOH$ $0.01 M$ ($molkg^{-1}$)) in a calorimetric cell. The experimental procedure consists of mixing a homeopathic dilution (that we suppose richer in H_2O molecular clusters than the 'standard' water solvent) with an alkaline solution. The pH variation seems to reflect breaking of hydrogen-bonded H_2O clusters, determining a transition order \rightarrow disorder. This is experimentally evidenced by the increased heat of mixing compared to 'normal' water containing few molecular clusters.

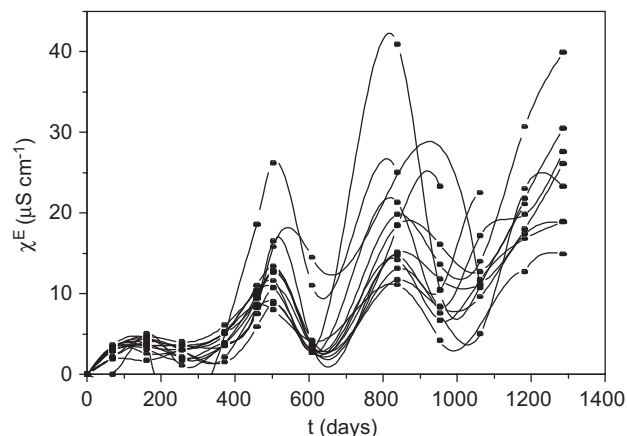


Figure 9 Specific excess conductivity, χ^E , vs the samples ageing, t . Each curve describes the temporal evolution of Arnica Montana (AM) samples in homeopathic dilutions prepared from the same mother tincture. There is no specific correlation between the χ^E behaviour and the degree of dilution (CH) of the samples.

and which remain or move away from equilibrium as a function of their ability to exchange energy with the external environment? We have already emphasised that the hypothesis of systems slowly evolving towards new equilibrium states is not compatible with our experimental findings. In particular, the hypothesis of systems evolving towards a minimum, even very slowly, contrasts with two new and very unexpected experimental phenomena characterising homeopathic dilutions:

- (a) the presence of a maximum in the physicochemical parameters with sample age (Figure 9);
- (b) the dependence of the physicochemical parameters (apart from age) also on the volume in which the homeopathic dilution is stored (Figure 10).

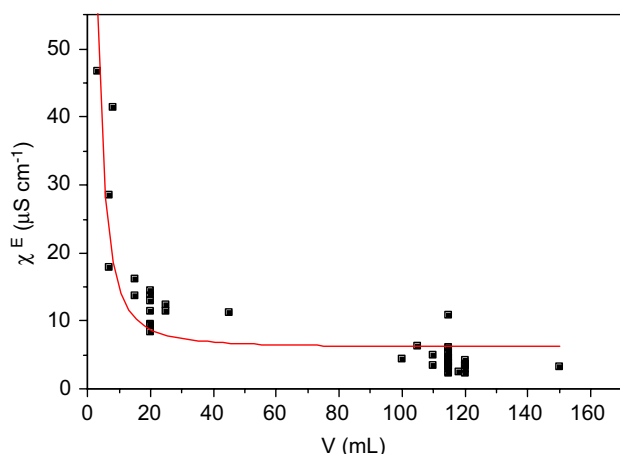


Figure 10 Excess specific conductivity, χ^E , vs ageing volume, V . Each point represents the value of χ^E for each single dilution, experimentally determined at the same age. There is a very strong variation of the parameter, about one order of magnitude, for the systems aged in very small volumes. This volume dependence cannot be explained in the frame of the classical physico-chemistry.

Phenomenon (b) is absolutely anomalous and inexplicable in the current paradigm,¹³ it appears to be in sharp contradiction with the classical concept that an intensive physical quantity cannot depend on the volume.

The temporal evolution of the excess specific conductivity of four sample systems is shown in Figure 11. The samples were obtained as follows: a highly diluted aqueous system was divided into three smaller volumes at a certain 'age'. As the figure shows, the excess specific conductivity (χ^E) behaviour across time of small volume samples is very different from that of larger volume samples. The larger volume sample does not display relevant modifications across time, while each new system of smaller volume evolves in a different way, with an overall common behaviour characterised by the presence of a maximum. This means that the evolution over time depends on the initial state (in this case: large or small volume), in a sense the systems have a 'memory' of the initial conditions.

Another example of such 'memory' of the system is apparent in the experimental data displayed in Figure 12, which shows the temporal evolution of the excess specific conductivity for samples made from the same mother tincture diluted in double distilled water without succussion, in different dilution ratios.¹² Again, the system's evolution in time is strongly conditioned by the initial conditions, with temporal variations characterised by very different maximum and slope values: past history influences the evolution of the 'pure water' system.

The apparent contradiction between the concept of intensive quantity, such as specific conductivity and heat of mixing, and the experimental evidence of dependence on volume may be solved by considering

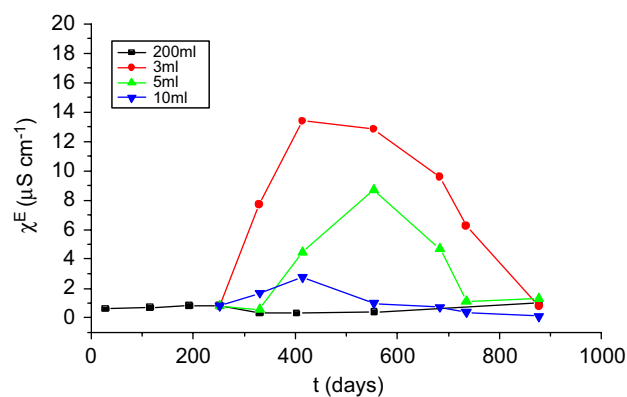


Figure 11 Excess specific conductivity, χ^E , vs sample age, t . In this experiment, a homeopathic dilution of Arnica Montana was left to age for about 250 days in a volume of about 200 ml. At this time point 18 ml were removed and divided into three different vessels of equal shape, containing 10, 5 and 3 ml. The four obtained samples, 182, 10, 5 and 3 ml, were studied vs time. Their temporal evolution was dramatically influenced by the perturbation induced by the repartition into smaller volumes. In particular the higher volume of 182 ml did not experience particular temporal variations, while in the case of the smaller volumes, a large temporal evolution was observed, depending strongly on the starting point.

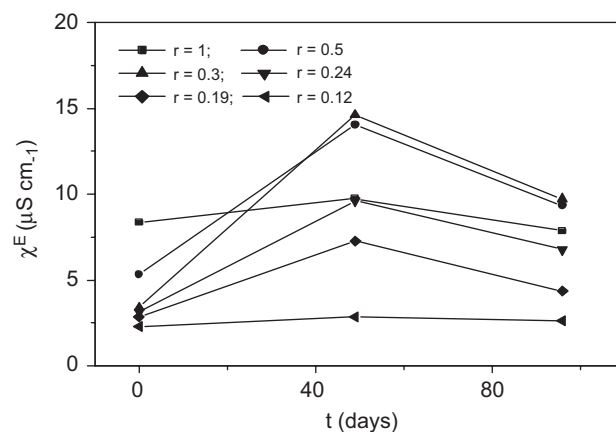


Figure 12 Excess specific conductivity, χ^E , vs age, t , of samples obtained by a simple dilution of the 'mother tincture' with double distilled water without succussion, in different dilution ratios (r). The volumes of the studied solutions were the same. The temporal evolution of the various systems, perturbed only by the simple dilution without succussion, is strongly dependent by the new starting state. In particular, the system with dilution 1:1, $r = 0.5$ (final volume is twice the initial one) exhibits an initial χ^E value markedly lower than the solution from which it was obtained, then, in about 45 days, exhibiting χ^E values much higher with respect to the 'mother tincture', reaching a sharp maximum. In this case, the applied perturbation, determines a strongly different starting point, as well as different temporal evolution.

that, within the solutions there are molecular clusters consisting of water molecules connected by hydrogen-bonds, in far from equilibrium conditions. They can remain in, or move away, from their unstable equilibrium state, dissipating energy derived from the

external environment: they are 'dissipative structures' as described by Prigogine.³

The spontaneous formation of molecular clusters in water is foreseen by the Coherent Quantum Electrodynamics (Coherent QED) without introducing the existence of hydrogen-bonds. This theoretical formulation, due to G. Preparata, E. Del Giudice, *et al* predicts the physicochemical properties of the water,^{2,26–29} much better than other theories. The introduction of the 'arrow of time' into this theoretical framework should yield very interesting results.

Conclusion

We propose a simplified empirical model that in principle seems able to explain the unexpected dependence of the physicochemical parameters on the volumes used.

A first hypothesis to explain the experimental results is to suppose that the solutions, after strong agitation (succussion), enter a far from equilibrium state, remaining there or getting even farther by dissipating energy in the form and amount necessary to stay in the far from equilibrium state. Then, assuming that radiant energy is exchanged, we can further suppose that, for a given flux of dissipated energy ($W\text{ cm}^{-2}$), the same number of dissipative structures would be formed, even if contained in different volumes. In this frame, on average, at any given age, small volumes of water will contain a higher 'concentration' of dissipative structures in comparison with larger volumes (Figure 13). The physicochemical parameters electrical conductivity and heat of mixing are in fact functions of the number, size and shape of the dissipative structures.

We conclude the following:

- the parameters whose values results 'in excess' (in general: variable with the history of the solvent in time) are correlated with the dynamics of supermolecular (mesoscopic) structures in the water solvent;
- the temporal evolution of the parameters is not connected to the tendency to seek an energetic minimum;
- an empirical interpretation, consistent with all current experimental data, is based on the presence of dissipative structures.

Succussion may be the trigger for the spontaneous formation of dissipative structures, that is the emergence of new dynamics. The temporal evolution may be connected to the variation of the number, dimension or the shape of the dissipative structures. It is well known, in Thermodynamics of Irreversible Processes, that the temporal evolution of the systems depends on the initial conditions and on the way the systems evolve.

Much new experimental data converge towards the validation of the statement that water, at least in the context of the procedure of the homeopathic medicine production, really has a 'memory'. That is to say: the

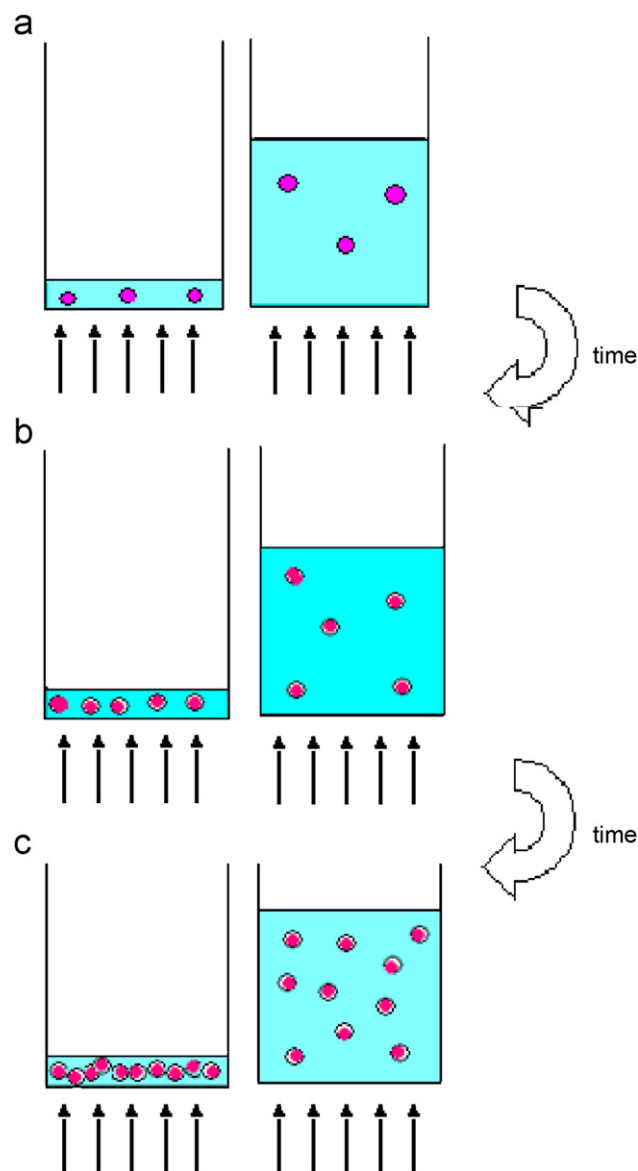


Figure 13 Schematic representation of a possible temporal evolution of a homeopathic solution, showing a variation of intensive quantities such as χ^E ($\mu\text{S cm}^{-1}$) and Q_{mix}^E (J kg^{-1}) vs the storage volume. At the time zero, the two vessels, of equal shape and volume, contain two identical homeopathic dilutions (same age, same active principle, same CH dilution) with no experimentally measurable effects determined by dissipative structures, because of their small number (Figure 13a). Assuming the same formation efficiency (and/or increase in size) for the dissipative structures in the two systems (small volume and large volume), with the same conditions of energetic flux, the number and/or size of the dissipative structures is almost the same in the two containers, at any given time (Figure 13b and c). So, when dissipative structures are numerically increasing, their concentration is much higher in the small volume than in the large one. Consequently, intensive quantities such as those measured, χ^E ($\mu\text{S cm}^{-1}$) and Q_{mix}^E (J kg^{-1}), sensitive to the structure concentration, will show a temporal behaviour dependent on the volume.

water solvent shows experimentally measurable physicochemical properties that vary as a function of the 'lived path', of the solute previously dissolved, and of elapsed time.

Without doubt liquid water has an extended and 'ordered' dynamics involving the whole body of the liquid. It is much more complex than the normal idea of a banal and chaotic cluster of 'molecular balls'.

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