Homeopathy (2007) 96, 183–188
© 2007 The Faculty of Homeopathy
doi:10.1016/j.homp.2007.03.007, available online at http://www.sciencedirect.com

Long term structural effects in water: autothixotropy of water and its hysteresis

Bohumil Vybiral* and Pavel Voráček

Department of Physics, Faculty of Pedagogy, University of Hradec Králové, Rokitanského 62, CZ-500 03 Hradec Králové, Czech Republic

We discovered a previously unknown phenomenon in liquid water, which develops over time when water is left to stand undisturbed, and which made precise gravimetric measurement impossible. We term this property autothixotropy (weak gel-like behaviour developing spontaneously over time) and propose a possible explanation. The results of quantitative measurements, performed by two different methods, are presented. We also report the newly discovered phenomenon of autothixotropy-hysteresis and describe the dependence of autothixotropy on the degree of molecular translative freedom. A very important conclusion is that the presence of very low concentration of salt ions, these phenomena do not occur in deionized water. Salt ions may be the determinative condition for the occurrence of the phenomena. *Homeopathy* (2007) **96**, 183–188.

Keywords: water; autothixotropy; core-ions; deionization; hysteresis

'Autothixotropy' of water

Qualitative laboratory observations

From 1978 to 1986 we performed a series of measurements[1,2] to verify the gravitational law in fluids as deduced by Horák.[3] Originally, in 1978 we observed a peculiar phenomenon in the measurements which compelled us to use another method. A series of experiments focusing on this phenomenon were conducted. In the Department of Physics in University of Hradec Králové, an experimental apparatus was constructed (Figure 1) to observe the phenomenon.[4]

After objects immersed in the water have been at rest for one or more days, seven qualitatively different phenomena are observed, using this method:

(1) When the hanger is rotated by a certain angle, the plate immersed in the water remains in practically the same position, in spite of the twisting tension arising in the thin filament. When a certain critical angle is reached, the plate will rotate, relatively quickly, to a new neutral position determined by the hanger, to the position where the filament is relaxed (ie with no torsion). If the rotation of the hanger is interrupted before the critical angle is reached, a 'creep' toward a new neutral position is observed over some days or weeks. When a smooth-surfaced cylinder, capable of rotating around its own axis, is used instead of the plate, these phenomena are not observed.
(2) Another, weaker phenomenon, is also observed: an immediate rotation of the plate in the direction of the rotation of the hanger; nevertheless, the angle of the immediate rotation is one or two orders of magnitude less than the angle in phenomenon (1).
(3) The critical angle of rotation in phenomenon (1) is dependent on the period of time the water has been at rest. This angle increases with time, starting from virtually zero. (The critical angle can reach values of several tens of degrees)
(4) If the plate is only partially immersed, the critical angle is significantly greater than when it is immersed completely.
(5) In the case of partial immersion, the phenomenon analogous to (2) is much more prominent. Phenomena (4) and (5) are time-dependent as described in (3) despite the time-invariance of the surface tension which we also tested.

---

*Correspondence: Bohumil Vybíral, Department of Physics, Pedagogical Faculty, University of Hradec Králové, Rokitanského 62, CZ-500 03 Hradec Králové, Czech Republic.
E-mail: bohumil.vybiral@uhk.cz

Received 14 March 2007; accepted 27 March 2007

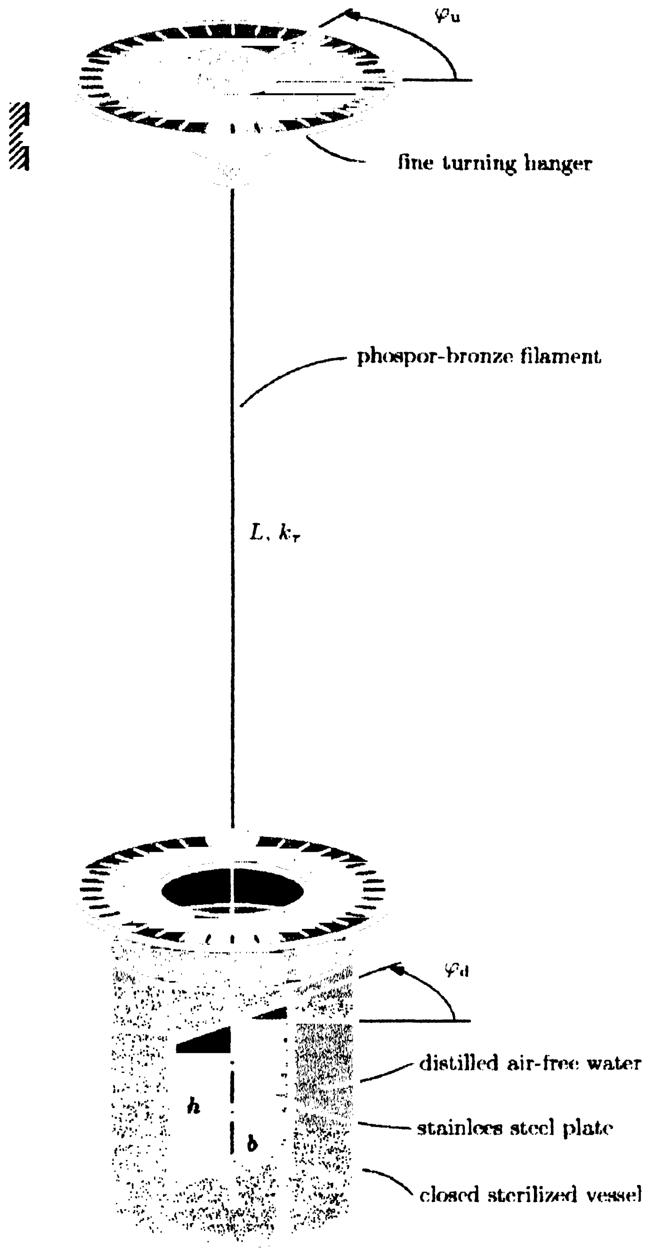

Figure 1 Experimental setup for the static method of measurement.

(6) If the water is stirred after having been at rest for several days, then, when again at rest, the critical angle increases from zero more quickly than when new 'fresh' water is used.

(7) The critical angle is significantly increased and the phenomenon appears earlier if the (distilled) water is boiled (thus substantially deaerated) before the experiment is started.

Our first attempts to carry out quantitative experiments with any acceptable precision were not successful. This was due to a too large dispersion of the measured values; much more sophisticated laboratory equipment than we could acquire, as well as stricter measurement conditions than those we could guarantee, were necessary. In spite of many problems, such experiments have since been performed,[5] and showed that the phenomena did not appear in deionized water. In accordance with the generally accepted terminology, we named this complex of phenomena *autothixotropy* of water.

Proposed explanation

In terms of explanation, a hypothesis based on 'ephemeral polymerisation' of water seems plausible. The existence of such a weak polymerisation was suspected decades ago, both defended and denied by experts. If ephemeral polymerisation of water is the cause of the observed phenomena, it suggests that water molecules are establishing chains or a network; first as minute complexes and thereafter combining successively with one another. The structure then becomes increasingly dense while oscillating at a certain amplitude on a scale of molecules. Such a structure will be relatively fragile, susceptible to differences in the concentration of materials dissolved in water, at different points inside the vessel. Brownian motion can be observed in the case of a conglomerate of molecules having a non-polar character, owing to collisions with molecules of water oscillating in the established network. Weak stirring of 'old' water seems to leave parts of the network intact, making the subsequent 'dipole polymerisation' quicker than it would be in the 'fresh' (ie well stirred) water. Further, the structure has some elasticity. If the water is boiled, no dissolved air (gases) disturb either on the developing process or integrity of the structures; consequently, the phenomenon appears earlier and is more pronounced.

One can expect that the described water structure can be important in biophysics for description and influence on cell characteristics (see eg Pollack[6]). Our observations are consistent with the recently published results of Wernet *et al.*[7]

Autothixotropy and molecular translative freedom

The autothixotropy of water depends, among other things, on the degree of freedom of the translative motion of its molecules. The freedom is limited close to the boundary between the water and some other environment, eg a solid body or the atmosphere over the surface of the water. The freedom of the molecular motion is then limited relatively very deep into the body of the water, perhaps on the scale of several hundred molecular layers or more. The limited degree of freedom, depending on the number of free space-dimensions being less than three, appears as follows:

(1) When the free motion is limited to two space-dimensions ie more or less to a plane, one can find its relevant manifestation in phenomena (4) and (5) described above.

(2) If a thin capillary tube were used, the free molecular transitive motion would be limited in practice to just one dimension. This explains the phenomenon of

polywater, observed decades ago, and claimed to be a sensational discovery, but which soon proved to be false.

(3) When the transitive freedom is limited in all possible directions, ie in all three space-dimensions, the manifestation of the autothixotropy must logically become very prominent and influential. Such a situation occurs in small cellular spaces and possibly significantly influences, or even determines, the rigidity of the cytoskeleton. It is presumed, however, that the cells aresufficiently static in relevance to the autothixotropy.

Salt ions

Currently two diametrically sets different of results supported by serious observations exist concerning the duration of structures in liquid water. According to one[8], molecular clusters in water have a duration of less than one hundred femtoseconds. According to ours, clusters grow to webs on a time scale of days. Since these webs do not arise in deionized water, we believe the purity of the water to be a decisive factor. The distilled water we used was not perfectly pure and could have been significantly contaminated by salt ions, even if only to a very minute degree. From a comparison of experiments with distilled water and deionized distilled water, it is possible to deduce that cores of macroscopic clusters of water molecules are salt ions contained in water.

Moral: If two different observations seem to be mutually incompatible within the frame of an established theory, the most probable explanation is not that one of the observations is wrong, but that the theory is wrong or at least incomplete, and that the observations merely discovered that it was not self-consisrent.

Quantitative experiments on autothixotropy and its hysteresis

Two different, independent strategies were used for quantitative experimental research on the autothixotropy of the water:

1. The static method of torsion.
2. Two dynamic methods: the method of torsion oscillations and the method of small balls falling in water under condition of laminar flow.

The results have been published by Vybiral,[5,9] and are summarised below.

Static torsion method

Principle

A stainless steel plate is suspended on an elastic filament of torsional rigidity k_τ, and immersed in the studied water (Figure 1). The water is in a steady state and the ideal fluid model is assumed. Thus, if we twist the upper end of the filament by angle ϕ_u, we expect that the plate will follow the rotation, so that $\phi_d = \phi_u$, ϕ_d being angle of rotation of the plate. According to our experiments, this equality was not achieved. In the static experiment, a series of increasing values of angle ϕ_d is observed, following a very slow, 'step by step', change of angle ϕ_u. One can specify the moment of force M_w, arising when the plate influences the water: $M_w = k_\tau (\phi_u - \phi_d)$. If angle ϕ_u reaches a *critical value* $(\phi_u)_{crit.}$, the rotation of the plate (ie ϕ_d) becomes quick.

Experimental device

The equipment that was used for the experiment is illustrated in Figure 1. The phosphor–bronze filament had a length $L = 465$ mm and a cross-section of 0.20×0.025 mm^2. The torsional rigidity of the filament was determined experimentally from torsion oscillations of the plate hung in non-perturbed air:[5] $k_\tau = (1.01 \pm 0.02) \times 10^{-7}$ Nm/rad. After reduction to the unit length (1 m), we get $k_{\tau 1} = (4.69 \pm 0.07) \times 10^{-8}$ Nm^2/rad. The results shown here are related to an experiment with a flat stainless steel plate of width $b = 38.5$ mm, height $h = 60.5$ mm, thickness 0.50 mm and mass 8.50 g. Angles ϕ_u and ϕ_d were read with an accuracy of ~0.5°. Water used for the experiment was distilled and then boiled for 3 min before the experiment began. In the course of the experiment, the temperature of the water was kept between 24 and 25°C. Water with volume of approx. 350 ml was in a glass vessel with an inner diameter of 80 mm and a height of 110 mm. The vessel was closed with a paper lid with a small opening for the filament. The lid was removed only briefly to read the scale.

Quantitative experimental results

Measurements of the critical angle $(\phi_u)_{crit.}$. The critical angle is the angle ϕ_u at which, when reached by the hanger the plate began to rotate (relatively quickly, in a time scale of tens of seconds) in the same direction. Some prominent results of repeated measurements[5] are:

1. The plate immersed with 65% of its surface in water, which had been standing for seven days: $(\phi_u)_{crit.} = (398 \pm 3)°$.
2. With the water boiled for a short time, but otherwise the same configuration of system (immersion 65%). After cooling (~24°C): $(\phi_u)_{crit.} \approx 30°$, after two days: $(\phi_u)_{crit.} \approx 115°$.
3. With the water boiled, the plate entirely immersed (the upper edge 10 mm below water level), the critical angle measured on the second and third day was $(\phi_u)_{crit.} = (356 \pm 3)°$.
4. The plate immersed only 50%: $(\phi_u)_{crit.} = (343 \pm 8)°$.
5. The influence of plate immersion on the critical angle $(\phi_u)_{crit.}$ is small: for plate immersion in the range 100–23%, the difference is $\Delta(\phi_u)_{crit.} \approx 14\%$.

6. The period of a water-standing influences the magnitude of the critical angle. For example, with immersion of 85% of the surface of the plate and a long period of standing (17 days), we observed $(\phi_u)_{crit.} = 1800°$. As a consequence of the 'rupture' which followed, the plate rotated through the angular interval $\Delta\phi_d = 1430$. With total immersion such a great critical angle was never reached.
7. After stabilization of the position of the plate (ie, $\Delta\phi_d = 1430$), a slow change of angle ϕ_d (*'creep'*) was observed: 4 in 5 min and, another 32°, in the subsequent 70 hours.

Reproducibility. The results for a given configuration of the measurement system have good reproducibility. For example, if the water was boiled and stood for 24 h, with the plate totally immersed in water for 14 days, six measurements of angle ϕ_d were performed. For the same set of angles ϕ_u : 60, 120, and 180°, the measured respective average angles ϕ_d were: (19.6 ± 0.7), (34.4 ± 0.6), (52.3 ± 1.3). When $(\phi_u)_{crit.} = (239 \pm 2)$ was reached, the plate quickly rotated (tens of seconds) and reached a new equilibrium position $(\phi_d)_0 = (198 \pm 2)$.

Hysteresis. Hysteresis means that a system does not instantly follow forces applied to it, but reacts slowly or does not return completely to its original state: its state depends on its history. Measurements for a cyclical change of angle ϕ_u were carried out. The results of three measurements are shown in Figure 2 and 3. Figure 2 shows the results for the plate entirely immersed in the water which was thoroughly stirred 17 h before. While changing angle ϕ_u from the starting equilibrium position $\phi_u = \phi_d = 0$, the change of angle ϕ_d did not follow an ideal straight line $\phi_u = \phi_d$, but the curve O–A. At point A the critical value $(\phi_u)_{crit\,1}$ was reached and then the plate rotated to a new equilibrium position – point B. With decreasing angle ϕ_u, angle ϕ_d changed according to curve B–C, until it reached the second critical value, denoted $(\phi_u)_{crit.2}$, then the plate rotated to another equilibrium position – point D. When angle ϕ_u was decreased again, the position of the plate went through the origin O to the third critical position – point E, with the third critical value, denoted $(\phi_u)_{crit.3}$. Another equilibrium position corresponded to point F and the fourth critical position corresponded to point G, where $(\phi_u)_{crit\,4} \cong (\phi_u)_{crit\,2}$. As the plate rotated further, a fourth equilibrium position point H, approximately identical with point D, was reached. From there, with decreasing angle ϕ_u, the position of the plate followed the previous section H–O and for $\phi_u = 0$ it returned to the original equilibrium position $\phi_d \cong 0°$.

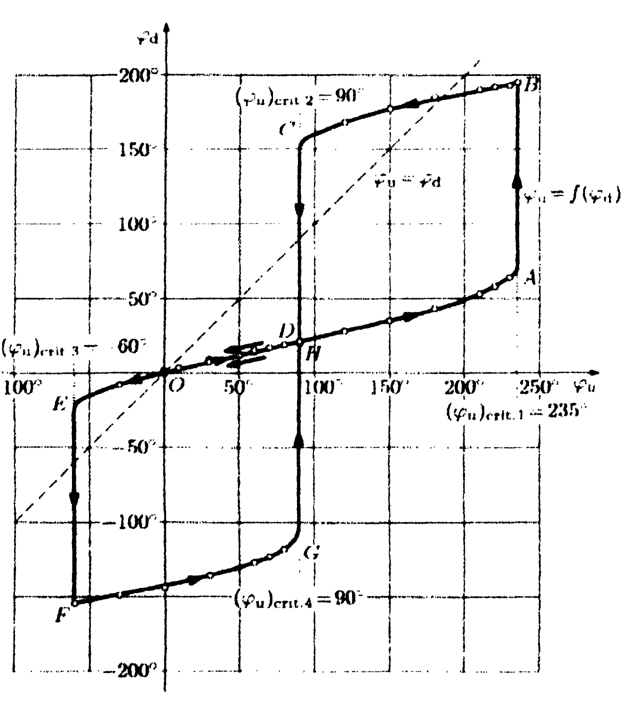

Figure 2 Results of the experiment with the completely immersed plate: loop of the changes of angle $\phi_d = f(\phi_u)$.

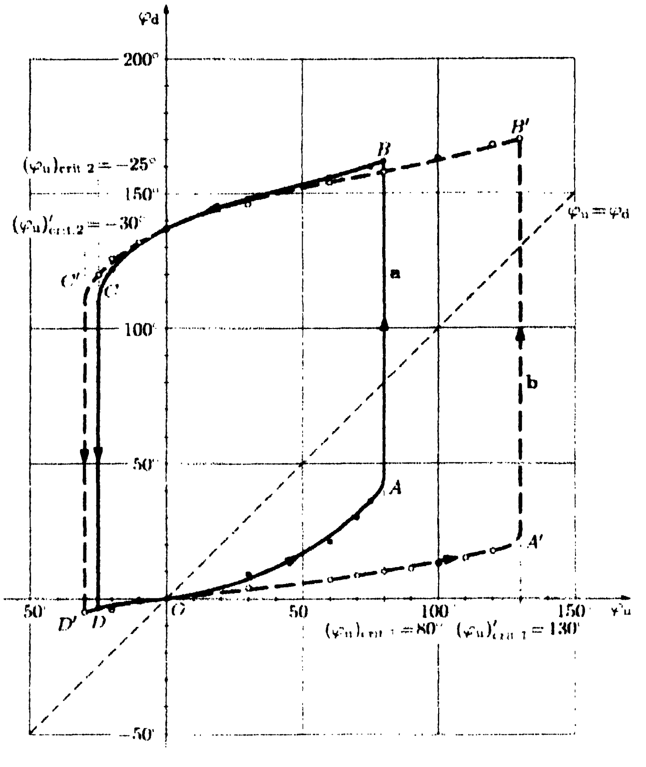

Figure 3 Results of two experiments (loops of the changes of angle $\phi_d = f(\phi_u)$): with the completely immersed plate (loop **a**) and with half-immersed plate (loop **b**).

In Figure 3, the results of two other experiments with water standing for one week are presented: Loop *a* refers to experiment with the plate totally immersed, loop *b* to the experiment with the plate half-immersed; the effect is more pronounced for the half-immersed plate. The loops in Figure 3 are simpler than those in Figure 2 and the respective values $(\phi_u)_{crit}$ are lower. This can be explained on the microscopic level: The plate probably deformed clusters of water molecules of various dimensions and rigidity.

These experiments suggest that the mechanical properties of clusters of water molecules display hysteresis. The hysteresis is however limited; in our experiment, for instance it does not appear in situations when the critical angle is not reached. For example, if a position of the plate corresponds to a point in the section O–A of the graph in Figure 2, before point A is reached, and if we begin to decrease angle ϕ_u, the character of the change of angle ϕ_d will follow the same curve O–A backwards. In these situations, the cluster seems to behave like an ideal elastic body. The dynamics of the phenomenon are similar to those of synovial fluid lubricating the joints of section, which is determined by the thixotropy of the hyaluronic acid present.

Additional measurements. During the experiment, some additional measurements were made to eliminate possible influences on the observed phenomena:

1. The pH of the sample of water was determined by potentiometric measurement. It did not change significantly over a long period; in the range of temperature from 24 to 25°C the pH moved in the range 7.1–6.9.
2. The electrical conductivity of entirely fresh water was 5.6 μS/cm, and after five weeks it increased to 30.5 μS/cm at 25°C. A dependence of the observed water properties on this change was not noted.
3. *Surface tension*: Using a Du-Noüyho apparatus (with an accuracy of ~1%), no measurable change of the surface tension was found.

Experiments with deionized water. In the second phase of these experiments, water, which was first distilled and then deionized, was used. These experiments showed that in deionized water the phenomenon of autothixotropy and its hysteresis was absent. The same equipment (Figure 1) was used for the experiment and the plate was immersed both to one half and entirely as well. The water stood for 10 days before the measurement. The rotation angle ϕ_d of the plate, which passed through the interval $\phi_d \in (0°, 360°, 0°)$, was equal to angle of torsion ϕ_u of the upper end of the filament, with accuracy of ~1.5°, as evaluated from the repeated measurements. Neither the existence of critical angles $(\phi_u)_{crit.}$, nor the phenomenon of hysteresis, were found. From this experiment, we arrived at the important conclusion that the autothixotropy of water, characterized by a nonzero critical angle and hysteresis is caused by the presence of ions in the water.[5]

Method of torsion oscillation

Principle

A plate hangs on a filament (with torsional rigidity k_τ) with their axes of symmetry aligned. The moment of inertia of the plate, relative to its axis, is I. We immerse the plate in the water (Figure 1) and measured its torsion oscillations in two situations:[5]

- In 'fresh' water (ie with negligible autothixotropy), under assumption of a viscous damping of the water, the period of free damped oscillations is T_1.
- In 'stood' water (ie with autothixotropy and viscous damping of the water), we suppose that it is necessary to add, to the quantities related to the elasticity of the filament with torsional rigidity k_τ, the elasticity parameter of putative clusters of water molecules in the considered situation, represented by torsional rigidity k_w. Then period of free damped oscillations is T_2.

By measuring the periods of oscillation T_1 and T_2, we can determine the moment of inertia I (eg, from the plate dimensions and its mass), and calculate the equivalent torsional rigidity:

$$k_w = 4\pi^2 I\left(\frac{1}{T_2^2} - \frac{1}{T_1^2}\right).$$

Quantitative experimental results

For the measurement, an aluminium plate with a thickness of 2.95 mm, width $b = (47.59 \pm 0.03)$ mm, height $h = (50.59 \pm 0.02)$ mm and mass 18.70 g, was used. Its moment of inertia was calculated from its dimensions and mass: $I = (7.518 \pm 0.001) \times 10^{-6}\,\mathrm{kg\,m^2}$. The plate was hung along its longitudinal axis of symmetry on a phosphor–bronze filament of cross-section of $0.025 \times 0.2\,\mathrm{mm^2}$ and length of $L = 569$ mm. The filament had a torsional rigidity $k_\tau = (8.25 \pm 0.12) \times 10^{-8}$ Nm/rad.[5]

The plate was immersed in distilled and boiled water so that the upper edge of the plate was 14 mm above the level of the water surface. The water with a volume of approximately 400 ml was in a glass vessel with an inner diameter 80 mm and height 110 mm; the experiment was carried out at a temperature of 23°C. The period of the damped torsion oscillations was measured three times.

First in fresh water. The period of oscillation was $T_1 = (101.7 \pm 1.2)$ s. Then the system was left at rest for seven days. Then plate was carefully rotated from this equilibrium position by ~45°, and at that position it stayed. Then, the plate was given a torsional pulse, initiating damped torsion oscillations. The period of oscillation was measured ten times; resulting in $T_2 = (5.34 \pm 0.06)$ s. The torsional rigidity of this system with autothixotropy was determined to be $k_w = (1.04 \pm 0.03) \times 10^{-5}$ Nm/rad. The degree of the level of autothixotropy of the system, is ascertainable by means of the measurement of critical angle $(\phi_u)_{crit.}$. For our system this was $\approx 340°$.

Conclusions

On this basis, it is possible to formulate some additional hypotheses about clusters of water molecules:

1. Clusters of water molecules may be of macroscopic dimensions, on scale of centimeters.

2. Clusters of water molecules may be destroyed by boiling or intense stirring or shaking.
3. Clusters of water molecules have certain mechanical properties analogous to the properties of solid substances, such as elasticity/rigidity and strength, but these properties are much smaller than for solid substances with a relative magnitude of 10^{-6} or less.
4. Mechanical properties of clusters of water molecules show a certain hysteresis.
5. Water slightly deviates from an ideal Newtonian viscous fluid, because autothixotropy also appears in the form of internal static friction, although very weak.
6. From comparison of experiments with natural distilled water and deionizated distilled water it is possible to deduce that the cause of macroscopic clusters of water molecules are the ions contained in water.

References

1 Vybíral B. Experimental verification of gravitational interaction of bodies immersed in fluids. *Astrophys Space Sci* 1987; **138**: 87–98.

2 Vybíral B. K experimentálnímu ověření gravitační interakce těles ponořených do tekutin. In: *Sborník Pedagogické fakulty*, 54 (*Fyzika*). Praha: SPN, 1989, pp 307–318 (in Czech).

3 Horák Z. Gravitational interaction of bodies immersed in fluids. *Astrophys Space Sci* 1984; **100**: 1–11.

4 Vybíral B, Voráček P. '*Autothixotropy' of Water – an Unknown Physical Phenomenon*. Available via 〈http://arxiv.org/abs/physics/0307046〉; 2003.

5 Vybíral B. The comprehensive experimental research on the autothixotropy of water. In: Pollack G, *et al* (eds). *Water and the Cell*. Dordrecht: Springer, 2006, Chap 15, pp 299–314.

6 Pollack G. *Cells, Gels and the Engines of Life*. Seattle, WA: Exner and Sons Publisher, 2001.

7 Wernet Ph, *et al*. The structure of the first coordination shell in liquid water. *Science* 2004; **304**: 995–999.

8 Cowan ML, *et al*. Ultrafast memory loss and energy redistribution in the hydrogen bond network of liquid H_2O. *Nature* 2005; **434**: 199–200.

9 Vybíral B. Experimental research of the autothixotropy of water. In: *Proceedings of the Conference New Trends in Physics – NTF 2004*, Brno: University of Technology, Czech Republic, pp 131–135.