Autothixotropy of Water and its Possible Importance for the Cytoskeletal Structures

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Abstract. The article deals with newly observed phenomenon in water that was called ‘the autothixotropy of water’. A history of the phenomenon is connected with very fine gravimetric measurements which were made by author in years 1978 till 1986. Autothixotropy is a very weak macroscopic phenomenon that is observed in water that has remained at rest for a certain time (in tens of hours or days). For the experiments distilled water was used, re-boiled before use. The phenomenon is manifested by force (mechanical resistance) to the immersed body in water with a tendency of changing its position. The static and dynamic methods were used for the research. With the static method a moment of force is measured that is necessary for a very distinctive turn of a stainless steel plate hung up on a thin filament and immersed in standing water. With a certain angular torsion of the filament a certain moment of force is achieved when a state of stress reaches a critical value in water which is demonstrated with an expressive changing of angular position of the plate. When the direction of moment of force is changed, it is reflected in the differential rotation of the angle plate (the phenomenon of hysteresis was observed). The phenomenon of autothixotropy expires after a certain time, if water is mechanically mixed or re-boiled. It is significant that the phenomenon of the autothixotropy is not present in deionised distilled water. When using a dynamic method both oscillations of the plate and a very slow fall of a small ball in standing water was observed. The autothixotropy of water can be explained by a hypothesis of a cluster formation by H\textsubscript{2}O molecules in standing water. As the phenomenon of autothixotropy is not present in deionised water, it may be primarily caused by a presence of ions in water, as it was demonstrated in the experiment with a salt solution (NaCl). Biophysical applications of the autothixotropy of water remain open. Some possible applications of the phenomenon in an organization of water in cytoskeletal structures are outlined.

1. Introduction – Phenomenon ‘Autothixotropy of Water’

Phenomenon in water that was called the autothixotropy of water: it is a very weak macroscopic phenomenon that is observed in distilled water that has remained at rest for a certain time (in about tens of hours or days) i.e. ‘standing’ water. For experiments distilled water was used, re-boiled before use. The phenomenon is manifested by very small force (definite mechanical resistance) against a body immersed in water and arising when it should change its position. If standing water is vigorously mixed or re-boiled again, the phenomenon disappears and appears again in a matter of about in tens of minutes or hours (or days). Macroscopically therefore standing water acts as a very weak gel, which has approximately tixotropical properties. Because it arises spontaneously a phenomenon was called ‘the autothixotropy of water’.
2. History of Discovery

2.1. Problems by Gravitational Measurements
The history of discovery of this phenomenon is connected with very fine gravimetric measurements which I made in the years 1978 till 1986. The subject of research was an experimental verification of gravitational interaction of bodies immersed in fluids – theoretically formulated by Z. Horák [1]. As fluids I used distilled water. First experiments that I proved were made in the year 1978 by an application of Cavendish static torsion – balance method (Figure 1a). The experiments were not successful: during the experiments unknown molecular properties of water were shown disturbingly and superimposed the expected gravitation of the phenomena very strongly. After a period longer than about ten hours it was not possible to control the torsion pendulum with a rotation of the filament hangings – molecular forces of unknown origin took effect against small gravitational forces. That’s why in 1986 I decided to use the dynamic Boys method (Figure 1b) which helped to verify the gravitational phenomena in fluids with an accuracy 0.4% [2], [3]. In this case the torsion pendulum was situated in more diluted air and the pendulum oscillated in gravitational field of system of two cast iron spheres immersed in water. Herewith an influence of phenomenon of autohixotropy was excluded.

![Figure 1](image-url)

*Figure 1. Diagram of apparatus for gravitational measurements: a) Apparatus for static measurement of Cavendish method (1978); b) Apparatus for dynamic measurement of Boys method (1986).*

2.2. Qualitative and Quantitative Analyse of Newly Observed Phenomenon in Water
At the recommendation of P. Voráček from University Lund, Sweden, I returned in years 1991 and 1992 to my failed experiments from 1978 and I made series of special experiments leading to an examination of new observed phenomena in water. The results of qualitative analysis were sent in year
1993 for a publication in *Annalen der Physik* (D), *Nature* (GB) and *Science* (USA), but without any positive feedback. Only in 2003 the text was published at http://arxiv.org/abs/physics [4]. The positive acceptance, especially by G. Pollack (USA, WA) and M. Chaplin (GB), brought me to a quantitative measurement of the phenomena in the years 2003 till 2005. The results of this research were published at [5], [6].

3. **Static Torsion Method of Experimental Research of Autothixotropy**

3.1. Principle of the Method and the Apparatus

Principle of the static torsion method is this ([5], [6]): an experimental body was a flat plate made of stainless steel is hanged up on an elastic filament and immersed into the distilled air-free water (photo of apparatus is on Figure 2). During the experiment it comes to a twisting of the upper filament end and a reaction of the hanged up plate is being watched.

![Figure 2. Photo of apparatus for measurements of authotixotropy of water.](image)

**Parameters of apparatus:**

- A flat stainless steel plate with dimensions: width 38.5 mm, height 60.5 mm, thickness 0.50 mm and mass 8.50 g.
- Phosphor-bronze filament with a cross-section of $(0.20 \times 0.025)$ mm$^2$ (as used in the clock industry), length $L = 465$ mm. The torsional rigidity of filament for length $L = 465$ mm was measured with a dynamic method (method of oscillations) $k_\tau = (1.01 \pm 0.02) \times 10^{-7}$ N.m/rad.
- Distilled air-free water: it was re-boiled for about 3 minutes before the experiment its temperature was kept within the values $24^\circ$C – $25^\circ$C. Before beginning experiment the water is in the steady state more than about ten hours or days.
- Water with volume of approx. 400 ml was in a glass vessel (beaker) with an inner diameter of 80 mm and a height of 110 mm. The vessel was closed with a paper lid when not in use.
- Accuracy of measurement of angles $\varphi_u, \varphi_d: \sim 0.5^\circ$. 
3.2. Measurements of the Phenomenon of Autotixotropy of Water

3.2.1. Ideal fluid model of water was assumed first. Then, if we turn the upper end of the filament by angle $\varphi_u$, we expect that the plate will follow the rotation of upper, so that approximately is $^1 \varphi_d = \varphi_u$, where $\varphi_d$ being angle of rotation of the plate (this equality is true for i.e. ‘fresh’ water).

3.2.2. Observations. According to our experiments with ‘standing’ water, equality $\varphi_d = \varphi_u$ was not achieved. In the experiment, a series of increasing values of angle $\varphi_d$ is observed, following a very slow – ‘step by step’ – change of angle $\varphi_u$. We measured function $\varphi_d = f(\varphi_u)$. One can specify the action moment of force $M_w$, arising when the plate influences the water: $M_w = k(\varphi_u - \varphi_d)$. If angle $\varphi_u$ reaches a critical value $(\varphi_u)_\text{crit}$, the rotation of the plate (i.e. $\dot{\varphi}_d$) becomes relatively quick.

3.2.3. Quantitative results. Typically experimental functions $\varphi_d = f(\varphi_u)$ are on Figures 3 and 4, in which there are two examples from many repeatedly measured values [5]. The important points of the graphs are:
- Starting point of equilibrium position $\varphi_u = \varphi_d = 0^\circ$.
- The critical angle $(\varphi_u)_\text{crit}$, i.e. the angle $\varphi_u$ at which – if reached – the plate began (for a few tens of seconds) to rotate in direction of turning of hanger (angle $\varphi_u$), during which time the angle $\varphi_d$ has been changed considerably.

![Figure 3. Loop of hysteresis – results of the experiment with the completely immersed plate: loop of the changes of angle $\varphi_d = f(\varphi_u)$; see [5].](image)

$^1$ See too subsection 5.2 and Figure 5 (the curve $a$).
3.3. Phenomenon of Hysteresis

When the direction of moment of force is changed, it is reflected in the different rotation of the hanged plate - the phenomenon of hysteresis is observed [5]. Measurements for a cyclic change of angle $\varphi_u$ were carried out – see Figures 3 and 4.

In Figure 4 we see the results of two other experiments: Loop $a$ is relevant for the experiment with the plate totally immersed in water, while loop $b$ is related to the experiment with the plate only half-immersed. The effect is clearly more pronounced for the half-immersed plate than for the completely immersed one.

Figure 4 (see loop $a$ for the experiment with the plate totally immersed in water): we starting in the equilibrium position $\varphi_u = \varphi_d = 0^\circ$ and slowly changing angle $\varphi_u$, than the change of angle $\varphi_d$ did not follow an ideal straight line $\varphi_u = \varphi_d$, but the curve $O-A$. At point $A$ the critical value $(\varphi_u)_{\text{crit.1}} = 80^\circ$ was reached and then the plate turned into a new equilibrium position – point $B$. The action moment of force is $M_w = k(\varphi_u - \varphi_d) \approx 7.1 \times 10^{-7}$ N.m. With now decreasing angle $\varphi_u$, than angle $\varphi_d$ changed according to curve $B-C$, until it reached the second critical value, denoted $(\varphi_u)_{\text{crit.2}} = -25^\circ$. Afterwards the plate turned to another equilibrium position – point $D$. When angle $\varphi_u$ was increasing again, the position of the plate went to origin $O$ with the original equilibrium position $\varphi_d \approx 0^\circ$. Herewith the loop of cyclical changes of the angle $\varphi_d$ of a rotation of the plate is closed. The loops in Figure 4 are simpler than those in Figure 3 and the respective values $(\varphi_u)_{\text{crit.}}$ are lower.

![Figure 4. Loops of hysteresis – results of the other two experiments (loops of the changes of angle $\varphi_d = f(\varphi_u)$): with the completely immersed plate (loop $a$) and with the half-immersed plate (loop $b$); see [5].](image-url)
3.4. Additional Measurements
To eliminate other eventual influences on the observed phenomena of autothixotropy, we realized additional measurements [5].

3.4.1. Acidity (pH) of the sample of water was determined by a potentiometric measurement of the pH factor. It did not change considerably during a long period of time (in the range of temperature from 24°C to 25°C the acidity moved in the range 7.1 to 6.9).

3.4.2. Electric conductivity of entirely fresh water was 5.6 μS/cm, and after 5 weeks it increased to 30.5 μS/cm at 25°C. A dependence of the observed water properties on this change was not noted.

3.4.3. Surface tension. With use of a Du-Noüyhy apparatus (with an accuracy of ~1%), no measurable change of the surface tension was found.

4. Dynamic Method of Torsion Oscillation
4.1. Principle of the Method
We have a plate with an axis of symmetry along with the filament (with torsional rigidity $k_\tau$) on which the plate hangs. We immerse the plate into the water (Figure 2) and consider its torsion oscillations in two situations (see [5]):

- In the ‘fresh’ water (i.e., with non-exist or negligible autothixotropy); the period of free damped oscillations is $T_1$.
- In the ‘standing’ water (i.e., with autothixotropy), we suppose that it is necessary to add, to the quantities related to the elasticity of the filament with torsional rigidity $k_\tau$, the elasticity parameter of putative clusters of water molecules in the considered situation, being represented – for the plate which we used – by a respective torsional rigidity $k_w$. The period of damped oscillations is $T_2 < T_1$.

- If we measure the periods of oscillation $T_1$ and $T_2$, we can calculate [5] the equivalent torsional rigidity:

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

where $I$ is the moment of inertia of the plate (calculated from the plate dimensions and mass).

4.2. Quantitative Experimental Results of Oscillations
For the measurement (see [5]), an aluminium plate with a thickness of 2.95 mm, width $b = 47.59$ mm, height $h = 50.59$ mm and mass 18.70 g, was used. Its moment of inertia was calculated from its dimensions and mass: $I = 7.518 \times 10^{-6}$ kg.m$^2$. The plate was hung along its longitudinal axis of symmetry on a phosphor-bronze filament with a cross-section of $(0.025 \times 0.2)$ mm$^2$ and a length of $L = 569$ mm (the filament had a torsional rigidity $k_\tau = 8.25 \times 10^{-8}$ N.m/rad). The plate was immersed in distilled and re-boiled water in such a way that the upper edge of the plate was 14 mm above the level of the water surface. The experiment was carried out at a temperature of 23°C. The period of the damped torsion oscillations was measured three times – the first one 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, so that a high level of autothixotropy could be reached. Afterwards, the plate was carefully rotated from this new equilibrium position by $\phi_d \approx 45^\circ$ and at that position it stayed. Then, the plate was given a torsional pulse, by which damped torsion oscillations were initiated. The period of oscillation was measured ten times, the period was distinctively shorter; resulting $T_2 = (5.34 \pm 0.06)$ s. The equivalent torsional rigidity of this system with autothixotropy was determined to be $k_w = (1.04 \pm 0.03) \times 10^{-5}$ N.m/rad. It is possible to say, that the degree of the level of autothixotropy of the system, is ascertainable by means of the measurement of critical angle $(\phi_u)\text{crit.}$ For our system it was determined to be $(\phi_u)\text{crit.} \approx 340^\circ$ (i.e. the action moment of force $M_w \approx 4.0 \times 10^{-7}$ N.m).
5. Measurements of the Phenomenon by Changed State

5.1. Deionised Distilled Water

In second phase of experiments [5], water, which was first distilled and then deionised, was used. The experiments showed that in deionised water the phenomenon of autothixotropy and its hysteresis was absent. The same equipment (Figure 2) was used for the experiment and the plate was immersed both to one half and entirely as well. The period that water was standing still before the measurement was almost ten days. It was found from the measurements that the rotation angle $\phi_d$ of the plate, which passed thru the interval $\phi_d \in (0^\circ, 360^\circ, 0^\circ)$, was equal to angle of torsion $\phi_u$ of the upper end of the filament. The ideal equation $\phi_d = \phi_u$ was achieved with accuracy of $\sim 1.5^\circ$, as evaluated from the repeated measurements. Neither the existence of critical angles ($\phi_u_{\text{crit}}$), nor the phenomenon of hysteresis, were found. From the performed experiment, we arrived at the important conclusion that the autothixotropy of water, characterized by a non-zero critical angle, and hysteresis, is caused by the presence of ions in the water.

Note: It results from the described experiments with deionised distilled water that in case of an application of deionised distilled water in a gravitational experiment according to Figure 1a, the measurement would be probably successful. This experiment was not made.

5.2. Salt Solution

The observation and newly finding from an experiment in June 2011: During a time period of 10 days deionised distilled water had been observed. The autothixotropy did not appear. Afterwards a salt solution with a concentration of 1 per mille NaCl was prepared from this water (0.400 g salt was dissolved in 400 ml of the examined deionised distilled water). For a period of 7 days no autothixotropy appeared, more to the contrary for angles inequality $\phi_d > \phi_u$ was valid (see the curve a in the Figure 5). Thus the angle $\phi_d$ of turning of plate is bigger than the angle $\phi_u$ of a primal turning of the upper hanging of filament. It is a remarkable finding because the plate shows a certain additional automotoric movement. The measurement shows that for the certain angle $\phi_u$ is the angle of turning $\phi_d$ the same (with an accuracy of $\sim 0.5\%$) both for angles $\phi_u$ going both up and down too; see the two-way curve a in the Figure 5. The described situation is valid until autothixotropy appears.

In the observed case with the salt solution the phenomenon of autothixotropy appeared after 7 days from a preparation of the solution. Afterwards, on the contrary, the measurement results $\phi_d < \phi_u$. After the critical angle $\phi_u = (\phi_u)_{\text{crit},1} = 70^\circ$ is achieved, the angle $\phi_d$ of plate turning starts increasing, even if $\phi_u$ is invariable how it is evident from the Figure 5, loop b. If we keep decreasing of the angle $\phi_u$ from this status (point A), the angle $\phi_d$ of plate turning starts decreasing gradually to the upper part of branch of the loop till the second critical angle $(\phi_u)_{\text{crit},2} = -40^\circ$ is achieved when the angle $\phi_d$ of plate turning decreases spontaneously to negative value of $\phi_d$ in comparison with the initial status (point B). During the angle $\phi_u$ increasing the loop b closes up in the initial position $\phi_d = 0$. A status of hysteresis happened. The status of changes $\phi_d = f(\phi_u)$, described with the curves a and b in Figure 5, were running in a different way.

5.3. Magnetization

For experiments deionised distilled water was used, partly not magnetized, partly strongly magnetized (magnetized induction of degree 1 T). The magnetization did not have any influence on the results – the autothixotropy did not appear.

5.4. Ethyl-alcohol

An inspectional measurement was also made when distilled water was substituted with ethyl-alcohol. But according to expectations the phenomena of the autothixotropy did not appear with the ethyl-alcohol usage.
6. Possible Microscopic Interpretation of the Phenomenon

The autothixotropy of water can be explained by a hypothesis of a cluster formation by H₂O molecules in ‘standing’ water. In the literature about water (collectively see M. Chaplin [7]) we can find various kinds of binding molecules links in a cluster formation on the basis of a hydrogen bridge in case of a water molecule. In case of explication of autothixotropy of water it could deal with a network of icosahedral water super clusters – see [7], [8] and Figure 6. As the phenomenon of autothixotropy is not present in deionised water, it may be primarily caused by a presence of ions in water. This hypothesis I was reached by me in 2005 based on the experiments made that time [5]: “From comparison of experiments with natural and deionised distilled water it is possible to deduce that kernels of macroscopic clusters of water molecules are the ions contained in water”. In case of the experiment with salt solution NaCl dissociation happens when molecule NaCl breaks up to ions Na⁺, Cl⁻ around which the clusters of molecule H₂O are formed.

Figure 6. Diagram of a super cluster in water (see [7] and [8]). Thirteen complete but overlapping icosahedral clusters form this super-icosahedral structure containing 1820 water molecules (an outer shell of an additional 360 water molecules is shown).
During the experiment with a rotating hanged up plate immersed in water, mechanical characters of a structure of molecules bound in super clusters are probably shown (its elasticity and strength are demonstrated).

7. Possible Importance of the Phenomenon for the Cytoskeletal Structures
Biophysical applications of the autothixotropy of water remain open, but it is expected that the described water structure can be important in biophysics influence on cell characteristics (see eg Pollack [9]). Our observations are consistent with the published results of Wernet et al. [10]. Some possible applications of the phenomenon in an organization of water in cytoskeletal structures are outlined.

We thought about this problem in previous work [11] and found the following conclusion: “A manifestation of the autothixotropy of water depends – among others – on the degree of freedom of the transitive motion of its molecules. When the transitive freedom is limited in all possible directions, i.e. in all three space-dimensions, the manifestation of the autothixotropy must – quite 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 of living cell”.

8. Conclusions
The experiments with ‘standing’ distilled water and salt solution proved that after certain time of standing (approximately in about tens of hours or in days) a status of autothixotropy of water is achieved.

- A state of autothixotropy of water is macroscopically demonstrated with very weak mechanical properties analogous to the properties of solid substances, such as a certain internal static friction, elasticity and strength. It was shown by means of the experiments with a plate hanged up on an elastic filament and immersed into the exploring water. During a twisting of upper filament end the plate starts rotating progressively when a certain critical angle has been reached. Then it starts spinning and reaches a new relatively equilibrium position.
- During a cycling rotation of the plate (i.e. forth and back) a phenomenon of hysteresis was found. It is manifested with a delayed reaction on the filament twisting.
- Water slightly deviates from an ideal Newtonian viscous fluid, because the autothixotropy appears in the form of a certain internal static friction, although very weak.
- Experimentally it has been found out that a phenomenon of autothixotropy does not originate in deionised distilled water.
- There was proposed a hypothesis that the existence of autothixotropy is caused by super clusters that are created in standing water after a certain time and evidently fill up the whole vessel with the exploring water. The kernels of these macroscopic clusters can be ions present in water, as it was demonstrated in the experiment with a salt solution (NaCl).
- The super clusters are affected with stirring and shaking of water and are broken with re-boiling over.
- The time interval during which the autothixotropy arises (in case of fresh water or solution or after its re-boiling) has not been definitely determined yet. Obviously it depends on the structure and amount of ions contained in water.
- A hypothesis was expressed that the autothixotropy can have a considerable significance for biophysics because with the aid of autothixotropy the rigidity of cytoskeleton of living cell could be explained.

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