

Physicochemical Properties Inherent in Thaw Water

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Abstract

A comparative analysis was carried out for some physicochemical parameters of initial and thaw water obtained as the result of freezing-unfreezing procedure: concentration of organic, inorganic impurities, the particles of disperse phase, as well as of heavy isotopes of water, hydrogen peroxide, dissolved gases such as oxygen and carbonic gas. It has been shown that according to these parameters thaw water differs from initial water and these differences are both of permanent and temporal character.

Key words: thaw water, medical properties, recrystallization, purity of water, isotope composition, hydrogen peroxide concentration, oxygen, carbonic gas

INTRODUCTION

Academician V. V. Voevodsky in his time attached a great significance to the studies on physicochemical properties of water since he realized that this information should promote solving numerous fundamental problems in the fields of physics, chemistry, biology and medicine. It was no casual that a spectroscopic group for studying the properties of water (where the author of the present paper worked being a student) has been organized in his laboratory.

The phenomenon of thaw water was known in the beginning of 60ths of the last century. The term “thaw water” (except as otherwise noted) means water formed as the result of melting of the ice prepared by freezing ~50 % of initial water volume. The phenomenon of thaw water is meant as a combination of particular physicochemical properties of water due to those the thaw water exhibits a stronger effect on the growth of plants, breeding performance of animals, healing properties of medical preparations, *etc.* as compared to usual water [1–3]. In all the communications devoted to thaw water a question is discussed concerning the period of time when non-standard properties of thaw water persist, *i.e.* its “memory”.

The lifetime parameter of thaw water is one of the central questions in the discussion be-

tween the supporters and opponents of the existence of the thaw water phenomenon. In the academic literature, only several works are devoted to the discussion of the phenomenon of thaw water «memory». So, the authors of [4] have established that the value of dielectric constant (Σ) for freshly thawed water in the course of time becomes close to that for initial water. However, the conditions of the experiments they carried out allowed one to ambiguously interpret the data obtained. In this connection, to consider these data as an argument in the dispute on special properties inherent in thaw water from the point of contemporary knowledge is quite impossible. Nevertheless, according to [4], the lifetime value for thaw water amounts to 750 s. In [5] data were presented carried out for measuring the lifetime of thaw water and initial (reference) water films. It was revealed, that the lifetime value a thaw water film is several times higher than that for initial water. However, this effect disappeared in 24 h. In fact, these data on the lifetime of thaw water exhaust all the information available in the literature of 1960–1970th.

Last time the quality of potable water has considerably worsened, therefore the studies on obtaining pure water suited for drink are of currently central value. From this point thaw water is of a particular interest, as evidenced

by numerous discussions on Internet websites. High activity in discussing the phenomenon of thaw water could be, to all appearance, connected with the fact that the use of thaw water seems to be promising for solving the problems connected with human health.

At the same time there are no publications in the contemporary literature devoted to a detailed discussion of the thaw water phenomenon in the context of physical chemistry. As a result, there is a paradoxical situation: biologists are quite sure that the phenomenon of thaw water (the increase in the productivity of agricultural crops, in the fertility of animals, *etc.*), physicians advise to drink thaw water and to use it for inhalations, and physical chemists (or chemical physicists), taking changes in water structure as the phenomenon of thaw water, on the contrary, declare, that there is no phenomenon, since any non-equilibrium structures arisen in water must disappear within such a short time as several picoseconds [6]. The paradoxicality of the situation is aggravated also with the fact that the persons engaged in production are already showing a considerable interest in the release of equipment for preparing thaw water [7–11], being guided by the reasons often based on doubtful data.

The purpose of the present communication consists in a comparative analysis of reliably measured physicochemical parameters of thaw (freshly thawed) and usual water.

This work is devoted to the consideration of the following parameters of thaw and initial water:

- 1) the concentration of organic, inorganic impurities and particles of a disperse phase;
- 2) the concentration of heavy isotopes;
- 3) hydrogen peroxide concentration;
- 4) the concentration of dissolved gases (oxygen, carbonic gas).

It is obvious that the variation of the first parameter value as the result of the freezing-unfreezing procedure (thaw water obtaining) occurs due to water purifying from various impurities (pollutants) basing on the method of the directional crystallization [12]. The difference of this method from common ones consists only in the direction of solid-melt interface movement. So, if in the case of the direc-

tional crystallization the solid-melt interface moves along one of the axes (for example, along a lengthy cylinder), then as water freezing in usual vessels the movement solid-melt interface occurs simultaneously along several directions (from top to down, from below upwards, from the edge to the centre).

The second parameter reflects a difference in the segregation level for deuterium isotope in thaw and initial water.

The third parameter reflects an increase in the concentration of hydrogen peroxide in water after freezing and the subsequent unfreezing. It should be noted that the dependence of thaw water stimulating biological properties on the concentration of hydrogen peroxide is hypothesized for the first time in the present publication.

Finally, changing in the concentration of dissolved gases (as a matter of fact, “the concentration of air”) is of temporal character: thaw water is being saturated with gases from air during a certain period of time. With no stirring the equilibration time is determined by the diffusion rate and depending on water volume it can be estimated amounting to several hours.

It follows already from the listed above physicochemical parameters bound together by the concept of “water purity”, “water composition”, *etc.* that the fresh-thaw water phenomenon (if that really takes place) could be explained only within the framework of the idea of the concentration of the listed substances (the list of substances being obviously incomplete). The concentration of the aforementioned substances in the freshly thaw water could differ from those in usual water, and the occurrence of the freshly thaw water phenomenon might be connected just with these differences. The listed above properties (parameters) specify the impurities those are always present in natural and technical water. Hence, as discussing the phenomenon of thaw water one should consider, first of all, a possible connection between the concentration of various impurities and healing properties of water and only in the case when such dependence is not observed one should involve the concepts of a specific “ice-like” structure of thaw water exhibiting “memory”.

COMPARISON OF PHYSICOCHEMICAL PROPERTIES FOR THAW AND INITIAL WATER

Concentration of organic, inorganic impurities and disperse phase particles

Organic impurities. Until recent time the publications on the efficiency of water purification from soluble organic impurities with the use of freezing method were limited by [13, 14]. The authors of [13] using a chromatography technique investigated the efficiency of the freezing method for water purifying from aldehydes and revealed the dependence of the water purification level on the type of aldehyde. The authors of [14] using the techniques of fluorescence and UV optical absorption experimentally investigated the application possibility for the freezing-unfreezing method for purifying tap water from soluble organic impurities at the temperature of $-17\text{ }^{\circ}\text{C}$. Under these conditions the water purification level under one-fold freezing amounted to 25–30 %. As the result the authors of [14] had concluded on a practical applicability of the freezing method both for water treatment and for the determination of water purification quality.

In order to further understand the mechanism of water purification we carried out additional experiments with organic impurity characterized by various hydrophobic properties. We used aqueous solutions of pyrene derivatives (the concentration amounting to $\sim 10^{-6}$ mol/L, the volume under freezing being of ~ 300 mL, $T = -17\text{ }^{\circ}\text{C}$, fluorescence registration technique, $\lambda = 390$ nm). It was observed that the efficiency of using the freezing method for water purification from dissolved organic impurities depends on the hydrophobic properties of the latter (Table 1). In particular, it was shown that this method is rather efficient in the special case that the molecules of an organic impurity are characterized by a low hydrophobicity level (the hydrophobicity level is meant as a quantitative ratio of non-polar to polar groups in the molecule under consideration). On the contrary, for water purification from organic impurities whose molecules exhibit a high level of hydrophobicity, this method appeared inefficient.

It is known that certain molecules under freezing are characterized by high concentra-

TABLE 1

Dependence of the separation coefficient on the number of atoms in the substituents within pyrene derivative molecules (water freezing level being of 50 %)

Molecule	Separation coefficient
Py	2.2
PyOH	3.4
PyHCO	5.2
Py-CH ₂ (CH ₃) ₂ COOH	13.3

Note. Separation coefficient represents the ratio of fluorescence intensity values for pyrene derivative molecules in non-frozen water to that in thaw water ($\lambda = 390$ nm).

tion factors, which is widely used for a long time for practical purposes in order to concentrate humic substances (HS) [15]. In view of the aforementioned hypothesis this well-known empirical approach could be explained as it follows: the HS molecules contain enough polar groups, capable to form hydrogen bonds, and, in addition, they contain enough ionic fragments [16]. Owing to this fact the HS molecules demonstrate a significant hydrophylicity level.

Thus, the values of separation coefficients for such molecules would be high in the magnitude since under freezing it is thermodynamically more favourable for the molecules to stay within the aqueous phase, rather than to be adsorbed onto the surfaces of ice microcrystallites [17]. It is this circumstance that could be connected with successful results of water treatment experiments using the freezing method carried out in Petergof [18]. Water of this region contains a lot of humic substance coming from peat; therefore water can be well purified with the help of the freezing method.

From the standpoint of the concept on the role of hydrophobic and hydrophylic interactions in water treatment by freezing the data obtained earlier concerning the purification of double-distilled water from aldehydes become understood [13]. So, according to the data presented in [13], the separation level amounts to 90 for formaldehyde, to 3 for acetaldehyde, to 2 for propyl aldehyde (*i.e.* as an aldehyde hydrophobic contribution increased the level of water purification decreases). Within the framework of the concepts we suggested these results are easy to interpret as it follows: when increasing hydrophobic contribution of an al-

dehydrate the level of aldehyde adsorption onto ice increases then the efficiency of freezing procedure for water purification decreases.

Thus, in view of the concept on hydrophobic interactions occurred in water one can non-contradictorily explain data concerning the interdependence between the level of water purification due to freezing method from organic impurities and the type of organic compounds belonging to different series (HS, aromatic hydrocarbons, aldehydes). The essence of this concept consists in the fact that under freezing the most hydrophobic molecules are most efficiently adsorbed onto the solid-melt interface of water, *i.e.* they are as though "sticking" to ice. On the contrary, less hydrophobic molecules mainly stay in water.

Inorganic impurities. One of the physico-chemical parameters that can be considered as differing thaw water from usual one, represents a decrease in the concentration of not only organic, but also inorganic impurities dissolved, since the solubility of these impurities in ice is much lower, than in water. However, it was demonstrated in [19] with the use of conductometry technique that under the conditions usually recommended for obtaining thaw water (the volume of water under freezing being ~0.3–0.5 L and the temperature at -17°C), NaCl concentration in water after one freezing-unfreezing cycle becomes 15–20 % lower as compared to those for initial water.

Similar results were obtained for CaCO_3 in [20] where, using the method of weighing, it was found out that the application of the freezing-unfreezing procedure for water purification from soluble amounts of CaCO_3 under the conditions recommended for the preparation of thaw water (the volume of water under freezing being about 0.5 L and the temperature at -17°C) is inefficient. It could be connected with a very good "freezing-in capability" of this salt with respect to ice [21] as well as with a high rate of freezing. Indeed, in order to obtain "good" ice single crystals under directional crystallization of saline aqueous solutions the velocity of solid-melt interface movement should be $<0.05\text{ cm/h}$ [22, 23], whereas under the conditions the experiment described in [20] this velocity value amounted to about 0.5 cm/h .

Disperse phase particles. The authors of [24] studied the dependence of the concentration of disperse phase (DP) particles in thaw water on the number of freezing-unfreezing cycles (all the volume was frozen). The effect of water purification from insoluble impurities consists in the following. During the water freezing process the insoluble particles cannot built in the structure of ice being oppositely displaced towards separate regions, since the difference in the size of particles and water molecules is at the least four orders of magnitude. In these regions, while the phase is still liquid the process of coagulation occurs and DP particle undergo coarsening [25]. Thus, under the freezing-unfreezing procedure the initial water transforms into a suspension from the colloid state.

As a result, frozen water (ice) represents the structure where one could observe both the areas with pure ice and areas with coagulated initial DP particles. Accordingly, under unfreezing the coagulated particles (being larger in size than ones within initial water and capable of faster sedimentation) should form a deposit. Thus, after separating the deposit the concentration of DP particles in thaw water must be lower. The authors of [25] carried out the freezing of tap water at -17°C . The amount of the deposit was determined using a gravimetric method. It was found that the application of one freezing cycle for water purification from suspended particles is inefficient. The level of purification amounted to about 30 %, and full precipitation of the deposit stopped after 18 cycles.

Thus, the comparative analysis of the data available with respect to the concentration of organic and inorganic impurities as well as the DP particles in thaw and initial water allows one to draw the following conclusions.

1. The concentration of all the impurity types in thaw water is always lower as compared to the concentration of impurities within initial water notwithstanding the fact that there is an individual mechanism for water purification from each type of impurity.

2. The impurity concentration decrease amounts to about 20 % for dissolved inorganic molecules, about 30 % for insoluble impurities, and about 20 % for organic impurities in usual tap water. At the same time for certain impurities this value could exceed 1000 %.

3. The efficiency of water purification after one freezing-unfreezing cycle under standard freezing conditions (T being at about $-17\text{ }^{\circ}\text{C}$, volume of $\sim 300\text{ mL}$) is low. In this connection a popular belief that the purity of thaw water prepared using single freezing-unfreezing cycle is close to that of distilled water is wrong. Thus, the question on the impurity concentration decrease should be removed from the discussion concerning the benefit of thaw water application, since the content of impurities is comparable to that for usual water and according to this parameter thaw water is close to initial one. Hence, popular ideas about desalination of an organism and other negative consequences of using thaw water for a long time [26] are, to all appearance, exaggerated.

Concentration of heavy isotopes

The effect of the isotopicity on physicochemical properties of liquids (in particular, water) are reliably established long ago [27]. Let us recall that the content of the lightest protium-based water H_2^{16}O amounts to 99.727 vol. % in natural water. Thus, 0.273 % falls on heavier isotopes, the content of the three water isotopes only being almost H_2^{18}O (73.5 %), H_2^{17}O (14.7 %) and HD^{16}O (11.5 %) [28]. It therefore follows that the amount of water with heavy oxygen is greater, than with heavy hydrogen. Meanwhile, as discussing water isotopic composition effect of on various biological processes, most often one consider the changes of deuterium concentration only, but the influence of variations in the concentration of the most widespread heavy water isotope such as H_2^{18}O is not under investigation [29]. Within the framework of this work the kind of a heavy water isotope is not crucial, therefore further by heavy water we mean only HD^{16}O and D_2^{15}O .

The freezing point of H_2O is at $0\text{ }^{\circ}\text{C}$, that for D_2O being at $3.98\text{ }^{\circ}\text{C}$ [27]. Thus, under freezing first of all a heavier ice (HDO) should be formed. When this ice is further to isolated and melt, the content heavy isotope in thaw water to be higher as compared to its content in initial water. The results of [30] indicate this fact, too. According to the opinion of the author of [29], the decrease in the heavy isotope con-

centration is of fundamental importance in the discussions concerning the usefulness of thaw water.

This conclusion is based on the well-known facts concerning the depression of plants growth due to increasing the concentration of heavy water [2] and, on the contrary, the stimulation of the weight gain of animals, when they were watered using lightened water (water where the deuterium content is by 20 % reduced) [28]. Comparative clinical testing of light water in the groups of patients have shown that it can be used as an auxiliary means in a complex treatment of arterial hypertension, carbohydrate metabolism disorder and energy imbalance in an organism [28].

Thus, the isotopic composition of thaw water differs from that of initial water, which is likely caused by the differences in the freezing temperature values for light and heavy water. Rather detailed analysis of the hypothesis concerning possible interconnection between the segregation of hydrogen isotopes and useful properties of thaw water is presented in [29].

Hydrogen peroxide concentration

Hydrogen peroxide takes part in all the vitally important processes, therefore the researchers pay much attention to the studies on its role, for example, in the homeostasis of a human organism [30, 31], in photosynthesis [32, 33] or in the self-purification of natural aquatic environment [34].

The data on the presence of hydrogen peroxide in natural snow water have been for the first time published in 1949 [35]. The authors of [36] investigated the effect of water freezing-unfreezing procedure on the concentration of hydrogen peroxide and have revealed that H_2O_2 concentration considerably higher than that in the initial specially prepared water ($\sim 10^{-12}\text{ mol/L}$) amounting to $\sim 3 \cdot 10^{-6}\text{ mol/L}$.

In order to explain the causes of the occurrence of a considerable amount of hydrogen peroxide the authors of [36] used the concept about water as a polymer, wherein chemical bonds might be broken mechanochemically. With reference to water it means the formation of OH^{\bullet} radicals those can further form a molecule hydrogen peroxide according to the following reaction:

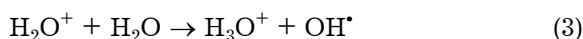


Within the framework of this model the authors of [36] have quantitatively estimated the concentration of hydrogen peroxide in thaw water ($\sim 10^{-6}$ mol/L); the value appeared close to the experimentally measured concentration. However, in our opinion, this model is ambiguous. Indeed, the nature of OH bond breaking is presumably fluctuation-based; therefore for explaining the increased concentration of hydrogen peroxide in thaw water one should assume a higher level of fluctuations as compared to initial water. However, there is no such information available now.

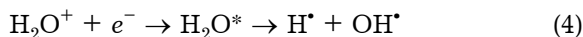
The most probable reason of the occurrence of OH[•] radicals as well as of the subsequent reactions could be presented by a Workman–Reynolds effect [37]. The essence of the effect consists in the fact that the formation of ice results in the formation of a potential about 100 V across water-ice interface. A similar effect is observed when thawing ice, too. This energy is sufficient, for example, in order to ionize a molecule of water. Further the processes could occur those are studied well enough for water radiolysis and electrochemistry [38]:



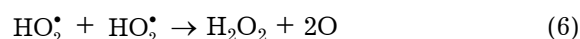
An ionized water molecule reacts with another neutral water molecule to result in the formation of a highly reactive hydroxyl radicals OH[•]:



those reacting then according to the reaction (1) to produce a molecule of hydrogen peroxide. Another possible way to form OH[•] radicals and, accordingly, to produce a hydrogen peroxide molecule is due to the reaction of an ionized water molecule of with electron (see eq. (2)) with the formation of the two radicals:



In the presence of oxygen the other radiolysis products those exhibit oxidizing properties are formed too: peroxide radical HO₂[•], atomic oxygen O and, finally, hydrogen peroxide H₂O₂ [38]:



As a matter of principle, a similar assumption about the mechanism of OH radical formation in thaw water could be made taking into account data concerning the energy that accompany the process of ice cracking. So, according to [39], this value is likely amounting to 1–10 keV [39], which is more than sufficient for the homolytic OH bond breakage within a water molecule, formation of an OH[•] radical with the further generation hydrogen peroxide according to reaction (1). Moreover, in this case all the mentioned processes (see eqns. (2)–(6)) can take place, too.

Thus, one of physicochemical parameters those cause thaw water to differ from usual water consists in the increased content of hydrogen peroxide. Taking into account numerous data on positive effect of hydrogen peroxide at certain concentrations on biological objects [32], one can assume the phenomenon of thaw water to be connected with an increased concentration of hydrogen peroxide. Within the framework of this assumption the occurrence of ostensibly observed effect of thaw water lifetime (a phenomenon in the phenomenon) might be explained basing on a reliably established fact of the spontaneous decomposition of hydrogen peroxide [34]. This process depends on temperature, the intensity of light irradiation, pH, medium properties, etc. and can proceed for many hours. It is obvious, that in this case the “memory” exhibited by thaw water one can explain attracting no ideas about the presence of so-called structural memory in water.

Concentration of dissolved gases

Now it is established, that the concentration of dissolved gases such as oxygen and carbonic gas in thaw water is lower as compared to these values for initial water [1]. This is not surprising since the solubility of gases in ice is much less, than in water [41, 42]. Thus, the concentration of gases in thaw water right after ice thawing is lower than the equilibrium value. On keeping and due to the contact with atmospheric air thaw water can absorb various gases, which results in their concentration growth. This is, for example, indicated by pH reduction due to increase in carbonic gas con-

centration and its chemical reaction with water resulting in the formation of carbonic acid. It is not excluded that just the acidity increased with respect to initial water determines healing properties of thaw water. Hence, in this case the concentration of gases plays a crucial role.

It should be noted that notwithstanding the fact the concentration of dissolved gases vary with time, this value for natural water is always distinct from zero, and therefore its influence should be taken into account as considering the parameters of thaw water. The nature of the thaw water phenomenon of is not conclusively established; therefore it is not inconceivable that just the change of the concentration of gases determines the manifestation of this phenomenon. The fundamental differences between thaw and usual water might consist, for example, in the fact that thaw water containing no gases, as a matter of principle, cannot form clathrate-like structures.

On the contrary, usual water saturated with gases can form clathrate-like structures [41, 42], and only in the presence of air molecules within water there is possibility of the formation of so-called bubstone clusters [43]. (A bubstone cluster represents a spontaneously arisen stable gas bubble within the liquid under equilibrium with respect to external gas environment.)

It is obvious that the nature of occurrence of "memory" of thaw water can be explained within the framework of the "gas" hypothesis, if the "memory" is considered as the time of natural establishing the equilibrium concentration of various gases in water after thawing ice or snow. In this case there is no necessity to involve the ideas concerning the "structural memory" of water.

CONCLUSION

Thus, the comparative quantitative analysis has demonstrated that thaw and usual water differ in a number of physicochemical parameters. However, to establish which of parameters determines useful properties of thaw water is impossible now, since regular and complex studies in this field were not carried out.

In this connection the attempts to develop the equipment for obtaining thaw water with

maximum useful physiological properties [8–12] seem to be doomed for failure since the optimum physicochemical conditions for obtaining such thaw water are not known till now. Whereas the effects of purification and light water obtaining can be combined [9, 10] in a single operating cycle freezing at first a heavy fraction and then after separating the residuary non-frozen water through the further freezing of its main part, the character of variation in the concentrations of hydrogen peroxide and gases during these processes is not established yet.

It is not unlikely that the curative properties of thaw water those were written so much in press, are caused only by one of the parameters, and in that case it is possible to optimize obtaining water with preset properties using the variation of this parameter. However, the question on determining this (or these) crucial parameter(s) remains still an open question up till now. Hence, it is prematurely to offer any technologies for optimum conditions in order to obtain thaw water. In this connection, it is rather difficult to fancy how the numerous recommendations and recipes for thaw water preparation, those wander from newspapers to magazines with the description of rather approximate (and often inconsistent) conditions of its preparation, can really promote obtaining thaw water with real healing properties.

Making such recommendations, the writers should not forget the precept: "don't do much harm". Indeed, water freezing and the subsequent ice thawing unequivocally cannot worsen the properties of water. It is not improbable, by the way, that just this circumstance combined with the effect of "placebo" results in the observed "positive" influence of thaw water.

In conclusion it should be emphasized once again, that really observed and ostensibly particular properties of thaw water can be explained by the presence of numerous impurities at certain concentrations in water. Thus, substantively there is always the presence of dilute aqueous solutions of electrolytes and non-electrolytes instead of imaginary pure water. Correspondingly, one should attract the ideas about a special structure of thaw water only within the framework of available knowledge of the structure of aqueous solutions, rather than pure undoped water.

In this sense the phenomenon of thaw water differs in nothing from the phenomenon of soda water or aerated water wherein the presence of the increased concentration of corresponding gas (oxygen, carbon dioxide, etc.) with respect to the equilibrium value is trivial and does not cause any questions.

Finally, it should be noted that questions on the properties of thaw water are intimately connected not only with the problem of obtaining pure potable water (indisputably one of the central ones for modern civilization), but also with the problem of obtaining water with curative properties. The last circumstance considerably emphasizes the urgency of regular studies concerning physicochemical properties of thaw water. These studies can (and must) finish with the chaos that is reigning in the field of the information on thaw water.

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