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Influence of Microwave Energy and Agitation on the Physicochemical Properties of Natural Mineral Water

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ABSTRACT

This study investigates the modifications in the physicochemical properties of natural mineral waters induced by microwave radiation and agitation. Mineral waters from Svyatoi Istochnik, Aqua Minerale, Karachinskaya, Arkhyz, and Serebrianyi Kliuch were subjected to these two activation methods. The pH and redox potential parameters were meticulously evaluated to observe alterations. Experimental studies provide evidence that microwave radiation and agitation significantly alter the physical and chemical characteristics of the selected mineral waters. A standard microwave oven operating at a frequency of 2.45 GHz was utilized for microwave radiation while agitation was performed using established techniques. The changes in pH and redox potential were carefully measured post-treatment. Technologically significant properties of the treated water were identified. These include a reduction in acidity due to an increase in pH value and a near tripling of the water's redox potential following a total stirring time of 2 minutes compared to an untreated control sample. The mechanochemical technique employed during agitation caused water molecule destruction due to friction forces, plastic deformation, impact compression, and mass transfer intensification. Conversely, the microwave radiation effect is attributed to the resonance effect resulting from the absorption of microwaves by water molecules. Moreover, it was experimentally determined that the activation effect of agitation persisted in the mineral waters for one hour, while the effect of microwave radiation lasted for one day. This longevity indicates a more profound influence of microwaves on the electronic systems of the water's components. In conclusion, the study demonstrates the feasibility of altering the physicochemical parameters of mineral waters with varying compositions and degrees of mineralization using short-term microwave radiation and agitation. The successful modification of the water samples' hydrogen index and redox potential signifies the potential utility of these techniques in manipulating the properties of mineral water.

1. INTRODUCTION

The therapeutic and preventive potential of mineral water consumption in addressing numerous ailments, including respiratory, skin, liver, and intestinal diseases, is well-acknowledged [1-3]. To gain insights into the inherent properties of mineral waters, their classifications were critically examined.

Significant disparities are observed in the therapeutic efficacy of different drinking mineral waters, which can be attributed to their unique physical and chemical compositions. These include variations in the degree of mineralization (typically ranging from 3-12 g/l) and the proportions of single, divalent, and monovalent ions. Notably, mineral waters possessing a salt concentration within the 9-12 g/l range, dominated by univalent cations and anions (such as sodium ions, chlorine, and bicarbonate), exert the most significant influence on the hormonal regulation of metabolism.

Beyond chemical composition, various physical parameters also serve as criteria for the classification of mineral waters. For instance, based on pH, mineral waters can be categorized into acidic (pH<7) or alkaline (pH>7) types. Temperature at the spring source further distinguishes mineral waters into cold (<20°C), hypothermal (20-30°C), mesothermal (30-40°C), and hyperthermal (>40°C) classes. Hardness, indicative of alkaline earth metal content, classifies mineral waters into very soft (0-

100 mg/l $CaCO_3$), soft (100-200 mg/l $CaCO_3$), or hard (200-300 mg/l $CaCO_3$) types [4-6].

Mineral water, sourced from natural springs, is typically rich in elements such as calcium carbonate, magnesium sulfate, potassium, and sodium sulfate. Nevertheless, mineral water is also artificially produced by incorporating salts into distilled water or infusing it with carbon dioxide. The mineral composition of both naturally occurring and artificially prepared mineral water displays considerable variation, and in certain instances, may even be inferior to that of conventional tap water [7-9].

Water, indispensable for life as we know it, plays a pivotal role in the crucial process of photosynthesis ($6CO_2+6H_2O+672kcal \rightarrow C6H_{12}O_6+6O_2$). Representing the most fundamental and vital chemical reaction on Earth, photosynthesis provides primary nutrients, directly or indirectly, to all living organisms, and serves as the principal source of atmospheric oxygen. Remarkably, water is the only substance present in all three physical states - solid, liquid, and gaseous - under natural Earth conditions.

Key physical attributes of water, including transparency, density and temperature responsiveness, high specific heat capacity, high heat of vaporization, viscosity, and surface tension, contribute to its exceptional versatility. To comprehend the reasons behind these unique properties of water and their importance for terrestrial life, it becomes crucial to examine the molecular structure and bonding patterns of water. Indeed, the extraordinary nature of water can be directly attributed to its distinct shape and bonding configurations [10-12].

In a water molecule, the central oxygen atom is surrounded by four pairs of valence electrons. Two of these pairs participate in covalent bonding with hydrogen atoms, while the remaining two pairs are unshared and exert the most significant repulsive effect [13, 14]. To fully appreciate the unique properties of water, a fundamental understanding of chemical bonding and the molecular structure of water is required. The water molecule, constituted by hydrogen and oxygen, features covalent bonds, with hydrogen atoms sharing electron pairs with oxygen [15].

A relatively strong attraction to the shared electron pairs is exhibited by the oxygen atoms, leading to the generation of electropositive regions around the hydrogen atoms and electronegative regions around the oxygen atoms. The resultant water molecule is classified as polar, as the positive and negative regions are not evenly distributed around the central point [16-18]. Due to its polar nature, a water molecule experiences electrostatic attraction towards other molecules and ions. The attraction between the electropositive hydrogen atom and the electronegative oxygen atom of an adjacent water molecule is defined as hydrogen bonding, which only contributes approximately 10% of the strength of a covalent bond [19].

When oxygen and hydrogen atoms form bonds, the disparity in their electronegativities is insufficient to generate ions; instead, a pair of electrons is shared, establishing a covalent bond. However, the higher electronegativity of the oxygen atom pulls the electron pairs closer to its nucleus [20]. Consequently, a concentration of negative charges accrues near the oxygen atom and farther from the positive charges of the hydrogen atom nuclei. The resulting bond is intermediate between a fully ionic and a covalent bond, with a partial, but not complete, charge separation as observed in ion formation [21].

As such, in a water molecule, the side encompassing the hydrogen atoms carries a partial positive charge, while the side with the lone pairs of electrons bears a negative charge. The molecule overall is thus polar, and the water molecule can be said to demonstrate a polar covalent bond. The inherent polarity of the water molecule facilitates mutual attractions among water molecules [22, 23].

The hydrogen bond formed between water molecules elucidates numerous essential properties of water. Foremost among these is the capability of water to remain liquid over a broad temperature range witnessed on Earth, a feature not observed in other molecules such as CO_2, which transition from liquid to gas at significantly lower temperatures [24]. Furthermore, hydrogen bonding is responsible for the substantial thermal energy required to vaporize water, transcending its liquid state. Hydrogen bonding also ensures the chemical stability of water molecules during reactions. While other compounds undergo chemical alterations, namely ionization, water maintains its chemical integrity, thus rendering it a weak conductor of electrical current. Interestingly, hydrogen bonding also accounts for the unusual phenomenon of ice having a lower density than water [25].

The inherent polarity of water designates it as a solvent, particularly compatible with other polar compounds, including salts, alcohols, and various carbonic compounds. As a solvent, water exhibits an unrivaled capacity to dissolve a wide spectrum of inorganic and organic substances, exceeding any other known substance [26]. Water persists in its liquid state over an extensive temperature range, beyond which biological processes either decelerate or halt entirely. Uniquely, water is the only substance wherein the solid state is less dense than the liquid state, a characteristic not shared by any other substance [27].

Water possesses the highest heat capacity among all common substances, necessitating more thermal energy for a given temperature increase than any other material. Conversely, water releases more heat upon cooling than any other substance. Notably, water's surface tension, the attractive force exerted by sub-surface molecules on those at the liquid-air interface, is surpassed only by mercury. This internal force inhibits the fluid's flow, with water's exceptionally high surface tension attributed to hydrogen bonding [28].

An abundance of low-mineralized waters is present within the spectrum of mineral waters found in Russia [29]. Consequently, enhancing the effects of these natural mineral waters by modifying their physical and chemical properties through methods such as microwave energy and agitation has become a topical issue.

In the current study, the effect of microwave energy and agitation on the physical and chemical properties of natural mineral waters is investigated. A theoretical analysis of the composition of Russian mineral waters, their biological impact on the human body, and their activation methods is conducted. The importance and practical implications of enhancing the activity of natural mineral waters are confirmed, thereby underscoring the need for novel and effective activation techniques. The most accessible non-specific mechanochemical and wave methods for influencing mineral waters are selected.

In the empirical section of the study, short-term exposure of three brands of mineral water (Svyatoi Istochnik, Aqua Minerale, Serebrianyi Kliuch) and two types of medical drinking mineral water (Karachinskaya, Arkhyz), each with diverse compositions and degrees of mineralization, to agitation and microwave treatment is undertaken. The potential for preserving the activation effect of mineral waters, depending on the type of external energy influence, is determined. The promising applications of activated mineral waters in various fields such as medicine, the food industry, and engineering, among others, are identified.

2. METHODS

Studies were conducted based on the Natural Nutraceuticals Biotesting Laboratory of Kemerovo State University from February 6 to February 20, 2023. The brands of mineral water examined, which are common in Russia, were:

- Svyatoi Istochnik;
- Aqua Minerale;
- Karachinskaya;
- Arkhyz;
- Serebrianyi Kliuch;
- Distilled water according to GOST R 51232-98.

The characteristics of the studied mineral waters are presented in Table 1.

Table 1 shows that the samples chosen for tests are well-known brands of drinking and medical drinking mineral water with different degrees of mineralization and containing particular cations and anions. These waters are commonly used for therapeutic and prophylactic purposes when consumed as drinks and for effective balneological therapeutic procedures in sanatoriums. Production of these waters for wide consumption in packaging is steadily established.

The conditions of the experiments were as follows:

1. Temperature of the experiment t=25℃.
2. Stirring time (at an angular speed of the stirrer 300 rpm) -2 min (manufacturer of the stirrer: UED Group, St. Petersburg; power: 350W; temperature range: 5-310℃).
3. Time of exposure to microwave radiation (microwaves) at minimum power in nonthermal mode -7 s.

The experiments were carried out three times under equal initial conditions to reduce the error in measurement results. At the first stage of the research, microwave treatment of natural mineral waters was carried out batch-wise in a microwave oven Daewoo Electronics KOR-6L7BS (Daewoo, South Korea), with microwave power 700 W, frequency 2.45 GHz. The assessment of the active reaction of water (pH value) in the mineral waters under study was carried out using an electronic pH meter Anion 4120 (Infraspark-Analit, Russia) with a combination glass electrode. The analyzed water sample was poured into a beaker. The electrodes, which should be kept in distilled water before measurement, were blotted with filter paper and lowered into the beaker with the analyzed water. After 0.5-1 min, when the readings stop changing, the pH value of the analyzed water was recorded. Then the electrodes were rinsed with distilled water three times and lowered into the beaker with distilled water.

The redox potential of mineral waters was measured using platinum electrodes EPV-1SR and ETP-02 and a glass redox electrode EO-01 (Gomel Plant of Measuring Equipment, Belarus). As a reference electrode for potentiometric measurements, we used a chlorosilver electrode EVL-1M3.1 (Gomel Plant of Measuring Equipment, Belarus). The redox potential of the solutions was recorded automatically on several indicator electrodes simultaneously using the Ecotest program (FARMEC Scientific and Production Company with Additional Liability, Belarus). The measurement time ranged from 10 to 150 minutes. To control the accuracy of redox electrode readings, a standard solution from HANNA (Hanna Instruments, US) with a redox potential of 240 mV was used. The potentials of all electrodes used in the standard solution did not deviate from the certified value by more than 3 mV.

Table 1. Characteristics of the studied mineral waters

No.	Mineral Drinking Water	Mineralization, g/dm^3	Water Components	Water Bottling Location	Water Type
1	Karachinskaya	2-3	bicarbonates, Mg, Ca, Na, K, F	Western Siberia	medical drinking
2	Arkhyz	1-1.5	K, Ca, Na, F, sulfates, chlorides, bicarbonates	Caucasus	medical drinking
3	Aqua Minerale	0.15-0.5	K, Ca, Na, sulfates chlorides bicarbonates	Solnechnogors, Moscow region	drinking
4	Svyatoi Istochnik	0.5-1.0	K, Ca, Na, sulfates chlorides bicarbonates	Novosibirsk	drinking
5	Serebrianyi Kliuch	0.5-1.0	K, Ca, Na, silver, silicates, sulfates chlorides bicarbonates	Altai	drinking

Table 2. Characteristics of mineral waters after exposure to external factors*

No.	Types of Water	Factor of Influence	pH	E, mV
1	Distilled water	No impact (control sample)	6.8	26.2
		Microwave	-	-
		Agitation	9.1/1.34	150.3/5.73
2	Aqua Minerale	No impact (control sample)	6.7	22.5
		Microwave	7.6/1.13	66.4/2.95
		Agitation	7.8/1.16	83/3.69
3	Svyatoi Istochnik	No impact (control sample)	7.7	76.4
		Microwave	8.0/1.04	91.5/1.2
		Agitation	8.3/1.08	108.1/1.41
4	Serebrianyi Kliuch	No impact (control sample)	7.0	99.5
		Microwave	8.2/1.17	101.2/1.02
		Agitation	8.3/1.19	108.2/1.09
5	Karachinskaya	No impact (control sample)	7.5	66.0
		Microwave	7.7/1.03	72.7/1.1
		Agitation	8.0/1.06	93.1/1.41
6	Arkhyz	No impact (control sample)	7.4	60.0
		Microwave	7.9/1.07	88.2/1.47
		Agitation	7.7/1.04	72.2/1.2

*numerator – measured value, denominator – degree of water activation

3. RESULTS

Indicators of the active reaction of mineral waters and redox potential determine the degree of activation of natural mineral waters. The results of the experiments are presented in Table 2.

It follows from Table 2 that the two energetically different activation methods change the physicochemical properties of the selected mineral waters.

Possible mechanisms of water activation are activation under mechanical and microwave influences.

As a result of agitation, cavitation collapse of nanobubbles of dissolved air, and the action of microwaves, there first occurs a breakage of the chemical bond in the molecule of water:

$$H_2O = \bullet H + \bullet OH = H^+ + OH^- \quad (1)$$

Then, due to the transfer of the electron from the hydrogen atom to the hydroxyl radical, hydrogen ions and hydroxyl ions are formed:

$$\bullet H = H^+ + e^- \quad (2)$$

$$\bullet OH + e^- = OH- \quad (3)$$

Recombination of hydroxyl radicals produces hydrogen peroxide, then hydrogen atoms (hydrogen molecules):

$$\bullet OH + \bullet OH = H_2O_2 \quad (4)$$

$$\bullet H + \bullet H = H_2 \quad (5)$$

An effective acceptor of the hydrated electron in water is dissolved oxygen to form a superoxide anion radical and, when protonated, a hydroperoxide radical:

$$O_2 + e^- = O^{2-} + H+ = \bullet HO_2 \quad (6)$$

$$\bullet HO_2 + \bullet HO_2 = H_2O_2 + O_2 \quad (7)$$

$$\bullet H + \bullet HO_2 = H_2O_2 \quad (8)$$

$$\bullet HO_2 + H_2O_2 = O_2 + H_2O + \bullet OH \quad (9)$$

4. DISCUSSION

From the physical standpoint, the mechanochemical technique used in agitation entails destruction due to the forces of friction, plastic deformation, impact compression, and intensification of mass transfer [30]. The effect of microwave radiation is driven by the resonance effect in the absorption of microwaves by water molecules. It can be assumed that the application of other frequencies can activate any ions included in the composition of mineral waters. Positive energetics of water is currently associated with its enrichment with hydroxyl ions, which stimulate many biological processes by increasing metabolic processes in cell membranes. These ions are considered to be natural doping. Water containing various amounts of hydrogen cations, salt ions, and hydroxyl anions regulates the assembly and self-assembly of all cellular components, cell division and function, and the growth, development, and aging of the body. Mechanisms of the therapeutic and prophylactic action of activated water are associated with this. We further experimentally established that the effect of activation after agitation is preserved in mineral waters for one hour, and the effect of microwaves – for one day. This is indicative of a more profound effect of microwaves on the electronic systems of the components of aqueous systems.

At present, the structure of aqueous electrolyte solutions in a wide range of concentrations is examined based on the model of formation of substitution and introduction structures of ions and long-lived hydrate complexes in the initial structure of water [31, 32]. This study was limited only to mineral water, in further studies it is necessary to continue the study of the effect of agitation and microwave energy on other types of water.

5. CONCLUSION

The paper reports the results of comparative experiments proving the change of properties of mineral water systems under the physical and chemical influence of the parameters of pH and redox potential. The study points to the leading pattern in the influence of the studied external factors on the activation effect: the increase in pH and redox potential is determined by the mineralization of natural waters. These two parameters decrease as the concentration of mineral components increases. The greatest activation is observed in distilled water. The limitation of the study lies in its focus on mineral waters produced in Russia.

Theoretical analysis and technological development of water activation and energization methods are of great practical importance for biology, medicine, agriculture, and industry. The biological importance of water is also determined by the fact that it is one of the essential metabolites, i.e., it participates in metabolic reactions. For example, water serves as a source of hydrogen in photosynthesis and also takes part in hydrolysis reactions. At present, it is possible to introduce methods of mineral water activation in the food industry. For example, in practice, the Karachinskaya mineral water is used as a base for sweet medicinal and table carbonated drinks. Therefore, the activation of mineral water under the influence of microwave energy and agitation contributes to improving human health and increasing the principles of environmental sustainability.

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