# POSSIBILITY OF ASSESSING CHANGES IN WATER pH DURING EXPOSURE TO RADIOACTIVE RADIATION

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Radioactive radiation affects water molecules and leads to the formation of a whole range of radicals, charged particles, and molecules. For example, high-energy neutrons break down water molecules and lead to the formation of additional protons; it would seem that this clearly leads to a decrease in the pH of water since the number of hydronium ions, or hydrated protons ($H_3O^+$), should increase. However, it is not that straightforward. Some experiments have shown that during irradiation from radon decay under natural conditions for drinking water, a pH increase has been observed, and the degree of increase is directly related to radon concentration. Another study showed a decrease in the pH of atmospheric precipitation also under the influence of radon, with the degree of decrease directly related to the reduction of the bismuth-214 isotope, which produces α-particles. Our long-term observations of recirculated water have not revealed any decrease in pH, even short-term; on the contrary, the trend is rather the opposite.

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## INTRODUCTION

When radioactive radiation acts on water, a large number of particles are formed, sometimes even several tens, which exist for up to picoseconds. The most stable particles are $e^-_{aq}$, $H^\bullet$, $^\bullet OH$, hydroperoxide radicals ($HO_2^\bullet$), and peroxide anion radicals ($O_2^{\bullet-}$), as well as molecules ($H_2$, $O_2$, and $H_2O_2$), as mentioned in work [1], which considers the effect of fast neutrons on subcritical (up to 350 $^0$C) and supercritical (SCW) (400...600 $^0$C) water. It is quite evident that hydronium ($H_3O^+$) and hydroxyl ($OH^-$) ions are also present. At a constant pressure of 25 MPa and increasing temperature, the density of water dec$^0$reases rapidly: 0.743 g/cm$^3$ at 300 $^0$C, 0.625 at 350 $^0$C, 0.167 at 400 $^0$C, 0.090 at 500 $^0$C; it seems that at the same time the concentration of hydrogen peroxide decreases with the reduction of water density. The formation of charged particles, radicals in supercritical water, as well as hydrogen and oxygen, is highly significant for the implementation of reactors with supercritical water, since these reactors will have an efficiency of 45% compared to the current 33% while saving on capital costs. However, the aggressive particles formed during the radiolysis of water intensify the corrosion of the reactor vessel and the fuel rod cladding. To reduce the corrosive activity of aggressive particles toward water, reducing agents are usually added: hydrogen, hydrazine, or ammonia. At a neutron energy of 2 MeV, which corresponds to the average neutron energy in a reactor, the range of such neutrons in water is 4 cm, while the range of recoil protons (3 units) and oxygen ions is on average 75 (from 230 to 14) and 1.5 µm, respectively. In the case of radiolysis, the water oxidation reaction $H^\bullet + H_2O \rightarrow {}^\bullet OH + H_2$ takes place, and the two hydroxyl radicals combine to form a hydrogen peroxide molecule $H_2O_2$; thus, an oxidizing agent and a reducing agent are simultaneously present in water. As indicated in [2], the set of particles formed does not depend on the type of radioactive irradiation. The presence of radiolysis products in the gas phase can form an explosive mixture. Regarding changes in pH during water irradiation, researchers generally do not pursue this as a goal.

## 1. GHANGE OF NATURAL WATER pH DURING IRRADIATION

It seems that only radon is responsible for the irradiation of natural water. In the work [3], the effect of radon on the pH of drinking water is considered. Radon decays as follows: $^{222}Rn \rightarrow {}^{218}Po + \alpha$. Then polonium decays, also giving an α-particle. High-energy α-particles knock protons out of water molecules. The authors of [3] suggest that these protons affect the water's pH and also believe that geochemical processes involving hydrated protons – hydronium ions – may occur. There is also a reaction involving the oxygen of water molecules: $^{16}O + \alpha \rightarrow {}^{19}F + p$. In one place, the authors write that the production of protons in water via radon promotes geochemical processes related to the acid dissolution of rocks, while in another place (on p. 4) they indicate that as for Monte Carlo modeling of proton production – "The findings demonstrated that proton production from radon interactions raises water pH, suggesting potential health benefits", and experimental verification shows an increase in pH. It's enough to drive one crazy; evidently, one should rely on the experiment.

The use of the Monte Carlo method for pH estimation involves having models that allow for the assessment of the interaction results of charged particles or neutrons with water molecules, so the obtained pH results will depend on the chosen models and, as a result, may not be reliable. A water phantom with dimensions of 1x1x1 cm was considered, and α-particle energies of 5.49, 6, and 7.69 MeV, emitted by radon, $^{218}$Po, $^{214}$Po, and $^{214}$Bi. We shell quote from article [3] – "Radon, a radioactive gas produced from the decay of uranium in soil, can decrease the pH of water by generating alpha particles and releasing protons," which is correct; the more protons there are, the lower the pH of the solution, by definition. Let's look at what they say next. For radon measurements in water, a device with a detection limit of 34 Bq/m$^3$ was used. The authors claim that low concentrations of radon (30 kBq/m$^3$) lead to an increase in water pH, while high concentrations (130 kBq/m$^3$) decrease pH. This is very interesting and questionable, but in our opinion, it is necessary to more closely examine the possibility of errors when measuring the pH of non-buffered systems before drawing conclusions. The pH meter was calibrated using two buffer solutions: 7.01 and 10.01, and all obtained values fall within this range. On page 8 of reference [3], the authors write that when α-particles interact with water molecules, a stable isotope of oxygen $^{16}O$ is obtained, if the water itself consists of this isotope, which shows some lack of awareness. The authors [3] write a lot about the effect of protons created during the impact of α-particles on water molecules, but protons reduce the pH of water (in the form of hydrated protons, hydronium ions $H_3O^+$). They give an empirical formula for calculating the hydrogen index $pH = pH_0 + 0.0017C$, where C is the radon concentration in (kBq)/m$^3$. From the formula, it is seen that radon increases the pH of water. Despite the fact that the study was conducted rather poorly, the authors should be respected for their courage in taking on such a difficult topic.

The effect of radioactive radiation on atmospheric precipitation also occurs in the presence of radon and its decay products. The primary source of radon is uranium, which is found in the soil. In work [4], a detailed scheme for obtaining radon and its decay is presented:

$^{238}U \rightarrow {}^{234}Th \rightarrow {}^{234}Pa \rightarrow {}^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow {}^{222}Rn$

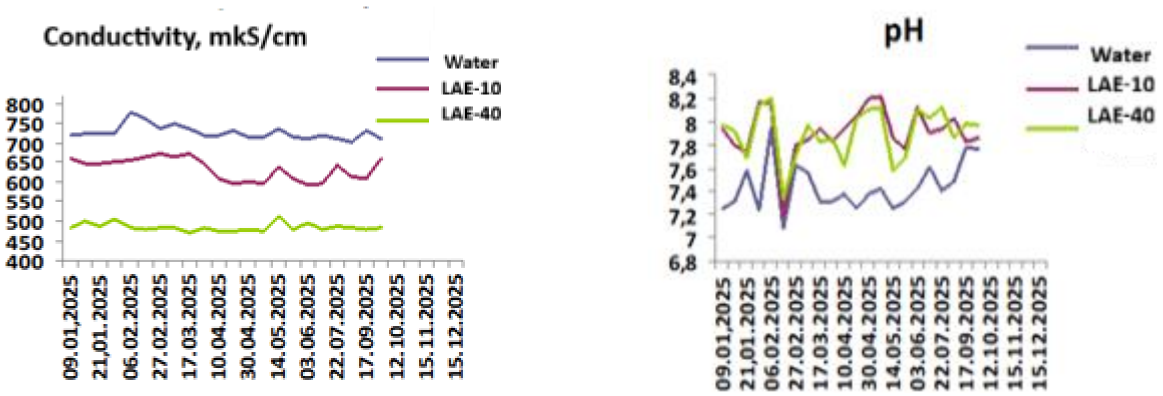
$^{222}Rn \rightarrow {}^{218}Po \rightarrow {}^{214}Pb \rightarrow {}^{214}Bi \rightarrow {}^{214}Po \rightarrow {}^{210}Pb \rightarrow$

$^{210}Bi \rightarrow {}^{210}Po \rightarrow {}^{206}Pb$.

As a result of the experiments, it was established that the pH of precipitation decreases during their passage, and the value is associated with the presence of $^{214}Bi$ in the atmosphere. The decrease in pH corresponds to the decrease in bismuth isotope activity in the atmosphere because α-particles produced during bismuth decay promote the production of additional protons. During rain, the pH can drop from 5.5 to 3.5. Attempts to calculate pH changes based on ionic balance and electroneutrality of the aqueous solution led to nothing, probably because there are quite a lot of ions present in the solution, making it difficult to identify them all. The decrease in pH under radiation is explained by the appearance of additional free protons; the authors demonstrated with their experiment that this occurs due to radon and its derivatives.

## 2. THE EFFECT OF RADIATION ON THE COOLING WATER OF ELECTRON ACCELERATORS

For an extended period in 2025, the changes in electrical conductivity and pH of the water in the cooling circuits of the LAE-10 and LAE-40 electron accelerators were studied [5].




*Graph of changes in conductivity and pH of water in the cooling circuit of LAE-10 and LAE-40 electron accelerators in 2025*

The data on conductivity and pH shown in Figure can be explained as follows – the conductivity of tap water is higher than that of the water in the accelerator cooling circuits due to the presence of a large amount of bicarbonates in the water, which then decompose in the cooling circuits, leading to a decrease in the total salt content and conductivity. The high bicarbonate content can explain the lower pH of tap water. The decomposition of bicarbonates and the release of carbon dioxide leads to the alkalization of the circulating water, and thus its higher pH. Moreover, during the cooling process of the accelerator systems, the water is subjected to high-frequency radiation, which can result in a decrease in electrical resistance and an increase in the water's pH.

The fluctuations of pH and conductivity in the cooling circuits of the two accelerators are similar. The effect of radiation on pH change may be masked by the high salt content of the water.

## CONCLUSIONS

The data presented in works [3] and [4] on the change in pH due to irradiation contradict each other. An experiment with drinking water after irradiation due to radon showed an increase in pH [3], whereas pH measurements of rainwater [4] showed a noticeable decrease in pH, which is directly related to a decrease in the concentration of the radon decay product $^{214}Bi$. Our experiments showed that the effect of irradiation on the pH of cooled water can be small and difficult to identify. From a theoretical standpoint, it is clear that the presence of additional protons produced during water irradiation leads to an increase in the concentration of hydronium ions ($H_3O^+$), which should lead to a decrease in pH, but this is ambiguous. Likely, some unaccounted factors are at play, possibly including the behavior of hydrogen formed during radiolysis. To reduce the influence of the salt background on changes in the pH of water under irradiation, experiments should be continued with distilled water.

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## МОЖЛИВІСТЬ ОЦІНКИ ЗМІНИ pH ВОДИ ПІД ДІЄЮ РАДІОАКТИВНОГО ВИПРОМІНЮВАННЯ

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Радіоактивне випромінювання діє на молекули води і призводить до утворення цілої низки радикалів, заряджених часток і молекул. Наприклад, високоенергетичні нейтрони руйнують молекули води і ведуть до утворення додаткових протонів; здавалось би це однозначно веде до зниження pH води, оскільки повинна збільшуватись кількість оксоній-іонів, інакше – гідратованих протонів ($H_3O^+$). Однак не все так однозначно, деякі експерименти показали, що під дією опромінення при розпаду радону в природних умовах для питної води зафіксовано підвищення pH, причому ступінь підвищення напряму пов’язана з концентрацією радону. Інше дослідження показало зменшення pH атмосферних осадів також при дії радону, причому ступінь зменшення напряму пов’язана із зменшенням ізотопу вісмуту-214, якій продукує α-частинки. Наші довгострокові спостереження за оборотною водою не виявили жодного, навіть короткострокового, зниження pH, скоріше навпаки.