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RADIOLYSIS OF KBr SOLUTIONS UNDER THE ACTION OF PROTONS WITH AN ENERGY OF 660 MeV

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In recent years, works have appeared in which the radiolysis of aqueous solutions under the action of protons, deuterons, and helions of various energies has been studied ($^{1-4}$). It was confirmed that, with an increase in the ionization density produced by the particle, or with an increase in the energy loss per unit path length–dE/dx–the yields of the molecular products of water radiolysis increase, while the yields of radical products decrease.

It was of interest to carry out an investigation with heavy radiation of such high energy that the value of dE/dx would be close to the value characteristic of light radiation. In this case, the difference between heavy and light radiation may arise from the effect caused by nuclear collisions of the particles with nuclei. Under the action of electron radiation, nuclear collisions cannot produce any additional effect. In the case of protons possessing energies of hundreds of MeV, nuclear collisions will cause the formation of multiply charged ions. These ions are formed by the knocking out, from molecules, of nuclei carrying remnants of the electron shell. In this case the multiply charged ions possess sufficiently high energy to ionize and excite molecules along their path. Having a high charge and a relatively low velocity, the ions will create a very high ionization density and thus influence radiation-chemical processes. For heavy particles with not very high energy, such an effect is impossible.

The present work was carried out with protons accelerated to 660 MeV, for which the effect of nuclear collisions may be expected. In terms of the value of $-dE/dx$, they are close to electrons with an energy of 1 MeV. It is known that 1 MeV electrons and γ-radiation from Co^{60} produce the same radiation-chemical processes. Therefore, in our work a comparison was made of the radiolysis of aqueous solutions under the action of γ-radiation from Co^{60} and protons with an energy of 660 MeV.

The main difficulty in working with protons of the indicated energy is the determination of the energy absorbed by the solution. To determine this quantity, the total flux of protons incident on an area of 1 cm^2 was measured. It is known that 660 MeV protons lose in water 2.1 MeV of energy per 1 cm of path (^{5}).

Fig. 1. Dependence of the concentration of Fe^{3+} on dose

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Fig. 2. Dependence of the hydrogen yield on log[KBr]: a—protons with energy 660 MeV, b—γ-radiation of Co^{60}

Figure 2: Fig. 2. Dependence of the hydrogen yield on log[KBr]: a—protons with energy 660 MeV, b—γ-radiation of Co^{60}

From this one can calculate the dose absorbed by 1 cm^3 of solution, knowing the total proton flux. Two methods were used to determine the proton flux. One method consisted in placing a graphite plate of known size and thickness in the path of the proton beam. Under the action of protons, radioactive carbon C^{11} is formed, with a half-life of 20 min. By measuring the activity of the graphite after irradiation with an end-window counter, one can calculate the total proton flux from the following formula:

$$I = \frac{n e^{\lambda \tau}}{\sigma N_c \eta \left(1 - e^{-\lambda t}\right)},$$

where n is the number of particles registered by the counter per unit time, λ is the decay constant of C^{11}, τ is the time from

the end of irradiation to the moment of measurement, σ is the reaction cross section, equal to 34 mbarn, η is the efficiency of positron registration, t is the irradiation time, N_c is the number of carbon nuclei in the layer in which there is no self-absorption and from which the same number of β-particles emerges as from a layer of infinite thickness. For the plates used, $N_c = 96 \cdot 10^{21}$ atoms.

A second method for determining the proton flux was its measurement by means of an ionization chamber calibrated against a Faraday cylinder. The results obtained by the different methods differ by 25–30%.

Fig. 1. Dependence of the concentration of Fe^{3+} on dose

Fig. 2. Dependence of the hydrogen yield on log[KBr]: a—protons with energy 660 MeV, b—γ-radiation of Co^{60}

Dosimetry was carried out with graphite plates; experiments with the ionization chamber were used to check the correctness of these results. The study was performed with the dosimetric solution $FeSO_4$, with evacuated KBr solutions, and with pure water. In the $FeSO_4$ solutions, $G(Fe^{3+})$ was determined. The concentration of Fe^{3+} was measured spectrophotometrically at a wavelength of 304 mμ. The results obtained are presented in Fig. 1. The straight line depicts the accumulation of trivalent iron with increasing dose, if it is assumed that for proton radiation $G(Fe^{3+})$ is equal to 15.6, i.e., has the same value as for

γ-radiation of Co^{60}. The points are the results of experiments in which the dose was determined from the activation of graphite plates. It is seen from Fig. 1 that, within the experimental error, $G(Fe^{3+})$ has the same value both for γ-radiation and for 660 MeV protons. The mean value of $G(Fe^{3+})$ obtained in these experiments is 16.9. The somewhat elevated value of $G(Fe^{3+})$ in comparison with the data for γ-rays of Co^{60} is probably caused by the nonuniformity of the proton beam, which is more intense at the center. Irradiation of the $FeSO_4$ solutions was carried out in glass spheres with a diameter smaller than the dimensions of the graphite plates. Therefore the mean proton flux per 1 cm^2 for the graphite plates, by which the dose was determined, was lower than the true flux on the sphere. The evolution of hydrogen from evacuated KBr solutions was also investigated. The amount of hydrogen formed was measured in a vacuum analytical apparatus by combustion on a platinum wire at a temperature of 180-200°. The results obtained are presented in Fig. 2. The curve shown in this figure gives the dependence of the hydrogen yield on log[KBr]. $G(H_2)$ increases when the KBr concentration is increased up to 10^{-3} M, remains constant in the concentration interval 10^{-3}–10^{-2} M, and again increases with further increase in the KBr concentration. A similar course of the dependence of $G(H_2)$ on log[KBr] was obtained for γ-radiation of Co^{60} (6). The higher yield values obtained for γ-radiation are explained by the different conditions under which irradiation was carried out. In experiments with γ-radiation, measurements were made by the membrane-cell method, in which the hydrogen liberated in the solution is partially transferred into the gas phase, whereas during irradiation with protons all the hydrogen remained in the solution. To confirm the correctness–

the correctness of this assumption, experiments were carried out with γ-radiation according to the procedure described above. The values obtained for $G(H_2)$ (Fig. 2,b) coincide with the results for protons with an energy of 660 MeV.

Thus, a study of the radiolysis of aqueous solutions of $FeSO_4$ and KBr showed that radiation-chemical phenomena are determined by only one characteristic of the radiation–the magnitude of the energy loss per unit path length, dE/dx.

However, determination of the hydrogen yield from pure evacuated water gave a discrepancy for γ-rays and protons. The water for these experiments was purified in the following way. Twice-distilled water was irradiated with Co^{60} γ-radiation to destroy organic impurities and with ultraviolet rays to destroy H_2O_2 formed under the action of γ-rays. The water was then distilled in a quartz apparatus. The electrical conductivity of water purified by this method was $0.7—0.6 \cdot 10^{-8}$ $\Omega^{-1} \cdot cm^{-1}$. The yield of hydrogen from water irradiated with γ-rays is 0.07 molecule/100 eV. The concentration of H_2 in this case is $5—6 \cdot 10^{-9}$ mole/cm^3. For 660 MeV protons, $G(H_2)$ is 0.15—0.19. The difference in hydrogen yields under the action of 660 MeV protons and Co^{60} γ-radiation may be explained in two ways. It is possible that this discrepancy is caused by the effects of head-on collisions. The multiply charged ions that arise in the water in this case produce processes characteristic of heavy radiation, for which the hydrogen yield is considerably higher than for γ-rays. However, this small difference may also be associated with traces of impurities, to which the hydrogen yield is very sensitive. Therefore it may be considered that 660 MeV protons and Co^{60} γ-radiation produce identical radiation effects.

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