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On the Reaction of Superoxide Ion with Organochloric Compounds

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Abstract—Superoxide ions were generated by the action of γ-radiation on water-alcohol solutions saturated with oxygen. The reaction of superoxide ions with sodium 2,4-dichlorophenoxyacetate and pentachlorophenol was studied. The reaction of $O_2^{-\bullet}$ with chloroaromatic compounds results in elimination of chloride ions and formation of phenyl radicals active in elimination of hydrogen atoms. The rate constant of the reaction between $O_2^{-\bullet}$ and 2,4-dichlorophenoxyacetate was calculated using the method of mathematical simulation.
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$O_2^{-\bullet}/HO_2$ radicals play the main role in many oxygen-containing radiolytic systems. They are generated in a number of electrochemical, chemical, and enzymatic reactions [1]. Superoxide ion is often formed in various biochemical processes. The processes of free-radical oxidation followed by formation of $O_2^{-\bullet}$ and its derivatives normally proceed in all tissues of aerobic organisms and represent one of the types of normal metabolic processes [2]. The acceleration or retardation of free radical processes in organism causes development of free radical pathology. The acceleration of free radical processes in organism may be due to the action of various physical and chemical factors. The ionizing radiation can result in the formation of $HO_2/O_2^{-\bullet}$ and other radicals and intensify free radical processes in organism with participation of $O_2^{-\bullet}$. Especially dangerous for living organisms are compounds capable of modification of primary radical processes in biosystems and transformation of superoxide ions inactive in the reactions of elimination of hydrogen atoms into active radicals, which can trigger further free radical damages of biomolecules in the organism. Chloroorganic compounds may act as such. It was shown [3, 4] that chloroform and tetrachloromethane enhance the radiation-induced damage of proteins and lipids of erythrocyte membranes. However, the mechanism of these processes remains unclear. The hepatotoxicity of CCl_4 was suggested [5, 6] to be caused by the CCl_4-induced peroxidation of unsaturated fat acids by means of the $CCl_3OO^{\bullet}$ radical. Formation of radicals $CCl_3OO^{\bullet}$ is the result of the reaction of superoxide ion with CCl_4. The reaction of $O_2^{-\bullet}$ with aliphatic halogen-containing compounds in aprotic medium has been studied in [5]. In the present work the reaction reaction of $O_2^{-\bullet}$ with chloroaromatic compounds in aqueous medium is studied.

Superoxide ions were generated by the action of γ-radiation on aqueous 2 M solutions of propanol-2 saturated with oxygen. The concentration of oxygen in the solutions was 0.0022 mol l^{-1}. The radiation causes the formation of active products of radiolysis of water: OH, e_{aq}, H_3O^+, H and H_2O_2 in radiation-chemical yields of 2.7, 2.7, 2.7, 0.55 and 0.7 molecules/100 eV, respectively [7, 8]. The formation of $O_2^{-\bullet}/HO_2^{\bullet}$ in the studied systems took place directly by reactions (1) and (2) presented in the Table as well as due to oxidation of hydroxyisopropyl radicals [reactions (8, 9)] and radical-anions $(Me)_2CO^{-\bullet}$ [reaction (12)].

We have studied the possibility of the reaction of $O_2^{-\bullet}$ with 2,4-dichlorophenoxyacetate ($ArCl_2$) and pentachlorophenol (PCP). The occurrance of these reactions was judged from the formation of Cl^- at the smallest concentrations of $ArCl_2$ and PCP. The concentration of $ArCl_2$ in solutions was varied from 0.01 to 0.0002 mol l^{-1} and the radiation-chemical yields of chloride ions (G_{Cl^-}) and acetone were followed. Figure 1 illustrates the dependence of the yield of the chloride ions and acetone on the $ArCl_2$ concentration. At high concentrations, $ArCl_2$ competes with oxygen for the hydrated electron (e_{aq}), and Cl^- is

Reactions and their rate constants used in mathematical simulation of the processes of radiolysis of aqueous 2-propanol with 2,4-dichlorophenoxyacetate

Reaction no.	Reaction	Rate constant, l mol^{-1} s^{-1}	References
1	$H + O_2 \rightarrow HO_2^{\bullet}$	2.1×10^{10}	[9, 10]
2	$e_{aq} + O_2 \rightarrow O_2^{-\bullet}$	1.8×10^{10}	[9, 10]
3	$H + HO^- \rightarrow e_{aq} + H_2O$	2.2×10^{7}	[9, 10]
4	$e_{aq} + H_2O_2(HO_2^-) \rightarrow HO^- + {}^{\bullet}OH(O^{-\bullet})$	4.7×10^{9}	[10, 11][a]
5	$e_{aq} + O_2^{-\bullet} \rightarrow H_2O_2 + 2\,HO^-$	1.3×10^{10}	[8–10]
6	$(Me)_2CHOH + H \rightarrow (Me)_2{}^{\bullet}COH + H_2$	8.2×10^{7}	[8]
7	$(Me)_2CHOH + {}^{\bullet}OH(O^{-\bullet}) \rightarrow (Me)_2{}^{\bullet}COH + H_2O(HO^-)$	1.7×10^{9}	[8][b]
8	$(Me)_2{}^{\bullet}COH + O_2 \rightarrow (Me)_2C(OH)OO^{\bullet}$	4.0×10^{9}	[12]
9	$(Me)_2C(OH)OO^{\bullet} \rightarrow (Me)_2CO + HO_2^{\bullet}$	683 s^{-1}	[1, 12]
10	$(Me)_2C(OH)OO^{\bullet} + HO^- \rightarrow (Me)_2CO + O_2^{-\bullet} + H_2O$	5.2×10^{9}	[12, 13]
11	$(Me)_2{}^{\bullet}COH + HO^- \rightarrow (Me)_2CO^{-\bullet} + H_2O$	7.0×10^{9}	[8]
12	$(Me)_2CO^{-\bullet} + O_2 \rightarrow (Me)_2CO + O_2^{-\bullet}$	4.0×10^{9}	[12]
13	$HO_2^{\bullet} + HO^- \rightarrow O_2^{-\bullet} + H_2O$	1.0×10^{10}	[10]
14	$O_2^{-\bullet} + O_2^{-\bullet} + 2\,H_2O \rightarrow H_2O_2 + O_2 + 2\,HO^-$	0.15	[1, 14, 15]
15	$HO_2^{\bullet} + O_2^{-\bullet} + H_2O \rightarrow H_2O_2 + O_2 + HO^-$	1.02×10^{8}	[1, 15]
16	$O_2^{-\bullet} + H_2O_2(HO_2^-) \rightarrow HO^- + {}^{\bullet}OH(O^{-\bullet}) + O_2$	15	[8, 15, 16]
17	$H_3O^+ + HO^- \rightarrow 2\,H_2O$	1.4×10^{11}	[8, 10]
18	$H_3O^+ + O_2^{-\bullet} \rightarrow HO_2^{\bullet} + H_2O$	5.1×10^{10}	[10, 17]
19	$(Me)_2CO + e_{aq} \rightarrow (Me)_2CO^{-\bullet}$	6.6×10^{9}	[8]
20	$H_2O_2 \rightarrow {}^{\bullet}OH + {}^{\bullet}OH$	1.33×10^{-7}	[10]
21	$ArCl_2 + e_{aq} \rightarrow Cl^- + {}^{\bullet}ArCl$	3.7×10^{9}	
22	$ArCl_2 + O_2^{-\bullet} \rightarrow Cl^- + {}^{\bullet}ArCl + O_2$	1.0	
23	${}^{\bullet}ArCl + (Me)_2CHOH \rightarrow (Me)_2{}^{\bullet}COH + HArCl$	2×10^{7}	[18][c]
24	${}^{\bullet}ArCl + O_2 \rightarrow {}^{\bullet}OOArCl$	1.6×10^{9}	[18][d]
25	${}^{\bullet}OOArCl + O_2^{-\bullet} + H_2O \rightarrow HOOArCl + O_2 + HO^-$	1×10^{8}	[18][e]
26	${}^{\bullet}ArCl + O_2^{-\bullet} + H_2O \rightarrow HOOArCl + HO^-$	1.6×10^{9}	[18][f]
27	$HOOArCl + O_2^{-\bullet} \rightarrow {}^{\bullet}OArCl + O_2 + HO^-$	5	[18][g]
28	$HOOArCl + e_{aq} \rightarrow HO^- + {}^{\bullet}OArCl$	5×10^{9}	[18][h]
29	${}^{\bullet}OArCl + O_2^{-\bullet} \rightarrow {}^{-}OArCl + O_2$	1.2×10^{9}	[18][i]
30	${}^{-}OArCl + O_2^{-\bullet} + H_2O \rightarrow {}^{\bullet}OArCl + HO_2^- + HO^-$	580	[18][j]
31	${}^{\bullet}OOArCl + (Me)_2CHOH \rightarrow HOOArCl + (Me)_2{}^{\bullet}COH$	100	[19][k]
32	${}^{\bullet}OArCl + O_2 \rightarrow OAr(Cl)OO^{\bullet}$	3×10^{8}	[20][l]
33	$HArCl + O_2^{-\bullet} \rightarrow Cl^- + {}^{\bullet}ArH + O_2$	0.2	

[a] Averaged rate constant corresponding to the ratio of concentrations H_2O_2/HO_2^- at pH = 12.8; [b] averaged value corresponding to the ratio of forms ${}^{\bullet}OH/O^{-\bullet}$ at pH = 12.8; [c] averaged value for reactions of radicals ${}^{\bullet}C_6H_5$ (k = 1.2×10^{7} l mol^{-1} s^{-1}) and 4-HO$^{-\bullet}C_6H_4$ (k = 3×10^{7} l mol^{-1} s^{-1}) with 2-propanol; [d] for reaction of radical ${}^{\bullet}C_6H_4$–CO_2^- with O_2; [e] for reaction of radical $CH_3CH(OCH_3)OCH_2OO^{\bullet}$ with $O_2^{-\bullet}$; [f] similar to reaction (24); [g] for reaction of $O_2^{-\bullet}$ with $(CH_3)_3COOH$; [h] for reaction e_{aq} + $(CH_3)_3COOH$; [i] for reaction 4-O_2C-2,6-$(CH_3O)_2$·$C_6H_2O^{\bullet}$ + $O_2^{-\bullet}$; [j] for reaction C_6H_5OH + $O_2^{-\bullet}$; [k] radicals $C_6H_5OO^{\bullet}$ are much more reactive than $CH_3OO^{\bullet}$ or substituted alkylperoxy radicals but less reactive than $CCl_3OO^{\bullet}$ [19]. Radical $(CH_3)_3COO^{\bullet}$ abstracts H atom from 2-propanol with k = 9×10^{-3} l mol^{-1} s^{-1} [1], while for $CCl_3OO^{\bullet}$ this constant is $\leq7\times10^{3}$ l mol^{-1} s^{-1} [18]; [l] rate constant for addition of oxygen to phenoxyl radicals.

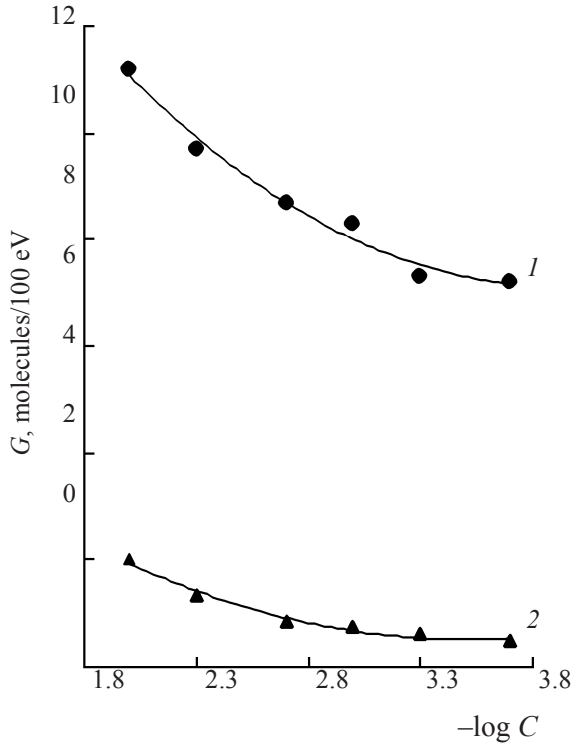

Fig. 1. Dependence of the radiation-chemical yield of (*1*) acetone and (*2*) chloride ions on the logarithm of concentration of sodium 2,4-dichlorophenoxyacetate in 2 M solution of 2-propanol saturated with oxygen (pH = 6.7).

mainly formed by reaction (21) (see table). The rate constant for reaction (21) was calculated by the method of competitive kinetics with respect to reaction (2) from the increase in the yield of the chloride ions with concentration of $ArCl_2$ (Fig. 1). The obtained value of $k_{21} = 3.7\times10^9$ l mol^{-1} s^{-1} is close to the rate constant $k = 4.7\times10^9$ l mol^{-1} s^{-1} for the reaction 1,2-$C_6H_4Cl_2 + e_{aq}$ [19]. The $^{\bullet}$ArCl radical formed in reaction (21) predominantly reacts with the molecule of propanol-2 [reaction (23)] to form the $(Me)_2C^{\bullet}OH$ radical, which is oxidized by oxygen to acetone. This is the reason for the rise in the yield of acetone with concentration of $ArCl_2$. With decrease in the $ArCl_2$ concentration the rate of reaction (21) decreases and the yields of the chloride ions and acetone drop sharply. For the chloride ions such a drop is related to a higher rate of reaction (2) as compared to that of reaction (21). In the range of $ArCl_2$ concentrations of 0.001–0.0002 mol l^{-1} the decrease in the yield of the chloride ions is substantially slower. The formation of chloride ions under these conditions is provided by reaction (22). The rate of reaction (2) of e_{aq} with O_2 at $ArCl_2$ concentration of 0.0002 mol l^{-1} is ~40 times as fast as that of reaction (21), and the radiation chemical yield of the chloride ions formed in reaction (21) should not exceed 2.7/40 ≈ 0.07 molecules/100 eV. However, the observed yield of the chloride ions G_{Cl}^{-} provided by reaction (22) is 0.48±0.05 molecules/100 eV (pH = 6.7). The effect of the pH of the solution on the yield of Cl^- ions proves the possibility of reaction (22) to occur. Thus, in an acidic medium, $O_2^{-\bullet}$ is protonated [reaction (34)] and the rate of reaction (22) decreases. The yield of the chloride ions decreases to 0.13± 0.01 molecules/100 eV at pH = 2.4.

$$O_2^{-\bullet} + H^+ \rightleftarrows HO_2^{\bullet}, \quad (34)$$
$$pK\ 4.7\ [1].$$

For calculation of the rate constant for reaction (22) mathematical simulation of the processes of radiolysis was employed. The simulation was performed in two steps. In the first step a mathematical model was developed for the radiolysis of the 2 M solution of propanol-2 saturated with oxygen [reactions (1)–(20)] at pH of the solution 12.8, which adequately described the process compared with the experimental data. In the second step, the developed model of radiolysis of the 2 M solution of propanol-2 was complemented by introduction of $ArCl_2$ in concentration of 0.0002 mol l^{-1}, reactions involving $ArCl_2$ and their rate constants. Variation of the rate constants for reactions (22) and (33) allowed to attain the coincidence of the calculated kinetics of accumulation of the radiolysis products with the experimentally obtained values. The choice of the pH value was determined by the following reasoning: (1) at this pH the $HO_2^{\bullet}$ radicals are completely converted into the superoxide ion [reaction (13)]; (2) radical $(Me)_2^{\bullet}COH$ has the value of pK = 12.2 [1] and to more than 50% exists in the anionic form, which is oxidized by oxygen to acetone with the formation of the superoxide ion [reaction (12)] at a high rate; (3) an alkali rapidly destroys hydroperoxide radicals $(Me)_2C(OH)OO^{\bullet}$ with elimination of the superoxide ion [1] [reaction (10)]. Therefore, at the chosen value of pH, the radical products of radiolysis of water, OH, e_{aq} and H, provide maximal generation of superoxide ions due to fast reactions, whereas the reactions of recombination of radicals $(Me)_2^{\bullet}COH$, $(Me)_2CO^{-\bullet}$, and $(Me)_2C(OH)OO^{\bullet}$ proceed with negligible rate because of their low concentration.

The reactions and their rate constants which were used in the calculations are presented in the table. The experimental rate constants for practically all radical

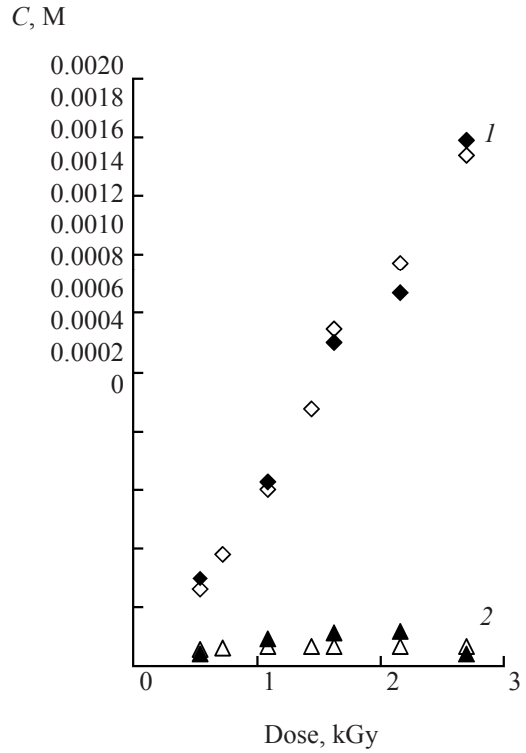

Fig. 2. Experimental (♦, ▲) and calculated (◊, Δ) values of concentration of (*1*) acetone and (*2*) hydrogen peroxide as a function of the dose of irradiation of 2 M solution of 2-propanol saturated with oxygen (pH = 12.8).

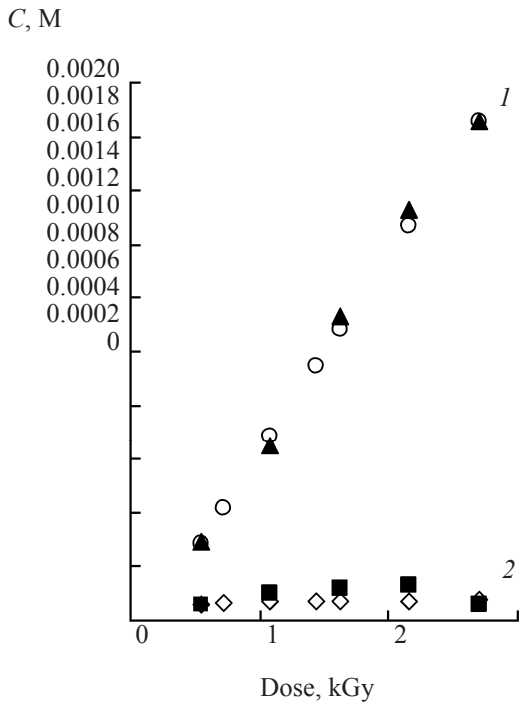

Fig. 3. Experimental (▲, ■) and calculated (○, ◊) values of concentration of (*1*) acetone and (*2*) hydrogen peroxide as a function of the dose of irradiation of 2 M solution of 2-propanol saturated with oxygen in the presence of 0.0002 mol l^{-1} of sodium 2,4-dichlorophenoxyacetate (pH = 12.8).

reactions proceeding at the radiolysis of aqueous solution of propanol-2 in the presence of oxygen are available in the literature. The largest scatter in the data on the rate constant was found for reaction (16): from 0.01 to 16 l mol^{-1} s^{-1} [16, 17]. This reaction (the Haber–Weiss reaction) was intensively studied since it plays an important role in biosystems being a source of hydroxy radicals. Due to scatter of the k_{16} values, only this value was varied when developing the mathematical model of radiolysis of the 2 M solution of propanol-2, other constants remained unvaryed. Only for k_{16} =15 l mol^{-1} s^{-1} the results of calculations were the most close to the experimental data. In Fig. 2 the calculated and experimental kinetics of accumulation of acetone and hydrogen peroxide obtained for aqueous–propanol solution at pH = 12.8 are presented. The optimized value of k_{16} = 15 l mol^{-1} s^{-1} refers to the summary reaction (35), since the pK for hydrogen peroxide is 11.6 [21].

$$O_2^{-\bullet} + HO_2^-(H_2O_2) \rightarrow O_2 + HO^- + O^{-\bullet}(^{\bullet}OH) \qquad (35)$$

When developing the mathematical model of radiolysis of aqueous–propanol solution with $ArCl_2$ reactions (1)–(33) were taken into consideration. True values for the rate constants of reactions (23–32) are lacking in the literature. Therefore, for calculations we used the literature values for the most similar reactions. The rate constants for reactions (22) and (33) were varied in the calculations. For k_{22} = 1 l mol^{-1} s^{-1} the best agreement between the calculated and experimental dependences of concentration of the chloride ions, acetone and peroxides on the dose of irradiation was achieved (Figs. 3 and 4). Reaction (22) provides 90% of the chloride ions. Taking into account reaction (33) allowed the achievement of a better agreement between the calculated and experimental data on concentration of Cl^- ions at large doses of irradiation. The obtained value of k_{22} is almost by one order of magnitude less than the rate constant for the reaction of aliphatic compound with two chlorine atoms, dichloromethane, with superoxide ion equal to 9 l mol^{-1} s^{-1} [5].

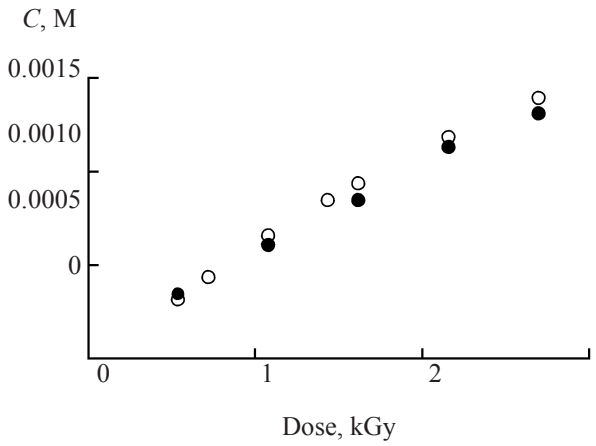

Fig. 4. Dependence of concentration of chloride ions on the dose of irradiation of the 2 M solution of 2-propanol saturated with oxygen in the presence of 0.0002 mol l^{-1} of sodium 2,4-dichlorophenoxyacetate (pH = 12.8). (●) Experiment and (○) calculations.

Formation of chloride ions was also observed in the radiolysis of the oxygen-saturated 2 M solution of 2-propanol with addition of PCP (0.0001 mol l^{-1}). At this low concentration of PCP practically all hydrated electrons react with oxygen. The formation of chloride ions is probably due to reaction (36) at pH 4.8–6.

$$O_2^{\bullet-} + C_6Cl_5OH \rightarrow O_2 + Cl^- + {}^{\bullet}C_6Cl_4OH, \quad (36)$$

$$HO_2^{\bullet} + C_6Cl_5OH \rightarrow H_2O_2 + C_6Cl_5O^{\bullet}, \quad (37)$$

$$O_2^{\bullet-} + C_6Cl_5O^- \rightarrow O_2 + Cl^- + {}^{\bullet}C_6Cl_4O. \quad (38)$$

The yield of chloride ions also depends on the pH of the solution. The largest yield was observed at pH = 4.8 and 6 and comprised 0.68±0.07 and 0.79± 0.07 molecules/100 eV, respectively. At pH = 2 G_{Cl^-} = 0.37± 0.04, and at pH = 7.6, G_{Cl^-} = 0.49± 0.05 molecules/100 eV. Decrease in the yield of the chloride ions in an acidic medium (pH = 2) is caused by the protonation of PCP and $O_2^{\bullet-}$ and by prevalence of reaction (37). The decrease in the yield of Cl^- ions in an alkaline medium is caused by deprotonation of PCP (pK = 4.7 [22]) and a lower rate of reaction (38) as compared to reaction (36).

Thus, superoxide ion can react with chloroaromatic compounds in water–alcohol media. The reaction leads to elimination of chloride ions and formation of phenyl radicals active in reactions of hydrogen atom abstraction. The effect of ionizing radiation on living bodies (under the conditions of enhanced radiation background) will promote the formation of $O_2^{\bullet-}$ and increase its stationary concentration. It is presumable that the presence of chloroaromatic impurities in living systems in conjunction with their irradiation will favor the development of radical processes damaging the biomolecules.

EXPERIMENTAL

The solutions were prepared from double distilled water and 2-propanol of "chemically pure" grade. The solutions were saturated with oxygen in a syringe so that the air in the solutions was replaced by oxygen. Sodium 2,4-dichlorophenoxyacetate and pentachlorophenol of "pure" grade were recrystallized. The irradiation was performed in sealed tubes. The power of the absorbed dose measured by ferrous sulfate dosimeter was 0.3 Gy s^{-1}. Analysis on chloride ions was made using the method of potentiometric titration with silver nitrate. Acetone was analyzed by GC with a flame ionization detector, carrier gas hydrogen. The concentration of oxygen in solutions was determined by the method of gas-adsorption chromatography, detector catharometer, carrier gas helium. Hydrogen peroxide was determined by the iodide method of Allen [23]. The extinction coefficient was 24240± 2640 l mol^{-1} cm^{-1} at 350 nm. Radiation chemical yields of the products of radiolysis were calculated from the dependences of their concentrations on the absorbed dose. The error in determination of the yields was 11% for chloride ions, 7% for acetone, and 12% for peroxides. Computer simulation of the kinetics of accumulation of the products of radiolysis was performed using the package of specialized programs POMSR [24, 25], in which the model of the process of radiolysis is approximated by the system of differential equations. The fourth-order Runge–Kutta method was used for numerical integration of these equations.

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