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Kinetics of radiation-stimulated processes on the surface of oxide materials

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The analysis of the experimental data shows that the processes of gas adsorption and radiation defects accumulation in metal oxides correlate with each other and most likely can be described in terms of equivalent kinetic equations. Given this circumstance, the kinetics of accumulation of radiation defects in oxides of different metals was analyzed. The obtained equations were used to analyze: a) the kinetics of accumulation of radiation defects in different oxide compounds; b) the data on the destruction of radiation-induced defects in the atmosphere of different gases, and on the kinetics of absorption by oxides of oxygen, hydrogen, and carbon dioxide molecules. The results of such analysis are systematized and are given in the form of a table. The following conclusions were made: 1. The quantum yield of radiation defects increases monotonically with growth of the temperature of processing, tending to a certain limit value. 2. The constant of destruction of radicals from ionizing radiation increases as well. 3. The ratio of the number of surface and bulk defects in different oxides can be arranged in the following series: silicon oxide > beryllium oxide > aluminum oxide. Thus, the most optimal (convenient) material for creating absorbing systems by energy intensity is silicon dioxide, and by adsorption efficiency is beryllium oxide.

Keywords: kinetics, adsorption, defects, radiation, oxides.

Introduction

The study of adsorption in the field of ionizing radiation opens new perspectives for establishing the nature of electronic factors, free and localized carriers of electric charges in elementary acts of surface processes. Ionizing radiation reveals the ability of adsorbed molecules to serve as electron and hole traps and to compete with some other traps – structural defects and admixtures located on the surface of solids [1-9].

Another feature observed with radiation with γ -quanta and some other types of high-energy particles is possible radiation sintering and structural transformations at high doses. When irradiated with heavy particles (α -rays, protons, neutrons, helium ions), radiation adsorption is accompanied with cascade processes and can cause electronic excitation, ionization, and atomic displacement in a solid. As a result of such interactions, the number of formed radiation defects significantly exceeds the number of defects arising from γ -irradiation, and, consequently, the number of adsorption centers. When high energy ions penetrate solids, the material along the path of the ion beam changes, atoms change their positions, molecules can break into pieces, and ordered structures – such as, for example, crystalline ones – are destroyed [10-25].

As a result of all the above factors, we can observe, in experiments on the study of radiation adsorption, pictures of the type shown in Figures 1 and 2 with saturation and, sometimes, decrease (bakeout) of a number of adsorbed molecules. In our case, we consider only processes with saturation.

The aim of this work is an attempt to describe, using kinetic equations, radiation-adsorption processes on the surface of some oxides.

Results and discussion

The combined data of numerous experiments show that the limiting number of radiation adsorption centers does not strongly depend on the type of radiation and on the energy of the bombarding particles [12-17]. The difference, sometimes reaching one exponent, is, apparently, due to structural features of oxides. As an example, the article presents experimental data obtained with the electronic-paramagnetic resonance method (EPR) (Figure 1) and the volume-adsorption method (Figure 2).

The dependence of the number of adsorbed molecules on the temperature of the preliminary heat treatment with the minimum of 673 K was discovered, which can be associated, on the one hand, with curing of adsorption centers, and, on the other hand, with increase of the surface of samples and associated with this growth of adsorption centers (Figure 2). At the same time, the dependence of paramagnetic centers on the temperature is somewhat different – at the temperature of 673 K, on the contrary, the maximum number of paramagnetic centers is observed (Figure 1). Further increase of the annealing temperature is accompanied by decrease of paramagnetic centers, which, possibly, is associated with anneal of paramagnetic centers.

The analysis of the experimental data shows that the processes of gas adsorption and accumulation of radiation defects in metal oxides correlate with each other and most likely can be described in terms of equivalent kinetic equations. Taking this into account, the kinetics of accumulation of radiation defects in oxides of different metals was analyzed.

The equation for accumulation of radiation defects has the form [9]:

$$dn/dt = GP - k_1Pn - k_2Pn^2 - \sum k_n(f_n), \quad (1)$$

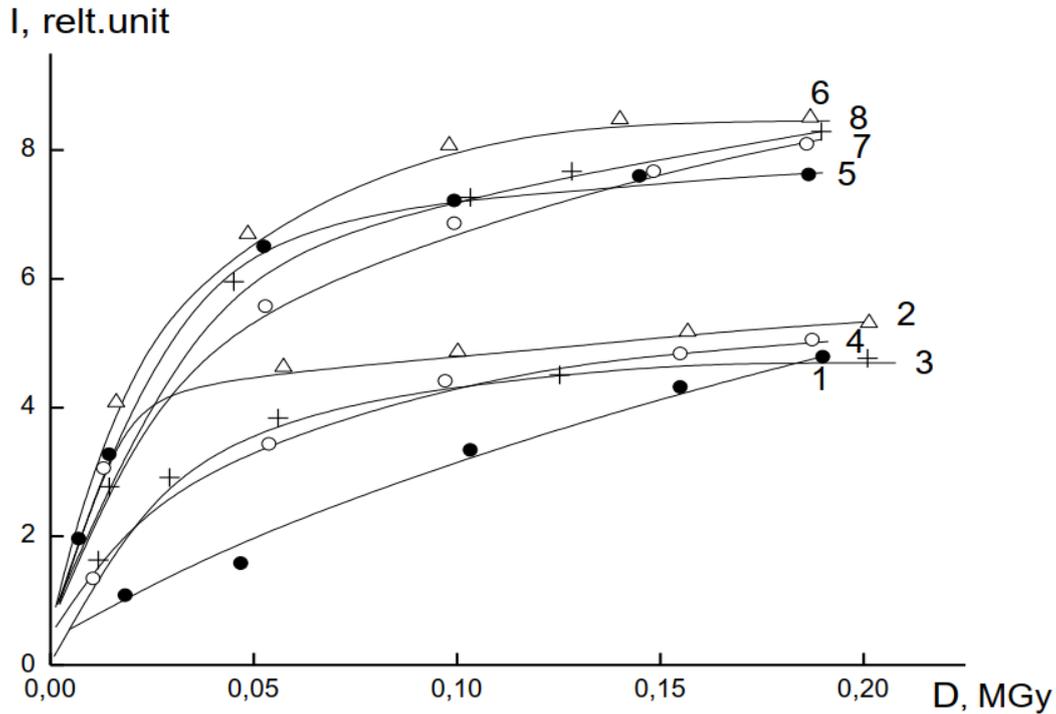


Figure 1. Dependence of paramagnetic centers on SiO_2 on the radiation dose.
 1.5- $T_{\text{heat}} = 473$ K, 2.6-673 K, 3.7-873 K, 4.8-1073 K; 1-4-vacuum-processed samples;
 5-8- O_2 bleeding in ($P_0 = 16$ Pa).

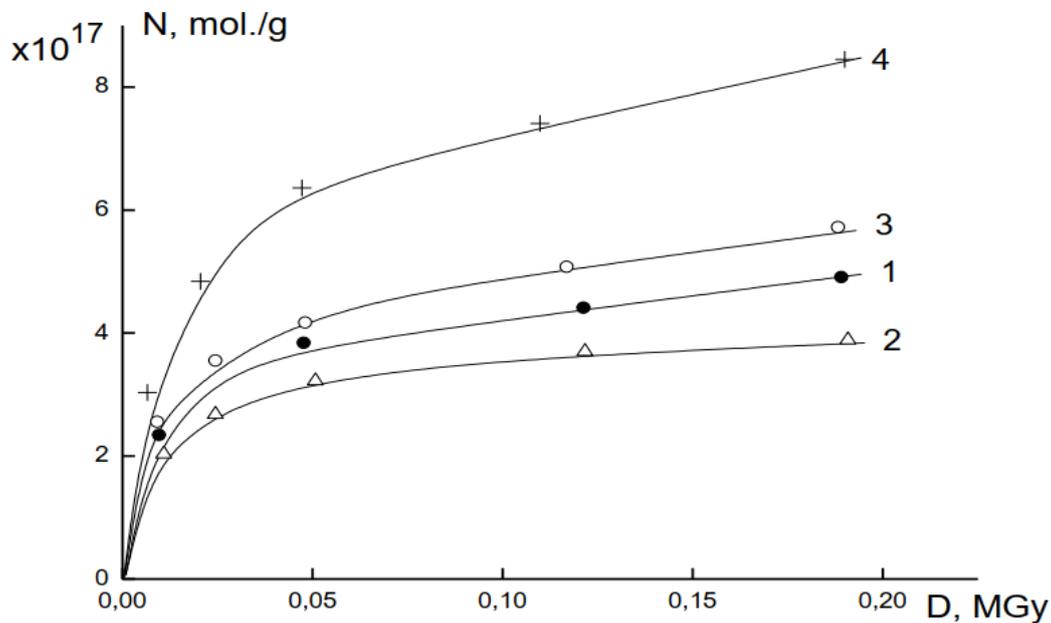


Figure 2. Dependence of adsorption of O_2 on SiO_2 on γ -radiation dose.
 1- $T_{\text{heat}} = 473$ K, 2-673 K, 3-873 K, 4-1073 K ($P_0 = 9.3$ Pa).

where: n is concentration of radiation defects (in our case, paramagnetic centers of adsorption); P – radiation power (Pt -radiation dose); k_1 – the constant of radiation-induced defects (paramagnetic centers) destruction rate; k_2 – the constant of defect recombination rate; $\sum k_n (f_n)$ are terms of the equation that take into account any other possible mechanisms of radiation defects destruction

(for example, owing to interaction of defects with oxygen dissolved in metals and with some other impurities).

The first term on the right side of equation (1) is responsible for the formation of defects. So, in the initial section of the curve of accumulation of radiation defects (where the loss of defects can be neglected), the solution of equation (1) will have the form:

$$n = GPt = GD, \quad (2)$$

Therefore, the initial section of the curve of accumulation of radiation defects is a straight line, the slope ratio of which corresponds to the quantum yield of radiation defects:

$$\operatorname{tg}\alpha = G = n_{lim}/D_{lim}, \quad (3)$$

where, n_{lim} and D_{lim} are concentration and dose values at any point we choose on a straight-line section of the accumulation curve.

If we consider the destruction of defects by the mechanism of their interaction with the radiation field, then the solution of equation (1) will take the form:

$$n = n_{lim}[1 - \exp(-k_1Pt)], \quad (4)$$

where $n_{lim} = G/k_1$ is the limit value of concentration of radiation defects.

It is seen from (4) that with $k_1 Pt = k_1 D \geq 3$ the accumulation curve goes on the plateau; the concentration of defects reaches its maximum (limit) value:

$$n = n_{lim} = G/k_1. \quad (5)$$

Thus, if from the experiment n_{lim} is known and the quantum yield G is determined from the initial section of the accumulation curve, then, according to (5), the constant of destruction of defects under the influence of ionizing radiation can be calculated as follows:

$$k_1 = G/n_{lim}, \quad (6)$$

If destruction of radical defects occurs as a result of their recombination, then the solution of equation (1) takes the following form:

$$n = n_{lim} \operatorname{th}(n_{lim}k_2D), \quad (7)$$

where

$$n_{lim} = \sqrt{G/k_2}, \quad (8)$$

Now, let us consider the process of destruction of radiation defects when oxygen molecules enter the ampule with an irradiated sample. In this case, the kinetics of destruction of radiation defects will be described by the following equation:

$$n(t) = n_{max} \exp(-k_4 \tau), \quad (9)$$

where τ is the time of interaction of the studied sample in the oxygen atmosphere the obtained equations were used for the analysis of:

- a) the kinetics of accumulation of radiation defects in different oxide compounds;
- b) the data of destruction of radiation defects in the atmosphere of different gases and kinetics of adsorption of oxygen, hydrogen, and carbon dioxide molecules by oxides.

The results of such analysis are systematized and are presented in the following table.

Table 1.

The Key Kinetic Parameters of Oxygen Radiation Adsorption on Oxides.

Oxide	$G, 10^{-6} \text{ rad}^{-1}$	$k, 10^6 \text{ rad}^{-1}$	N_{sur}/n_{vol}	$N_{ads} 10^{15} \text{ mol/g}$	Optimal condition	
					$D, \text{ Mrad}$	$T_{heat}, \text{ K}$
silicon	1.3÷4.0	0.2÷0.7	≈1.6	2.8	20÷40	1200±100
beryllium	2.8÷8.5	0.2÷2	1.0±0.2	≈23	8÷15	700±50
aluminum	2.7÷5.5	0.5÷1.2	≈0.2	2.7	≥10	700±50

Conclusion

Radiation-adsorption processes (Figure 1) and accumulation of irradiation defects (Figure 2) on the surface of oxides are satisfactorily described by a kinetic equation of the exponential nature. The quantum yield of radiation-induced defects increases monotonically with an increase in the treatment temperature, tending to a certain limiting value (Table 1). The constant of destruction of radicals by ionizing irradiation also increases (and the growth rate k_1 exceeds the growth rate G). The ratio of the number of surface and bulk defects in different oxides $n_{sur}/n_{bulk} = \gamma$, characterizing the absorbing activity of these materials, can be placed in the following row: silicon oxide > beryllium oxide > aluminum oxide (Table 1). Thus, the most optimal (convenient) material for creating absorbing systems by energy intensity is silicon dioxide, and by adsorption efficiency is beryllium oxide.

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