

Modeling the Processes of the Dissolution of Chromium(III) and Copper Oxides in an Acidic Environment

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Abstract—Based on the experimental studies of the dissolution of Cr₂O₃ and CuO in an acidic environment, as well as investigation of the effect of acids, pH, and concentration of anions on the rate of dissolution of the oxides, the nature of the limiting stage is determined which consists in the formation of surface compounds (depending on the acidity of the environment) of the MeOH_s^{x+} and MeHSO_{4s}^{x+} types and their subsequent transition into the solution of the electrolyte. The comparative analysis of the dissolution of the oxides of two *d* metals is performed.

Keywords: chromium(III) oxide, copper oxide, heterogeneous kinetics, acid–base equilibria

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Oxides of many *d* elements can be used as catalysts as well as additives for improving the corrosion resistance of stainless steel. Chromium oxide and copper oxide are no exception. Particles of CuO of 10^{−9} m can be used in agriculture as fertilizers or pesticides. To model the processes of dissolution of chromium(III) and copper oxides in acids, they should be analyzed from the perspective of heterogeneous kinetics, the conditions of the existence of a kinetic region where the rate of removal (feeding) of the dissolution products slightly affects the kinetics of the process should be identified, and the factors affecting the kinetics of the interaction of the oxides with electrolytes should be found. In this respect, the need emerged to determine the nature of the limiting stage, model the kinetics of dissolution, provide the scheme of the process of dissolution of Cr₂O₃ and CuO in acids, and derive mathematical equations describing the dependence of the rate of dissolution of the oxides on different parameters based on the obtained data.

MATERIALS AND METHODS

To study the kinetics of dissolution, fractions of powder chromium(III) and copper oxides with particles of 10^{−5}–10^{−4} m were used. The composition and structure of the samples were verified via X-ray phase analysis (an HZG4/2 diffractometer, Cu α radiation) and thermogravimetry (a PH Q-1500D derivatograph). The specific surface area was determined by

the Brunauer–Emmett–Teller (BET) method on a Sorpti-1750 adsorption instrument by the adsorption of the standard gas (nitrogen) at 77.4 K. The surface determined by BET was 5 and 100 ± 10 m²/g for α -Cr₂O₃ and CuO, respectively.

A 500-mg sample was introduced into a reaction vessel containing 1 L of an aqueous solution of sulfuric, nitric, hydrochloric, chloric, or phosphoric acid with a known concentration and pH. The experiments were performed in a thermostated reactor at 298 to 343 K. To find the order of the rate of dissolution with respect to the hydrogen ions, the kinetic curves of the process of dissolution were studied at different pH values as well as constant values of the concentration of the anions and temperature. The pH of sulfuric acid solutions was adjusted by adding H₂SO₄ to the solution. The concentration of chromium(III) ions was determined by the procedures based on the application of the diphenylcarbazide method. The concentration of copper was determined spectrophotometrically using formaldoxime [1–3]. Specimens with a volume of 1 to 5 mL were periodically collected using a glass Schott filter no. 4 by generating a vacuum in the tube intended for the collection of the filtrate. The fraction of the dissolved oxide was determined as the ratio of the concentration of the metal ions (optical density of the solution *D*) at the time of collection of the specimen (*C_t*) to the concentration of the metal ions upon complete dissolution of the oxide (*C_∞*).

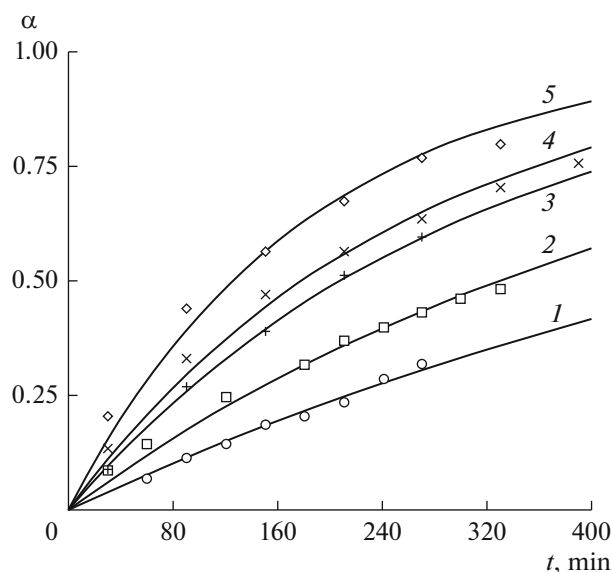


Fig. 1. Dependence of the fraction of dissolved Cr_2O_3 on time ($T = 333 \text{ K}$, pH of 2) at different concentrations of sulfuric acid: (1) 0.10, (2) 0.40, (3) 0.50, (4) 1.00, and (5) 2.00 mol/L.

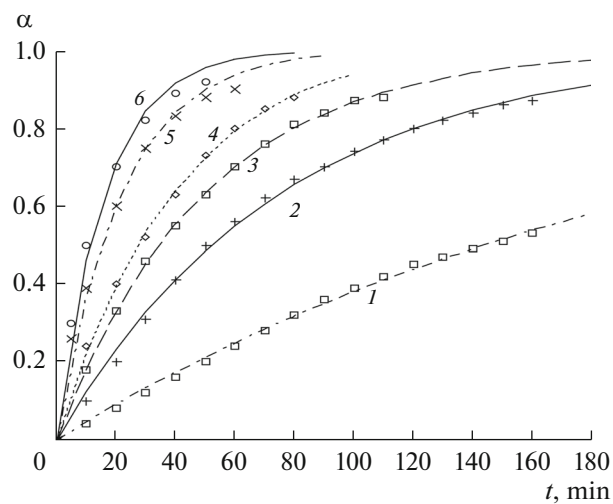


Fig. 2. Dependence of the fraction of dissolved CuO on time ($T = 293 \text{ K}$, pH of 2.05) at different concentrations of sulfuric acid: (1) 0.25, (2) 0.5, (3) 1.0, (4) 2.5, (5) 5.0, and (6) 7.5 mol/L.

RESULTS AND DISCUSSION

Investigation of the Effect of the Concentration of Acids on the Kinetics of Dissolution of Cr_2O_3 and CuO

The obtained experimental data on the effect of the concentration of the acid on the kinetics of dissolution of chromium(III) and copper oxides are presented in Figs. 1 and 2 (the points are the experimental data and

the lines are the graphical solution of the equation of chain mechanism).

The analysis of the data presented in Figs. 1 and 2 shows that the fraction of the dissolved oxide increases with the increase in the concentration of the acid. At the initial time point, an acceleration of the process of dissolution of the oxides, which is associated with the etching of the surface, was observed [4]. At the final time point, incomplete dissolution of the oxide was noted, which is probably associated with the equilibration in the oxide–water–acid anion system. Similar results were obtained for chromium(III) oxide in solutions of nitric, phosphoric, and oxalic acids, and for copper oxide, in solutions of hydrochloric and chloric acids. The kinetics curves of dissolution of chromium(III) oxide and copper oxide in the acids under study have a σ -shaped character. The nature of the acid substantially affects the rate of dissolution. Under comparable experimental conditions, the rate of dissolution of copper oxide grows in the following series of inorganic acids [5]:



For chromium(III) oxides, the experimental kinetic data upon dissolution in HNO_3 , H_3PO_4 , H_2SO_4 , and $\text{H}_2\text{C}_2\text{O}_4$ are presented in Fig. 3.

Under comparable experimental conditions, the rate of dissolution grows in the following series of acids [6]:



The slowness of the rate of dissolution in nitric acid can be explained by the fact that it passivates the surface of chromium(III) oxide.

Investigation of the Effect of pH on the Kinetics of Dissolution of Cr_2O_3 and CuO

Figures 4 and 5 present the experimental data on the dissolution of Cr_2O_3 and CuO at various values of pH (the points are the experimental data and the lines are the graphical solution of the equation of chain mechanism). As follows from the data presented in Figs. 4 and 5, an increase in the rate of dissolution of chromium(III) and copper oxides is observed in the case of an increase in the concentration of hydrogen ions. The analysis of the data has shown that, for copper oxide, a decrease in the concentration of hydrogen ions to a pH of ~ 8.5 leads to the deceleration of dissolution; here, a change in the order of the rate of dissolution with respect to hydrogen ions from 0.5 ($0 < \text{pH} < 2$) to 1.0 ($\text{pH} > 3$) is observed.

Investigation of the Effect of the Temperature of the Solution on the Kinetics of Dissolution of Cr_2O_3 and CuO

The effect of temperature on the kinetics of dissolution of the oxides is presented in Figs. 6 and 7. It has been shown that, with the growth in the temperature

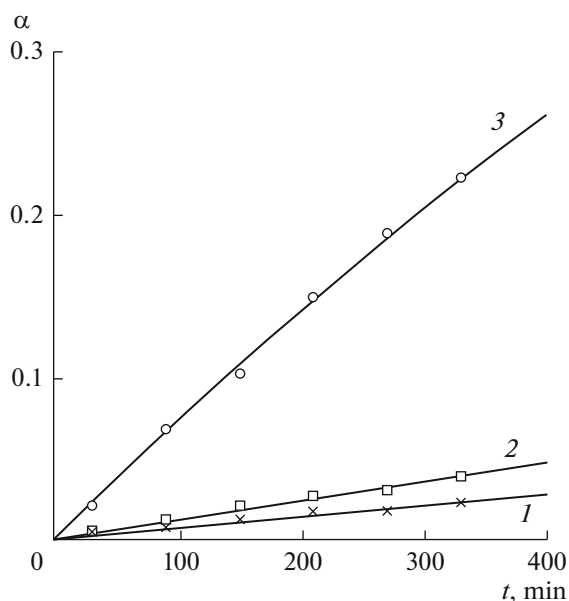


Fig. 3. Dependence of the fraction of dissolved chromium(III) oxide on time ($T = 333$ K) in different acids ($C_{\text{acid}} = 1$ mol/L): (1) H_3PO_4 , (2) HNO_3 , and (3) H_2SO_4 .

(from 293 to 343 K), the fraction of the dissolved oxide increases. The value of the apparent activation energy of dissolution of copper(II) oxide in acidic environment is 50–80 kJ/mol, while this parameter is 40–60 kJ/mol for chromium(III) oxide. Such a value of the activation energy makes it possible to judge about the fact that the process is limited by kinetic processes (probably, by the breaking of the Cu–O–Cu and Cr–O–Cr bonds) rather than by diffusion processes (in the solution and solid phase) as is often considered.

Analysis of the Kinetic Data with the Use of the Perceptions of Formal Heterogeneous Kinetics [7–15]

As is seen from Figs. 1–7, the kinetic curves have a σ -shaped character, which is indicative of a great number of active dissolution sites. To determine the specific rate of dissolution (W_i), effective activation energy (E_a), and orders of the reaction with respect to different ions (n_i), the kinetic curves were analyzed by the method of affine transformations. The obtained invariability of the $\alpha - \tau/\tau_{0.5}$ curves in relation to the type of the oxide, values of pH and temperature, and composition of the solution is indicative of a single mechanism of the process of dissolution is independent of the parameters under study; i.e., the sequence of dissolution stages remains unchanged.

The principle of the theory of dissolution of chromium and copper oxides in aqueous solutions proposed by us, which is based on the use of a model of

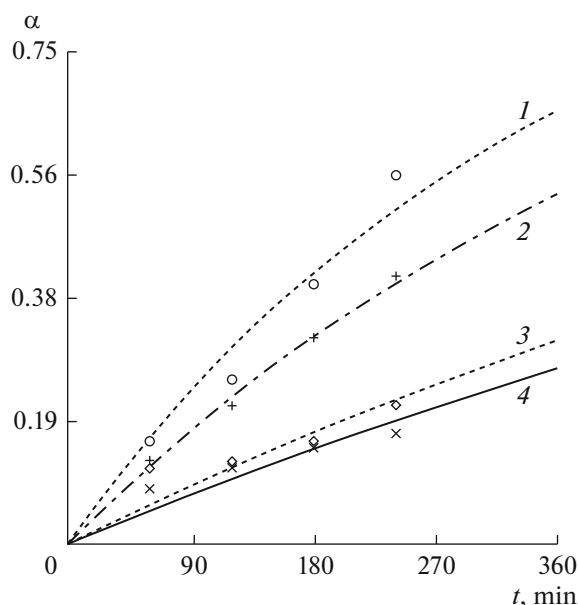


Fig. 4. Dependence of the fraction of dissolved chromium(III) oxide on time upon dissolution in H_2SO_4 at different values of pH: (1) -0.12 , (2) 0.125 , (3) 0.72 , and (4) 1.35 ($T = 333$ K, $C_{\text{acid}} = 0.1$ mol/L).

acid-based equilibria consists of the following points: particles $-\text{M}-\text{OH}_{2\text{Ssol}}^+$ and $-\text{M}-\text{OH}_2^+ \dots A_{\text{Ssol}}^-$, which can be considered as the intermediate compounds of the process of dissolution, are mainly present on the surface of the oxides in acidic solutions. The dissolution sites possess acid–base properties. Depending on the coordi-

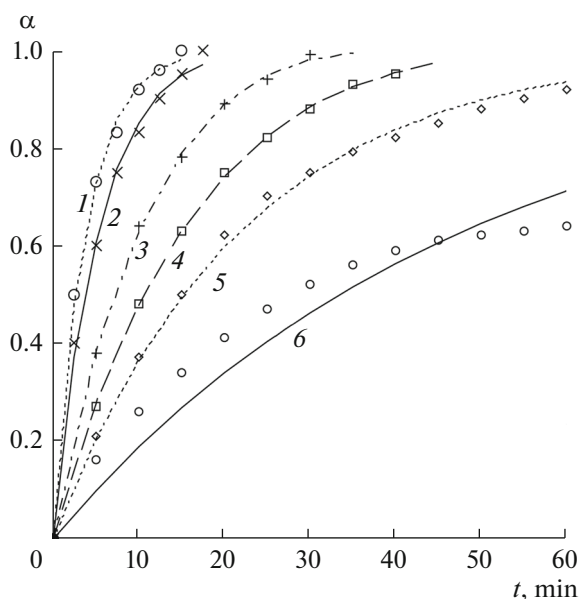


Fig. 5. Dependence of the fraction of dissolved copper oxide on time upon dissolution in H_2SO_4 at different values of pH: (1) -0.40 , (2) 0.08 , (3) 0.70 , (4) 0.95 , (5) 1.16 , and (6) 1.47 ($T = 333$ K, $C_{\text{acid}} = 0.1$ mol/L).

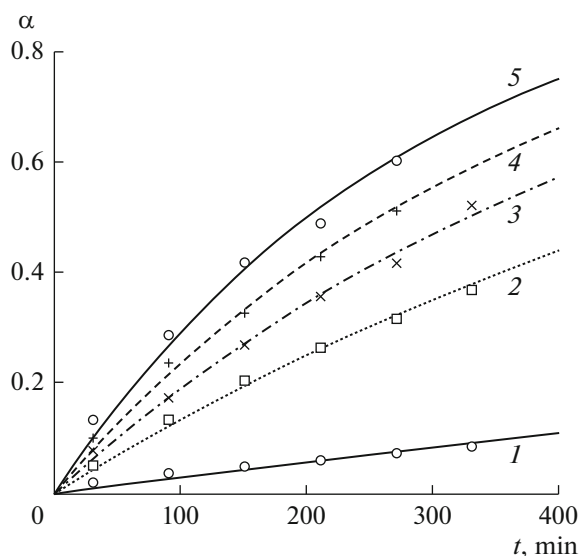


Fig. 6. Dependence of the fraction of dissolved Cr_2O_3 on time in 0.1 mol/L H_2SO_4 at different values of temperature: (1) 298, (2) 313, (3) 323, (4) 333, and (5) 343 K.

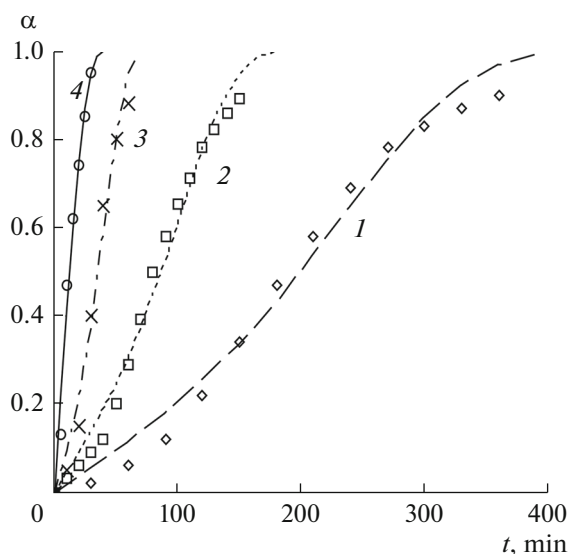


Fig. 7. Dependence of the fraction of dissolved CuO on time upon dissolution in 0.1 mol/L H_2SO_4 at different values of temperature: (1) 293, (2) 303, (3) 313, (4) 333, and (5) 323 K.

nation of metal ions by oxygen atoms, bond types of the addition of H^+ ions from a solution by the $\text{M}-\text{O}-\text{M}$, MOH^+ , etc., bonds are distinguished. These effects are associated with the different electronegativities of the oxygen ions on the surface of the oxide phase.

The addition of a proton induces the breakup of the $-\text{M}-\text{O}-\text{M}-$ bridge bond, which leads to the appearance of not only Brønsted but also Lewis sites (of the $-\text{M}^+$ type). As a result of this, dissociative adsorption of water molecules and appearance of a double electric layer occur. In the ideal case, fast dynamic equilibria appear which proceed two-to-three orders of magnitude more intensively than the process of dissolution with respect to the rate. To answer the question whether the process of dissolution is electrochemical or chemical, let us quantitatively consider a model based on the chemical perceptions only.

The process of the chemical dissolution of oxides can be represented by Scheme 1.

In this process, the limiting stage is the transition of the intermediate complex from the surface of the solid phase to the solution with a constant speed k_2 :

$$W_i = k_2 \theta, \quad (1)$$

where θ_1 is the degree of coverage of the surface of the oxide by the complexes of the $-\text{M}-\text{OH}_2^+ \dots \text{A}_S^-$ type.

By means of the joint solution of Eqs. (3)–(5) with the use of the method of kinetic analysis based on the application of the methods of stationary approximation and detailed analysis of equilibrium to the stages preceding the limiting stage, we find that

$$d\theta_0/dt = k_1[\text{H}^+][\text{A}^-]\theta_0 + k_{-1}\theta_1 \approx 0; \quad (3)$$

$$d\theta_1/dt = k_1[\text{H}^+][\text{A}^-]\theta_0 - (k_{-1} + k_2)\theta_1 \approx 0; \quad (4)$$

$$\theta = \theta_1 + \theta_0, \quad (5)$$

where θ_0 and θ_1 are the degrees of coverage of the surface of the oxide by the particles $-\text{M}-\text{OH}_S^0$ and $-\text{M}-\text{OH}_2^+ \dots \text{A}_S^-$, respectively.

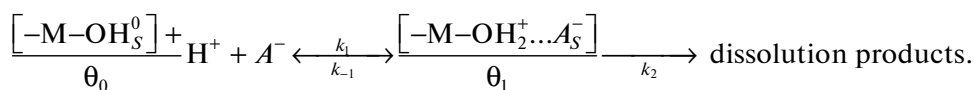
The values of θ_1 and W_i were calculated. The final expressions for θ_1 and W_i is as follows:

$$\theta_1 = \theta_0[\text{H}^+][\text{A}^-] \frac{k_1}{k_{-1} + k_2} = \theta \frac{[\text{H}^+][\text{A}^-]}{[\text{H}^+][\text{A}^-] + K_{\alpha 1}}, \quad (6)$$

where $K_{\alpha 1} = (k_{-1} + k_2)/k_1$;

$$W_i = W_{\max}^0 \frac{[\text{H}^+][\text{A}^-]}{K_{\alpha} + [\text{H}^+][\text{A}^-]}, \quad (7)$$

where $W_{\max}^0 = k_2 \theta$.



Scheme 1.

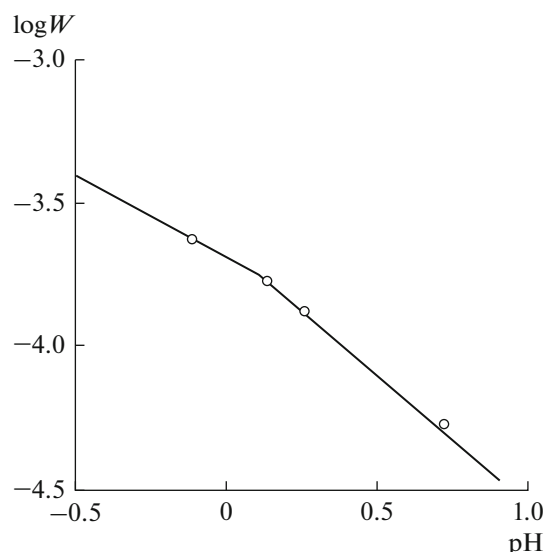


Fig. 8. Dependence of the logarithm of the rate of dissolution of chromium(III) oxide on pH ($T = 333$ K, $[\text{SO}_4^{2-}] = 0.135$ mol/L).

To analyze the experimental results on the kinetics of dissolution of the oxides, it is better to use the modified equation

$$\frac{1}{W_i} = \frac{1}{W_{\max}^0} + \frac{K_\alpha}{W_{\max}^0} \frac{1}{[\text{H}^+][\text{A}^-]}, \quad (8)$$

The coefficient W_{\max}^0 corresponds to the maximum rate of dissolution of the oxide; and the quantity K_α , to the reverse dissolution constant $K_{\alpha 3}$.

Let us consider the applicability of the found equations by way of example of the effect of sulfuric acid on the rate of dissolution of chromium(III) oxide (Fig. 8). The acid-based model makes it possible to describe the kinetic data on the dissolution of chromium oxides in sulfuric and other acids. Modeling using the Hougen–Watson method made it possible to calculate the dependence of the rate of dissolution on the pH and concentration of the solution [7–12, 16–17, 19–24]. According to the existing perceptions, the process of dissolution occurs on the sites of the oxides which adsorbed the protons or $\text{H}^+ \dots \text{A}^-$ or $\text{OH}^- \dots \text{K}^+$ ionic pairs occurring as a result of the acid-based equilibrations at the oxide/electrolyte boundary.

According to the data of infrared spectroscopy, the particles determining the rate of dissolution such as Cr^{3+} , CrOH^{2+} , $\text{Cr}(\text{OH})_2^+$, $\text{Cr}(\text{OH})_3^0$, and $\text{Cr}(\text{OH})_4^-$ can be present on the surface of chromium(III) oxide preliminarily exposed in the solutions of electrolytes (at different values of pH) [6]. For copper oxide, according to the data of infrared spectroscopy, the presence of copper ions and hydroxo complexes with the compositions Cu^{2+} , CuOH^+ , $\text{Cu}(\text{OH})_2$, $\text{Cu}(\text{OH})_3^-$, and $\text{Cu}(\text{OH})_4^{2-}$

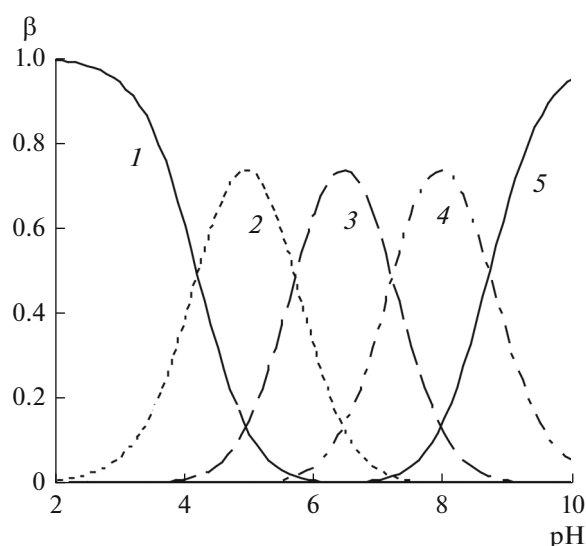


Fig. 9. Distribution of chromium(III) hydroxo complexes at different values of pH: (1) Cr^{3+} , (2) CrOH^{2+} , (3) $\text{Cr}(\text{OH})_2^+$, (4) $\text{Cr}(\text{OH})_3$, and (5) $\text{Cr}(\text{OH})_4^-$.

has been found on the surface of copper(II) oxide [13, 18].

When modeling the mechanism of dissolution of copper oxide, it was assumed that one of the aforementioned copper(II) ions, being an active dissolution site, can enter the solution. For this, the distribution of copper and chromium hydroxo ions in the solution at different values of pH was calculated. A system of equations of the material balance with respect to the base components of the hydrolysis reaction (hydrogen and copper ions or hydrogen and chromium ions), as well as systems of equations of stepwise hydrolysis, were used for the calculation. The relative distribution of the hydrolysis ions on pH was found [25]. The results of the calculation are presented in Figs. 9 and 10.

It is seen from Fig. 9 that a chromium(III) ion is susceptible to hydrolysis, and all the main forms are within pH values of 3 to 10.

It can be found from the analysis of the calculation which particles can be present on the surface of the oxide being dissolved. Let us present an example of the calculation based on copper oxide. For the quantitative description of the kinetic regularities, their surface concentration, which is expressed through the bulk concentration, should be known. Assuming that the surface concentration of the aforementioned ions is associated with the bulk Langmuir isotherm, we calculated the dependence of the rate of dissolution of the oxide on the concentration of the adsorbed ions (including hydrogen ions). In this case, the rate of dissolution, taking into account the surface concentra-

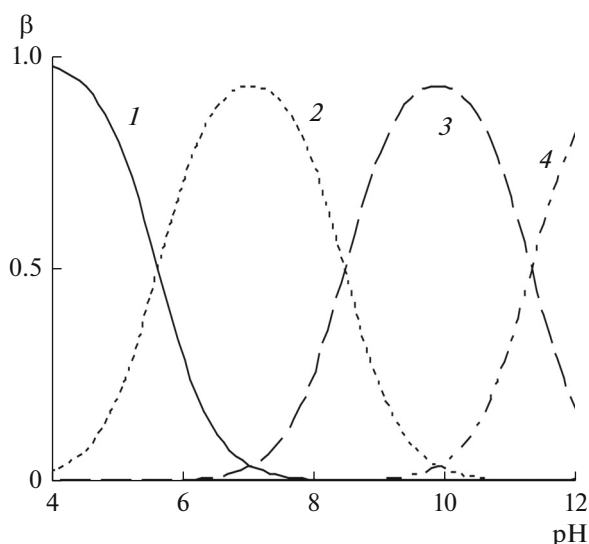


Fig. 10. Distribution of copper hydroxo ions in solutions at different values of pH: (1) Cu^{2+} , (2) $\text{Cu}(\text{OH})^+$, (3) $\text{Cu}(\text{OH})_2^0$, and (4) $\text{Cu}(\text{OH})_3^-$.

tion of the particles $\text{Cu}(\text{OH})_n^{2-n}$ will be expressed by the equation

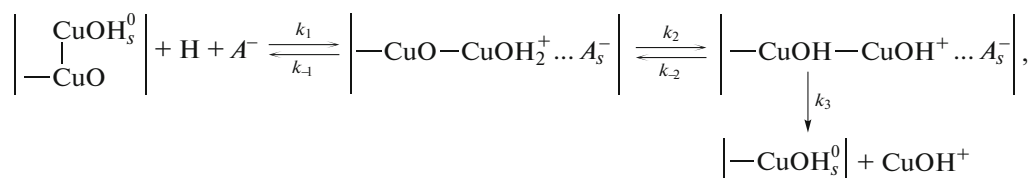
$$W = W_1^0 G(\text{H}^+) G(\text{Cu}(\text{OH})_n^{2-n}). \quad (9)$$

The dependence of the rate of dissolution W on the bulk concentration of the surface-active particles can be obtained by expressing the surface concentration of hydrogen ions and copper(II) or chromium(III) hydroxo ions through the bulk concentrations using the Langmuir equation [10].

The following expressions are used depending on the surface-active ion:

Cu^{2+} ions

$$W = W_{\max} \left(\frac{[\text{H}^+]}{[\text{H}^+] + K_1} \right) \left(\frac{\alpha_0}{\alpha_0 + K_2} \right) [\text{Cu}_{\text{total}}^{2+}]. \quad (10)$$



Scheme 2.

When performing the modeling for chromium(III) oxide using similar equations, it was obtained that the equation

$$W_2 = W_2^0 \left(\frac{[\text{H}^+]}{[\text{H}^+] + K_1} \right) \left(\frac{[\text{CrOH}^{2+}]}{[\text{CrOH}^{2+}] + K_2} \right), \quad (13)$$

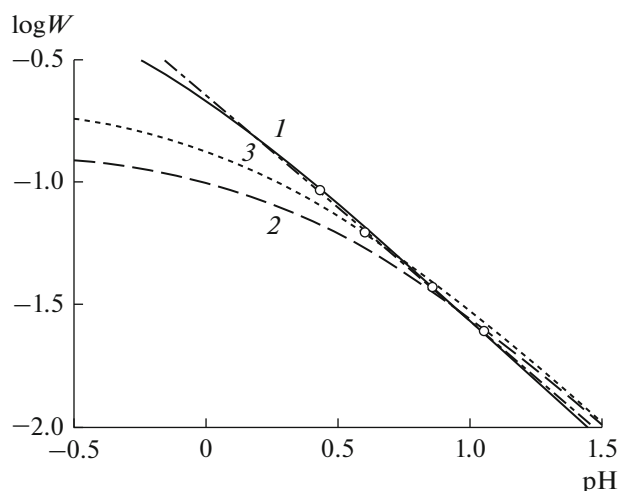


Fig. 11. Model dependence of the logarithm of the rate of dissolution of copper(II) oxide on pH of the solution of sulfuric acid under the condition that the following particles go into the solution: (1) CuOH^+ , (2) Cu^{2+} , and (3) $\text{Cu}^{2+} + \text{CuOH}^+$ (the points denote the experiment).

CuOH^+ ions

$$W = W_{\max} \left(\frac{[\text{H}^+]}{[\text{H}^+] + K_1} \right) \left(\frac{\alpha_1}{\alpha_1 + K_2} \right) [\text{Cu}_{\text{total}}^{2+}], \quad (11)$$

CuOH^+ and Cu^{2+} ions

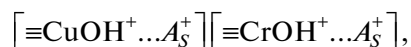
$$W = W_{\max} \left(\frac{[\text{H}^+]}{[\text{H}^+] + K_1} \right) \left(\frac{\alpha_0 + \alpha_1}{\alpha_0 + \alpha_1 + K_3} \right) [\text{Cu}_{\text{total}}^{2+}], \quad (12)$$

where K_1 , K_2 , and K_3 are the adsorption constants equal to the reciprocal value of the adsorption constant in the isotherm and α_0 and α_1 are the relative fractions of the Cu^{2+} and CuOH^+ ions, respectively.

The results of the modeling of Eqs. (10)–(12) presented in Fig. 11 showed that, on the surface of copper oxide, CuOH^+ was the most probable particle which was a surface intermediate compound and went into the solution of the electrolyte.

was correlated in the best way with the experimental data. In this case, the rate of dissolution of the oxide was determined by the transition of the surface-active particle CrOH^{2+} into the solution. The calculations showed that, in the case of a low concentration of hydrogen ions, CrOH^{2+} was the surface-active particle

determining the rate of dissolution. A CrHSO_4^{2+} ion becomes the surface-active particle determining the rate of dissolution with the growth in the concentration of sulfuric acid. In this case, the equation of the process of dissolution occurring at the hydrated copper(II) oxide/electrolyte boundary in the acidic region can be described by the reaction (Scheme 2) (similarly for chromium(III) oxide) where only the active sites of the type



participate in the process of dissolution, to which the surface complex $\text{CuOH}^+(\text{CrOH}^{2+})$ corresponds. For the scheme, the dependence of the specific rate of dissolution of chromium(III) or copper oxide on various factors is expressed by the equation

$$W = W_{\max} \frac{[\text{H}^+][\text{A}^-]}{K_{\text{ol}} + [\text{H}^+][\text{A}^-]}, \quad (14)$$

$$\frac{1}{W} = \frac{1}{W_{\max}} + \frac{K_{\text{ol}}}{W_{\max}} \frac{1}{[\text{H}^+][\text{A}^-]}, \quad (15)$$

where $W_{\max} = k_2\theta$, K_{α} is the quantity reciprocal to the adsorption constant, k_2 is the dissolution rate constant, and θ is the degree of coverage by the intermediate compounds.

As is seen from Figs. 10 and 11, Eqs. (14) and (15) accurately describe the experimental data of the dependence $\log W$ –pH.

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COMPLIANCE WITH ETHICAL STANDARDS

Humans or animals were not used as research objects in this work.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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