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Liquid-Phase Oxidation of Inorganic Sulfides in Aqueous Medium in the Presence of a Catalyst Based on 3,3',5,5'-Tetra-*tert*-Butyl-4,4'-Stilbenequinone

H. Y. Hoang[a], *, R. M. Akhmadullin[b], F. Yu. Akhmadullina[a], R. K. Zakirov[a], and A. G. Akhmadullina[b]

[a]*Kazan National Research Technological University, Kazan, 420015 Russia*
[b]*AhmadullinS – Science and Technology, Kazan, 420029 Russia*
**e-mail: nhochieny@gmail.com*

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Abstract—The kinetics of liquid-phase oxidation by oxygen from sodium sulfide was studied in an aqueous medium in the presence of a catalyst based on 3,3',5,5'-tetra-*tert*-butyl-4,4'-stilbenequinone dissolved in kerosene fraction. It was found that 3,3',5,5'-tetra-*tert*-butyl-4,4'-stilbenequinone selectively oxidizes sodium sulfide to sodium thiosulfate. The reaction orders in sodium sulfide, oxygen, and the catalyst were determined. The reaction mechanism was proposed.

Keywords: liquid-phase oxidation of sulfide, 3,3',5,5'-tetra-*tert*-butyl-4,4'-stilbenequinone, reaction mechanism, kinetics

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INTRODUCTION

Oil refineries and the enterprises of petrochemical industry are sources of a large amount of sulfur-containing waste, including sulfur–alkaline wastewater (SAWW). These aqueous solutions contain highly toxic concentrated sulfide compounds that must not be discharged into the environment.

To clean SAWW from sulfide sulfur in industry, liquid-phase catalytic oxidation by air is used. Various heterogeneous and homogeneous catalysts based on oxides, hydroxides, spinels, transition metal phthalocyanines, and other catalysts [1–3] are used in SAWW treatment. Other countries, mostly the United Kingdom and Japan, make use of the methods of treatment to remove sulfide sulfur SAWW that are based on benzoquinone, anthraquinone, or naphthoquinone. These are called quinone-based methods in Russia. The disadvantages of quinone-based methods are the low catalytic activity, substantial loss of the catalyst in the course of the process, and complications related to catalyst separation from the reaction medium when the oxidative treatment of waste is complete [4–6].

In this work, we propose to use 3,3',5,5'-tetra-*tert*-butyl-4,4'-stilbenequinone (henceforth for the sake of brevity, stilbenequinone or $C_{30}H_{42}O_2$) as a catalyst dissolved in the kerosene fraction. It shows high catalytic activity and stability in alkali media. The choice of the kerosene fraction is due to its weak solubility in water, high volatility, the ability to dissolve stilbenequinone well enough, and the ease of its separation from waste. Our goal was to study the kinetics of liquid-phase oxidation of sodium sulfide in the presence of stilbenequinone as a catalyst and determine the mechanism of reaction.

EXPERIMENTAL

3,3',5,5'-tetra-*tert*-butyl-4,4'-stilbenequinone was synthesized as described earlier [7]. The solution of sodium hydrosulfide was obtained by the method described in [8]. Sodium sulfide (GOST 2053-77) was dissolved in water. Oxygen (GOST 5583-78), argon (GOST 10157-79) in gas cylinders, and kerosene fraction (GOST 10227-2013) were used.

The catalytic oxidation of inorganic sulfides by oxygen were carried out in a three-necked glass tank reactor with a volume of 150 cm^3. The reactor was filled with the solution of inorganic sulfides (40 cm^3) and kerosene (20 cm^3) and added the necessary amount of the catalyst. Oxygen was supplied from a gas cylinder at a space velocity of 300 h^{-1}. The solution was stirred by a thermally controlled magnetic stirrer with 500–1400 rpm. Temperature was kept constant in the range 50–90°C.

Oxidation by stilbenequinone in the inert medium was carried out in the stainless steel autoclave

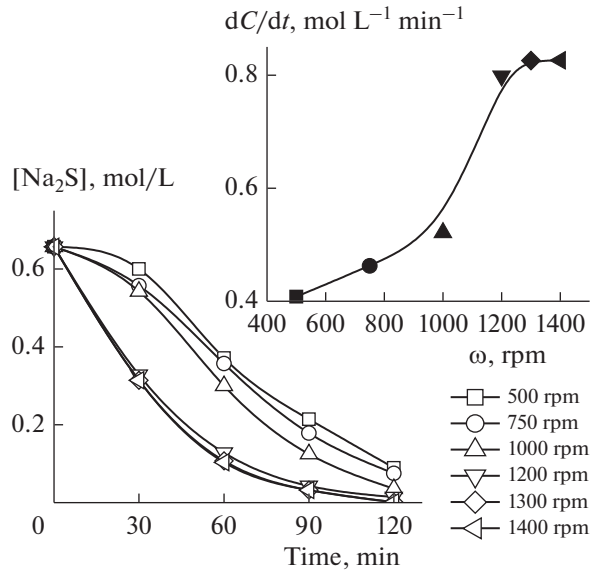


Fig. 1. Kinetic curves of sodium sulfide oxidation in the presence of the catalyst based on stilbenequinone at different stirring rates. The reaction temperature was 80°C; the concentration of stilbenequinone was 0.029 mol/L; the oxygen space velocity was 300 h^{-1}.

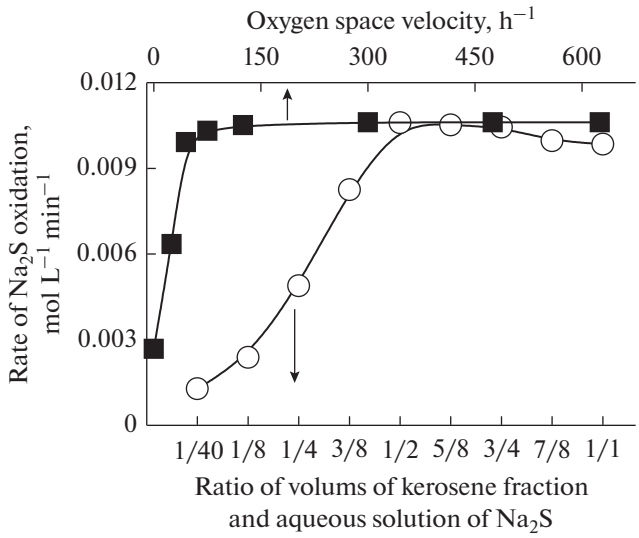


Fig. 2. The dependence of the rate of sodium sulfide oxidation on the ratio of the volume of sodium sulfide solution to the volume of kerosene fraction at an oxygen space velocity of 300 h^{-1} and its dependence on oxygen space velocity. The reaction temperature was 90°C; the initial concentration of sodium sulfide was 0.7 mol/L; the concentration of stilbenequinone was 0.052 mol/L.

(150 cm^3) into which 40-cm^3 solution of inorganic sulfides, 20 cm^3 of kerosene, and stilbenequinone were added. Temperature was maintained at 70–110°C and a stirring rate was 1400 rpm.

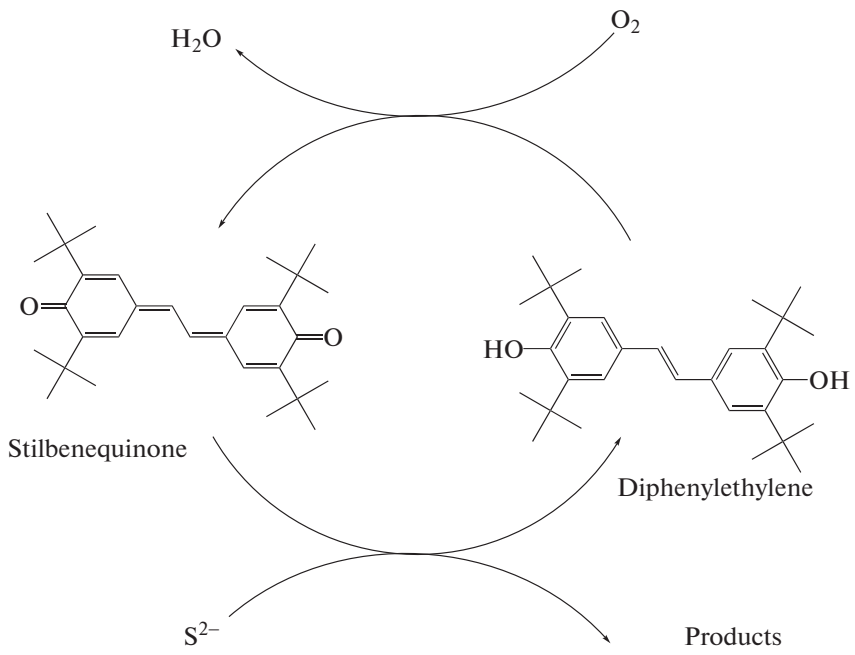
In the course of experiment, stirring was switched off at regular intervals and the samples of aqueous phase were withdrawn. The amount of sulfides was determined by potentiometric titration according to GOST 22985-90. The concentration of sodium thiosulfate and sulfite were calculated from iodometric data according to the technique described in [9]. The concentration of sodium sulfate was determined by spectrophotometry [10]. To determine the concentration of 3,3',5,5'-tetra-*tert*-butyl-4,4'-stilbenequinone the sample withdrawn from the kerosene phase was subjected to hundredfold dilution with toluene and its absorbance was measured using a PE5300V spectrophotometer (Ekros, Russia) at a wavelength λ of 500 nm in a 10-cm wide cell. Then, we used a calibration curve.

RESULTS AND DISCUSSION

The oxidation of sulfide sulfur in the presence of the catalyst based on stilbenequinone occurs in a three-phase system oxygen–kerosene fraction–aqueous alkali solution of sulfide sulfur. To determine where the reaction occurs, we carried out a series of experiments with varying the rate of stirring, the ratio between the volumes of kerosene fraction and the volume of the aqueous solution of sodium sulfide, and the amount of oxygen at a constant initial concentration of reactants and catalysts. The inset in Fig. 1 shows that the rate of sodium sulfide oxidation depends on the intensity of stirring in the range from 500 to 1200 rpm, but above 1200 rpm, it remains constant. Further experiments were carried out at a stirring rate of 1400 rpm.

The chosen ratio of the volume of kerosene fraction to the volume of the solution of sulfide sulfur (1 : 2) in the presence of stilbenequinone is optimal. It is likely that, at this ratio, the surface of interphase interaction reaches the maximum value (Fig. 2). It was experimentally found that at an oxygen space velocity of >45 h^{-1}, oxygen does not affect the catalytic oxidation of sulfide sulfur. The results obtained allow us to conclude that the reaction is not controlled by diffusion.

The oxidation of sodium sulfide in the presence of the catalyst based on stilbenequinone occurs via a complex mechanism. We have shown earlier [7, 11] that the catalytic cycle of sulfide sulfur oxidation in the reaction under study includes two steps: first, sulfide sulfur oxidation to thiosulfate by stilbenequinone with the reduction of the latter to 3,5,3',5'-tetra-*tert*-butyl-4,4'-dihydroxy-1,2-diphenylethylene (henceforth for the sake of brevity, diphenylethylene or $C_{30}H_{44}O_2$), and then, catalyst recovery by diphenylethylene oxidation to stilbenequinone in an alkali medium (scheme 1):



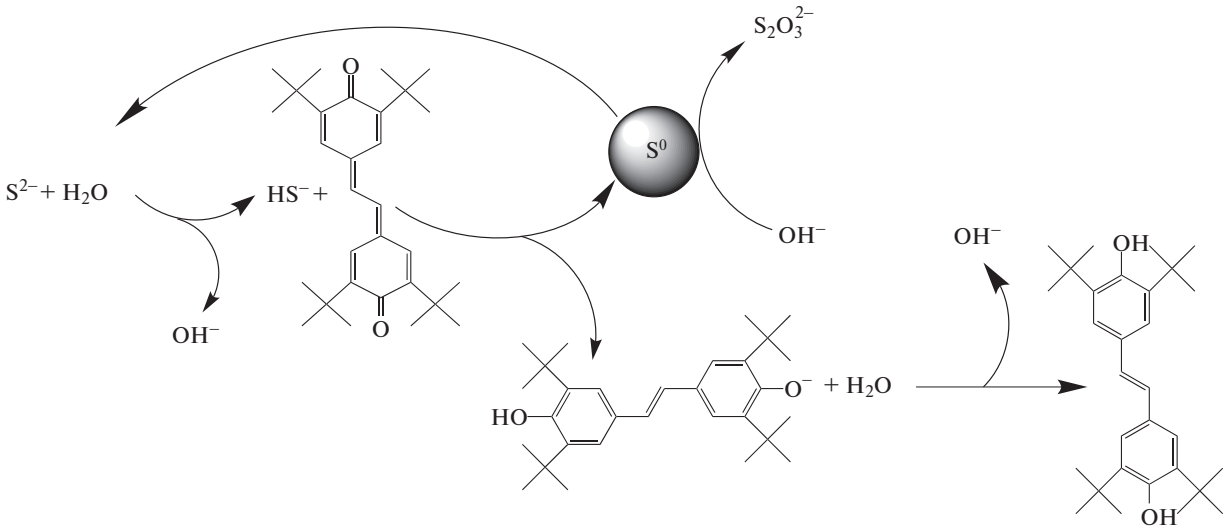
Scheme 1.

Quinones are usually strong oxidants under certain conditions [12]. In a stilbenequinone molecule, two cyclohexadiene fragments are separated by two methylene groups, which stipulates the extension of a conjugated system and increases electron affinity [13, 14]. Therefore, stilbenequinone is good two-electron oxidant capable of oxidizing sulfide sulfur with the participation of two protons. In doing so, stilbenequinone is reduced to diphenylethylene.

Shanina et al. [15] showed that, in the liquid-phase oxidation of sodium sulfide by an organic substance, first sodium sulfide is oxidized to sulfur, which is then converted to thiosulfate via the following equation

$$6OH^- + 4S \rightarrow S_2O_3^{2-} + 2S^{2-} + 3H_2O. \quad (I)$$

According to the proposed mechanism, the formation of thiosulfate molecule in oxidation to sulfide sulfur by stilbenequinone may occur as follows (scheme 2):



Scheme 2.

Table 1. Accumulation of stilbenequinone in diphenylethylene oxidation at different temperatures in the presence of aqueous 10% solution of NaOH

Temperature, °C	60	70	80	90
Stilbenequinone concentration, mol/L	0.00550	0.00765	0.00917	0.01043

The initial concentration of diphenylethylene was 0.013 mol/L. The space velocity of oxygen was 300 h^{-1}. The stirring rate was 1400 rpm. The volume of kerosene was 50 cm^3. The time of regeneration was 90 min.

Stilbenequinone oxidizes HS^- to S^0 and transforms to phenoxy anion, which reacts with water and forms diphenylethylene and hydroxy anion. Then, S^0 reacts with hydroxy anions and forms thiosulfate. In the course of this reaction two species of S^{2-} are formed. Thus, to form one molecule of thiosulfate, the participation of four stilbenequinone molecules and two S^{2-} species is needed.

Like diphenols, diphenylethylene is a hydroquinone, and thus it is sensitive to the action of oxidants. In the alkali medium that plays the role of a catalyst, hydroquinones are oxidized by oxygen to the corresponding quinones [16, 17]. The study of stilbenequinone regeneration at different temperatures (Table 1) showed that, with an increase in temperature, the rate of diphenylethylene oxidation by oxygen increases. The role of stilbenequinone reduces to the creation of new, more efficient way of electron transfer from a reducing agent (sulfide sulfur) to the oxidant (oxygen). Kerosene in this case serves as a catalyst carrier and absorbs oxygen from the reaction medium, because the coefficient of oxygen solubility in kerosene at 20°C (0.13) is higher than in water (0.016) [18].

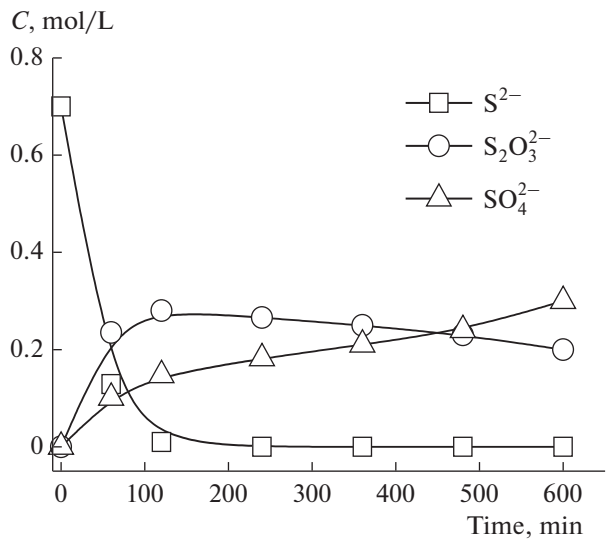


Fig. 3. Changes in the concentrations of sodium sulfide and the products of its catalytic oxidation in the course of the reaction in the presence of stilbenequinone. The reaction temperature was 90°C; the initial concentration of sodium sulfide was 0.7 mol/L; the concentration of stilbenequinone was 0.052 mol/L; the oxygen space velocity was 300 h^{-1}; the stirring rate was 1400 rpm.

Anasysis of intermediate and final products of sulfide sulfur oxidation in the presence of stilbenequinone catalyst showed that the main products are sodium thiosulfate and sodium sulfate (Fig. 3). Sulfite ions were not formed.

According to literature data [19], in the catalytic oxidation of sulfide sulfur using quinones as a catalyst, noncatalytic oxidation may also occur. To check this assumption, we studied how the concentration of products of sodium sulfide oxidation change if stilbenequinone is available in the autoclave at different temperatures in the inert medium. As can be seen from Table 2, sodium thiosulfate is selectively formed in this reaction. Sodium sulfate and sodium sulfite were not detected. In our opinion, a sulfate ion formed in Na_2S oxidation in the presence of the catalyst based on stilbenequinone is the product of noncatalytic oxidation of thiosulfate and sulfide ions by oxygen, because sulfide ions may be oxidized to sulfate ions in strong alkali media by the following reactions [20]:

$$2HS^- + 2O_2 \rightarrow S_2O_3^{2-} + H_2O, \quad \text{(II)}$$

$$HS^- + 2O_2 + OH^- \rightarrow SO_4^{2-} + H_2O, \quad \text{(III)}$$

$$2S^{2-} + 2O_2 + H_2O \rightarrow S_2O_3^{-2} + 2OH^-, \quad \text{(IV)}$$

$$S^{2-} + 2O_2 \rightarrow SO_4^{2-}, \quad \text{(V)}$$

$$HS^- + O_2 + 2OH^- \rightarrow SO_3^{-2} + H_2O, \quad \text{(VI)}$$

$$2S^{2-} + 3O_2 \rightarrow SO_3^{2-}, \quad \text{(VII)}$$

$$2SO_3^{2-} + O_2 \rightarrow SO_4^{2-}, \quad \text{(VIII)}$$

$$S_2O_3^{2-} + 2O_2 + 2OH^- \rightarrow 2SO_4^{2-} + H_2O. \quad \text{(IX)}$$

To confirm that selective synthesis of thiosulfate is possible in the catalytic oxidation of sulfide sulfur, we compared the yield of sodium sulfate formed in $Na_2S_2O_3$ oxidation both in the presence and in the absence of stilbenequinone. It follows from Fig. 4 that stilbenequinone does not affect the oxidation of sodium thiosulfate.

Based on the above data, we may consider that the catalytic cycle of sulfide oxidation to thiosulfate includes the following sequence of reactions:

Table 2. The temperature dependence of the concentrations of sodium sulfide and the products of its oxidation in the presence of stilbenequinone

Temperature, °C	Concentration, mol/L	
	Na_2S	$Na_2S_2O_3$
70	0.046	0.022
80	0.033	0.029
90	0.022	0.034
100	0.013	0.039
110	0.002	0.044

The initial concentration of sodium sulfide was 0.09 mol/L. The concentration of stilbenequinone was 0.358 mol/L. The stirring rate was 1400 rpm. The volume of kerosene was 20 cm^3. The volume of the aqueous solution of sodium sulfide was 40 cm^3. The reaction time was 120 min. Na_2SO_4 and Na_2SO_3 were not found in the reaction products.

Table 3. The catalytic activity of stilbenequinone in consecutive cycles of sodium sulfide oxidation at different reaction times

Cycle	Reaction time, min				
	30	60	90	120	150
	Na_2S conversion, %				
Blank run	4.082	12.245	16.327	20.408	27.551
First	65.306	93.367	99.133	99.993	100.000
Fifth	63.750	92.500	99.050	99.992	100.000
Nineth	63.265	92.347	99.184	99.991	99.997

The reaction temperature was 90°C. The initial concentration of sodium sulfide was 0.7 mol/L. The concentration of stilbenequinone was 0.052 mol/L. The oxygen space velocity was 300 h^{-1}.

$$S^{2-} + H_2O \rightarrow HS^- + OH^-, \quad (X)$$

$$C_{30}H_{42}O_2 + HS^- + H_2O \rightarrow C_{30}H_{44}O_2 + S + OH^-, \quad (XI)$$

$$4S + 6OH^- \rightarrow S_2O_3^{2-} + 2S^{2-} + 3H_2O, \quad (XII)$$

$$2C_{30}H_{44}O_2 + O_2 \rightarrow C_{30}H_{42}O_2 + 2H_2O. \quad (XIII)$$

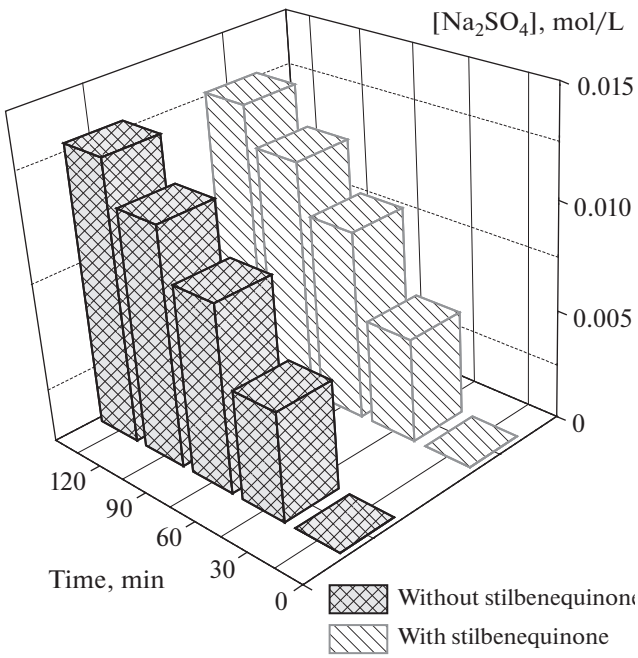


Fig. 4. The accumulation of sodium sulfate in thiosulfate oxidation by oxygen in the presence and in the absence of stilbenequinone. The reaction temperature was 90°C; the initial concentration of sodium sulfide was 0.1 mol/L; the concentration of stilbenequinone was 0.052 mol/L; the oxygen space velocity was 300 h^{-1}; the amount of solid NaOH was 0.75 g; the volume of kerosene was 20 cm^3; the volume of the aqueous solution of sodium thiosulfate was 40 cm^3.

In the aqueous medium, sulfide is hydrolyzed to hydrosulfide by reaction (X), and the latter is oxidized by stilbenequinone to sulfur by reaction (XI). Stilbenequinone is thus reduced to diphenylethylene. Sulfur is converted to thiosulfate by reaction (XII). The regeneration of stilbenequinone occurs by reaction (XIII) as a result of diphenylethylene oxidation by oxygen in an alkali medium.

Stilbenequinone has a high catalytic activity in reaction of sulfide sulfur oxidation. The rate of the catalytic reaction at 90°C is 17 times higher than the rate of noncatalytic oxidation. One of the important characteristics of the catalyst is its stability. The experiments confirmed that the activity of stilbenequinone remains constant after nine reaction cycles (Table 3). The turnover number, that is, the ratio of the number of moles of oxidized sulfide to the number of moles of stilbenequinone was 121.15.

The differential method was used to determine the order of liquid phase reaction of sulfide sulfur oxidation in the presence of stilbenequinone at different initial concentrations of sodium sulfide. Figure 5 shows how the concentration of Na_2S changes in the course of the reaction. The inset shows the logarithmic dependence of the initial rate of sodium sulfide oxidation on its initial concentration. As can be seen, this dependence is linear and the slope is 0.95. Thus, the reaction has nearly the first order in Na_2S. This conclusion can be confirmed by the integral method: the linear dependence of the logarithm of Na_2S concentration on time (Fig. 6) means that the liquid-phase catalytic oxidation of sulfide sulfur in the presence of stilbenequinone is described by the first order in Na_2S.

To estimate the effect of oxygen concentration in the gas/oxidant and the catalyst concentration on sulfide sulfur oxidation, we carried out a series of experiments, in which we varied the initial concentration of stilbenequinone in the kerosene fraction and the space velocity of oxygen by diluting it with argon (Figs. 7, 8).

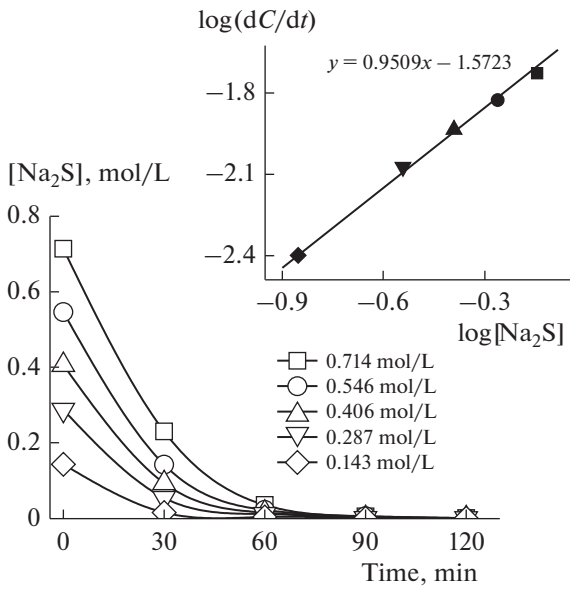


Fig. 5. Kinetic curves of sodium sulfide oxidation for different initial concentrations of sodium sulfide. The inset shows the logarithmic dependence of the initial rate of Na_2S oxidation on its initial concentration. The reaction temperature was 90°C; the stilbenequinone concentration was 0.052 mol/L; the oxygen space velocity was 300 h^{-1}.

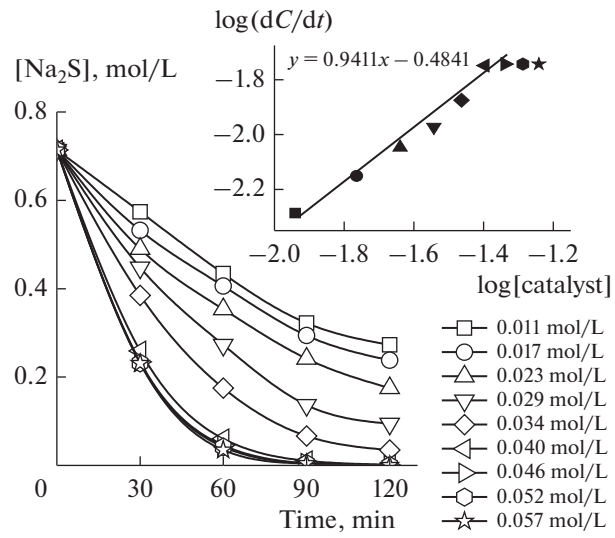


Fig. 7. Kinetic curves of sodium sulfide oxidation at different initial concentrations of the catalyst. The inset shows the logarithmic dependence of the initial rate of Na_2S oxidation on the initial concentration of the catalyst. The reaction temperature was 90°C; the initial concentration of sodium sulfide was 0.69 mol/L; the oxygen space velocity was 300 h^{-1}.

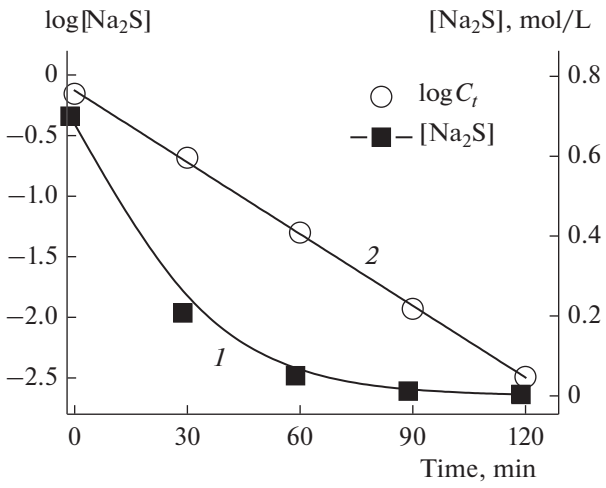


Fig. 6. Changes in (*1*) the concentration of Na_2S and (*2*) its logarithm with reaction time. The reaction temperature was 90°C; the initial concentration of sodium sulfide was 0.7 mol/L; the stilbenequinone concentration was 0.052 mol/L; the oxygen space velocity was 300 h^{-1}.

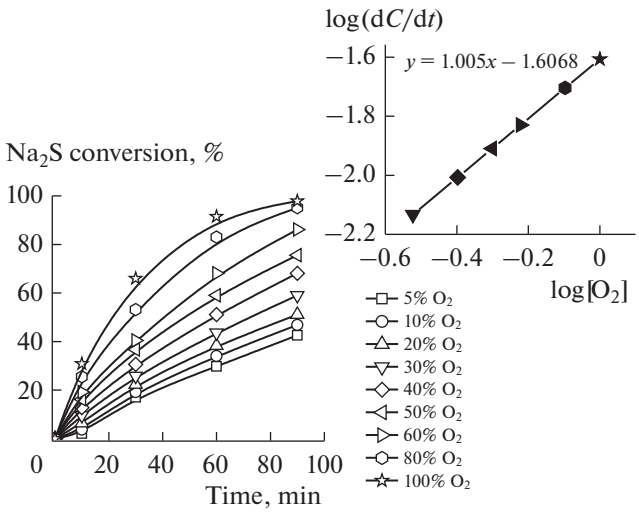


Fig. 8. Kinetic curves of sodium sulfide oxidation at different initial concentrations of oxygen. The inset shows the dependence of the initial rate of Na_2S oxidation on the initial concentration of oxygen. The reaction temperature was 90°C; the initial concentration of sodium sulfide was 0.69 mol/L; the stilbenequinone concentration was 0.052 mol/L; the space velocity of an oxygen–argon mixture supply was 100 h^{-1}.

It follows from Fig. 7 that, with an increase in the concentration of the catalyst in the kerosene fraction from 0.011 to 0.04 mol/L, the initial rate of sodium sulfide oxidation increases. With a further increase in the amount of stilbenequinone, it remains constant. The linearity of the logarithmic dependence of the initial rate of sodium sulfide oxidation from the initial concentration of the catalyst points to the first-order reaction with respect to stilbenequinone.

Figure 8 shows how the concentration of oxygen in the gas phase affects the oxidation of sodium sulfide in the presence of stilbenequinone. When the concentration of O_2 is lower than 20%, the reaction occurs with an induction period. The slope of the linear depen-

dence of the initial rate of Na_2S oxidation on log[O_2] is 1.00. That is, the reaction is of the first order with respect to oxygen. As shown above, in the catalytic oxidation of sulfide sulfur, noncatalytic oxidation of sulfur-containing compounds by oxygen leads to the formation of sulfate. It has been shown in [21, 22] that the order of sulfide sulfur liquid-phase oxidation in oxygen was 0.56. The rate of sodium sulfide oxidation in the presence of stilbenequinone is much higher than the rate of its noncatalytic oxidation. Therefore, we can be sure that the first order with respect to sulfide sulfur characterizes the catalytic reaction, and the contribution from noncatalytic oxidation is insignificant.

Thus, sulfide sulfur oxidation in the presence of a catalyst based on stilbenequinone has the first order in oxygen, stilbenequinone, and catalyst. These results confirm the proposed mechanism according to which one stilbenequinone molecule oxidizes one sulfide ion to sulfur S^0 to form diphenylethylene. The latter is oxidized by one oxygen molecule in the course of catalyst regeneration. Then, S^0 thus formed reacts with a hydroxide anion to form a thiosulfate molecule.

CONCLUSIONS

Thus, we found that, at a stirring rate of >1200 rpm, oxygen space velocity >45 h^{-1} and a ratio of the kerosene fraction to the volume of the sulfide sulfur solution of 1 : 2, the catalytic oxidation of sulfide sulfur is not controlled by diffusion.

The kinetic study of sulfide sulfur oxidation in the presence of the catalyst based on stilbenequinone showed that the reaction has first order in sodium sulfide, oxygen, and a catalyst. The only product of the catalytic oxidation of sodium sulfide is sodium thiosulfate. Sodium sulfate is formed only on the noncatalytic oxidation of Na_2S.

A sequence of reactions is proposed that occur in the oxidation of sulfide sulfur in the presence of the catalyst based on stilbenequinone.

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