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The Mechanism of Selective NO_x Reduction by Hydrocarbons in Excess Oxygen on Oxide Catalysts: II. Spectral and Kinetic Characteristics of Intermediate Complexes on the Commercial NTK-10-1 Catalyst

V. A. Matyshak*, V. F. Tret’yakov, T. N. Burdeinaya**, and Yu. P. Zakorchevnaya****

** Semenov Institute of Chemical Physics, Russian Academy of Sciences, Moscow, 119991 Russia*
*** Topchiev Institute of Petrochemical Synthesis, Russian Academy of Sciences, Moscow, 117912 Russia*

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Abstract—Quantitative spectrokinetic *in situ* measurements showed that the nitrate complex is an intermediate species in the process of the selective reduction of nitrogen oxides by propane on the commercial NTK-10-1 catalyst at $T > 150°C$. A nitroorganic compound is formed via the next step and is capable of transforming into the products of complete oxidation and into nitrogen oxides in the oxidative medium. At $T < 150°C$, the reaction occurs via the formation and further transformation of the nitrite complex. This process explains the unexpectedly high activity of the catalyst at low temperatures.

INTRODUCTION

An increase in the process selectivity can be achieved by choosing the optimal composition of a multicomponent catalytic system for various steps of the complex process [1]. One of the most, effective methods for creating such catalysts for various reactions is the use of the synergistic effect. Our experimental studies showed that the approach based on the preparation of binary mechanical mixture of commercial oxide catalysts with various concentrations of components is very promising [2, 3]. The synergistic effect is due to the mutual influence of system components on the oxidation–reduction reactions in the process of selective catalytic reduction of nitrogen oxides by hydrocarbons in excess oxygen. When mechanical mixtures are used, the principle can be realized whereby a complex reaction can be separated into stages, each of which is accelerated by one of the components of catalyst composition.

It is likely that the synergistic phenomenon is rather common [4], but there is no unified mechanism for it. Earlier, based on kinetic and thermal desorption measurements [5, 6], we determined experimental conditions under which synergism is observed in the reaction of selective reduction of nitrogen oxides by hydrocarbons on commercial oxide catalysts, and a hypothesis on the mechanism of this phenomenon was advanced. In this work, we used the spectrokinetic method proposed for the mechanistic study of this reaction, viz. for elucidating the role of surface complexes formed in the course of NO interaction with C_3H_8 and O_2 on the commercial NTK-10-1 catalyst, one of the components of the mechanical mixture.

EXPERIMENTAL

The spectrokinetic method is based on the simultaneous measurement of the catalyst activity and registration of the IR spectrum of surface compounds [7]. Such experiments in steady-state and non-steady-state regimes make it possible to compare the rate of surface complex transformation with the reaction rate.

Transmittance and diffuse-reflectance IR spectra were recorded using a Perkin-Elmer Spectrum RX I FT–IR system spectrometer (resolution, 4 cm^{-1}; scan time, 4.2 s). The IR spectra were processed using the Spectrum program developed by Perkin-Elmer. We used cell/reactors that made it possible to record IR spectra at elevated temperatures [8]. The weight of a sample in recording the diffuse-reflectance spectra was 1000–1200 mg; in recording transmittance spectra it was 40–60 mg. The intensity of absorption bands was measured in Kubelka–Munk units for diffuse reflectance spectra and in absorbance units (D) for transmittance spectra.

Non-steady-state spectrokinetic experiments for measuring the rate constants of surface species transformations were carried out as follows: after adsorption complexes had been formed on the catalyst surface; the inlet and outlet of the cell/reactor were closed. Then, the gas supply lines were purged with a reaction mixture and when the constant composition of gases was reached, the inlet was open.

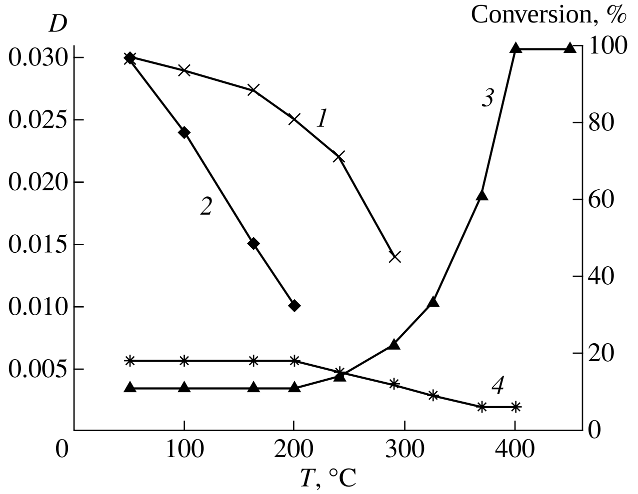


Fig. 1. Temperature dependences of the intensities of absorption bands at (*1*) 2970 and (*2*) 2930 cm^{-1} and for (*3*) propane and (*4*) NO_x conversion.

The concentrations of reactants in the initial mixtures was varied in the following limits (vol %): NO, 0–0.22; C_3H_8, 0–0.67; O_2, 0–3.5; carrier gas, N_2, The flow rate of the reaction mixture 150 ml/min (9000 h^{-1}). All gases were purified using anhydrone and ascarite traps. To analyze gases before and after the cell/reactor, a Beckman-951A chemiluminescent NO/NO_x analyzer, a Beckman-590 HC/CO analyzer, and a chromatograph with a thermal conductivity detector and Porapak-Q adsorbent were used.

The reaction temperature was controlled using a MINITERM-300.31 programmable thermal controller (deviation of the temperature from the preset value was <5°C).

RESULTS

The Effect of Temperature on the Properties of Surface Compounds and the Catalyst Activity

In the course of interaction of the reaction mixture (vol %) 0.1 NO + 0.5 C_3H_8 + 2.5 O_2/N_2 with the catalyst surface, *in situ* diffuse reflectance spectra at 25–300°C contained intense absorption bands at 2970 and 2930 cm^{-1} corresponding to adsorbed propane (Fig. 1, curves *1*, *2*). Note that with the addition of NO to the reaction mixture containing propane and oxygen, the concentration of adsorbed propane species increased. The activated adsorption of propane is possibly associated with the formation of surface species oxidized by oxygen or NO, analogous to those detected in [9]. Figure 1 also shows the temperature dependences of propane conversion (curve *3*) and NO_x conversion (curve *4*). The greatest decrease in the intensity of bands corresponds to an increase in the propane conversion, which reached ~100% at ~ 400°C. The maximal conversion of NO was 15%. Figure 2 shows the results of an analogous experiment using transmittance spectroscopy. With an increase in the temperature, the band at 1215 cm^{-1} corresponding to the nitrate complexes rapidly disappears (curve *1*). The concentration of nitrate complexes (the band at 1280 cm^{-1}) passes through a maximum (curve *2*). The value of *D* for the band at 2970 cm^{-1} weakly depends on the temperature (curve *3*), and the intensity of the band at 1500 cm^{-1} increases with an increase in temperature (curve *4*). Note also the presence of bands at 1760 cm^{-1} in the spectrum in the temperature range 50–120°C (curve *5*).

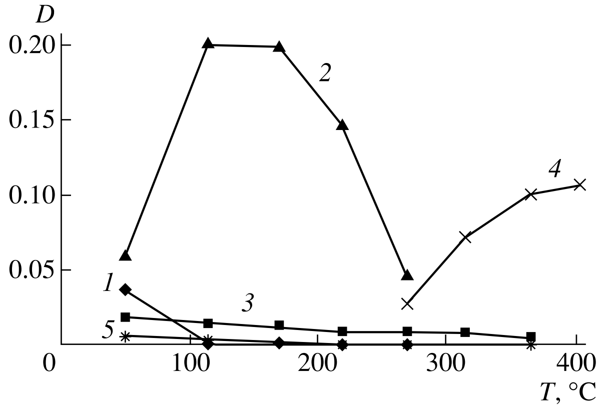


Fig. 2. Temperature dependences of the intensities of absorption bands at (*1*) 1215, (*2*) 1280, (*3*) 2970, (*4*) 1500, and (*5*) 1760 cm^{-1}.

The Effect of the Reaction Mixture Composition on the Properties of Surface Compounds and Catalyst Activity

Experiments were carried out at 235°C because under these conditions the noticeable conversion of nitrogen oxides and propane is accompanied by the absorption bands corresponding to propane adsorption and nitrite–nitrate surface complex formation.

The effect of propane concentration. The conversion of propane increases with an increase in its concentration in the reaction mixture, and the conversion of NO remains constant (Fig. 3a). In the transmittance spectra, the intensity of the band at 2970 cm^{-1} increases with an increase in the concentration of propane, whereas the intensity of the band at 1280 cm^{-1} decreases (Fig. 3b). The ratio of the NO and NO_x concentrations in the flow at the outlet from the cell/reactor is close to unity and depends on the propane concentration in the reaction mixture (Fig. 3b), pointing to the absence of NO_2 in the reaction products.

The effect of oxygen concentration. Under the chosen conditions the conversion of propane is independent of the amount of oxygen in the reaction mixture. In contrast to this, the conversion of nitrogen oxide passes through a maximum as the concentration of oxygen increases. This maximum corresponds to ~2.5% O_2 (Fig. 4a). The intensity of the band at 2970 cm^{-1} is inde-

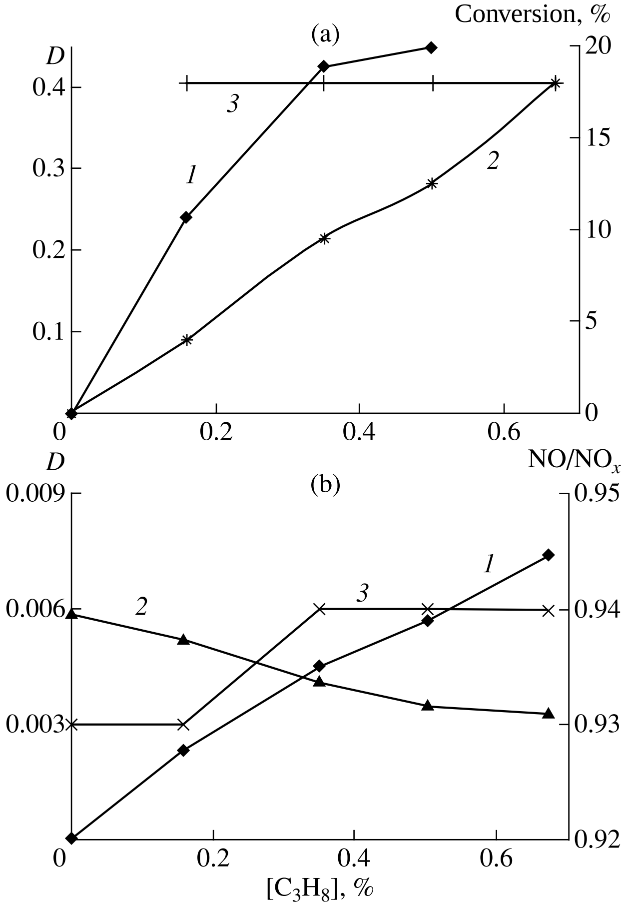


Fig. 3. C_3H_8 concentration dependences in the interaction of mixtures with compositions (0.1% NO + 2.5% O_2)/N_2 and 0–0.67% C_3H_8: (a) (*1*) the intensity of the band at 2970 cm^{-1} in the diffuse-reflectance spectrum, (*2*) propane conversion, and (*3*) NO_x conversion; (b) the intensity of the band at (*1*) 2970 cm^{-1}, (*2*) 1280 cm^{-1}, and (*3*) the NO/NO_x ratio.

pendent of the concentration of oxygen, but the intensity of the band at 1280 cm^{-1} increases when 1% O_2 is added and then remains constant in the range of O_2 concentrations from 1 to 3.5%. The NO/NO_x value in a flow at the outlet of the cell/reactor passes through a maximum at ~ 2% O_2 (Fig. 4b).

The effect of NO concentration. The values of propane and NO conversion are practically independent of the concentration of nitrogen oxide in the flow of the reaction mixture (Fig. 5a). The intensities of bands corresponding to adsorbed propane and nitrate complexes change slightly with an increase in the NO concentration (Fig. 5b). The NO/NO_x value at the outlet of the cell/reactor is close to unity and does not depend on the concentration of NO in the reaction mixture.

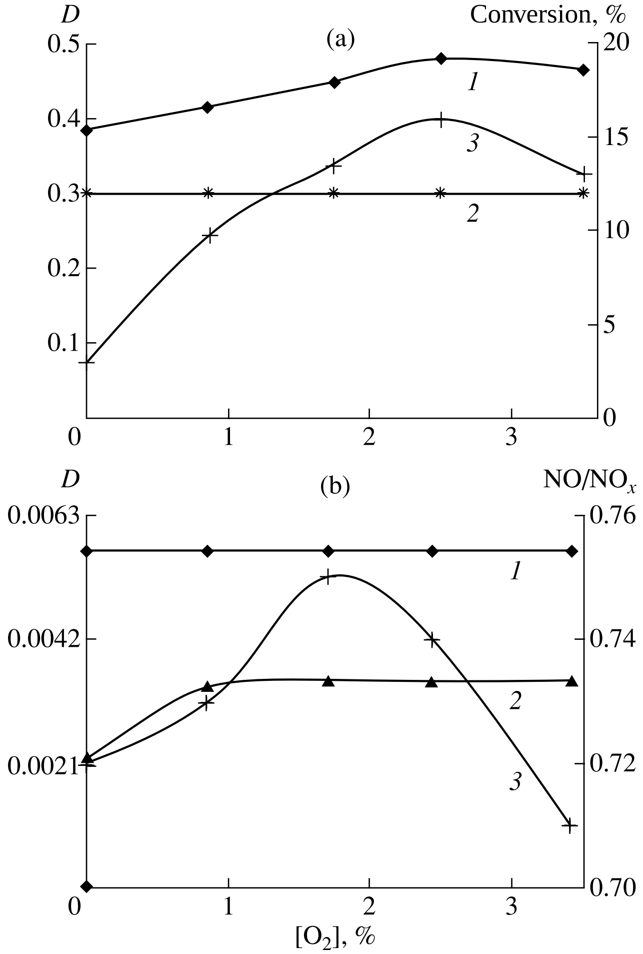


Fig. 4. O_2 concentration dependences in the interaction of mixtures with compositions (0.1% NO + 0.5% C_3H_8)/N_2 and 0–3.5%O_2: (a) (*1*) the intensity of the band at 2970 cm^{-1} in the diffuse-reflectance spectrum, (*2*) propane conversion, and (*3*) NO_x conversion; (b) the intensity of the bands at (*1*) 2970 cm^{-1} and (*2*) 1280 and (*3*) the NO/NO_x ratio.

The Reactivity of Surface Complexes

The results presented above show that the complexes of adsorbed propane, nitrite, and nitrate surface compounds are present on the catalyst surface under reaction conditions. In this part of the work, we present data on the reactivity of surface nitrite and nitrate complexes toward propane.

Experiments were carried out using transmittance spectroscopy. The intensities of absorption bands were obtained by subtracting the spectrum of the sample in a flow of nitrogen, from the spectrum of the sample in a flow of the reaction mixture under the same conditions.

Surface complexes were formed by treating the catalyst with a gaseous mixture containing 0.1% NO/N_2 at room temperature (15 min), and then at 70, 100, 115, 220, 270, and 315°C. At each temperature, three experiments were carried out to determine the dependence of

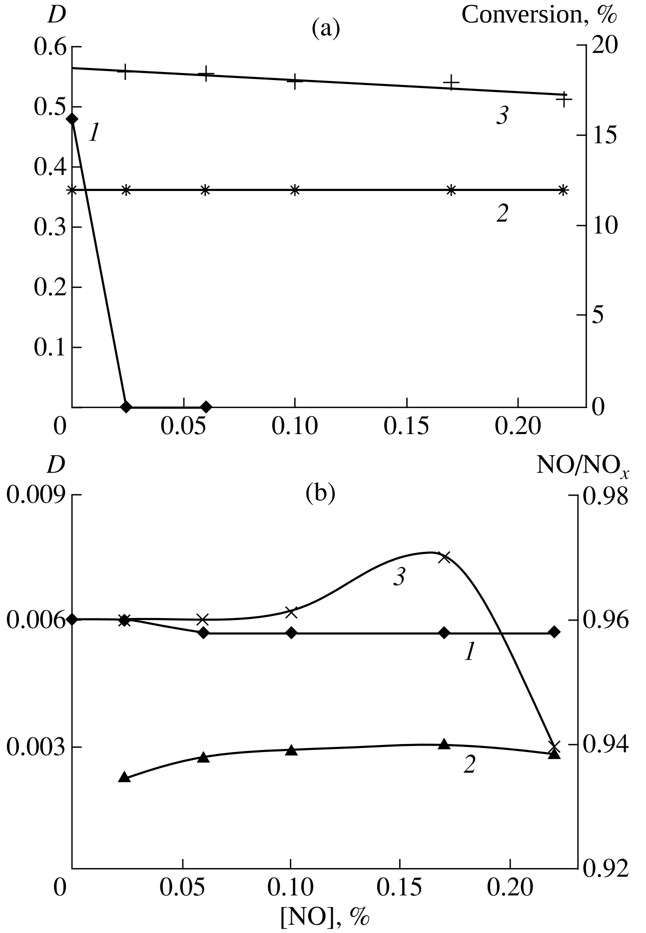


Fig. 5. NO concentration dependences in the interaction of mixtures with compositions (0.5%C_3H_8 + 2.5%O_2)/N_2 and 0–0.22% NO: (a) (*1*) the intensity of the band at 2970 cm^{-1} in the diffuse-reflectance spectrum, (*2*) the conversion of propane, and (*3*) the conversion of NO$_x$; (b) (*1*) the intensity of the band at 2970, (*2*) 1280 cm^{-1}, and (*3*) the NO/NO$_x$ ratio.

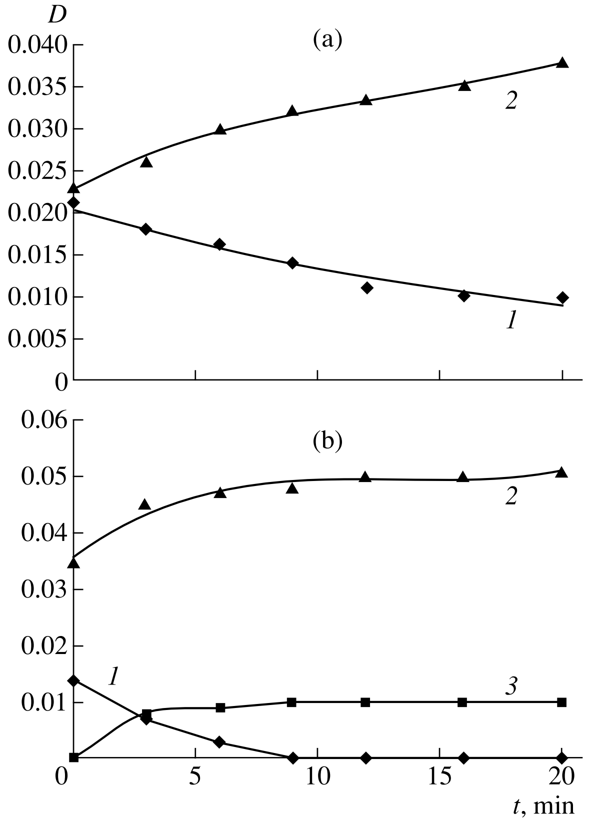


Fig. 6. Dependence of the intensity of the bands at (*1*) 1215, (*2*) 1280, and (*3*) 2970 cm^{-1} on the duration of catalyst treatment with (a) nitrogen and (b) a mixture of propane and oxygen at 115°C.

the band intensities corresponding to nitrite and nitrate complexes on time.

Figure 6 shows the results of one of the experiments at 115°C. It is seen that with an increase in the duration of catalyst treatment with nitrogen (Fig. 6a), the amount of nitrate complexes on the surface increases (the band at 1280 cm^{-1}), while the concentration of nitrites decreases (the band at 1215 cm^{-1}). Upon the admission of the mixtures C_3H_8/N_2 and $C_3H_8 + O_2/N_2$ (Fig. 6b), the band at 2970 cm^{-1} appears in the spectrum and its stationary value is established rather rapidly. As in a flow of nitrogen, the intensity of the band at 1280 cm^{-1} increases with time, and the intensity of the band at 1215 cm^{-1} decreases. Note that the rate of increase of the intensity of the band at 1280 cm^{-1} is lower in this case and the rate of decrease of the intensity of the band at 1215 cm^{-1} is much higher.

In the course of catalyst treatment with nitrogen at 270°C, nitrate complexes are not consumed (Fig. 7), whereas in the run with the propane–oxygen mixture their amount substantially decreased with a noticeable rate. It is important that the rate of transformation of complexes practically coincide when the catalyst is treated by propane or by a propane–oxygen mixture.

At 315°C the rate of nitrate complex transformation in a flow of the propane–oxygen mixture noticeably increases with an increase in the temperature (Fig. 8). The spectra start to contain a broad band at 1500 cm^{-1}, and the bands at 1795, 2510, and 2590 cm^{-1} belonging to the vibrations of bulk $CaCO_3$. Their presence means the occurrence of the complete oxidation of propane to CO_2, which reacts with calcium in the catalyst and forms the corresponding carbonate.

The dependences of the intensities of absorption bands on the reaction time were used to calculate the apparent rate constants of the corresponding processes. The values of the apparent rate constants of NO_3^- and NO_2^- consumption in the gaseous mixtures of the following compositions were calculated: N_2; 0.5% C_3H_8/N_2; 0.5% C_3H_8 + 2.5% O_2/N_2. The dependence of a change in the band intensities in the course

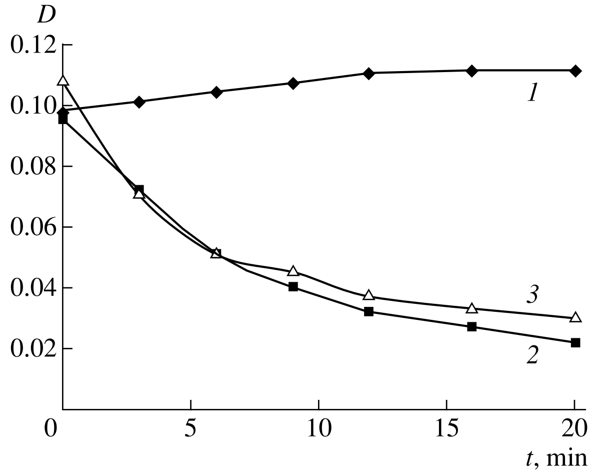


Fig. 7. Dependence of the band intensity at 1280 cm^{-1} on the duration of catalyst treatment with (*1*) nitrogen, (*2*) propane, and (*3*) a mixture of propane and oxygen at 270°C.

of the reaction was approximated by the rate law that is first-order in NO_3^- and NO_2^- concentrations with an approximation degree R^2 of at least 0.9.

The results of determination of rate constants from changes in the band intensity at 1280 cm^{-1} are presented below:

The values of the apparent rate constants of nitrate complex transformation in the reaction with the $C_3H_8 + O_2$ mixture

T, °C	220	270	315
k, min^{-1}	0.02	0.08	1.1

Surface nitrite complexes also show a rather high reactivity. Figures 9 and 10 show the results of experiments carried out at 70 and 100°C. It can be seen that there is no increase in the intensity of the band corresponding to the nitrate complex in the presence of propane. Moreover, the rate constant of nitrite complex transformation at 100°C is equal to the rate constant of surface nitrate complex formation (Table 1).

DISCUSSION

Comparison of thermal desorption [10] and spectral data shows that the high-temperature desorption peak is associated with the decomposition of nitrate complexes. Data shown in Fig. 6 provide further evidence. The high reactivity of surface nitrites is notable (Fig. 6b). In the presence of propane in the gas phase, a decrease in the intensity of the band corresponding to the nitrite complex is much faster then in a flow of nitrogen (Fig. 6a). The rate of increase in the nitrate concentration is low. The relatively high conversion of NO at low temperatures is probably associated with nitrite complexes (Fig. 1).

The concentration of nitrates $[NO_3^-]$ on the surface of the catalyst for selective NO_x reduction by hydrocarbons was calculated using the proportionality coefficient A_0 (transmittance IR spectroscopy), which relates $[NO_3^-]$ with the integral intensity (A) of the band at 1280 cm^{-1}:

$$[NO_3^-] = A_{1280}/A_0. \qquad (1)$$

The values of these coefficients were obtained by the combined use of *in situ* IR spectroscopy and TPD NO_x. At 250°C, when we can neglect the decomposition of surface nitrates and the formation of any nitrogen–oxygen complexes other than $NO_{3(ads)}$, NO_x adsorption was carried out. Then, the cell was purged with nitrogen for 20 min, the IR spectrum was

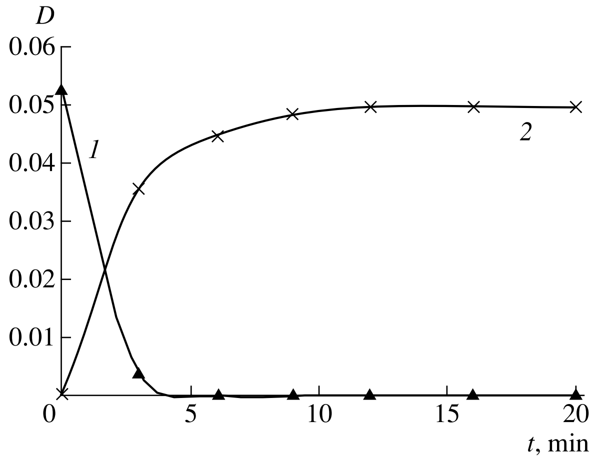


Fig. 8. Dependence of the band intensities at (*1*) 1280 and (*2*) 1500 cm^{-1} on the duration of the catalyst treatment with a mixture of propane and oxygen at 315°C.

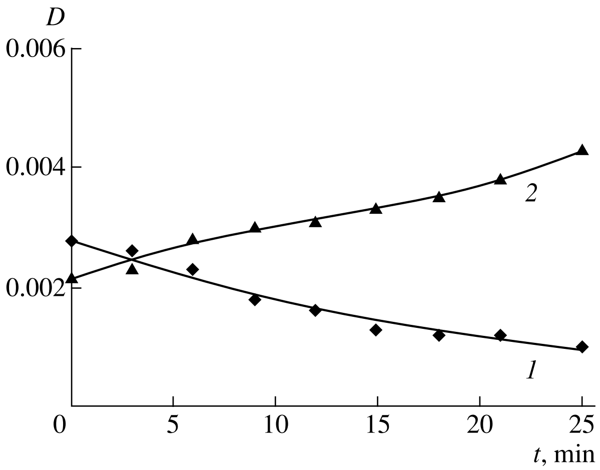


Fig. 9. Dependence of the band intensities at (*1*) 1215 and (*2*) 1280 cm^{-1} on the duration of the catalyst treatment with nitrogen at 100°C.

recorded, and NO_x was desorbed. The value of A_0 was calculated from the amount of desorbed nitrogen oxide molecules ($N_{NO_{x(des)}}$) and the band intensity at 1280 cm^{-1} (A):

$$A_0 = A_{1280}/N_{NO_{x(des)}}\ (\text{cm}^{-1}\ \text{molecule}^{-1}). \qquad (2)$$

This resulted in a value of A_0 equal to 3.36×10^{-18} cm^{-1}/molecule. We note the closeness of this value to that determined in [11]. Using this value and the intensity of the band at 1280 cm^{-1} under reaction conditions, one can determine the steady-state amount of nitrate complexes. The extinction coefficient was determined from the height of the band in a similar way (4.8×10^{-20} cm^2/molecule).

To determine the concentration of NO_3^- on the catalyst surface under reaction conditions, the same samples as for the determination of the A_0 value were used.

The values of the rate constants of nitrate complex transformation (see above) determined in non-steady-state spectrokinetic experiments and steady-state amounts of nitrate complexes make it possible to calculate the rate of nitrate complex transformation and compare it to the rate of the reaction of selective NO_x reduction by hydrocarbons.

The rate of nitrate complex transformation (w) was calculated from the formula

$$w = kN, \qquad (3)$$

where k is the rate constant of transformation of the nitrate complex and N is the amount of nitrate complexes on the catalyst surface (per unit weight of the sample).

The activation energy of the nitrate complex transformation determined from the temperature dependence the rate constant is 20 kJ/mol.

The results of comparing the rates obtained at different temperatures are shown in Table 2.

Data in Table 2 show a strong correlation between the rate of nitrate complex transformation and the rate of the reaction at 220 and 270°C. At 315°C, the rate of nitrate transformation is substantially higher than the reaction rate. Note that an increase in the temperature from 270 to 315°C is accompanied by a decrease in the reaction rate (Table 2). The correlation of the rates [12] means that the nitrate complex is an intermediate species in the process of selective NO_x reduction. The fact that the rate of nitrate complex is higher than the rate of the reaction can be explained by the idea that the next step in the process is the formation and consumption of a nitroorganic compound. These steps were observed and discussed as intermediate steps in a number of papers [9, 13–18]. When studying the pathways of transformation of surface nitroorganic compounds [16], it was found that

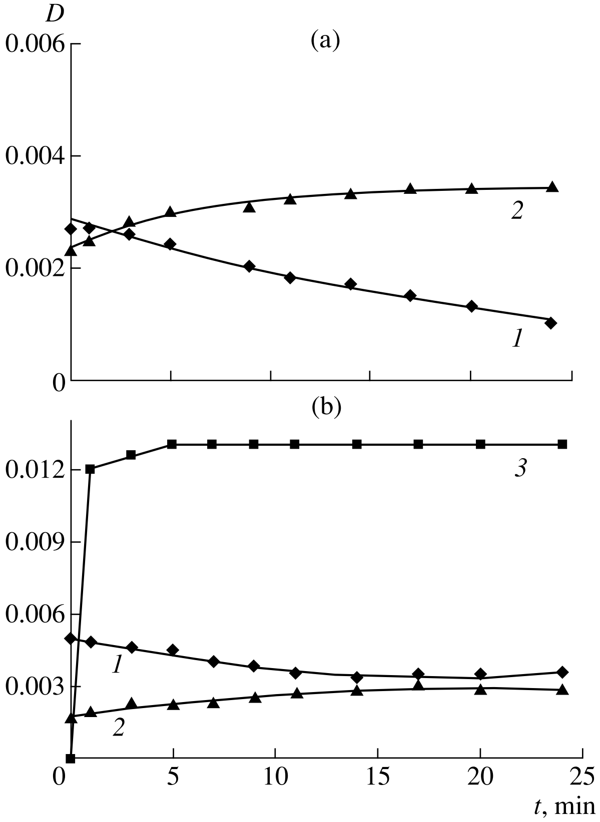


Fig. 10. Dependence of the band intensities at (*1*) 1215, (*2*) 1280, and (*3*) 2970 cm^{-1} on the duration of the catalyst treatment with (a) nitrogen and (b) a mixture of propane and oxygen at 70°C.

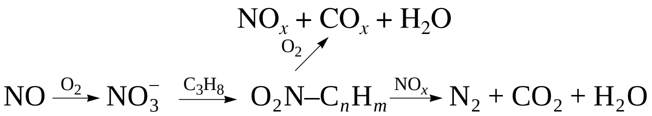
Table 1. Characteristics of transformation of nitrite and nitrate complexes

T, °C	k_{cons} for the nitrite complex in a flow of N_2, min^{-1}	k_{form} for the nitrite complex in a flow of N_2, min^{-1}	k_{cons} for the nitrite complex in a flow of C_3H_8/N_2, min^{-1}	Intensity of the band at 1215 cm^{-1}
115	0.05	0.07	0.18	–
100	0.04	0.05	–	–
70	0.04	0.17	0.05	0.07

Table 2. Comparison of the rate of nitrate complex interaction with the rate of selective NO_x reduction

T, °C	k, min^{-1}	A	$N \times 10^{-18}$, molecule	$w_{\text{complex}} \times 10^{-17}$, molecule/min	$w_{\text{reaction}} \times 10^{-17}$, molecule/min
220	0.02	0.095	35	7.0	6.8
270	0.08	0.032	8.4	6.7	4.1
315	1.1	0.019	2.0	22.0	2.8

they can give products of selective oxidation and can be oxidized further with the formation of gaseous CO_x and NO_x in the presence of oxygen. This is apparently our case. The nitrate complex forms an intermediate nitroorganic compound in the reaction with the activated propane fragment. The formation of this compound under reaction condition is confirmed by the presence of the band at 1760 cm^{-1} (Fig. 2). This band is absent from the spectra only when both propane and NO are present in the mixture. Based on the data, we propose the following scheme of the process on the NTK-10-1 catalyst:



$$NO \xrightarrow{O_2} NO_3^- \xrightarrow{C_3H_8} O_2N\text{–}C_nH_m \xrightarrow{NO_x} N_2 + CO_2 + H_2O$$
$$O_2N\text{–}C_nH_m \xrightarrow{O_2} NO_x + CO_x + H_2O$$

We estimate the rate of the side process of nitroorganic complex decomposition into CO_x and NO_x proceeding from this scheme and the data in Table 2 and determine the activation energy from the temperature dependence (24 kJ/mol).

Note that the proposed scheme is valid at $T > 150$–200°C when the concentration of nitrite complexes on the surface is negligibly small. At lower temperatures one should take into account data on the high reactivity of nitrite complexes (Fig. 6). Their interaction with propane explains the activity of the catalyst at relatively low temperatures (Fig. 1).

The processing of kinetic curves (Figs. 6, 9, 10) shows that at 100°C (Table 1) the rate constants for the formation of the nitrate complex (the band at 1280 cm^{-1}) and its transformation (the band at 1215 cm^{-1}) are equal. Therefore, at this temperature the nitrite complex indeed transforms into the nitrate complex. The desorption of the nitrite complex is negligible in this case. This fact makes it possible to estimate the extinction coefficient ε of the nitrite complex:

$$\Delta A_{1280} = \varepsilon_{1280}\Delta N_{NO_3}, \quad (4)$$

$$\Delta N_{NO_3} = \Delta N_{NO_2}, \quad (5)$$

$$\varepsilon_{1215} = \Delta A_{1215}/\Delta N_{NO_2}, \quad (6)$$

where ΔA_{1280} is a change in the absorption band intensity at 1280 cm^{-1} and ΔN is a change in the amount of molecules.

From the value of ε_{1215}, the intensity of the absorption band of the nitrite complex under reaction conditions, and the rate constant of transformation of this complex in the reaction with a mixture of propane oxygen (see above), we can calculate the rate of transformation of the nitrite complex and compare it with the rate of reaction at $T < 115$°C, in the region where the nitrate complex is practically absent.

It follows from the data presented at Fig. 9 that an increase in the absorbance of the nitrate complex band is equal to a decrease in the absorbance of the band of the nitrite complex. This means that the extinction coefficients of the nitrite and nitrate complexes are equal (4.8×10^{-10} cm^2/molecule). Data in Table 2 make it possible to compare the rate of transformation of nitrites with the reaction rate at 70°C (at 100°C, the steady-state concentration of the nitrite complex is low and is masked by the absorbance of the nitrate complex). The rate of nitrite complex transformation is $w = kN = 1.1 \times 10^{18}$ complex/min. The reaction rate at this temperature is 1×10^{18} molecule/min. The equality of these values suggests that the surface nitrite complex is an intermediate species in the reaction of selective reduction at low temperatures. The data presented in Table 1 need some comments. The rate constant of nitrite complex transformation (k_{cons}) calculated from a change in the band intensity at 1215 cm^{-1} only coincides with the rate constant of formation (k_{form}) within the experimental accuracy at 100 and 115°C. At 70°C the value of k_{cons} for the nitrite complex is much lower than k_{form} for the nitrate complex. This is due to the formation of the low-frequency state of the nitrite complex, which exists at $T \leq 100$°C. Its transformation into the nitrate complex is confirmed by data in Table 1, which gives the value of the rate constant of nitrate formation at 70°C.

Some process detail can be refined when analyzing the effect of the composition of the reaction mixture on the catalytic activity and properties of the surface species (Figs. 3–5). An increase in the concentration of propane in the reaction mixture is one of the reasons for a decrease in the surface coverage with nitrate complexes, which is due to an increase in the surface coverage by propane complexes (Fig. 3). The constant value of NO means that propane activation and nitrate complex formation occur on the same regions of the catalyst surface. In this case, the surface concentration of nitrates decreases and the surface coverage with surface propane increases. This conclusion support the data shown in Figs. 3–5. In the presence of propane in the gas phase, the intensity of the band of nitrate complexes is no longer increased, although in a flow of an inert gas this process is rather efficient.

An increase in propane conversion in this case is associated not only with the reaction of deep oxidation but also with the participation of gaseous and weakly adsorbed propane molecules (with an increase in temperature in the range below 250°C the intensity of the band corresponding to adsorbed propane increases). The calculation shows that under reaction conditions (flow rate, 150 cm^3/min; concentration of propane in the flow of the reaction mixture, 0.5 vol %; the conversion of C_3H_8, 15%; duration of an experiment, 200 min; catalyst weight, 1 g; specific surface area, 75 m^2/g) the maximal surface coverage with propane complexes is 10–15%. That is, in this case, C_3H_8 adsorption may provide the apparent conversion of propane.

The constant ratio of the NO and NO_x concentrations at the outlet of the cell/reactor with a change in the C_3H_8 concentration in the reaction mixture (Fig. 3b) means that an increase in surface coverage with propane does not affect the process of nitroorganic compound transformation.

The initial increase in NO conversion with an increase in the oxygen concentration in the reaction mixture is associated with an increase in the surface coverage with propane as follows from Fig. 4. A decrease in the NO conversion when the concentration of O_2 in the reaction mixture is >2.5% correlates with the NO/NO_x ratio (Fig. 4b) and is associated with an increase in the rate of nitroorganic complex oxidation to CO_2 and NO_2. This means that oxygen accelerates the process of nitrate complex formation and participates in the oxidation of nitroorganic complexes.

The constant conversion of nitrogen oxide despite an increase in its concentration in the reaction mixture points to a linear increase in the rate of NO transformation. This can be explained by an increase in the surface coverage with nitrate complexes formed from the more reactive weakly bound forms, nitrates, when the surface concentration of propane essentially remains constant. The constant NO/NO_x ratio at the outlet from the cell/reactor with a change in the concentration of NO in the reaction mixture (Fig. 5b) means that nitrogen oxide does not participate in the process of nitroorganic compound transformation into CO_2 and NO_2.

CONCLUSIONS

Quantitative spectrokinetic measurements showed that the nitrate complex is an intermediate species in the selective NO_x reduction by propane at $T > 150$°C. At a further stage, a nitroorganic compound is formed that is capable of transforming into the products of deep oxidation and nitrogen oxides in the oxidative medium. Nitrogen does not participate in this stage. Only under the reaction conditions (i.e., in the presence of NO and oxygen) is a complex found that involves an oxidized fragment of propane.

At $T < 150$°C, when nitrate complexes are practically absent from the catalyst surface, the reaction occurs through the formation and further transformations of nitrite complexes. This process explains the unexpectedly high activity of the catalyst at low temperatures.

The high reactivity of nitrite complexes can be used to design low-temperature catalysts for the selective reduction of NO_x by hydrocarbons in the presence of excess oxygen.

When evaluating the properties of the NTK-10-1 catalysts from the standpoint of synergy theory, we may conclude that NO_x activation on the surface occurs easily, whereas the formation of the activated propane is difficult. Therefore, to develop efficient catalytic compositions, one would have to add an additional component to the composition of a mechanical mixture that would be capable of activating a hydrocarbon.

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