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Study of the Oxidation Process of Nitrogen Oxides by Ozone

The need to reduce emissions of pollutants, in particular nitrogen oxides, as required by regulations in Ukraine, requires the use of modern technologies and methods for waste gas treatment at industrial enterprises. This is especially true of thermal power plants, which are powerful sources of nitrogen oxide emissions. The technological part of the wet or semi-dry method of purification is the area for the oxidation of nitrogen oxides to obtain easily soluble compounds. The paper presents the results of a study of the process of ozone oxidation of nitrogen oxides in a chemical reactor. Data for the analysis of the process were obtained by performing physical experiments on a laboratory installation and related calculations on a mathematical model. Studies of the oxidation process have shown that the required amount of ozone depends not only on the content of nitrogen monoxide, but also on the content of nitrogen dioxide. The process of conversion of nitrogen monoxide to a satisfactory level occurs at the initial value of the molar ratio of ozone to nitrogen monoxide in the range of 1.5...2. The conversion efficiency of nitrogen monoxide reaches 90 % at a gas temperature less than 100 °C. To achieve high conversion efficiency at gas temperatures above 100 °C, it is necessary to increase the initial ozone content when the molar ratio exceeds 2. The analysis shows that the conversion efficiency of nitric oxide largely depends on the residence time of the gas mixture in the reaction zone. Due to lack of time under certain conditions, the efficiency decreases by approximately 46 %. To increase it, it is necessary to accelerate the rate of oxidation reactions due to better mixing of gases by turbulence of the flow in the oxidizing reactor. *Bibl. 6, Fig. 6, Tab. 3.*

Keywords: nitrogen oxides, ozone, reactor, experiment.

Introduction

Industrial enterprises emit large amounts of pollutants into the atmosphere. Among them, nitrogen oxides (NO_x) occupy one of the first places from an ecological point of view. The order of the Ministry of Ecology and Natural Resources of Ukraine [1] establishes Technological standards for permissible emissions of nitrogen oxides from thermal power plants, the nominal heat capacity of which exceeds 50 MW. When using semi-dry or wet methods to reduce emissions of nitrogen monoxide (NO) and dioxide (NO_2) there is a problem due to their low solubility in water as a solvent for chemical reagents. Moreover, nitrogen monoxide is almost insoluble. To solve the problem, you need to convert NO into more soluble substances. This can be achieved by oxidation. The process of oxidation by ozone (O_3) is considered in the work. As a result of oxidation of nitrogen monoxide, a few chemical compounds are formed: dioxide (NO_2) and trioxide (NO_3) of nitrogen and trioxide (N_2O_3), tetroxide (N_2O_4) and pentoxide (N_2O_5) of dinitrogen. The latter compound is the most soluble in water, so it is a target in the conversion of nitrogen oxides. To obtain the data necessary to study the process of conversion of nitrogen oxides, an experimental laboratory-scale installation was built. For a more detailed study of the chemical process in the oxidizing reactor using a mathematical model, a series of calculations were performed. The result is a distribution of the concentrations of

compounds formed during the passage of the gas mixture along the reactor.

Laboratory Installation

The central object of the laboratory installation is the oxidation reactor, in which the process of conversion of nitrogen oxides takes place. Reactor height is 510 mm. The volume of the reaction zone of the reactor is 2 liters. The average inner diameter of the reaction zone is 71 mm. The length of the reaction zone is 500 mm. Reactor cover thickness is 10 mm. The outlets of the gas supply pipes are located at a distance 50 mm from the bottom of the reactor. The gas mixture in the oxidizing reactor is heated by an external spiral heater. The temperature of the gas mixture is measured by thermocouples, which are located at a distance 50 mm from the bottom and lid of the reactor. Figure 1 shows a schematic diagram of a laboratory installation. The experiments were performed on a mixture of model gas and ozonated atmospheric air. Model gas with a $\text{NO} + \text{N}_2$ fixed composition is fed into the reactor through a pipe into the lower part of the reactor. Ozonated moist air is supplied there through another pipe. The flow rates of both gas streams are measured using two flow meters. Ozonation of air is carried out in the O_3 generator. The concentration of ozone in the air is measured by a spectrometer. After the oxidation reactor, the gases pass through the destructor O_3 to prevent the entry of ozone into the gas analyzer and to damage its sensors.

Figure 1. A Schematic Diagram of a Laboratory Installation.

According to certificate of the model gas tank, the concentration of nitrogen monoxide is 1000 ppm, and the rest is molecular nitrogen. However, measurements with a gas analyzer showed the presence of nitrogen dioxide in small quantities.

Physical Experiments

According to [2, 3] during the oxidation of nitrogen oxides by ozone in the oxidizing reactor, the following chemical reactions preferably occur:

The rate of any chemical reaction depends on the concentration of the reagents and the reaction rate constant. The value of the chemical reaction rate constant at a certain gas temperature is calculated according to the kinetic parameters [4]. For example, Table 1 shows the values of the rate constants of reactions (1)–(9) at a gas mixture temperature 25 °C.

The dinitrogen trioxide is unstable. This fact is confirmed by the fact that for reactions (3) and (6) the values of the rate constants are close. The situation is similar for reactions with the dinitrogen tetroxide. The rate constants for reactions (4) and (7) are also close. Therefore, these two compounds can be ignored in the calculations. For reaction (5), the rate constant is almost on 11 orders of magnitude greater than for reaction (8). Therefore, reaction (8) can also be ignored. Reaction (9)

Table 1. Reaction Rate Constants

Reaction	Value	Unit
(1)	$1.123 \cdot 10^{+7}$	L/(mole·s)
(2)	$2.160 \cdot 10^{+4}$	L/(mole·s)
(3)	$4.710 \cdot 10^{+8}$	L/(mole·s)
(4)	$5.000 \cdot 10^{+7}$	L/(mole·s)
(5)	$1.140 \cdot 10^{+9}$	L/(mole·s)
(6)	$3.617 \cdot 10^{+9}$	1/s
(7)	$4.496 \cdot 10^{+8}$	1/s
(8)	$6.559 \cdot 10^{-2}$	1/s
(9)	$1.239 \cdot 10^{-4}$	L/(mole·s)

is slow, as indicated by the value of its rate constant. Thus, to calculate the concentration of N_2O_5 at the outlet of the oxidant reactor, the use of balance equation (10) is legitimate:

$$[\text{N}_2\text{O}_5]'' = [([\text{NO}]' - [\text{NO}]'') + ([\text{NO}_2]' - [\text{NO}_2]'')]/2, \quad (10)$$

where $[\text{X}]'$ and $[\text{X}]''$ – the concentration of gas at the inlet and outlet of the reactor, respectively, ppm.

To study the process of conversion of nitrogen oxides, a series of experiments were performed on the laboratory installation described above. The composition of the model gas in the tank is fixed, and the power of the ozone generator is constant. Therefore, to vary the molar ratio of O_3/NO , the consumption of model gas and ozonated air changes. The consumption of model gas was set at 0.25 L/min, 0.5 L/min, 1.0 L/min and 2.0 L/min. Atmospheric air flow was set at 0.5 L/min and 1.0 L/min. The temperature of the gas mixture in the reaction zone of the reactor varied from 20 °C to 150 °C.

The NO and NO_2 concentrations were measured during the physical experiments at the outlet of the chemical oxidizing reactor. The concentrations of monoxide and nitrogen dioxide at the reactor inlet were calculated from the known composition of the model gas and the volumetric flow rates of both gas streams at a given temperature were measured. The concentration of N_2O_5 is calculated by formula (10). Table 2 shows the values of meas

Table 2. Concentrations of Gases, ppm

t, °C	NO'	NO ₂ '	O ₃ '	O ₃ '/NO'	NO''	NO ₂ ''	N ₂ O ₅ ''
20	742	44	162	0.218	605	134	24
20	627	43	170	0.271	486	132	26
20	588	62	271	0.461	383	173	47
20	469	40	249	0.531	277	145	44
20	323	26	337	1.043	74	120	78
20	212	28	486	2.292	0	48	96
20	206	30	518	2.515	0	71	83
75	740	37	172	0.232	592	156	15
75	610	36	179	0.293	460	148	19
75	618	48	287	0.464	386	221	30
75	464	31	267	0.575	247	182	33
75	317	18	357	1.126	34	182	60
75	226	25	440	1.947	15	41	98
150	743	28	167	0.225	677	86	4
150	623	28	178	0.286	503	115	17
150	621	29	273	0.440	500	124	13
150	478	25	271	0.567	331	129	22
150	325	12	345	1.062	164	122	26
150	275	31	424	1.542	96	101	55

ured and calculated concentrations of individual gases at the inlet and outlet of the reactor.

Table 2 shows that with increasing initial concentration of ozone in the gas mixture, the concentration of nitrogen monoxide at the outlet of the reactor gradually decreases. The concentration of nitrogen dioxide fluctuates slightly. But it has a general tendency to decrease at the output. As a result of the oxidation process of nitrogen oxides from the table it is seen that the concentration of dinitrogen pentoxide increases.

The main attention in the study of the oxidation process is paid to the conversion of nitrogen oxides. In addition, the efficiency of ozone use must be closely monitored because the presence of ozone outside the reactor in a gaseous medium makes it aggressive. The equipment of the laboratory installation does not allow simultaneous measurement of ozone at the inlet and outlet of the oxidizing reactor. The concentration of ozone at the reactor outlet was determined by a mathematical model [5]. Figure 2 shows the efficiency of NO conversion because of physical experiments in a laboratory setting. The conversion efficiency of nitrogen oxide is defined as $1 - \text{NO}''/\text{NO}'$.

The process of conversion of nitrogen oxides can be divided into three ranges: $\text{O}_3/\text{NO} < 1$, $\text{O}_3/\text{NO} = 1 \dots 2$ and $\text{O}_3/\text{NO} > 2$. The figure shows that at a ratio of $\text{O}_3/\text{NO} < 1$ the efficiency of conversion of nitrogen monoxide does not exceed 70 %. Whereas for most experimental points the efficiency of ozone use is more than 90 %. The concentration of nitrogen dioxide is gradually increasing.

Figure 2. The NO Conversion Efficiency.

Moreover, the behavior of the parameters weakly depends on the temperature of the gas mixture below 100 °C. This is due to the fact that with increasing temperature, the reaction rate increases, but it reduces the residence time of gases in the reaction zone, i.e. the reaction time. When the ratio of O_3/NO from 1 to 2, the conversion efficiency of nitrogen monoxide is in the range of 70...90 %, and the efficiency of ozone is more than 90 %. The concentration of nitrogen dioxide at the outlet of the reactor remains almost at the previous level because more nitrous oxide is formed. When the ratio $\text{O}_3/\text{NO} > 2$, the conversion efficiency of nitrogen monoxide is in the range of 90...100 %. The efficiency of ozone use is reduced to 70...50 %. However, the concentration of nitrogen dioxide decreases sharply due to the intensive formation of N_2O_5 .

At a gas mixture temperature greater than 100 °C and an O_3/NO ratio greater than 2, the conversion efficiency of nitrogen monoxide is markedly reduced. Under such conditions, the effect of gas temperature increases markedly. Temperature is influenced by two factors: the volumetric flow rate of the gas mixture and the reaction rate constant. The residence time of the gases in the reactor depends on the volumetric flow rate, namely the reaction time. To determine the effect of volumetric flow rate according to the mathematical model [5], calculations were made with a longer residence time of gases in the reactor conditionally with a higher height. It was found that in comparison with the regime at a gas mixture temperature of 20 °C, the conversion efficiency of nitrogen monoxide at a temperature of 150 °C is reduced by a maximum of 46 %. As the temperature increases, the reaction rate constant also increases, because of which the reaction rate also increases. But the rates of reactions (1), (2) and (5) increase more slowly than reaction (9). Now there is an effect of ozone concentration, which decreases faster. According to [6], up to 50 % of ozone can decompose at temperatures above 100 °C, depending on the concentration. Thus, due to the decomposition of ozone, the conversion efficiency of nitrogen monoxide decreases even more.

Numerical Experiments

The data of physical experiments are supplemented by the results of calculations according to the mathematical model [5]. Below, Figures 3, 4,

5 and 6 show the calculated gas concentrations for the characteristic modes of physical experiments at different ratios of the initial concentrations of ozone to nitrogen monoxide. The distribution of the content of individual gases in the reaction zone is constructed depending on the residence time of the gas mixture in the chemical oxidizing reactor.

At a low O_3/NO ratio, ozone is not enough to convert all nitrogen oxides. It is not enough, first, for the oxidation of nitrogen monoxide, but nitrogen trioxide and dinitrogen pentoxide have already begun to form. The conversion efficiency of nitrogen monoxide reaches 18.46 %, and the efficiency

of ozone use is 99.38 %.

At a ratio of O_3/NO slightly more than one, ozone is still not enough for the complete conversion of nitrogen monoxide because O_3 is spent on the oxidation of nitrogen dioxide. Increasing the concentration of the latter leads to more intense formation of dinitrogen pentoxide. Under such conditions, the conversion efficiency of nitrogen monoxide increases to 77.09 %, while the efficiency of ozone use becomes 97.03 %.

As the O_3/NO ratio approaches two, the behavior of the parameters is notable for the fact that the line of nitrogen dioxide content is bent with

Figure 3. Changes in the Gas Content over Time at $O_3/NO = 0.218$: 1 – NO; 2 – NO_2 ; 3 – N_2O_5 ; 4 – O_3 .

Figure 5. Changes in the Gas Content over Time at $O_3/NO = 1.947$: 1 – NO; 2 – NO_2 ; 3 – N_2O_5 ; 4 – O_3 .

Figure 4. Changes in the Gas Content over Time at $O_3/NO = 1.043$: 1 – NO; 2 – NO_2 ; 3 – N_2O_5 ; 4 – O_3 .

Figure 6. Changes in the Gas Content over Time at $O_3/NO = 2.515$: 1 – NO; 2 – NO_2 ; 3 – N_2O_5 ; 4 – O_3 .

the formation of a small "hill". Most of the nitrogen monoxide has been oxidized and there is enough ozone to oxidize the nitrogen dioxide. Dinitrogen pentoxide is actively formed. The conversion efficiency of nitrogen monoxide reaches 93.36 %, and the efficiency of ozone use decreased to 70.23 %. Ozone is not fully used due to the lack of residence time in the chemical reactor of monoxide and nitrogen dioxide at low concentrations.

When $O_3/NO > 2$, ozone is sufficient for the complete conversion of nitrogen monoxide. Ozone is sufficient for the oxidation of nitrogen dioxide, but there is not enough time for a deeper conversion. The level of inflection of the NO_2 content line increased almost one and a half times compared to the previous mode. Dinitrogen pentoxide is actively formed. Nitrogen monoxide is completely converted, although there is still a small amount of nitrogen dioxide. The efficiency of ozone use decreased to 55.79 % due to the low rate of oxidation of nitrogen dioxide and the lack of residence time in the chemical reactor of the gas mixture.

Discussion

Figure 3 shows that at a low relative concentration of ozone ($O_3/NO = 0.218$) the curves for NO and O_3 are almost equidistant, and the curve for NO_2 is mirror to NO relative to the conditional line 400 ppm. The formation of N_2O_5 is very sluggish. Thus, we can say that there is mainly an oxidation reaction of nitrogen monoxide to dioxide. As the relative concentration of ozone increases ($O_3/NO = 1.043$), the distance between the curves for NO and O_3 increases markedly (see Figure 4). Nitrogen dioxide is now oxidized to trioxide, which increases ozone consumption. The presence of NO_2 and NO_3 gases in the mixture leads to the formation of a more noticeable amount of N_2O_5 . At approach to the point $O_3/NO \approx 2$, the picture of the distribution of gas concentrations along the chemical reactor changes radically (see Figure 5). At the beginning of the process, the concentrations of ozone and nitrogen oxides change dramatically. Then the reaction of formation of dinitrogen pentoxide accelerates. Nitrogen dioxide is used not only for the formation of NO_3 , but also for the formation of N_2O_5 . The concentration of NO_2 begins to decrease, so the curve appears inflection, which forms a distinct "hill". Figure 6 ($O_3/NO = 2.515$) shows that all nitrogen monoxide is converted to dioxide. At the end of the process there is a signifi-

cant supply of ozone, but the nitrogen dioxide is not completely oxidized due to lack of time with decreasing of an oxidation reaction rate.

Analysis of the results of physical experiments obtained in a laboratory installation shows that in the oxidizing reactor the nitrogen oxides are not completely converted even with a significant excess of ozone. This mainly applies to nitrogen dioxide. The reason for this situation in the real process is the hydrodynamic regime, in which the individual gases in the reaction zone are poorly mixed. Under such conditions, the concentrations of gases that must to react in the local areas of the reactor are low. Therefore, the reaction rates in these zones are small, so there is not enough time to react to the end. It is calculated that the gas flow rate varies from 5 mm/s to 15 mm/s, and the Reynolds number is in the range of 20...55 units. That is, in the reaction zone, a laminar regime of gas motion is established, when the main component of mass transfer in volume is diffusion. It is also small due to the temperature, which does not exceed 150 °C. To adequately describe the process of oxidation of nitrogen oxides in the equation of the reaction rate of the mathematical model, correction factors are added, which consider the difference between the concentrations of reagents in real mixing from the ideal. When calculating the experimental points according to the mathematical model, the correction factors are determined for each mode separately.

For the characteristic experimental points, calculations were performed with ideal gas mixing in the reaction zone of the oxidizing reactor. Table 3 shows the results of physical and numerical experiments for these points at the outlet of the reactor.

Table 3. Comparison of Experimental Results (ppm)

O_3/NO	Real mixing				Ideal mixing			
	NO	NO_2	N_2O_5	O_3	NO	NO_2	N_2O_5	O_3
0.218	605	134	24	1	580	206	0	0
1.043	74	120	78	10	0	321	14	0
1.947	15	41	98	131	0	0	126	88
2.515	0	71	83	229	0	0	118	193

Calculations for ideal gas mixing, when $O_3/NO < 1$, ozone is spent entirely only on the formation of nitrogen dioxide. At $O_3/NO \approx 1$ ozone is also consumed all, but part of it goes to the oxidation of nitrogen dioxide to trioxide with the subsequent formation of dinitrogen pentoxide.

As the O_3/NO molar ratio approaches to 2 and more, nitrogen oxides are completely converted to dinitrogen pentoxide. By the way, there is so much ozone that it is present in the gas mixture at the outlet of the oxidizing reactor. Regardless of the mixing, less dinitrogen pentoxide is formed in the $O_3/NO > 2$ mode than in the $O_3/NO \approx 2$ mode because the initial nitrogen monoxide content is less (see Table 2), so less nitrogen dioxide is formed.

Conclusions

Studies have shown that the oxidation of both nitrogen monoxide and dioxide must be considered when determining the required amount of ozone. Intensive conversion of nitrogen oxides occurs at the initial molar ratio of ozone to nitrogen monoxide in the range of 1.5... 2. At a temperature of gases greater than 100 °C, the efficiency of NO conversion reaches 60 % or more, and at temperatures less than 100 °C – 90 %.

During actual mixing in a laboratory reactor, nitrogen oxides are converted almost completely when the O_3/NO molar ratio exceeds 2. This amount of ozone is required for the oxidation of nitrogen monoxide and most of the nitrogen dioxide formed, with some excess to compensate for imperfect gas mixing.

Lack of time for the conversion of nitrogen oxides in the reactor of the laboratory installation, even with a significant excess of ozone and the temperature of the gas mixture above 100 °C can reduce the conversion efficiency by 46 %.

The residence time of the gas mixture in the oxidizing reactor during physical experiments exceeds 50 seconds. In a real installation for the removal of nitrogen oxides from the flue gases, such a time is not acceptable. It should be much smaller.

Calculations of the oxidation process with perfect mixing of gases in the oxidizing reactor of the laboratory installation showed that nitrogen monoxide is completely converted into dioxide after 500 ms at $O_3/NO \approx 2$. Although complete oxida-

tion of NO at $O_3/NO \approx 1$ takes an order of magnitude longer.

The conversion efficiency of nitrogen oxides depends not only on the amount of ozone in the gas mixture, but also on the hydrodynamic regime of gas movement in the reaction zone of the reactor. To increase efficiency, it is necessary to better mix the gases by turbulence of the flow.

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Экспериментальное исследование процесса окисления оксидов азота озоном

Необходимость снижения выбросов загрязняющих веществ, в частности, оксидов азота в Украине по требованиям нормативных документов требует применения современных технологий и методов очистки отходящих газов на промышленных предприятиях. Особенно это касается тепловых электростанций, которые являются мощными источниками выбросов оксидов азота. Технологической частью мокрого или полусухого метода очистки является участок окисления оксидов азота с целью получения легкорастворимых соединений. В работе представлено исследование процесса окисления озоном оксидов азота в химическом реакторе. Данные для выполнения анализа процесса получены в результате проведения физических экспериментов на лабораторной установке и связанных с ними расчётов по математической модели. Исследования процесса окисления показали, что необходимое количество озона зависит не только от содержания монооксида азота, но и от содержания диоксида азота. Процесс преобразования монооксида азота до удовлетворительного уровня происходит при начальном значении мольного соотношения озона к монооксиду азота в диапазоне 1,5–2. Эффективность преобразования монооксида азота доходит до 90 % при температуре газов меньше 100 °С. Для достижения высокой эффективности преобразования при температуре газов выше 100 °С необходимо увеличить начальное содержание озона, когда мольное соотношение превышает 2. Анализ показывает, что эффективность преобразования монооксида азота в большей мере зависит от времени пребывания газовой смеси в реакционной зоне. Из-за недостатка времени при определённых условиях эффективность снижается приблизительно на 46 %. Для её повышения необходимо увеличить скорость реакций окисления за счёт улучшения перемешивания газов вследствие повышения турбулентности потока в реакторе-окислителе. *Библ. 6, рис. 6, табл. 3.*

Ключевые слова: оксиды азота, озон, реактор, эксперимент.

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Експериментальне дослідження процесу окислення оксидів азоту озоном

Необхідність зменшення викидів забруднюючих речовин, зокрема оксидів азоту, в Україні за вимогою нормативних документів потребує застосування сучасних технологій та методів для очищення відхідних газів на промислових підприємствах. Особливо це стосується теплових електростанцій, які є потужними джерелами викидів оксидів азоту. Технологічною

частиною мокрого або напівсухого методу очищення є ділянка по окисленню оксидів азоту з метою отримання легкорозчинних сполук. У роботі наведено дослідження процесу окислення озonom оксидів азоту у хімічному реакторі. Дані для проведення аналізу процесу одержано при виконанні фізичних експериментів на лабораторній установці та пов'язаних з ними розрахунків за математичною моделлю. Дослідження процесу окислення показали, що необхідна кількість озону залежить не тільки від вмісту монооксиду азоту, але й від вмісту діоксиду азоту. Процес перетворення монооксиду азоту до задовільного рівня відбувається при початковому значенні мольного співвідношення озону до монооксиду азоту в діапазоні 1,5–2. Ефективність перетворення монооксиду азоту доходить до 90 % при температурі газів менше 100 °С. Для досягнення високої ефективності перетворення при температурі газів вище 100 °С потрібно збільшити початковий вміст озону, коли мольне співвідношення перевищує 2. Аналіз показує, що ефективність перетворення монооксиду азоту значною мірою залежить від часу перебування газової суміші у реакційній зоні. Через брак часу за певних умов ефективність знижується приблизно на 46 %. Для її підвищення потрібно прискорити швидкість реакцій окислення за рахунок кращого перемішування газів шляхом збільшення турбулізації потоку у реакторі-окиснювачі. *Бібл. 6, рис. 6, табл. 3.*

Ключові слова: оксиди азоту, озон, реактор, експеримент.

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