



Absorption of Nitroses Gases Using a Solution of Ferrous Sulphate

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ABSTRACT: The article is devoted to the absorption capture of nitrous gases using a solution of iron sulfate in step absorbers for its further desorption and the production of sodium nitrite based on it or its use to produce nitric acid. The results of the study can be particularly widely used in industries characterized by intermittent emissions of nitrogen oxides with a wide range of nitrogen oxide contents. The dependence of the degree of absorption and desorption of nitrogen oxides on the temperature and concentration of the iron sulfate solution, as well as on the time during desorption, was determined.

KEYWORDS: Absorber, nitrous gases, ferrous sulfate, nitrogen oxides, nitric acid, ecology, purification of nitrous gases.

I. INTRODUCTION

It is known that the chemical industry makes a significant contribution to the development progress of our country. The most serious environmental problem in the industry is emissions of exhaust gases containing toxic substances, including nitrogen oxides. The maximum permissible average daily content of nitrogen oxides in terms of N_2O_5 in the atmospheric air of populated areas should not exceed $0,1 \text{ mg/m}^3$, while simultaneously complying with the requirement for a single maximum content of nitrogen oxides of no more than $0,3 \text{ mg/m}^3$. In the production of nitric acid, the main problem is the emission of nitrous gases and the development of methods to reduce their emission volume. There are various methods for neutralizing nitrous gases: purification of gases from nitrogen oxides in an equally flowing hollow tower; purification of gases from nitrogen oxides in combined type devices; purification of gases from weakly oxidized nitrogen oxides, absorption of nitrogen oxide by ammonium sulfite solutions; oxidation and absorption of nitrogen oxides by aqueous solutions of oxidizing agents; purification of gases from nitrogen oxides with aqueous solutions of hydrogen peroxide; absorption of weakly oxidized nitrogen oxides by aqueous solutions of potassium permanganate; absorption of highly oxidized nitrogen oxides; studies of the absorption of nitrogen oxides by organic sorbents; adsorption of nitrogen oxides by solid sorbents; purification of gases from nitrogen oxides and other impurities using peat-alkaline sorbents to produce peat-nitrogen fertilizers; absorption of nitric oxide by ferrous sulfate solutions [1].

II. SIGNIFICANCE OF THE SYSTEM

The article is devoted to the absorption capture of nitrous gases using a solution of iron sulfate in step absorbers for its further desorption and the production of sodium nitrite based on it or its use to produce nitric acid. The study of methodology is explained in section III, section IV covers the experimental results of the study, and section V discusses the future study and conclusion.

III. METHODOLOGY

Here is a method of absorption of nitrous gases by nitric acid condensate supplied from a refrigerator-condenser to the middle part of the absorption column; absorption is carried out with an additional supply of nitric acid taken from the breaker plate of the column, regenerated from unreacted nitrogen oxides and diluted to a concentration of 5-10 %, in two streams, one of which is directed to the first plate from above in the amount of 1,0-2,0 kg of liquid/kg of gas, and on



the other hand in the amount of 0,09-0,12 kg of liquid/kg of gas under the impact plate, and regeneration is carried out by supplying air into the regenerator in an amount of 0,04-0,1 kg of gas/kg of liquid [2].

IV. EXPERIMENTAL RESULTS

There is a method for complete capture of nitrogen oxides from nitrous tail gases of nitric acid plants. The method consists in the absorption of nitrous gases by lead hydroxide and its subsequent regeneration according to the following scheme: Nitrogen oxides are passed through a scrubber irrigated with a suspension of lead hydroxide in a saturated solution of nitrite and sodium nitrite; the resulting basic lead nitrite and nitrate are separated from the mother liquor and treated with nitric acid to convert the nitrite to nitrate. The nitrogen oxides released during this process are used to saturate the production nitric acid; After separating the crystals, the mother liquor is mixed with lead hydroxide and returned to the process; the resulting solution of lead nitrate, containing part of the nitrate in the form of crystals, is treated with a solution of ammonium nitrate to dissolve the crystals and then with ammonia to convert lead nitrate into ammonium nitrate. The released lead hydroxide is separated from the liquid and, after washing with water, is returned to the processes. The remaining liquid is a saturated solution of ammonium nitrate and is sent for evaporation [3].

There is a method [4] according to which the resulting solutions of nitrates and nitrites formed after the absorption of nitrous gases are treated with nitric acid in order to oxidize nitrites into nitrates, and the nitrogen oxides released in this case are used in the sulfuric acid system. With a soda or alkaline solution, nitrogen oxides in the form of various oxides are captured from the tail gases until they contain no more than 0,06 % NO, after which the gases are released into the atmosphere. The resulting solutions after alkaline absorption, which are a mixture of nitrite (80 %) and sodium nitrate (20%), containing an admixture of sodium sulfate, are sent to an inversion located at the head of the system, where they are converted into solutions of sodium nitrate, released after evaporation as ready-made nitrogen fat. In this case, per ton of sulfuric acid produces 15-25 kg of sodium nitrate. The loss of nitric acid with exhaust gases is no more than 5 g per 1 ton of sulfuric acid. Fresh nitric acid is used for inversion; the system is powered by nitrogen oxides released after the inversion. This will allow inversion to be carried out at almost no cost.

There is a method [5] according to which sulfur carbon obtained from wood is used as a sorbent, which makes it possible to obtain a high-quality sorbent that can be restored and replaces calcium or magnesium cations in solutions with sodium or hydrogen ions. The production of sulfonated coal from sawdust and its use is as follows. Sawdust, released to 8-10% moisture content and freed from impurities, is loaded into a reactor, where it is treated with concentrated sulfuric acid (sp. 1,84) at a ratio of 7-7,5 parts by weight of acid per one part of sawdust (4 liters of acid per 1 kg of sawdust). The mixture is stirred for an hour, and then the resulting sulfonate is washed from the acid. Further processing is carried out depending on the application: with a one percent H₂SO₄ solution or a ten percent NaCl solution. If the sorbent is to be stored or transported, it should be dried. The use of sulfonated charcoal obtained from sawdust can be done either by filtering solutions through it, or by mixing it with solutions when heated and filtering through filter presses. The dosage of sulfonated carbon depends on the content of salts to be removed, the consistency and color of the solutions. Regeneration of sulfonated coal is carried out with a one percent solution of ammonium sulfate or a ten percent solution of NaCl.

In [6], the effect of temperature on the degree of purification of exhaust gases from nitrogen oxide and nitrogen dioxide with a urea solution was studied. It has been shown that with increasing temperature the degree of purification increases. The degree of conversion of nitrogen dioxide in the temperature range 30–80 °C is 90–98 %, while the conversion of nitrogen oxide is from 13 to 58%. It has been established that the addition of an oxidizing agent, nitric acid, up to 10% to a urea solution increases the degree of conversion of nitric oxide by 15–20 %. The addition of oxygen increases the conversion of nitric oxide to nitrogen by 40 %. The influence of the content of active components of the reaction mixture on the degree of conversion of nitrogen oxides in the exhaust gases was studied. The optimal composition of the reaction mixture was selected: – 10–20 % urea; 5–10 % nitric acid. It has been shown that the optimal conditions for the conversion of nitrogen oxide and dioxide are: solution temperature 30–35 °C; consumption of urea solution for cleaning is no more than 4 m³/h; oxygen content in the gas for purification is at least 7%; the speed of the rotor blades is no more than 1500 rpm. It was shown that at a urea solution temperature of 30–35 °C, a urea solution flow rate of 4 m³/h, and a rotor blade speed of 1500 rpm, a degree of purification from nitrogen oxides of 98–100% was achieved, which is much higher than when using a sodium solution. alkalis.

The work [7] shows that the production of oxalic acid is based on the interaction of nitric acid with sucrose. As a result of this process, up to 200 kg of nitrogen oxides are released into the atmosphere per ton of product produced. This leads to excessive atmospheric pollution and deterioration of sanitary and hygienic working conditions in production. Based on theoretical and experimental research, a new method for the thermal decomposition of nitrogen oxides has been developed, providing sanitary purification of waste gases during the production of oxalic acid to

maximum permissible concentrations. When studying on a plasmatron, it was found that by thermal decomposition of nitrogen oxides, their concentration from any to the initial one can be reduced to equilibrium at a given temperature. By introducing gaseous reducing agents, the reaction of thermal decomposition of nitrogen oxides can be shifted towards their decomposition into their original elements. The concentration of nitrogen oxides after decomposition can be increased to 0,1-0,3 % with an initial concentration of 2-3 %. The thermal decomposition of nitrogen oxides in the presence of coke begins at a temperature of 500 °C. At 1000 °C, the degree of decomposition of nitrous gas approaches 100 %.

However, this problem has not yet found its complete solution. A detailed study and analysis of methods for the absorption of nitrogen oxides by water is a chemisorption process, leading to the formation of a multicomponent system containing initial NO, N₂O₄, NO₂, N₂O₃ and final products H₃O⁺, NO³⁻, NO²⁻ consists of the following stages: diffusion of nitrogen oxides (NO, NO₂) from the gas phase into liquid through a two-layer film at the gas-liquid interface; the formation of nitric and nitrous acids as products of the interaction of nitrogen oxides with water; decomposition, or rather disproportionation of nitrous acid and the return of the resulting nitrogen oxide to the gas phase [8].

In this work, solutions of ferrous sulfate having concentrations from 5 to 40 % were used for research. The purpose of this work is not only the absorption of nitrous gases but also its use to produce sodium nitrite. To carry out the experiments, we used a laboratory installation consisting of six reaction vessels, the first reactor for producing nitrogen oxides, five reactors - absorbers of nitrogen oxides. In the first reactor, a mixture of nitrogen oxide is formed by the interaction of copper with non-concentrated nitric acid (59 %). The remaining reactors absorb incoming nitrogen oxide from the first reactor. To carry out the experiment, 20 ml of ferrous sulfate solution were placed in five reactors (absorber volume 50 ml).

Nitrogen oxides obtained in the first reactor were passed through the solution using a water-jet pump. After the end of the experiment, all solutions were individually brought to the 100 ml mark, from where an aliquot was taken for nitrogen determination. In addition, the solution was regenerated and nitric oxide was obtained in concentrated form and it was absorbed using a sodium hydroxide solution. The total content of nitrogen oxides in solutions was determined by Kjeldahl with subtraction from the total nitrogen contained in the original nitric acid [9]. The results of the experiments are given in tables 1 and 2. Chemical analyzes of the solutions used for the absorption and desorption of nitrous gases established the degree of absorption and desorption of nitrous gases depending on temperature, solution concentration and also on time during desorption of nitrous gases.

Table 1
The degree of absorption of nitrous gases in solutions of ferrous sulfate depending on temperature and concentration

Concentration of ferrous sulfate solutions	Degree of absorption of nitrous gases in absorbers, in terms of N%					Total absorption rate, %
	Absorber № 1	Absorber № 2	Absorber № 3	Absorber № 4	Absorber № 5	
Solution temperature, 35 °C						
5 %	24,06	16,21	9,05	4,64	2,18	56,14
8 %	26,02	17,10	9,68	5,03	2,48	60,31
12 %	27,63	18,63	10,32	5,55	2,66	64,79
15 %	29,80	20,04	11,20	6,27	2,99	70,30
20 %	31,66	21,70	11,64	6,94	3,26	75,20
Solution temperature, 30 °C						
5 %	25,24	17,89	9,36	4,70	2,44	59,63
8 %	27,18	18,47	10,23	5,31	2,69	63,88
12 %	29,31	19,92	11,06	5,71	2,97	68,97
15 %	31,07	21,14	11,87	6,41	3,33	73,82
20 %	32,47	23,12	12,08	7,10	3,85	78,62
Solution temperature, 25 °C						
5 %	26,12	18,53	10,07	4,82	2,90	62,44
8 %	28,51	19,68	10,41	5,32	3,29	67,21
12 %	30,64	20,96	11,42	5,83	3,41	73,15
15 %	32,59	22,79	12,45	6,67	3,81	78,31
20 %	33,98	24,46	12,42	7,26	3,97	82,09
Solution temperature, 20 °C						

5 %	27,56	19,77	10,49	4,91	2,96	65,69
8 %	29,78	20,56	10,92	5,48	3,32	70,06
12 %	31,66	21,89	12,06	5,98	3,58	75,17
15 %	33,86	24,12	12,97	6,72	3,83	81,50
20 %	35,47	25,94	13,54	7,31	4,27	86,53

Table 2
Dependence of the degree of desorption of nitrogenous gases on temperature and time from FeSO₄ solution in terms of nitrogen

Indicators	Concentration of FeSO ₄ solution and molar ratios N : Fe ⁺²				
	5 %, 1,12:1	8 %, 1,12:1	12 %, 1,11:1	15 %, 1,10:1	20 %, 1,09:1
Desorption temperature, 75 °C					
Desorption degree, %	47,67	47,86	48,12	48,69	49,01
Equilibrium time, hour	7	7,2	7,3	7,35	7,5
Desorption temperature, 80 °C					
Desorption degree, %	64,31	64,72	65,04	65,32	65,76
Equilibrium time, hour	5,35	5,35	5,40	5,45	5,50
Desorption temperature, 85 °C					
Desorption degree, %	79,54	79,81	80,07	80,52	80,84
Equilibrium time, hour	4,00	4,05	4,05	4,10	4,10
Desorption temperature, 90 °C					
Desorption degree, %	95,28	95,46	95,88	95,97	96,12
Equilibrium time, hour	1,5	1,5	1,55	1,6	1,6
Desorption temperature, 95 °C					
Desorption degree, %	99,57	99,59	99,56	99,59	99,63
Equilibrium time, hour	0,45	0,45	0,50	0,55	0,6

Based on the results obtained, we can conclude that the absorption of nitrous gases by ferrous sulfate solutions is 56,14–86,53 %, depending on the purification conditions. At a ferrous sulfate concentration of 5 % and a temperature of 35 °C, the absorption degree is 56,14 %. Reducing the temperature by 15 °C and increasing the concentration of the solution by 15 % allows you to increase the degree of absorption of nitric oxide by approximately 30 % (Table 1). Preliminary analysis results and calculations have shown that using solutions of ferrous sulfate to absorb nitrogen oxides at normal temperatures and pressure in five absorbers, it is possible to absorb up to 86,53 % of nitrogen oxides and almost completely desorption of nitrous gases up to 99,63 % at a temperature of 95 °C from gland sulfate solution (Table 2). Nitrous gases after high concentration desorption can be used to produce nitric acid, and the ferrous sulfate solution can be reused to absorb nitrous gases.

V. CONCLUSION AND FUTURE WORK

Thus, the dependence of the degree of absorption and desorption of nitrogen oxides on the temperature and concentration of the ferrous sulfate solution, as well as on the time during desorption, was determined.

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