



The Catalysts For The Determination Of Ammonia

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ABSTRACT

The regularities of the oxidation of combustible substances in the presence of various catalytic systems have been established, and catalysts have been selected on their basis for the sensitive elements of a highly sensitive ammonia monitoring sensor. The use of the created sensors significantly reduces the analysis error, increases the service life and stability of the device, and at the same time improves the reproducibility and selectivity of determining the micro concentration of ammonia.

KEYWORDS

Sensing element, analysis error, thermal effect, selectivity, monitoring, sensor, analyzer, ammonia.

INTRODUCTION

From a theoretical point of view, the oxidation of ammonia is interesting in that it is one of the few oxidation reactions of inorganic substances that proceed in several directions. Therefore, when developing a catalyst for the selective oxidation of ammonia, not only its activity is important, but also its selectivity [1]. A detailed study of the kinetics of the mechanism of ammonia oxidation on oxide catalysts was carried out in [2, 3], where they

found that the rate of formation of N_2 and N_2O and selectivity at temperatures below $40^\circ C$ do not depend on the contact time of the reaction mixture.

The established regularity indicates the absence of the inhibition process by the reaction products, which is one of the important factors in the thermocatalytic determination of ammonia from a gas-air mixture. As you know, the oxidation of

ammonia is accompanied by a significant thermal effect. Therefore, one of the promising methods for the determination of ammonia may be the semiconductor and thermocatalytic method. The primary task in the development of a semiconductor and thermocatalytic ammonia sensor is the selection of gas-sensitive films and catalytic systems with improved operational properties.

MATERIALS AND METHODS

Known catalysts for the oxidation of ammonia, have significant drawbacks are insufficiently active, little stable and non-selective, in some cases, their preparation is expensive and difficult to implement, some of them include valuable and scarce raw materials. All this limits the use of known catalysts as catalytic active elements of a thermocatalytic and semiconductor ammonia sensor [4-7]. In the oxidation of ammonia, the main attention of researchers was paid to the selection of active catalysts, and very little attention was paid to the issues of selectivity of catalysts in the oxidation of ammonia in the presence of hydrogen, carbon monoxide, hydrocarbons, etc.

Moreover, due to the incomparability of data obtained under different conditions on catalysts of different nature, it is impossible even to judge the selective oxidation of

ammonia in the presence of other combustible gases and vapours [3, 5,10].

In this regard, the primary task dedicated to the development of an ammonia sensor is the creation of sensitive sensor elements, selective catalytic systems with increased operational parameters. To select a catalyst for a thermocatalytic and semiconductor sensor providing a selective determination of ammonia, the characteristics of catalysts based on mixtures of metal oxides were studied. The catalysts were prepared from metal oxides: Mn, Mo, Zr, V, Cu, Cr, Ga, Ni, Cd, Co, Sn, Bi, Fe, and Ir, which are characterized by high activity and selectivity in the oxidation of combustible gases. Experiments to identify the activity and selectivity of catalysts in the oxidation of ammonia were carried out in the presence of hydrogen, carbon monoxide, and methane, which are often found together with the emissions of various objects (as part of gaseous discharges, production of ammonia, mineral fertilizers, etc.).

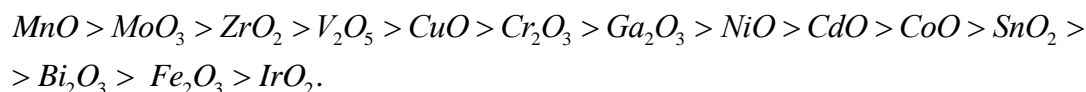
The activity of oxides with respect to ammonia was characterized by determining the content of ammonia in the reaction products. Judging by the decrease in the ammonia content, oxides of zirconium, molybdenum and manganese exhibit high activity at 150 °C, and the latter of them is capable of providing 100% conversion of ammonia (Table 1).

Table 1. The results of determining the activity of metal oxides during the oxidation of combustible substances (content in the mixture,% vol: H₂-1,55; Co-2,00; CH₄ -1,50; NH₃ -2,00 temp.

experience, 150 °C) No. p/p	Catalyst composition	Excess level, %			
		H ₂	Co	CH ₄	NH ₃
1	CoO	100,0	84,6	3,5	32,9
2	CdO	99,0	10,1	5,9	35,0
3	MnO	74,0	62,6	4,7	100,0
4	MoO ₃	66,6	82,8	3,5	84,8
5	SnO ₂	21,8	5,5	8,3	18,0
6	CuO	21,8	79,1	2,4	63,6
7	ZrO ₂	61,4	56,1	ZrO ₂	61,4
8	NiO	20,8	55,2	4,7	39,2
9	Ga ₂ O ₃	12,5	31,3	4,7	41,3
10	Fe ₂ O ₃	11,4	5,5	2,4	10,6
11	Bi ₂ O ₃	6,2	16,6	2,4	12,7
12	V ₂ O ₅	48,9	20,2	7,1	71,0
13	Cr ₂ O ₃	4,2	40,5	5,9	42,4
14	IrO ₂	2,6	12,0	8,3	4,2

In the results of the experiments, it was found that with an increase in temperature, the activity of copper and chromium oxides sharply increases; 100% conversion at 250 °C was observed in the presence of only V₂O₅. As can be seen from the results in Table 1, the oxides of gallium, cadmium and nickel showed moderate activity in the studied process (the conversion of ammonia from 35 to 42% at 150

°C). Oxides of tin, iron and bismuth exhibit low activity in the oxidation of ammonia. In their presence at 150 °C, the oxidation state of ammonia is at the level of 4.0-12.0%. According to experimental data, the catalytic activity of the investigated metal oxides during the oxidation of ammonia decreases in the following order:



Thus, the oxides of manganese, zirconium, molybdenum, and manganese are among the most active catalysts for the oxidation of ammonia. Vanadium, chromium, nickel, cadmium and cobalt oxides were moderately active. Oxides of iridium, bismuth and gallium are inactive. It should be noted that in all cases the oxidation of hydrogen and carbon monoxide was observed simultaneously with

ammonia, which excluded the possibility of using the studied individual oxides as catalysts in the development of a thermocatalytic sensor for the selective determination of ammonia in the presence of hydrogen. One of the possible methods for the development of a selective thermocatalytic ammonia sensor is also the use of thermosensitive (measuring and comparative) elements containing catalysts with different activity towards the

components of the gas mixture. Based on the results of studying the activity of a mixture of some metal oxides during the oxidation of ammonia, hydrogen, carbon monoxide and methane, a MnO-MoO₃ catalyst (80-20 wt.%) was selected for the measuring sensing element of the sensor. The best catalyst for the compensation sensitive element of the sensor under the studied conditions should be considered CdO-Bi₂O₃-ZrO₂ (50-30-20% of the mass), providing complete oxidation of hydrogen and carbon monoxide. In the presence of this catalyst, ammonia and methane are practically not oxidized. An increase and decrease in the catalyst components lead to a deterioration in selectivity [8-10]. The results of the effect of

temperature on the activity of the catalysts of the measuring and compensating sensor element of the selective thermocatalytic ammonia sensor are presented in Table 2. As follows from Table 2, in the temperature range 200-300 °C in the presence of catalysts of the measuring and compensating sensor element, identical oxidation of hydrogen and carbon monoxide is observed, i.e. the sensor is not sensitive to these gases. In this case, the catalyst of the measuring sensor ensures complete oxidation of the ammonia. Methane is practically not oxidized on the catalysts of the measuring and compensating elements at the same temperatures.

Table 2. The results of the effect of temperature on the activity of the catalyst of the thermocatalytic ammonia sensor (the content of the combustible component in the mixture, in% vol: H₂-1,55; Co-2,00; CH₄-1,50; NH₃-2,00).

№ п/п	Experiment temperature, °C	Oxidation state, in %			
		H ₂	Co	CH ₄	NH ₃
MoO-MoO ₃ (80 – 20%) measuring element					
1	150	86,0	92,0	-	94,0
2	200	100,0	100,0	-	99,0
3	250	100,0	100,0	-	100,0
4	300	100,0	100,0	2,7	100,0
5	350	100,0	100,0	17,0	100,0
CdO-Bi ₂ O ₃ -ZrO ₂ (50 – 30 - 20)- measuring element					
6	150	29,6	26,5	-	-
7	200	96,4	88,5	-	-
8	250	100,0	100,0	-	-
9	300	100,0	100,0	-	2,7
10	350	100,0	100,0	4,5	14,0

The composition of the reaction products of the catalytic oxidation of ammonia in the presence of a MnO-MoO₃ catalyst (80-20 wt.%) was determined by gas chromatography, where it was found that at low temperatures (up to 150 °C) molecular nitrogen is the only nitrogen-containing

reaction product. As the temperature rises (> 250 °C), nitrous oxide appears; at higher temperatures (> 300 °C), traces of NO are found. In all studied catalysts, the temperature of the onset of product formation in the N₂-N₂O-NO series was maintained. In the presence of catalysts for the measuring and comparative

sensitive element of the sensor at temperatures up to 300 °C, ammonia is oxidized almost only to elemental nitrogen. Thus, as a result of the experiments carried out, a catalyst was selected for the measuring and compensation elements of the thermocatalytic ammonia sensor.

In this case, the catalyst of the measuring element ensures the complete oxidation of ammonia, carbon monoxide and hydrogen, and carbon monoxide and hydrogen are oxidized on the catalyst of the compensating element, as a result, the output signal of the compensating element is proportional to the concentration of hydrogen and carbon monoxide, and the difference between the signals of the measuring and compensating sensor is proportional to the concentration of ammonia.

CONCLUSION

As a result of studying the activity of individual oxides and their mixtures during the oxidation of combustible gases, a thermocatalytic sensor was developed that provides selective determination of ammonia in various objects containing mixtures of combustible and explosive substances. The regularities of the oxidation of combustible substances in the presence of various catalytic systems have been established, and catalysts have been selected on their basis for the sensitive elements of a highly sensitive ammonia monitoring sensor.

The use of the created sensors significantly reduces the analysis error, increases the service life and stability of the device and at the same time improves the reproducibility and

selectivity of determining the micro concentration of ammonia.

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