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TECHNOLOGY OF SYNTHESIS OF EFFECTIVE CATALYSTS FOR NEUTRALIZATION OF WASTE GASES OF THE VEHICLES AND INDUSTRY

Abstract. The paper describes the technique of synthesis of catalysts on block metal carriers for neutralization of waste gases of the vehicles and the industry developed and improved by authors. For preparation of the secondary carrier there were used either aluminum oxide or aluminum oxide with the addition of a zeolite or Ce^{4+}, Ti^{4+}, Zr^{4+}, La^{3+}, Fe^{3+}. As the active phase were applied compounds of Mn, Ni, Co, Fe and the platinum group metals. The catalysts were tested in the process of complete oxidation of NO_x, CO, C_3H_8 and $NO_x+C_3H_6+O_2$ reaction. Activity of Pd-Mo of catalysts increases at addition in composition of the carrier of cations Ce^{4+} и Zr^{4+}. The activity of palladium catalyst in oxidation reactions CO - 90-100% and is independent of the concentration of metal, in complete oxidation of C_3H_8 at 473-623 K - 78-90%, for Pt -containing catalyst - 95-100%. For catalysts with base metals introducing of Ti^{4+} into the secondary carrier significantly increases the degree of reduction of nitrogen oxides with propylene.

Key words: catalyst, waste gases, ecology

Introduction

The problem of cleaning of motor transport exhaust gases and the industrial wastes - one of the most urgent problems of the modern world. The exhaust gases of motor transport contain in their composition the hydrocarbons, carbon monoxide, nitrogen oxides, carcinogenic substances which are toxic substances and represent a considerable threat for health of the population [1-6]. Increase in number of means of motor transport - one of the reasons of deterioration in a condition of ecology in the cities and large settlements. Today decrease in harmful emissions of motor transport and the industry to the international standards is possible only by catalytic methods [7-9]. As the catalysts of CO and hydrocarbons oxidation and decomposition of nitrogen oxides are used mainly noble metals on carriers having a high catalytic activity and heat resistance to poisons. Creation of new effective and stable catalysts of neutralization of waste gases of the industry and exhaust gases of motor transport and research of physical and chemical bases of improvement of the catalytic systems - an actual problem in the field of an ecological catalysis. One of the promising directions in this area - the partial replacement of noble metal on the oxide component [10-12]. The work purpose - development of technology of synthesis of samples of catalysts on block metal carriers and the study of their effectiveness in the laboratory processes and under real operating conditions by cleaning of toxic industrial and vehicle emissions.

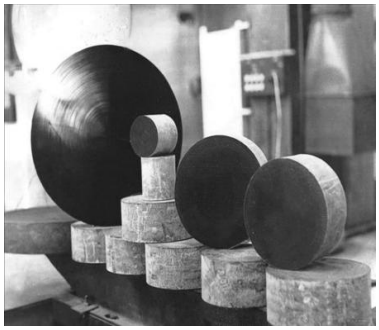
Experimental

The authors of this study on the results of many years of tests developed and perfected the technology of synthesis of highly stable catalysts for cleaning of motor vehicles exhaust gases and harmful emissions from industry on the basis of the monolithic metal block catalysts [14-19]. The monolithic block catalysts with the honey comb structure of channels, high thermal and mechanical stability maintain high efficiency of cleaning of exhaust gases from CO, hydrocarbons, nitrogen oxides and correspond to the EURO-3

standard. Degree of cleaning of exhaust gases of cars running on gasoline: CO-CH_x-90-100%, NO_x- 80-100%. The samples of block catalysts have a cylindrical shape which is convenient for placing at the source of toxic emissions. High durability of metal and high workability give the chance to make carrier walls rather thin that as a result provides the general significant increase in the geometrical area of a metal substrate. For preparation of the block carrier the heat-resistant foil 50 microns thick which is exposed to goffering is used, on a strip of a smooth foil the strip of a corrugated foil is imposed, then both strips of a foil are displaced in the cylindrical block and are fixed by welding.

The following stage in preparation of catalysts - the secondary carrier is applied on the prepared block metal carriers with the honey comb structure of channels. Figures 1, 2 show the laboratory and full-size samples of neutralizers, respectively.

Figure 1- The samples of catalysts on metal blocks for laboratory tests



(a)

(b)

Figure 2- Full-size catalytic neutralizers on block metal carriers for testing under real operating conditions: (a) general view, (b) cross-section

For preparation of full-metal catalysts on the block carriers there are trial installations on the pilot experimental plant of "D.V.Sokolski IFCE" (Fig.3).

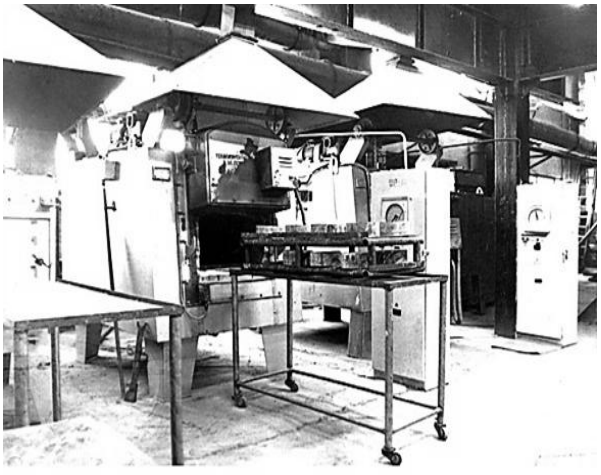
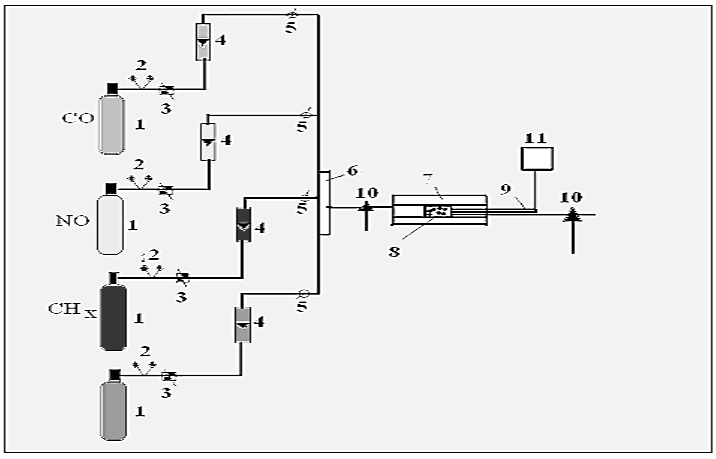


Figure 3- The kilns for full-size catalysts on metal block carriers

As a secondary carrier is used either alumina with the addition of zeolite or alumina modified by additives Ce^{4+}, Ti^{4+}, Zr^{4+}, La^{3+}, Fe^{3+}. For the preparation of solutions of the active components of the catalysts were applied oxides of Mn, Ni, Co, Fe, prepared from acetates and formiates. The previously weighed blocks were immersed in the required solution, then were shaken slightly from the excess solution between channels of the blocks, and were dried at 423 K for 2 hours in an oven. The blocks were calcined in an electric furnace at 873 K for 2 hours. The metal salts are decomposed to form metal oxides on the surface of the carrier the block. The blocks were weighed again and by a difference of masses before and after deposition the concentration of the deposited metal was determined. In case of use of catalysts on the basis of platinum metals in the reactions of oxidation of CO, hydrocarbons and nitrogen oxides decomposition platinum metals were transferred to a colloidal state. It was carried out by impregnation of catalysts by previously prepared solutions of polymers with the added solutions of active metals with the subsequent thermal decomposition. In the work platinum nano-size particles were prepared by reduction with hydrogen in an aqueous solution containing chloroplatinic acid and citric acid. As the stabilizer of colloid platinum particles is used isopropyl alcohol. The obtained colloidal platinum metal by such methods was coated on carrier block. The metal sols supported on the block carriers were dried in an oven for 4 hours at a temperature of 423 K. The catalysts were heated in a muffle furnace at 773 K for 2 hours. Also in the synthesis of catalysts based on platinum and palladium as active components of catalysts acetates of Pt and Pd and their π-complexes are used. Solutions of acetate of palladium were prepared by dissolution of Pd in the acetic acid containing 3% of HNO_3. Nitric acid was removed in the course of evaporation before complete finishing of release of nitrogen oxides. The π-complexes of Pd and Pt were prepared by reacting of allyl alcohol with the salts of these metals with subsequent drying of the catalysts at 423 K and calcination at T = 773 K for 2 h. For increase of thermal stability catalysts were modified with additives of the second metal and oxides of refractory metals. For test of catalysts the flowing catalytic installation with the tubular reactor of integrated type (fig. 4) was used.



1 - A gas bag; 2 - Manometer; 3 - Ventile of thin regulation; 4 - Rotameter; 5 - Crane; 6 - Mixer; 7 - Heating system; 8 - Catalyst; 9-Thermocouple; 10-Selection of tests before and after the catalyst; 11 –IRT

Figure 4 - Scheme of the flowing installation

Installation consists of the cylinders (1) containing researched gases (hydrocarbons, carbon oxide, nitrogen oxide, nitrogen). In system air was moved, gases were moved from cylinders, then through ventile of thin adjustment (3) entered to rotameter (4) individually calibrated under each gas and needed for gas speed regulation intended which then moved to the mixer (6) where gases were mixed up and entered to a quartz reactor (7) with diameter 10 mm. The reactor was heated with by tubular furnace, the temperature in which was measured with IRT (11) by chromele-alumele thermocouple (9).

The gas mixture was prepared by feeding of hydrocarbons from container and the compressed air from the line into the mixer. The hydrocarbon content of the mixture was about 0.5. %. The oxygen concentration was varied from 2 to 10 vol.%. The gas mixture was analyzed by GLC and OPTOGAZ gas

analyzer before and after the reaction. Crystal 2000M and Chrom 3700 chromatographs with the flame ionization detector are used. Time of analysis - 20-30 min. For testing it was used a sample of the 2 cm 3 colloidal catalyst on the metal carrier. Previously the catalyst was calcinated at 773 K within 4 h. on air in the muffle furnace. The activity of the catalysts was determined at temperatures of 423-773 K. When designing the optimal compositions and methods of preparation of colloid metals were varied its dispersion, the active metals content, their relative proportions and a temperature of the preliminary heat treatment.

Characteristic of activity of the catalyst was the degree of conversion (α) of initial reagent (hydrocarbon, carbon monoxide, nitric oxide), defined by the formula:

$$\alpha = C_{init.} - C_{fin.} / C_{init.} \cdot 100\ \%,$$

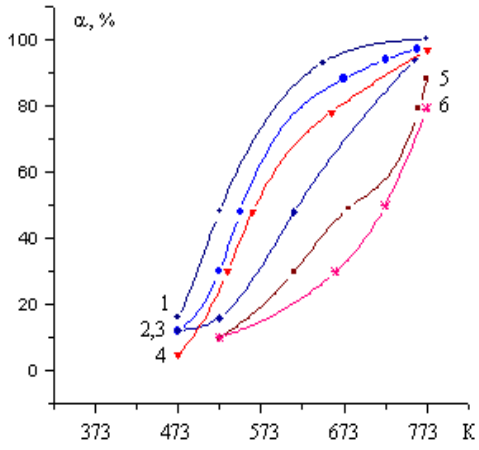
where $C_{init.}$ and $C_{fin.}$ - are the initial and final concentrations of a reagent in volume of a test.

Results and discussions

As an example, the results on studying of influence of concentration of Pt and Pd in π-allelic complexes on activity of block catalysts in reaction of oxidation of CO, C_3H_8 and reduction of NO_x at a volume speed of a stream of gases - 35000 h^{-1} are given in the Table.

Table - Influence of concentration of π- allylic complexes of Pt and Pd on a degree of conversion of CO, C_3H_8,NO_x

Catalyst	The active metal concentration, %	Conversion degree, % at the different temperatures of research of catalysts, K											
		CO				C_3H_8				NO_x			
		773	623	523	473	773	623	523	473	773	623	523	473
Pt π-allyl	0.01	100	100	89	16	95	85	50	0	20	20.7	13.9	0
Pt π-allyl	0.05	100	100	92	50	98	94	73	0	20.7	21.1	14.4	0
Pt π-allyl	0.1	100	100	100	87	100	99	73	17	34.2	36.7	17.1	1.2
Pd π-allyl	0.1	100	100	100	83	53	22	5.0	0	18.4	16.8	8.0	0
Pd π-allyl	0.15	100	100	100	90	59	32	18	0	24	26.6	16	0.5



1-0.1% Pt-initial Pt, 2- 0.1% Pt-50 h. of testing, 3-0.1% Pt- 100 h.,
4- 0.2% Pd-initial Pd, 5- 0.2% Pd-50 h. of testing , 6- 0.2% Pd-100 h. of testing
Figure 5- Dependence of stability of catalysts in reaction of 0.5 % of propane-butane in air oxidation

Data of the Table show that activity of palladium-containing catalyst in reaction of oxidation of CO does not significantly depend on concentration of metal and reaches 90-100% in the studied interval of temperatures. By the way the efficiency of Pd-catalyst in reaction of complete oxidation of C_3H_8 at low

temperatures (623-473 K) was much lower, than that of the Pt-catalyst, and was 78-90 %. The higher activity of the Pt-catalyst in reaction of NO reduction in comparison with Pd the catalyst was observed at contents of Pt equal to 0.1 % and T=773 K.

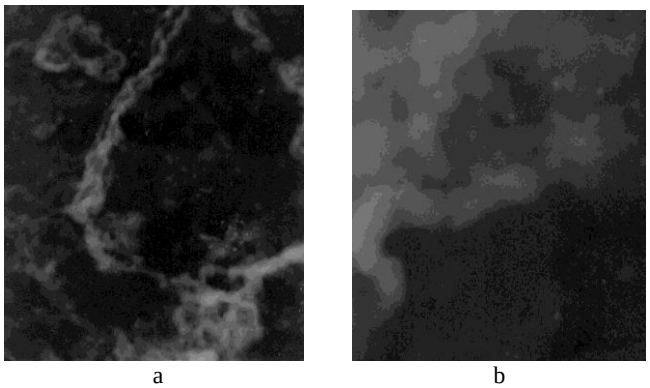
Research of Pt and Pd catalysts on thermo stability was carried out by maintaining of the catalyst with an interval of 5 hours at T=773 K in a reactionary gas mix with the contents of 0.5 % of propane-butane with the subsequent analysis of products of reaction. The total time of researches was 100 h. The most stable appeared the catalysts obtained from acetates Pt, less stable - on basis Pd.

It's known that by supporting of the secondary oxide covering on metal blocks of honey comb structure, as a rule, the carriers with the developed specific surface and porous structure are produced [20, 21], and high thermal stability of the secondary oxide covering is provided with introduction to it of the modifying additives. There are the works about inclusion of cerium, zirconium, lanthanum cations into the secondary alumina coating, which stabilize γ-Al_2O_3 phase and provide resistance to poisons or sintering of Pt, Pd, Rh- active components of the catalysts [22, 23]. In the work Pd-Mo-catalysts on Al_2O_3 carrier modified by additives of Ce^{4+}, Ti^{4+}, Zr^{4+}, La^{3+}, Fe^{3+} are prepared and investigated in reaction of $NO_x+C_3H_6+O_2$. Activity of Pd-Mo of catalysts increases when modifying the carrier with cations of Ce^{4+}, Zr^{4+}. Activity of Pt-Cu catalysts on the zeolite-containing carriers NaY, ZSM-5 and their hydrogen forms is studied. High activity of the catalyst is noticed when supporting of the active phase on the carrier in H^+-form. A number of oxidic catalysts with the differing compositions of the secondary carrier (γ-Al_2O_3,γ-$Al_2O_3+TiO_2$) and of the active agent (Co+Mn, Fe+Mn) promoted and not promoted by platinum is studied. Introduction of titanium dioxide into the secondary carrier considerably increases extent of reduction of nitrogen oxides by means of propylene on both compositions of oxidic catalysts in all interval of the studied temperatures (423-773 K) and was equal to 34% at 523 K on the Co-Mn-catalyst, to 29% -on Fe-Mn, and at 773 K-33% and 55%, respectively. Promotion with platinum improves the reduction ability of cobalt-manganese catalyst on Al_2O_3 only at temperatures higher than 673 K. On the titanium-containing sample in the presence of platinum degree of NO conversion is decreased.

The samples of catalysts based on platinum group metals were investigated by electron microscope EM-125K with single stage replica method. In a sample with Pt the small congestions of dense particles which don't grow together in units and are dispersed on a carrier surface are observed.

The particles sizes are predominantly 10.0 nm, 5.0 nm and less 9.0 nm (Fig.6, a). In the sample with Pd there are some isolated dense particles in size 12.0 nm-15.0 nm (Fig.6, b). The platinum obtained by reduction in solution has the sizes about 15.0-18.0 nm, at the same time, the platinum colloid obtained by reduction with lemon acid had almost monodisperse distribution, the average size of particles was equal to 8.0 nm. It is found that the organometallic complexes of Pd and Pt at magnification of 33,000 times represent translucent areas of the clots of polymer filled with dispersed particles of 3.0 nm. At higher magnification (in the 62,000 times) also small rare congestions of more dense particles of 5.0 nm in size are observed.

Investigation of Pt and Pd-containing catalyst by means of XPA showed X-ray scattering, which confirmed the high dispersion of catalysts obtained by thermal decomposition of organometallic complexes.



a b

Figure 6-EM - pictures of distribution of noble metals : (a) -Pt, (b) -Pd, on the metal carrier

Physical and chemical researches of catalysts on the basis of base metals were carried out by XRD method on x-ray diffractometer DRON-4.0.7 with the copper anode. Samples for research were prepared by mechanical destruction of the catalyst put on a block metal framework. The fallen part of the catalyst was crushed in an agate mortar up to 100 microns and was used for research by method XPA. It was found, that the oxide catalysts represented spinel with cubic lattice $NiMnO_4$ with peaks 2Å, 52Å, 148Å, 203Å. Also there were small intensive peaks of CeO_2 (308Å) and alumina (160Å, 256Å). The conducted research of supports and catalysts by means of XPA showed the formation TiO_2-anatase structure, peaks 3.52; 1.89; 2,38 Å. The carrier based on V_2O_5-WO_3-peaks 4.38, 3,4, 2,8 Å, it has an orthorhombic lattice. In the active phase NiO-crystal lattice is not formed. Catalysts based on base metals have been investigated with an electron microscope EM-on device 125M single by stage replica method. On a nickel-vanadium-tungsten sample the congestions of dense particles which don't grow together in units and are dispersed on a carrier surface are observed. The sizes of particles were mainly 15.0-20.0 nm.

Conclusions

The technology of synthesis of catalysts on block metal carriers for neutralization of waste gases of motor transport and the industry is developed and improved. Catalysts for neutralization of toxic gases of the industry and motor transport on metal carriers with the honey comb structure of channels are prepared. For preparation of the secondary carrier were used either aluminum oxide or aluminum oxide with the addition of a zeolite or Ce^{4+}, Ti^{4+}, Zr^{4+}, La^{3+}, Fe^{3+}. For preparation of solutions of the active components of catalysts are applied oxides of Mn, Ni, Co, Fe, obtained from acetates and formiates. Also were synthesized the samples of catalysts based on metals of the platinum group converted into colloidal state.

The activity of Pd-Mo catalysts carrier is increased by modifying with cations Ce^{4+}, Zr^{4+}. The activity of the palladium catalyst does not depend on the metal concentration in the CO oxidation reaction and is about 90-100%. However, the effectiveness of the Pd-catalyst in the C_3H_8 complete oxidation reaction at low temperatures (623-473 K) is significantly lower than of Pt-catalyst, and is 78-90%. For catalysts based on base metals introducing into the secondary carrier of Ti^{4+} significantly increases the degree of reduction of nitrogen oxides with propylene, at 523 K on a Co-Mn-catalyst - 34%, on Fe-Mn - 29%, at 773 K - 33% and 55%, respectively.

REFERENCES

[1] Awofeso N., *American Journal of Respiratory and Critical Care Medicine*, **2011**, 10, 1437. (In Eng.). http://dx.doi.org/10.1164/ajrccm.183.10.1437

[2] Trimm D.H. Catalytic Combustion (Review), *Appl.Catalysis*, 7, **1983**, 249-282. (In Eng.)

[3] Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990-1999. U.S. *Environmental Protection Agency*, 8 April 15, **2001**, Washington, DC, USA. (In Eng.)

[1] Yang S., He L-Y., *J.Energy & Environment.*, **2016**. (In Eng.)
http://dx.doi.org/10.1177/0958305x15627545.

[5] Lukanin V.N., Trofimenko Y.V. Industrial and transport ecology. M.: Visshay shkola, 2001, 273 p. (In Rus).

[6] Kalygin V.G. Industrial ecology. M.: *MNEPU*, 2000, 239 p. (In Russ).

[7] Medvedev V.T. Engineering ecology. M.: *Gardariki*, 2002, 687 p. (In Russ).

[8] Akhatov A.G. Ecology and International law. M.: *AST-PRESS*, **1996**, 512 (In Russ.).

[9] Vystrobets E.A. International cooperation in the field of environment and natural resources. M.: *Izdatelstvo MNEPU*, **2000**, 112 (In Russ.)

[10] Global change of climate. Kazakhstan: the steps to the Kyoto protocol. Project 70-242 *TASIS*. Astana, 2006. (In Russ.).

[11] Yadava O.P., Palmqvist A., Cruise N. and Holmberg K. Coll.&Surfaces A: Physicochemical and Engineering Aspects, **2003,** 221,1-3, 131-134 (In Eng.).

[12] Gryaznov V., Serov Ju., *12th Int.Congress on Catalysis*, Proceedings. Pt.B., Granada, Elsevier, **2000**, P.1583-1588. (In Eng.)

[13] Lucena P., Vadillo J.M., Joserna J.J., *J.Appl.Spectrosc.*, **2001**, 55, 3, 267-272 (In Eng.)

[14] Sassykova L.R., Massenova A.T. *ISCS 2012, Interrn.Symposium on Catalysis and Specialty Chemicals*, Tlemcen, Algeria,**2012**,121(In Eng.)

[15] Gilmundinov Sh.A., Sassykova L.R., Nalibayeva A.M., *Catalysis:Fundamentals and Application*, Novosibirsk, **2007**, Abstracts, II,532-534 (In Eng.)

[16] Sassykova L.R., Massenova A.T., Gil'mundinov Sh., Tel'baeva M.M., Bunin V.N., Komashko L.V., *15th International Congress on catalysis ICC15*, Germany, Munich, **2012**, PP.03,456 (In Eng.).

[17] Sassykova L.R., Massenova A.T., Sharifkanova G.N. *The Sixth Asia-Pacific Congress on Catalysis (APCAT-6)***,** Taipei, Taiwan , **2013,** P2-177, 255(In Eng.)

[18] Sassykova L.R., Massenova A.T., Gilmundinov Sh.A., Bunin V.N., Rakhmetova K.S., *Selective Oxidation and Functionalization: Classical and Alternative Routes and Sources*, Berlin, Germany. Preprints of the Conference, *DGMK Tagungsbericht*, **2014**, 3, 181-187 (In Eng.)

[19] Nalibayeva A.M., Sassykova L.R., Kotova G.N., Bogdanova I.O., *Izvestiy NAN RK, ser. Chimiy I technologiy*, **2016**, 5, 55-64. (In Russ.)

[20] Wang Qing Min, Shen Dongmin, Bulow Martin, Lau Miu Ling, Deng Shuguang, Fitch Frank R., Lemocoff Norberto O., Semanscin Jessica. *Microporous and Mesoporous Mater.: Zeolites, Clays, Carbons and Related Materials*,**2002**, 55, 2, 217-230 (In Eng.)

[21] Burke N.R., Trimm P.L., Howe R., *Proceedings. Pt.B. 12thInt.Congress on Catalysis*, Granada, **2000**, July 9-14: Elsevier 2000, P.1451-1456 (In Eng.)

[22] Mamede A.S., Leclerg G., Payen E., Grimblot J., Granger P., *Phys.Chem.*, **2003**, 5, 20, 4402-4406 (In Eng.)

[23] McGrath M. http://www.bbc.com/news/science-environment-38170794. (In Eng.)

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АВТОКӨЛІК ПЕН ӨНДІРІСТЕН ШЫҒАРЫЛАТЫН ГАЗДАРДЫ ТИІМДІ БЕЙТАРАПТАНДЫРУҒА АРНАЛҒАН КАТАЛИЗАТОРЛАРДЫ СИНТЕЗДЕУ ТЕХНОЛОГИЯСЫ

Аннотация. Мақалада автокөлік пен өндірістен шығарылатын газдарды залалсыздандыруға арналған блокты металл тасымалдаушылардағы катализаторларды, авторларымен әзірленген және жетілдірілген синтездеу әдістемесінің сипаттамасы. Екінші ретті тасымалдаушыларды дайындау үшін алюминий оксиді мен цеолит немесе Ce^{4+}, Ti^{4+}, Zr^{4+}, La^{3+}, Fe^{3+} қоспасы бар алюминий оксиді қолданылды. Активті фаза ретінде Mn, Ni, Co, Fe қосылыстары және платина группасының металдары қолданылады. Катализаторлар NO_x, CO, C_3H_8 қосылыстарының толық тотығу процесі және $NO_x+C_3H_6+O_2$ реакциясы барысында сынақталды. Pd-Mo катализаторларының активтілігі құрамына Ce^{4+}, Zr^{4+} катиондарын енгізген кезінде жоғарылай бастайды. Палладия негізіндегі катализаторлардың активтілігі CO-нің тотығу реакциясы кезінде 90-100%-ды құрайды және металл концентрациясына тәуелсіз, температурасы 473-623 К-дегі C_3H_8толық тотығу реакциясы кезінде - 78-90%, құрамында Pt бар катализаторлардікі - 95-100%. Асыл емес металдар негізіндегң катализаторлардың екінші ретті тасымалдаушы құрамына Ti^{4+}енгізу азот оксидінің пропиленмен тотықсыздандыру дәрежесін айтарлықтай арттырады.

Түйін сөздері: катализатор, экология, шығарылатын газдар.

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ТЕХНОЛОГИЯ СИНТЕЗА КАТАЛИЗАТОРОВ ДЛЯ ЭФФЕКТИВНОЙ НЕЙТРАЛИЗАЦИИ ОТХОДЯЩИХ ГАЗОВ ТРАНСПОРТА И ПРОМЫШЛЕННОСТИ

Аннотация. В статье описывается разработанная и усовершенствованная авторами методика синтеза катализаторов на блочных металлических носителях для обезвреживания отходящих газов транспорта и промышленности. Для приготовления вторичного носителя использовали оксид алюминия или оксид алюминия с добавлением цеолита или Ce^{4+}, Ti^{4+}, Zr^{4+}, La^{3+}, Fe^{3+}. В качестве активной фазы использовались соединения Mn, Ni, Co, Fe и металлов платиновой группы. Катализаторы испытывали в процессе полного окисления NO_x, CO, C_3H_8 и реакции $NO_x+C_3H_6+O_2$. Активность Pd-Mo катализаторов увеличивается при добавлении в состав носителя катионов Ce^{4+}, Zr^{4+}. Активность катализатора на основе палладия в реакциях окисления CO составляет 90-100% и не зависит от концентрации металла, в полном окислении C_3H_8 при 473-623 К - 78-90%, для Pt-содержащего катализатора - 95-100%. Для катализаторов на основе неблагородных металлов введение Ti^{4+} во вторичный носитель значительно увеличивает степень восстановления оксидов азота пропиленом.

Ключевые слова: катализатор, экология, отходящие газы

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