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Catalytic oxidation of CO in gases emitted by industrial processes and vehicle exhaust

A new type of catalysts not containing noble metal oxides have been developed and the possibilities of their application both for the complete neutralization of carbon monoxide in exhaust gases and the process of deep oxidation of volatile hydrocarbons are studied. It has been found that the activity of catalysts based on vanadium and phosphorus oxides supported on Al_2O_3, SiO_2, TiO_2 by and their modification with of 1–3 % oxides of Cu, Cr, Co, Zn enhanced the conversion of the deep oxidation process to 95–100 % at the temperatures of 673–693 K and volumetric velocities of 5000–10000 h^{-1}. During the simultaneous oxidation of CO and C_3H_8 at a CO conversion of 90 %, the C_3H_8 conversion was 70 %. It has been established that oxidation of CO and C1–C4 hydrocarbons, and especially propane, with the participation of synthesized catalytic series, occurs by stepwise and associative mechanisms. The oxidation of CO and C_3H_8 required a high oxygen content of 1:20–25 mol. Besides utilizing carbon monoxide in exhaust gases from motor vehicles, these catalytic systems can be successfully used to neutralize industrial gases, especially those emitted from oil refineries and thermal power plants. Preliminary research has shown that these catalytic systems can operate for about 50 000 hours without changing the activity.

Keywords: catalyst, organic compounds, catalytic systems, monoxide, propane, exhaust gases, aluminium oxide, cobalt, chrome, vanadium oxide.

Introduction

It is known that since the second half of the last century the exponential growth of industry, especially oil refining, petrochemicals, metallurgy, energy complexes, organic compounds, automotive industry, spontaneously led to rapid pollution of the environment. It should be noted that most of the toxic gases emitted into the environment fall on the share of metallurgy, thermal power plants (~50 %), and more recently in the field of rapid development of road transport (~20 %) in the world [1–6]. Automobiles are the most popular mode of transport used by people. During the operation of these vehicles, air pollution emitted from transportation contributes to emissions of air toxics and poor air quality, which has negative impacts on the health and welfare of mankind [7–9].

The amount of exhaust gases emitted into the environment from industrial plants and vehicles continues to increase from year to year. In particular, 0.3–12 % carbon monoxide, 0.5–1.0 % NO_x gases, 0.2–3 % C_xH_y, 0.1–3 % organic compounds, 3–10 % water vapour, l–15 % soot, and carbon dioxide from internal combustion engines, in addition to SO_2 from the vehicle engines that run on the diesel fuel is also released into the environment. Although many "euro" standards (EURO 4 to EURO 6) are set for the norms and amount of the emitted exhaust gases, they are often not followed, which is associated with many problems [10–14].

Recent research show that catalytic systems based on the most valuable non-ferrous and rare metals are currently used for the conversion and disposal of carbon monoxide, nitrogen gases, and volatile hydrocarbons. Depending on the neutralization process, catalytic methods are the most preferred ones in industrial waste disposal. [15].

Experimental

Initially, a vanadium-phosphorus catalyst was synthesized. Catalyst samples were obtained by known methods, precipitated, and impregnated on aluminium oxide and silicon oxide [16]. The catalysts were then heat-treated by known methods [17]. The catalyst is then cooled, crushed, sieved, and divided into the required 0.2–0.8 mm (or 0.2–0.4; 0.4–0.6; 0.6–0.8 mm) parts. Such catalyst samples are called "newly synthesized" samples. In this case, depending on the initial materials, catalyst samples are taken in ratios such as V:P = 1:10.

It should be noted that the main active mass precipitated or impregnated on the carriers is 8–10 % of the total catalytic system.

The activity and kinetic regularities of the synthesized catalysts in the conversion, oxidation reactions of CO, C_1–C_4 hydrocarbons, and NO_x gases were studied in open-flow and pulse system reactors.

Gas mixtures CO+He, O_2+He, CO_2+He, CO+O_2+He, NO+He, NO_2+He, CH_4+He, C_2H_4+He, C_3H_8+He, C_4H_{10}+He were prepared in certain volume concentrations (volume %) and filled into air balloons. Some of them were prepared by known methods and given to the system directly before the reactions. Gas mixtures are taken at CO (0.5–5.0 %), CO_2 (2–15 %), O_2 (1–20 %), C_xH_y(0.5–2.0 %), H_2O (l–20 %), NO (200–600 ppm), NO_2 (200–1000 ppm), which is close to the emissions of real automobiles, thermal power plants, oil refining, and petrochemical industries. Moreover, the gas mixtures were used separately as a model for kinetic studies. Analysis of CO, CO_2, NO_x, and hydrocarbons was performed on AMX (calorimeter) and SVET-500 chromatographs. The gas carrier was He (helium); separation of the CO, CO_2, and hydrocarbons was carried out in a column filled with “Poropak Q” (length 3.0 m) at a programmed temperature (3°C/min) 30–90 °C. It should be noted that carbon monoxide is obtained by the treatment of concentrated sulphuric acid with formic acid. O_2, N_2, and CO were analyzed in a parallel column using NaX zeolite (1.2 m length), where a rate of helium was 50 ml/min, and N_2O and NO_2 on activated carbon using CKT. Other organic compounds, (acetone, benzene, toluene, chlorobenzene, and others) were taken and analyzed using a carrier adsorbent containing SE-30 on a chromoton in a 3 m column.

Adsorption isotherms, pore distributions, surface area, and pore volumes information has been studied by using the BET method for synthesized and activated (I); promoters added (Cr_2O_3 or Co_2O_3) (II); deactivated samples (III). The results showed that the values of samples I and II do not differ significantly during both adsorption and desorption, but the distribution of the pore volume and surface area of sample III differs from other ones. It should be noted that for samples with added Co and Cr oxides the P/P_o-dependent distribution of the adsorption and desorption isotherms and the P/P_o-dependent distribution occur according to known regularities.

In the laboratory the experiments were carried out with a vertically aligned tubular quartz reactor operating at atmospheric pressure. The schematic of the experiment set up is shown in Figure 1.

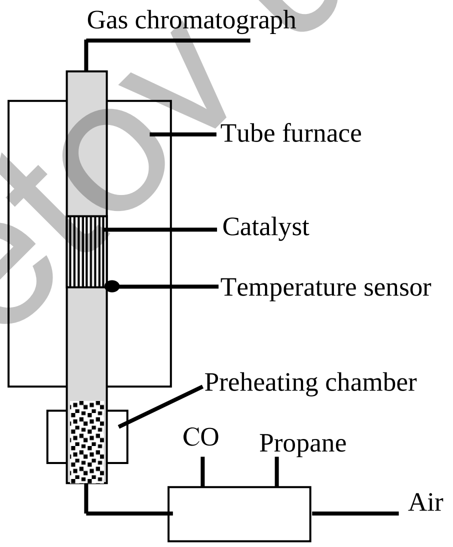


Figure 1. The experimental vertically-aligned reactor

Due to the corrosive reaction products, all materials used in the experimental setup were corrosion-resistant: quartz glass, heated Teflon pipes (T = 180 °C), and Teflon connectors.

Results and Discussion

The activity of the synthesized catalytic systems was initially tested at a temperature of 673–723 K, at a volume rate of 8000–10000 h^{-1}. In all cases, the effect of different promoters on their activity was studied by taking the catalytic systems based on V–P–O/Al_2O_3 and V–P–O/SiO_2 (Table 1).

Table 1

The activity of the catalytic systems synthesized for the oxidation reactions of carbon monoxide and propane depended on the nature of the carriers and added promoters

Catalysts	Promotor		T, K	V, h^{-1}	Conversion, %	
	P_1	P_2			CO	C_3H_8
V–P–O/SiO_2	–	–	573	8000	40	50
	–	Zn	573	8000	45	54
	Mg	–	673	8000	45	56
	Co	–	673	8000	50	58
	–	Cu	673	8000	68	62
	Cr	–	673	8000	68	61
	Ni	–	673	8000	52	48
	–	Zn	723	10000	50	55
	Co	–	723	10000	54	62
	–	Cu	723	10000	70	65
	Cr	–	723	10000	74	66
V–P–O/Al_2O_3	–	–	673	8000	42	52
	Mg	–	723	10000	46	60
	–	Zn	673	8000	48	61
	Ni	–	673	8000	80	54
	–	Cu	723	10000	55	60
	Cr	–	673	8000	70	65
	Cr	–	673	8000	70	66
	–	Cu	723	10000	76	70
			723	10000	72	68
V–P–O/SiO_2	Mg	Zn	673	8000	47	54
	Mg	Cu	673	8000	70	64
	Mg	Co	673	8000	52	56
	Mg	Co	723	10000	60	65
	Mg	Cr	673	8000	72	65
	Zn	Cu	673	8000	75	78
	Zn	Co	723	10000	60	60
	Zn	Cr	723	10000	62	60
	Ni	Co	723	10000	73	65
V–P–O/SiO_2	Cu	Co	673	8000	80	73
	Co	Cu	723	10000	82	73
	Cu	Cr	673	8000	92	90
	Cu	Cr	723	10000	96	96
	Co	Cr	673	8000	84	90
	Co	Mn	723	10000	78	70
V–P–O/Al_2O_3	Cu	Cr	673	8000	96	97
	Cu	Cr	723	10000	97	95
	Cu	Co	673	8000	88	83
	Co	Cr	673	8000	90	84
	Cr	Co	723	10000	92	84
	Mg	Cu	673	8000	70	68
	Mg	Co	723	10000	64	70

As can be seen from the results, one or two types of promoters were added to V–P–O/Al_2O_3 and V–P–O/SiO_2, and their activity in the process of CO and propane conversion was studied. The amount of added promoters is 1–3 %. Initially, MgO was added to V–P–O/Al_2O_3 and V–P–O/SiO_2 catalyst after which modified by Zn, Mn, Cu, Co, Cr, Ni oxides, and all catalysts showed 50–70 % catalytic activity. No significant change was observed when MgO was replaced by the NiO, although subsequent additions slightly increased the conversion of CO and C_3H_8 (70–76%).

As can be seen from the results in the table, catalytic systems obtained with the addition of 1–3 % CuO and then 1–3 % Co_2O_3 or Cr_2O_3 showed high (90–95 %) activity in both carbon monoxide conversion and propane conversion.

First we studied the effect of the ratio of CO:O_2 on the CO oxidation reaction.

Table 2

CO conversion on $CO:O_2$ ratios, T = 713 K, V = 10000 h^{-1}

Catalysts	CO conversion on $CO:O_2$ ratios				
	0.5:1	1:1	1:5	1:10	1:20
V–P–O/SiO_2	3	13	18	27	46
V–P–O/Al_2O_3	5	15	20	34	48
V–P–O/Al_2O_3 + 2 % Mg	6	15	31	53	61
V–P–O/Al_2O_3 + 2 % Cu	8	20	41	81	87
V–P–O/Al_2O_3 + 1 % Cu	7	18	37	73	86
V–P–O/Al_2O_3 + 2 % Cu + 2 % Cr	10	50	80	98	100
V–P–O/Al_2O_3 +2 % Cu + 1 % Cr	8	46	64	83	93

Some of the catalytic systems that were synthesized and showed high activity were selected, and the oxidation reaction on their surface in the range of $CO:O_2$ = 0.5–l:20 ratios was studied (Table 2). The results show that the conversion of CO is very low in the presence of V–P–O/SiO_2 and V–P–O/Al_2O_3 catalysts at $CO:O_2$ = 0.5:l and 1:1.

When a small amount of Mg, Cu is added into the catalyst, the conversion of CO does not increase much. However, CO conversion begins to increase significantly (20–50 %) in the presence of catalyst samples with the addition of Co and Cr. The catalytic oxidation process increases rapidly at a ratio of $CO:O_2$ = 1:5. In special cases, it reaches 60–80 %. At higher values of this ratio (1:10 and 1:20), the conversion of CO in the presence of all catalyst samples exceeds almost 50 %, and on the V–P–O/Al_2O_3 + 2 % Cu + 1 % Cr catalyst it reaches 90–92 %, and 100 % in the presence of V–P–O/Al_2O_3 + 2 % Cu + 2 % Cr catalyst. The effect of synthesis and preparation methods of catalytic systems on the oxidation of carbon monoxide and propane has also been studied (Table 3).

Table 3

The effect of catalysts preparation method on oxidation of CO and propane [1]

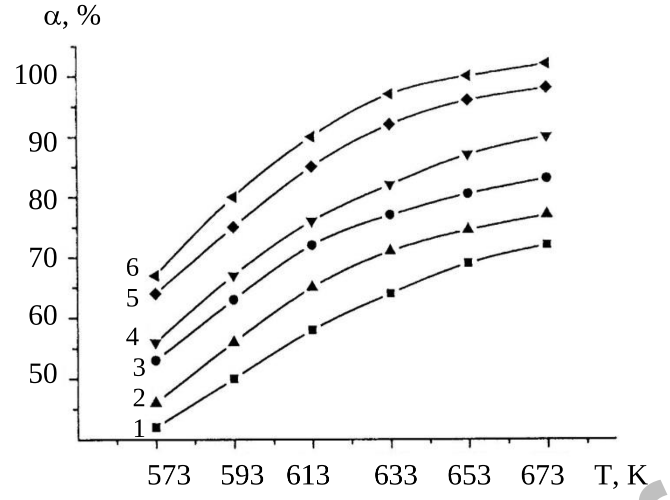
Synthesis method	Temperature, K	Volume rate, V, h^{-1}	CO conversion, α, %	C_3H_8 conversion, α, %
Co-precipitation	673	8000	100	70
Impregnation	693	8000	95	70
Shifting	673	8000	96	75
Mechanical-chemical	653	10000	95	75
CuO, Al_2O_3, ZnO industrial	573	7000	75	50
CuO, Cr_2O_3 HTK	473	7000	70	50
Perovskite	615	5600	95	100 %, 923 K
Pt (USA)	523	560	95	100 %, 953 K
V/P	$VOPO_4$		V/V = 1/45	

In the presence of catalysts obtained by precipitation and impregnation methods, the conversion of carbon monoxide was100 % and the conversion of propane was 70 % at a temperature of about 673–693 K and a volume rate of 8000–10000 h^{-1}[17].

Although the conversion of propane varies slightly under approximately the same conditions in the impregnated and mixed samples of catalytic systems, the conversion of carbon monoxide decreases slightly, to 95–96 %. These results can be applied to catalyst samples obtained by mechanochemical methods.

It is known, that the abovementioned methods are easier ways to prepare catalysts. While comparing the activity of the prepared catalysts with the catalyst samples (CuO, ZnO, CaO, Cr_2O_3) used in industry Pt (USA) in motor vehicles, it is observed that the results are nearly the same as (Pt (USA)) or catalysts are during the conversion of carbon monoxide (Table 3). However, the industrial catalyst can provide 100 % conversion of propane at very high temperatures at 933 K. Lowering the temperature by 200–400 K in such processes means saving a lot of energy.

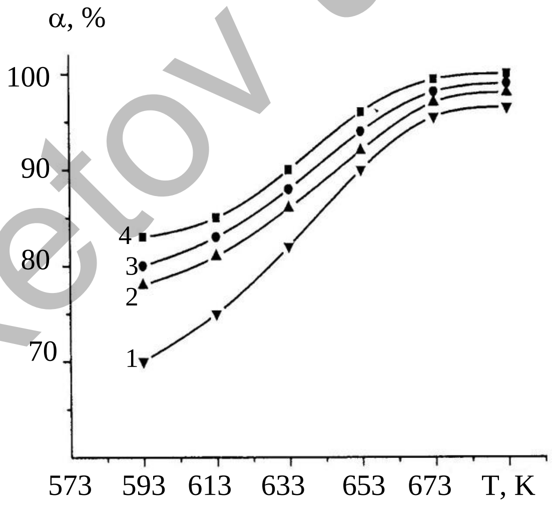
Furthermore, considering the presence of many organic compounds in exhaust gases emitted into the environment from industrial plants and vehicles, including isopropyl alcohol, acetone, benzene, mono- and dichlorobenzene, toluene, the study of their oxidation processes, along with other hydrocarbons, is a very important problem. The best results were also shown here by the V–P–O/Al_2O_3 + CuO + Cr_2O_3 catalytic systems (Fig. 2).



1 — V–P–O/SiO_2 + Mg; *2* — V–P–O/Al_2O_3 + CuO; *3* — V–P–O/Al_2O_3 + ZnO;
4 — V–P–O/Al_2O_3 + Cr_2O_3; *5* —V–P–O/Al_2O_3 + 1 % Cu + 1 % Cr; *6* — V–P–O/Al_2O_3 + 2 % Cu + 2 % Cr

Figure 2. The activity of catalytic systems in an oxidation process of different organic compounds (acetone, isopropyl alcohol, benzenes). CO:O_2:H_2O:CO_2:C_xH_y = 1:20:5:10:6

In addition to the hydrocarbons listed on these selected active catalytic systems, we have studied the conversion of other organic compounds. It is also clear that the 100 % conversion of CO at temperatures of 633–673 K and the conversion of organic compounds within the 95–96 % conversion of C_3H_8–C_4H_{10} are also high (97–98 %) (Fig. 3).



1 — CO; *2* — C_3H_8; *3* — C_4H_{10}; *4* — organics (acetone, isopropyl alcohol, benzenes, and toluene)

Figure 3. Temperature effect on the oxidation of hydrocarbon, CO, and organic compounds in the presence of V–P–O/Al_2O_3 + 2 % Cu + 2 % Cr

In the ratios of CO:O_2 = 1:1 and 1:2, the catalysis proceeds at a very slow rate and is practically stabilized after two pulses. Although the oxidation of CO doubles at a ratio of CO:O = 1:5, a very high rate is still not observed. However, the nature of the kinetic curve does not change at a ratio of CO:O_2 = l:10 and more 1:20, catalysis is observed to proceed at high speed.

Conclusions

Active catalytic systems and optimal conditions for the conversion of 95–98 % CO, 80–85 % C_xH_y have been identified, when the amounts of carbon monoxide, hydrocarbons, water vapor, carbon dioxide were in close proportions to real gas mixtures ($CO:O_2:C_xH_4:CO_2:H_2O$–1:15:6:12:6) in the oxidation. It has been shown that the catalytic systems we obtained and used for the oxidation were more active at temperatures 200–300K less, i.e., at 500–700 K, compared to catalysts containing precious metals such as Pt, Pd, Au.

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Өнеркәсіптік процестерден және автокөлік құралдарынан шығарылатын газдардағы CO-ның каталитикалық тотығуы

Құрамында асыл металл оксидтері жоқ, түрлі қоспалары бар оксидтер негізінде жаңа типті каталитикалық жүйелер әзірленді және оларды пайдаланылған газдардағы көміртегі тотығын толық бейтараптандыру және басқа ұшпа көмірсутектермен терең тотығу процесі үшін қолдану мүмкіндіктері көрсетілді. Al_2O_3, SiO_2, TiO_2–ге жағылған ванадий және фосфор оксидтері негізіндегі катализаторлардың белсенділігі олардың 1–3 % Cu, Cr, Co, Zn оксидтерімен модификациясы нәтижесінде 673–693 К температурада және 5000–10000 сағ$^{-1}$ көлемдік жылдамдықта терең тотығу процесін 95–100 %-ға дейін жеткізу кезінде жоғары нәтиже көрсететіні анықталды. Бір мезгілде CO және C_3H_8 тотығуы CO – 90 % конверсиясы кезінде C_3H_8 конверсиясы 70 % құрады. Сондай-ақ, судың белгілі бір концентрацияға дейін тотығу процесіне оң әсер ететіні анықталды, бірақ осы концентрациядан жоғары болғанда оның әсері, керісінше тежеуші фактор болып саналады. Автокөлік құралдарынан шығатын газдардағы көміртегі тотығын кәдеге жаратудан басқа, бұл каталитикалық жүйелер өнеркәсіптік газдарды, әсіресе мұнай өңдеу кәсіпорындары мен жылу электр станцияларынан шығарылатын газдарды залалсыздандыру үшін сәтті қолданылуы мүмкін екендігі көрсетілген. Алдын- ала зерттеу көрсеткендей, бұл каталитикалық жүйелер ұзақ уақыт аралығында, атап айтқанда, шамамен 50 000 сағат белсенділікті өзгертпестен жұмыс істей алады.

Кілт сөздер: катализатор, органикалық қосылыстар, каталитикалық жүйелер, монооксид, пропан, пайдаланылған газдар, алюминий тотығы, кобальт, хром, ванадий.

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Каталитическое окисление CO в газах, выделяемых промышленностью и автотранспортом

Разработаны каталитические системы нового типа на основе оксидов с различными добавками, не содержащие оксидов благородных металлов, и показаны возможности их применения для полной нейтрализации монооксида углерода в выхлопных газах и процесса глубокого окисления с другими летучими углеводородами. Установлено, что активность катализаторов на основе оксидов ванадия и фосфора, нанесенных на Al_2O_3, SiO_2, TiO_2, в результате их модификации 1–3 % оксидами Cu, Cr, Co, Zn показывает высокие результаты при доведении конверсии процесса глубокого окисления до 95–100 % при температуре 673–693 К и объемных скоростях 5000–10000 ч$^{-1}$. Одновременное окисление CO и C_3H_8 при конверсии CO — 90 %, конверсия C_3H_8 равна 70 %. Также установлено, что вода до ее определенной концентрации положительно влияет на процесс окисления, но выше этой концентрации оказывает тормозящее действие. Показано, что, кроме утилизации монооксида углерода в выхлопных газах от автотранспортных средств, эти каталитические системы могут успешно применяться для обезвреживания промышленных газов, особенно выбрасываемых нефтеперерабатывающими предприятиями и теплоэлектростанциями. Предварительное исследование показало, что данные каталитические системы могут работать длительное время, а именно около 50 000 ч, без изменения активности.

Ключевые слова: катализатор, органические соединения, каталитические системы, монооксид, пропан, выхлопные газы, окись алюминия, кобальта, хрома и ванадия.

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