Structure and activity of CuO catalysts promoted with CeO$_2$ and La$_2$O$_3$ for complete oxidation of VOCs

Aurimas Urbutis*,

Saulius Kitrys

*Department of Physical Chemistry,
Kaunas University of Technology,
Radvilėnų Rd. 19,
LT-50254 Kaunas,
Lithuania*

Catalytic activity of CuO/γ-Al$_2$O$_3$, xCuO-yCeO$_2$/γ-Al$_2$O$_3$ and CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ catalysts was investigated for complete oxidation of volatile organic compounds (VOCs) in this work. The aforementioned catalysts were characterized by X-ray diffraction (XRD), atomic absorption spectroscopy (AAS), flame atomic emission spectroscopy (FAES) and Brunauer Emmett Teller (BET) methods. Catalysts were prepared by γ-Al$_2$O$_3$ co-impregnation with metal nitrate solutions. During the experimental runs methanol and methyl acetate were used as common pollutants in air stream. Catalytic activity of catalysts promoted with CeO$_2$ and La$_2$O$_3$ was compared with basic CuO/γ-Al$_2$O$_3$ catalyst containing 13.51 wt.% of CuO. The obtained results indicate that the addition of CeO$_2$ increased catalytic activity for oxidation of investigated VOCs. However, CuO/γ-Al$_2$O$_3$ catalyst modified with La$_2$O$_3$ did not expose a significant increase in activity for oxidation of methanol and methyl acetate. Investigation of different compositions of CuO and CeO$_2$ on γ-Al$_2$O$_3$ support indicated that the optimal ratio of CeO$_2$ and CuO was 0.2–0.3. At this composition CuO-CeO$_2$/γ-Al$_2$O$_3$ was the most active among tested catalysts.

Key words: methanol, methyl acetate, volatile organic compound, catalytic oxidation

INTRODUCTION

Emission of volatile organic compounds (VOCs) from industrial pollution sources is a major concern worldwide. Strict environmental requirements demand low emissions of these pollutants. Abatement of VOCs in the effluent gas can be achieved by several methods of decontamination: adsorption, biofiltration, thermal combustion, photocatalytic and heterogeneous catalytic oxidation [1–5]. Low temperature catalytic oxidation is the most promising, convenient and effective technique. As compared to thermal combustion, catalytic oxidation has the advantage because there are no emissions of NO$_x$ (unless heating burner is used) and it is more energy efficient.

The most effective industrially used catalysts for VOC decontamination contain noble metals [6–8]. However, the aim of many researches is to substitute high cost noble met-als with transition metals of different compositions. Catalytic oxidation of various VOCs was investigated in a series of works performed in the KUT Department of Physical Chemistry. Jaskūnas, Dabrilaitė, Čičinskas, Brazlauskas have studied the catalytic oxidation process of VOCs using catalysts that contained transition metal oxides (CuO, Co$_3$O$_4$, MnO$_2$, Cr$_2$O$_3$, etc.) supported on γ-Al$_2$O$_3$, activated carbon and different types of zeolites [9–11]. It was found that CuO expressed the highest activity in the catalytic oxidation of VOCs. However, combination of CuO and M$_x$O$_y$ (M – transition metal) did not give any positive or significant results.

Incorporation of lanthanides for various catalytic processes has a great interest in the past few decades [12–15]. Trovarelli has reviewed catalytic properties of ceria and CeO$_2$ containing materials [16]. The redox properties and the high lability of the lattice oxygen are one of the most important factors determining the suitability of CeO$_2$ for use in complete oxidation of VOCs. Chuan et al. used copper(II) oxide and the ceria based binary catalytic system for the abatement

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* Corresponding author. E-mail: aurimas.urbutis@ktu.lt

of CO, NO and C_3H_6 [17]. The obtained results indicated that CuO dispersed on CeO_2/γ-Al_2O_3 increased thermal aging resistance. Though La_2O_3 is not a material considered to be an oxygen donor [18], Radwan claims that doping catalysts with La_2O_3 result in an effective decrease in the degree of crystallinity of other metal oxides, decrease particle size and increase a specific surface area [19].

The aim of this research was to study how the binary system containing CuO and CeO_2 or La_2O_3 affects the catalytic oxidation of VOCs, which is important for development of adsorbents-catalysts. An adsorbent-catalyst is a material representing adsorptive and catalytic properties in a heterogeneous system [10]. This type of system can be widely used for the catalytic oxidation of VOCs. The main idea and advantage of this system is that it can operate in a single apparatus. The process consists of two steps: the first step is the adsorption and concentration of VOC from the flow, the second step is the thermal regeneration of an adsorbent-catalyst and the catalytic oxidation of VOC. This work consists of two parts: activity comparison of CuO/γ-Al_2O_3 and copper(II) oxide catalysts promoted with CeO_2 and La_2O_3 at stationary conditions; activity tests of xCuO-yCeO$_2$/γ-Al_2O_3 catalysts containing different loads of CuO and CeO_2.

MATERIALS AND METHODS

Catalyst preparation

The supported metal oxide catalysts were synthesized by a conventional co-impregnation method, which is a general technique for the production of heterogeneous catalysts. As a precursors, $Cu(NO_3) \cdot 3H_2O$ (Reachim, Russia), $La(NO_3)_3 \cdot 6H_2O$ (Sigma Aldrich) and $Ce(NO_3)_3 \cdot 6H_2O$ (Sigma Aldrich) were used. Industrially produced γ-Al_2O_3 pellets (4 × 4–6 mm) were impregnated with a nitrate solution of Cu^{2+} or with a mixture of Cu^{2+}-Ce^{3+} and Cu^{2+}-La^{3+} solutions. Cu^{2+} and La^{3+} concentrations were constant in the solutions and consisted of 50 g/dm³ and 10 g/dm³, respectively. Ce^{3+} concentration varied from 5 to 20 g/dm³. Impregnated γ-Al_2O_3 was dried at 120 °C for 2 hours and calcined to drive off the volatile components in an air atmosphere at 450 °C for 6 hours, depositing CuO, CeO_2 and La_2O_3 on the surface of the support. Due to higher decomposition temperature of $La(NO_3)_3$, CuO-La_2O_3/γ-Al_2O_3 catalyst was prepared by calcining impregnated and dried γ-Al_2O_3 at 600 °C. A desired amount of active components was achieved only after three cycles of impregnation, drying and calcination. Catalysts with addition of ceria are denoted as xCuO-yCeO$_2$/γ-Al_2O_3, where x and y are an approximate content (wt.%) of CuO and CeO_2, respectively.

Catalyst characterization methods

Analysis of copper, cerium and lanthanum content in the catalysts was carried out with an AAnalyst 400 (Perkin Elmer, USA) by using atomic absorption (AAS) and flame atomic emission (FAES) spectroscopy methods. Content of copper and lanthanum was determined by dissolving crushed catalyst samples in 20% HCl solution. Catalysts containing ceria were dissolved in HNO_3 (65%) and H_2O_2 (35%) solution (HNO_3/H_2O_2 = 1 : 1). Pure metal oxides were found to completely dissolve in the mentioned solutions.

X-ray diffraction (XRD) analysis was performed with an X-ray diffractometer DRON-6 (Bourevestnik Inc., Russia) using Cu K_α radiation (λ = 0.154056 nm). The detector position step was 0.02°. The crystallite size D_{hkl} of copper(II) oxide was calculated from the line broadening using the Scherrer's equation ($D_{hkl} = (K \cdot \lambda)/(B_{hkl} \cdot \cos\theta)$), where λ is the wavelength of the Cu K_α radiation, θ is the Bragg diffraction angle, B_{hkl} is the full width at the half maximum intensity of the characteristic reflection peak (2θ = 35.5° for CuO) in radians and K is the Scherrer's constant (the value used in this study was 0.94) [20].

A specific surface area (S_{BET}) was determined by N_2 adsorption at –196 °C with a Sorptometer Kelvin 1042 (COSTECH instruments, Italy). Catalyst samples were outgassed for 3 h at 300 °C prior to N_2 adsorption.

Simultaneous thermal analysis (STA) was performed to identify $Ce(NO_3)_3$ and $La(NO_3)_3$ thermal decomposition temperatures and properly estimate calcination temperature (NETZSCH STA 409 PC Luxx, Germany). Differential scanning calorimetry (DSC) and thermogravimetry (TG) curves were recorded in the temperature range of 30–700 °C. Analysis was performed in an inert nitrogen atmosphere. Heating rate was 15 °C/min.

Catalytic activity tests

As common substances found in industrially vented humid air streams, methanol and methyl acetate were used for the oxidation activity tests. Catalytic oxidation reactions of methanol and methyl acetate were performed in a fixed-bed reaction system. This system can be divided into three main parts: an effluent gas simulation block, fixed-bed quartz reactor, and the inlet and outlet gas analyzing system. Vapours of VOCs were prepared in a vapour generator. Flow rates of dilution air were controlled with two parallel flow meters. A custom-made fixed-bed quartz reactor was loaded with 40 g of catalyst. Catalytic oxidation activity tests were performed at a gas hourly space velocity (GHSV) of 3 100 h⁻¹. Concentrations of methanol and methyl acetate were determined by Clarus 500 GC/MS (Perkin-Elmer, USA). COL-ELITE 5MS capillary column was used (length 30 m, internal diameter 0.25 mm). Temperature profile of GC column: isothermal for 5 minutes at 40 °C, then heated to 200 °C (heating rate 10 °C/min) and held at 200 °C for 9 minutes. Ionization by electron impact (EI) of 70 eV was used to meet NIST (National Institute of Standards and Technology) MS database requirements. Reactor inlet concentrations of methanol and methyl acetate were 800 mg/m³ and 3 000 mg/m³, respectively. All catalysts were pre-treated in an air stream (flow rate = 3 dm³/min) at 380 °C for 1 h before the catalytic activity tests were carried out.

Carbon monoxide concentration was monitored with an online gas analyzer TESTO 445 (Testo AG, Germany). Yield of by-products Y$_{by\text{-}p}$ is expressed as a ratio of by-product concentration (mg/m^3) at the outlet of the reactor and initial VOC (methanol or methyl acetate) concentration (mg/m^3) at the inlet of the reactor.

RESULTS AND DISCUSSION

Catalyst characterization

Decomposition temperature of Cu(NO$_3$)$_2$ was determined in previous works [11]. Catalysts containing CuO were produced by calcination of impregnated supports at 450 °C. Simultaneous thermal analysis (STA) (Fig. 1) was carried out to determine decomposition temperatures of Ce(NO$_3$)$_3$ and La(NO$_3$)$_3$. The first three peaks in DSC curves of both compounds were attributed to the loss of adsorbed, physical and crystallization water. Decomposition of Ce(NO$_3$)$_3$ starts at 258 °C. La(NO$_3$)$_3$ decomposes in three steps which start at 395, 464 and 513 °C, respectively. Intermediate products of thermal decomposition are not discussed whereas the desired decomposition product is La$_2$O$_3$. Since the γ-Al$_2$O$_3$ support was simultaneously impregnated with Cu^{2+} and Ce^{3+}, calcination was conducted at 450 °C. CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ was obtained at 600 °C.

Though the γ-Al$_2$O$_3$ support was impregnated with Ce(NO$_3$)$_3$, after calcination at 450 °C CeO$_2$ was formed. Thermal decomposition of Ce(NO$_3$)$_3$ is considered to be a redox reaction and can be written as follows [21]:

$$Ce(NO_3)_{3(s)} \rightarrow CeO_{2(s)} + \text{oxides of nitrogen}. \qquad (1)$$

According to the results of XRD analysis (Fig. 2), characteristic peaks confirm the formation of active components (CuO, CeO$_2$ and La$_2$O$_3$) in the supported catalysts. As the aim was to obtain catalysts with different CeO$_2$/CuO ratio, three solutions of different Ce^{3+} concentration (5, 10 and 20 g/dm^3) and constant Cu^{2+} concentration of 50 g/dm^3 were used to impregnate γ-Al$_2$O$_3$.

In this manner 11CuO-1CeO$_2$/γ-Al$_2$O$_3$, 9CuO-2CeO$_2$/γ-Al$_2$O$_3$ and 9CuO-4CeO$_2$/γ-Al$_2$O$_3$ catalysts with CeO$_2$/CuO ratio of 0.10, 0.21 and 0.42 were synthesized, respectively. Relative intensities of CuO (2θ = 35.5°) and CeO$_2$ (2θ = 27.6°) characteristic peaks in the XRD patterns of xCuO-yCeO$_2$/γ-Al$_2$O$_3$ catalysts correlate with the quantities of active components obtained by AAS and FAES methods (Table).
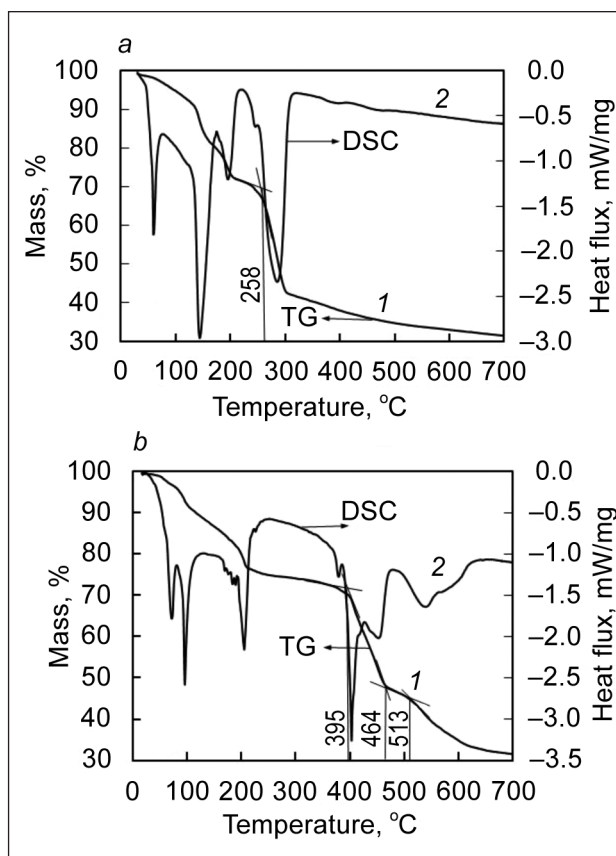

Fig. 1. STA of Ce(NO$_3$)$_3$ (*a*) and La(NO$_3$)$_3$ (*b*) decomposition: *1* – TG curve, *2* – DSC curve

The minimal average size of CuO crystallites was obtained in CuO-La$_2$O$_3$/γ-Al$_2$O$_3$, which corresponds well to literature data [19]. However, the addition of CeO$_2$ leads to the increase of the average size of CuO crystallites. The specific surface areas (S$_{BET}$) of synthesized CuO/γ-Al$_2$O$_3$ and xCuO-yCeO$_2$/γ-Al$_2$O$_3$ catalysts were about the same (~150–160 m^2/g). However, as was expected, CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ catalyst had the lowest value of S$_{BET}$, which can be attributed to thermal aging of the support during calcination procedure at 600 °C.

Catalyst activity

The preliminary tests started performing a feasibility study on three primarily synthesized catalysts. Activity tests of methanol and methyl acetate oxidation over CuO/γ-Al$_2$O$_3$, 9CuO-2CeO$_2$/γ-Al$_2$O$_3$ and CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ catalysts were carried out in order to compare their ability to convert VOCs to desired and nonhazardous products: carbon dioxide and water

Table. **Content of active components and catalyst surface characteristics**

Catalyst sample	Content, wt.%			S$_{BET}$, m^2/g	Average size of CuO crystallites, nm
	CuO	CeO$_2$	La$_2$O$_3$		
CuO/γ-Al$_2$O$_3$	13.51	–	–	156.6	53.4
CuO-La$_2$O$_3$/γ-Al$_2$O$_3$	9.56	–	1.74	134.5	50.0
11CuO-1CeO$_2$/γ-Al$_2$O$_3$	11.39	1.14	–	162.3	60.2
9CuO-2CeO$_2$/γ-Al$_2$O$_3$	9.32	1.94	–	147.2	60.6
9CuO-4CeO$_2$/γ-Al$_2$O$_3$	8.59	3.63	–	149.2	57.6

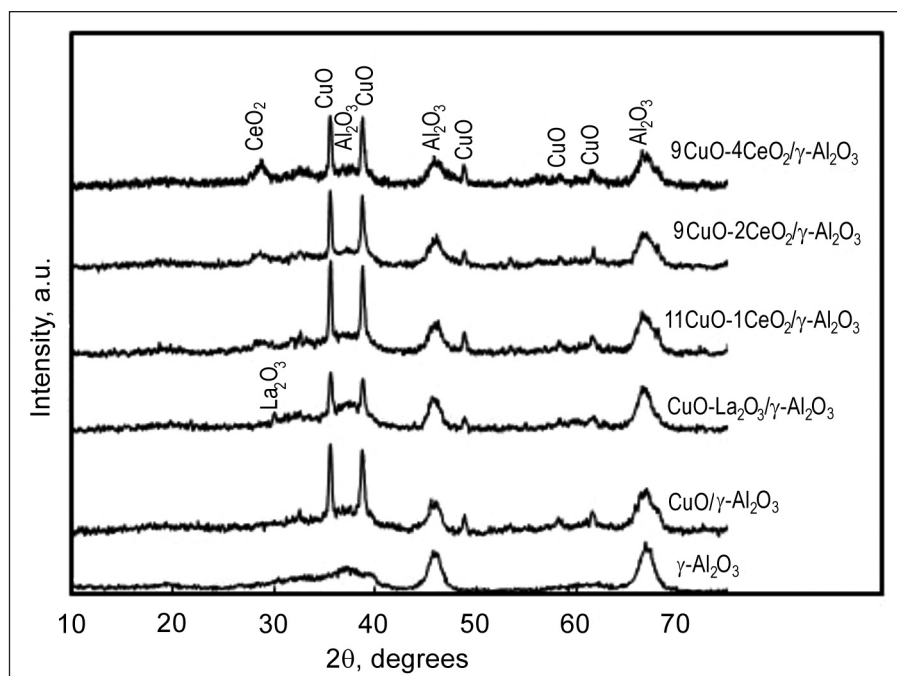**Fig. 2.** XRD patterns of synthesized catalysts and γ-Al$_2$O$_3$ support

vapour. The results of methanol and methyl acetate conversion are represented in Fig. 3. Comparing temperatures of 95% (T$_{95}$) methanol conversion in the activity test, 9CuO-2CeO$_2$/γ-Al$_2$O$_3$ catalyst showed the highest activity, reaching the mentioned conversion value at 192 °C. T$_{95}$ values for CuO/γ-Al$_2$O$_3$ and CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ were 205 °C and 227 °C, respectively. Thus, the temperature shift of T$_{95}$ between the ceria and lanthanum(III) oxide promoted catalyst was sufficiently high (35 °C).

The results of methyl acetate oxidation over supported catalysts revealed that activity of all three primarily tested catalysts is almost the same above 180 °C. However, the copper(II) oxide catalyst with addition of ceria performed better at lower temperatures.

It is clearly seen that the sum of the active components of three firstly tested catalysts is different (see Table). Therefore it was reasonable to compare specific rates of VOCs oxidation over CuO/γ-Al$_2$O$_3$, 9CuO-2CeO$_2$/γ-Al$_2$O$_3$ and CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ catalysts (Fig. 4). Oxidation specific rate is expressed as a quantity of VOC in μmol reacted per 1 s over 1 g of catalyst active components (μmol/(g$_{AC}$ · s)).
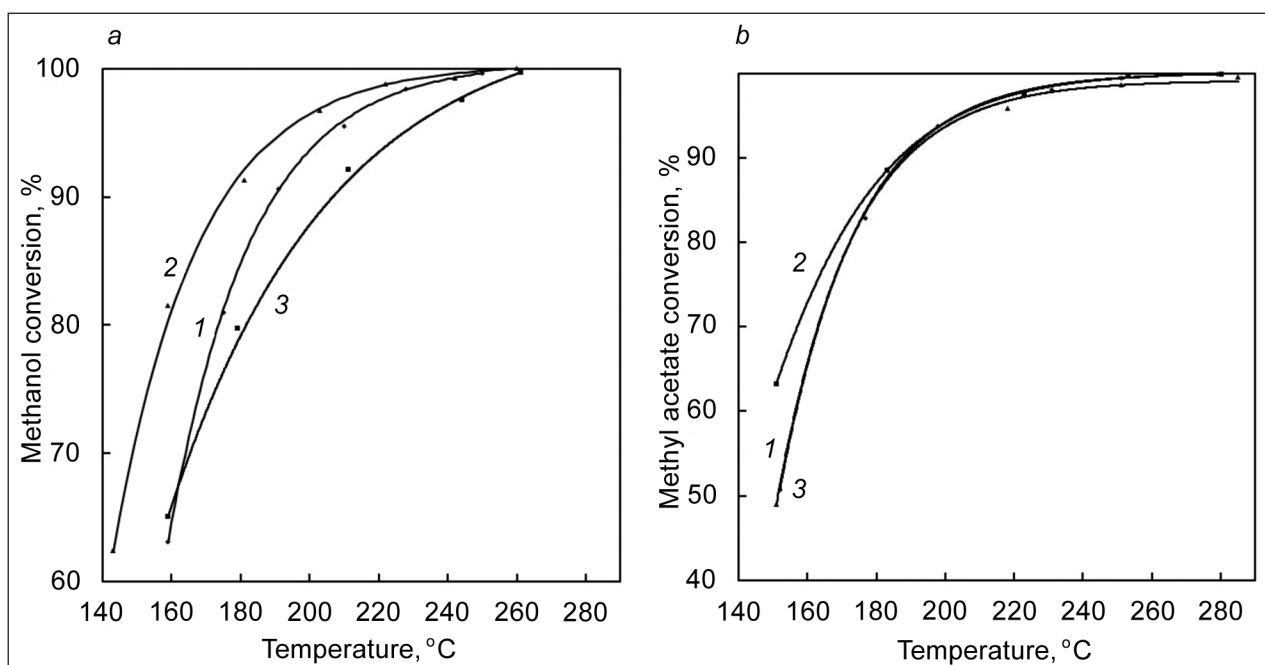
Fig. 3. Methanol (*a*) and methyl acetate (*b*) oxidation over supported catalysts: *1* – CuO/γ-Al$_2$O$_3$, *2* – 9CuO-2CeO$_2$/γ-Al$_2$O$_3$, *3* – CuO-La$_2$O$_3$/γ-Al$_2$O$_3$
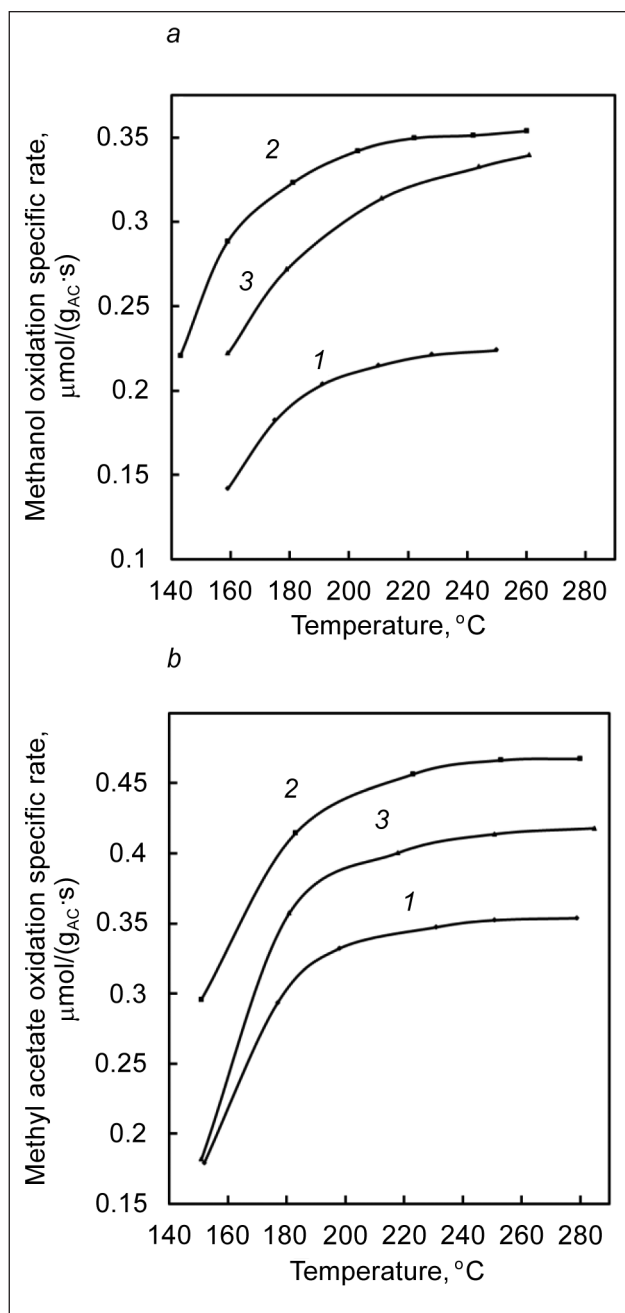
Fig. 4. Methanol (*a*) and methyl acetate (*b*) oxidation specific rates over supported catalysts: *1* – CuO/γ-Al_2O_3, *2* – 9CuO-2CeO_2/γ-Al_2O_3, *3* – CuO-La_2O_3/γ-Al_2O_3

Fig. 5. Carbon monoxide yield during methanol (*a*) and methyl acetate (*b*) oxidation over supported catalysts: *1* – CuO/γ-Al_2O_3, *2* – 9CuO-2CeO_2/γ-Al_2O_3, *3* – CuO-La_2O_3/γ-Al_2O_3

As results show, CuO-La_2O_3/γ-Al_2O_3 catalyst alters the previous sequence of catalytic activity for methanol oxidation (interchanges with CuO/γ-Al_2O_3 catalyst) and performs better for methyl acetate oxidation. CuO/γ-Al_2O_3 catalyst contained 13.51 wt.% of CuO, though the integral quantity of active components was 11.3 wt.% in CuO-La_2O_3/γ-Al_2O_3. Therefore, the calculated specific rates of both VOCs were higher in the analyzed temperature range. However, specific rates of methanol and methyl acetate oxidation were highest over CuO catalyst promoted with CeO_2. Comparing either fractional conversion or specific rates of VOCs oxidation, 9CuO-2CeO_2/γ-Al_2O_3 catalyst exhibited the highest activity.
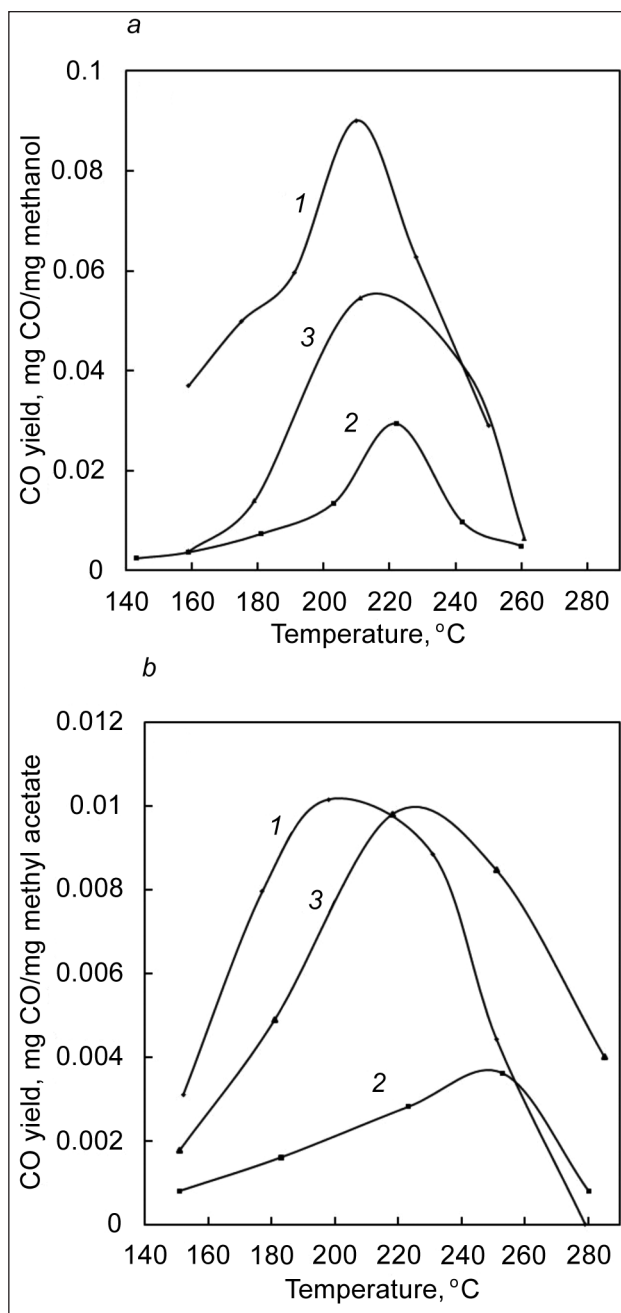
Formation of by-products

Since desired products of the complete oxidation are CO_2 and H_2O, it was necessary to verify whether by-products were formed or not during the catalytic oxidation of VOCs. The results of analysis show the formation of CO during methanol and methyl acetate oxidation and methanol formation during methyl acetate oxidation. The results represented in Fig. 5 show the yield of CO during methanol and methyl acetate oxidation over three primarily tested catalysts. CuO/γ-Al_2O_3 catalyst produced three times, CuO-La_2O_3/γ-Al_2O_3 produced two times more of CO per mg of methanol compared to
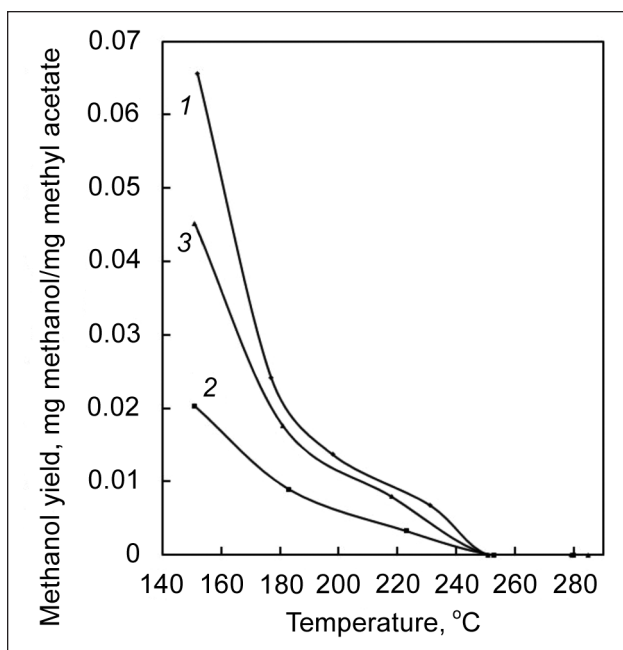
Fig. 6. Methanol yield during methyl acetate oxidation over supported catalysts: *1* – CuO/γ-Al$_2$O$_3$, *2* – 9CuO-2CeO$_2$/γ-Al$_2$O$_3$, *3* – CuO-La$_2$O$_3$/γ-Al$_2$O$_3$

Fig. 7. VOC oxidation specific rate over supported *x*CuO-*y*CeO$_2$/γ-Al$_2$O$_3$ catalysts at different temperatures: methanol – *1* at 180 °C, *2* at 225 °C; methyl acetate – *3* at 180 °C, *4* at 220 °C (where CeO$_2$/CuO=0 is CuO/γ-Al$_2$O$_3$)

9CuO-2CeO$_2$/γ-Al$_2$O$_3$ catalyst. The peak concentration of CO was observed in the range of 210–220 °C during methanol oxidation. The yield of CO was almost the same over CuO/γ-Al$_2$O$_3$ and CuO-La$_2$O$_3$/γ-Al$_2$O$_3$ catalysts during methyl acetate oxidation, however, 9CuO-2CeO$_2$/γ-Al$_2$O$_3$ produced the lowest amount of carbon monoxide.

According to the results obtained with GC/MS (Fig. 6), methanol formation has occurred during methyl acetate oxidation. The highest amounts of methanol were formed in the temperature range of 150–180 °C. However, in accordance with amounts of methanol formed during methyl acetate oxidation, catalysts may be arranged in the same sequence as it was presented in the comparison of their specific rates of VOC oxidation or CO yield.

Effect of CeO$_2$ and CuO ratio on VOC oxidation activity

Since CuO catalyst promoted with CeO$_2$ was the most active and yielded the lowest amounts of by-products among tested catalysts, it was reasonable to determine the optimal ratio of active components. Therefore, VOC oxidation specific rates over three catalysts with different loadings of CuO and CeO$_2$ were compared with CuO/γ-Al$_2$O$_3$ at two constant temperatures (Fig. 7). Composition of CuO/γ-Al$_2$O$_3$ and *x*CuO-*y*CeO$_2$/γ-Al$_2$O$_3$ catalysts was expressed as a ratio of CeO$_2$ and CuO in the horizontal axis. The highest VOC oxidation specific rates obtained in parabolic shape curves determine the optimal ratio of CeO$_2$ and CuO in the catalyst.

CONCLUSIONS

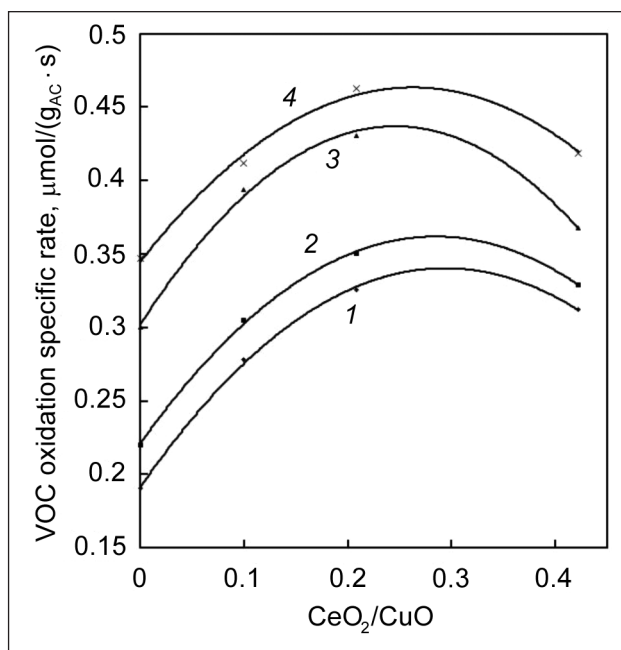CuO catalysts promoted with CeO$_2$ and La$_2$O$_3$ were more active during methanol and methyl acetate oxidation than single CuO supported on γ-Al$_2$O$_3$. However, mixed oxide catalysts of CuO-CeO$_2$ prepared by co-impregnation of γ-Al$_2$O$_3$ were the most efficient for the destruction of tested VOCs. Minimal formation of by-products was obtained over 9CuO-2CeO$_2$/γ-Al$_2$O$_3$ catalyst. According to the activity and the lowest yield of undesired by-products, promoted and pure CuO catalysts may be arranged in the following order – CuO-CeO$_2$>CuO-La$_2$O$_3$>CuO. Specific oxidation rates of VOCs over different composition *x*CuO-*y*CeO$_2$/γ-Al$_2$O$_3$ catalysts revealed that the optimal CeO$_2$ and CuO ratio was in a range of 0.2–0.3 for methanol and methyl acetate oxidation. Neither S$_{BET}$ nor average crystallite size of CuO were significantly affected by addition of CeO$_2$. Therefore, activity of *x*CuO-*y*CeO$_2$/γ-Al$_2$O$_3$ catalysts with CeO$_2$ and CuO ratio of 0.2–0.3 can be attributed to increased redox properties of mixed oxides.

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Aurimas Urbutis, Saulius Kitrys

CeO$_2$ IR La$_2$O$_3$ MODIFIKUOTŲ CuO KATALIZATORIŲ STRUKTŪRA IR AKTYVUMAS LOJ VISIŠKO OKSIDAVIMO REAKCIJOSE

S a n t r a u k a

Darbe nustatytas cerio(IV) ir lantano(III) oksidais modifikuotų užneštinių CuO katalizatorių aktyvumas metanolio ir metilacetato garų visiško oksidavimo reakcijose. Katalizatoriai buvo pagaminti tradiciniu impregnavimo metodu. Katalizatorių aktyviųjų komponentų (CuO, CeO$_2$ ir La$_2$O$_3$) nešikliu buvo panaudotos γ-Al$_2$O$_3$ granulės. Cerio(IV) ir lantano(III) oksidų priedais modifikuotų katalizatorių aktyvumas lygintas su baziniu CuO/γ-Al$_2$O$_3$ katalizatoriumi, kurio sudėtyje buvo 13,51 % vario(II) oksido. Pagaminti katalizatoriai charakterizuoti rentgeno difrakcinės analizės, atominės absorbcinės spektroskopijos, liepsnos atominės emisijos spektroskopijos bei Brunauerio Emeto Telerio specifinio paviršiaus ploto tyrimų metodais. Pirminiais tyrimais nustatyta, kad aktyviausias katalizatorius tirtų lakiųjų organinių junginių oksidavimo reakcijose buvo 9CuO-2CeO$_2$/γ-Al$_2$O$_3$. Šio katalizatoriaus darbo metu susidarė mažiausiai šalutinių, visiško oksidavimo reakcijose nepageidaujamų produktų kiekis. Atlikus tyrimus su skirtingos aktyviųjų komponentų sudėties xCuO-yCeO$_2$/γ-Al$_2$O$_3$ katalizatoriais, nustatyta, kad optimaliausias metanolio ir metilacetato oksidavimo reakcijų metu CeO$_2$ ir CuO santykis buvo nuo 0,2 iki 0,3.