Y-doped ceria-supported gold and palladium mono- and bimetallic catalysts for complete propene oxidation

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Monometallic Au and Pd as well as bimetallic Pd-Au particles deposited on Y-doped ceria supports were studied in complete propene oxidation (CPO), which was used as a probe reaction for aliphatic hydrocarbons abatement. Y-modified ceria supports (1 wt.% Y_2O_3) were prepared by impregnation and coprecipitation. Gold (3 wt.% Au) and palladium (1 wt.% Pd) containing samples were prepared by deposition-precipitation method. Bimetallic Pd-Au catalysts were prepared by adding palladium to already deposited gold. Catalyst samples were characterized by BET, XRD, TPR, and XPS techniques. In general, the catalytic activity in CPO did not differ significantly in the presence of Y dopant and because of synthesis method of mixed oxide support. Total propene conversion over gold catalysts was reached at 220 °C. Pd-based catalysts demonstrated a higher oxidation activity in comparison with Au-based samples showing 100% propene conversion at 200 °C. The lowest temperature of total propene oxidation of 180 °C was achieved over the bimetallic Pd-Au catalysts and long-term catalytic performance showed good stability with no loss of catalytic activity.

Key words: *Au, Pd, and Pd-Au catalysts, Y-doped ceria, complete propene oxidation.*

INTRODUCTION

Treatment of gaseous emissions containing volatile organic compounds (VOCs) has been an increasing concern in the last years. Catalytic combustion is considered a promising way to convert VOCs, especially of low concentration, into harmless CO_2 and water. Selection of catalytic materials for the reduction of VOCs is not easy because several factors such as support type, active sites distribution, and synthesis methods would influence catalyst activity and catalyst lifetime.

Gold-based catalysts are already well known as promising candidates for the total oxidation of VOCs at relatively low temperatures [1]. Rapid change in cerium oxidation state ($Ce^{4+}\leftrightarrow Ce^{3+}$) makes ceria a suitable reducible support, which is also able to stabilize finely dispersed gold particles. Due to the formation of oxygen vacancies, CeO_2 modification with metal dopant of proper oxidation state and concentration could lead to further increase of oxygen mobility in ceria-based catalysts. Own results of gold deposited on ceria doped with MeO_x (Me = Fe, Mn, Co, and Sn) have shown an effect of mixed oxide support composition and structure on the catalytic behaviour in complete benzene oxide-tion [2,3]. Recent results have shown a beneficial effect on the catalytic performance in complete benzene oxidation in the case of Au deposited on Y-doped ceria support (1 wt.% Y_2O_3) prepared by impregnation as compared to gold on bare ceria [4].

Catalysts containing both highly dispersed gold and palladium particles were reported as very effective in many reactions including organic compounds oxidation. A higher catalytic activity in the case of bimetallic formulation as compared to the corresponding monometallic Au and Pd catalysts for catalytic destruction of *p*-aniline [5] and toluene [6] was observed. An important role of metal loading sequence was established [7,8]. Catalysts prepared by palladium introduction to already deposited gold, containing Pd(shell)–Au(core) particles, exhibited the best catalytic activity in toluene and propene oxidation [7]. In accordance with these observations, the highest complete benzene oxidation activity has been reported for Pd-Au catalyst prepared by impregnation of Fe-doped ceria support when Pd loading followed gold deposition [9].

The present study is focused on the complete oxidation of propene, the latter being selected as a probe molecule for aliphatic hydrocarbon combustion. The activity of mono- (Au, Pd) and bimetallic Pd-Au particles (Pd added to deposited Au particles) supported on differently prepared Y-doped ceria (1

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wt.% dopant) in the complete propene oxidation (CPO) was investigated. Modification of Au, Pd, and Y oxidation state before and after catalytic work was elucidated

EXPERIMENTAL

Sample preparation

Three series of catalysts containing Au (3 wt.%), Pd (1 wt.%), and Pd (1 wt.%)-Au (3 wt.%) supported on bare ceria, Y-doped ceria prepared by impregnation (IM), and Y-doped ceria prepared by co-precipitation (CP) were synthesised. Yttria dopant amount was 1 wt.%.

Ceria was prepared by precipitation of aqueous solution of $Ce(NO_3)_3.6H_2O$ with K_2CO_3 at 60°C and constant pH = 9.0. The resulting precursor was aged at the same temperature for 1 h, filtered and carefully washed until NO_3^- ions removal, dried in vacuum at 80 °C, and calcined in air at 400 °C for 2 h. Using IM method ceria was impregnated with aqueous solution of appropriate amount of $Y(NO_3)_3.6H_2O$ under vigorous stirring at room temperature for 4 h. Then, the suspension was evaporated under vacuum at 70 °C in a rotary evaporator until water was completely removed. By CP method a solution of Y and Ce nitrates at an appropriate ratio were co-precipitated with a solution of K_2CO_3 at constant pH = 9.0 and temperature of 60 °C. The precipitates were aged at the same temperature for 1 h, filtered, and washed until removal of NO_3^- ions. All resulting precursors were dried in vacuum at 80 °C and calcined in air at 400 °C for 2 h. Before metal deposition, the support was dispersed in water and activated in an ultrasound disintegrator.

Gold was deposited by deposition-precipitation method from $HAuCl_4.3H_2O$ and K_2CO_3 precursors under vigorous stirring. The precipitation was carried out under full control of all parameters of preparation (constant pH = 7, T = 60 °C, stirring speed = 250 rpm, reactant feed flow rate = 0.15 $l.h^{-1}$, etc.). After filtering and careful washing, the solid was dried under vacuum and calcined in air at 400 °C for 2 h. The gold catalysts were denoted as AuCe, AuYCeIM, and AuYCeCP.

Pd-containing samples were prepared from $Pd(NO_3)_2.xH_2O$ precursor following aforementioned preparation method and calcination procedure for gold catalysts. These samples were denoted as PdCe, PdYCeIM, and PdYCeCP.

Sequential deposition-precipitation was applied for the preparation of bimetallic Pd-Au catalysts. Palladium was loaded on already prepared and calcined Au/Y-doped ceria. After Pd deposition calcination in air at 400 °C for 2 h was carried out. The samples were denoted as Pd-AuCe, Pd-AuYCeIM, and Pd-AuYCeCP.

Sample characterization

Sample BET surface area (S_{BET}) was evaluated by performing nitrogen adsorption/desorption experiments on a Carlo Erba Sorptomat 1900 instrument. Prior to the measurements, each sample was outgassed at 200 °C for 1 h under vacuum. Computerized analysis of the nitrogen adsorption isotherm at −196 °C allowed estimating the specific surface areas of the samples in the standard pressure range of 0.05–0.3 P/P_0.

X-ray powder diffraction (XRD) measurements were performed using a D5000 diffractometer (Bruker AXS), with Cu sealed tube operating at 40 kV and 40 mA. The setup employed Bragg-Brentano focusing geometry with 1° beam divergence and LynxEye strip detector. Data were analysed using PeakFit program (Jandel Scientific) and fityk (Copyright 2001–2014 Marcin Wojdyr) fitting XRD profiles to K_α 1,2 doublets having PEARSON VII analytical form. The CeO_2 phase was analysed on the basis of 14 well measured reflections and Williamson-Hall plot [10,11] to calculate the average crystal size. Gold particle crystal size was estimated based on the strongest (111) reflection as the only one detectable. The procedure is described in detail elsewhere [4].

X-ray photoelectron spectroscopy (XPS) analysis was performed by VG Microtech ESCA 3000 Multilab equipped with a dual Mg/Al anode [12]. The spectra were excited by unmonochromatized Al Kα source (1486.6 eV) run at 14 kV and 15 mA. The analyser was operated in the constant analyser energy (CAE) mode. For the individual peak energy regions, a pass energy of 20 eV set across the hemispheres was used. Sample powders were pelletized and mounted on a double-sided adhesive tape. The pressure in the analysis chamber was in the range of 10^{-8} Torr during data collection. Constant charging of the samples was removed by referencing all the energies to the C1s set at 285.1 eV arising from adventitious carbon. Peak analysis was performed with software provided by VG based on non-linear least squares fitting program using a weighted sum of Lorentzian and Gaussian component curves after background subtraction according to Shirley and Sherwood [13,14].

Temperature-programmed reduction (TPR) measurements were carried out by means of an apparatus described elsewhere [15]. A cooling trap (−40 °C) for removing water formed during reduction was mounted in the gas line prior to the thermal

conductivity detector. A hydrogen-argon mixture (10% H_2), dried over a molecular sieve 5A (−40 °C), was used to reduce the samples at a flow rate of 24 ml.min^{-1}. The temperature was linearly raised at a rate of 15 °C min^{-1}. The amount of sample used was 0.05 g based on a criterion proposed by Monti and Baiker [16]. Hydrogen consumption during the reduction process was calculated using preliminary calibration of the thermal conductivity detector performed by reducing different amounts of NiO to Ni^0, NiO – 'analytical grade' being calcined at 800 °C for 2 h to avoid presence of non-stoichiometric oxygen.

Catalytic activity measurements in CPO

The catalytic activity in CPO, expressed as degree of propene conversion, was evaluated in the temperature range of 100–300 °C after catalyst pretreatment using 50 ml.min^{-1} of 5% O_2 in He for 1 h at 350 °C. The measurements were carried out in U-shaped quartz reactor under reactive gas mixture containing 1500 ppm C_3H_6 + 4.2% O_2 (He as balance) at weight hourly space velocity (WHSV) of 60000 ml.h^{-1}.g^{-1}. The reactants and products were examined by IR analysers (ABB Uras 14) for CO and CO_2 species, while oxygen concentration in the feed was measured by ABB paramagnetic analyser. Propene conversion was evaluated taking into account the CO_2 formed during the reaction.

RESULTS AND DISCUSSION

Catalytic activity measurements

The catalytic results of CPO over the studied catalysts as a function of reaction temperature are compared in figure 1. In all cases, detected CO was negligible and no intermediate products of partial propene oxidation were registered. The effect of monometallic (Au and Pd) and bimetallic Pd-Au deposition on combustion activity is clearly seen in the figure. In general, the Pd-based catalysts exhibited a higher catalytic activity than the matching

Au-containing samples. A higher activity of Pd as compared to Au catalysts in total oxidation of propene and methyl ethyl ketone has been shown in a comparative study of Au and Pd deposited on titanium oxide nanotubes [17]. In a recent investigation of toluene catalytic oxidation over Au and Pd supported on macro-mesoporous metal-doped titania, it has been reported that Pd-containing systems also exhibited a better performance [18]. Formation of highly dispersed Pd^{2+} species on different oxides as supports was considered responsible for the propene oxidation at lower temperatures by Gil *et al*. [19].

The present results indicated the same catalytic behaviour in CPO over Pd and Pd-Au catalysts on bare ceria. The effect of Y was negligible for the monometallic gold and palladium catalysts and slightly positive with the bimetallic samples. However, a definitive positive effect of the palladium deposition after gold loading was observed for Y-doped ceria supports. The reason for higher catalytic activity over bimetallic Pd-Au catalysts as reviewed above is still not exactly clarified. Enache *et al*. [20] supposed that gold influenced electronically the catalytic properties of Pd. Hosseini *et al*. [7] have proposed a Langmuir-Hinshelwood mechanism for toluene and propene oxidation over Pd-Au/TiO_2 catalysts, suggesting that a VOC molecule and oxygen from a [Pd–O] complex compete for adsorption. The authors explained the higher activity of the Pd(shell)–Au(core) as compared to the Au(shell)–Pd(core) system by easier [Pd–O] formation and low adsorption of oxygen on gold due to its lower ability to polarize the O=O bond as compared to Pd.

In the present study, the lowest temperature of total propene conversion was achieved at 180 °C over both Pd-AuYCeIM and Pd-AuYCeCP catalysts. Bimetallic Pd-Au entity on IM support demonstrated better activity at a lower temperature (150 °C) as compared to Pd-Au on CP support (see Fig. 1). Long lasting tests (72 h) in CPO at 180 °C performed over Pd-AuYCeIM sample showed that the catalyst was stable with no loss of oxidation activity.
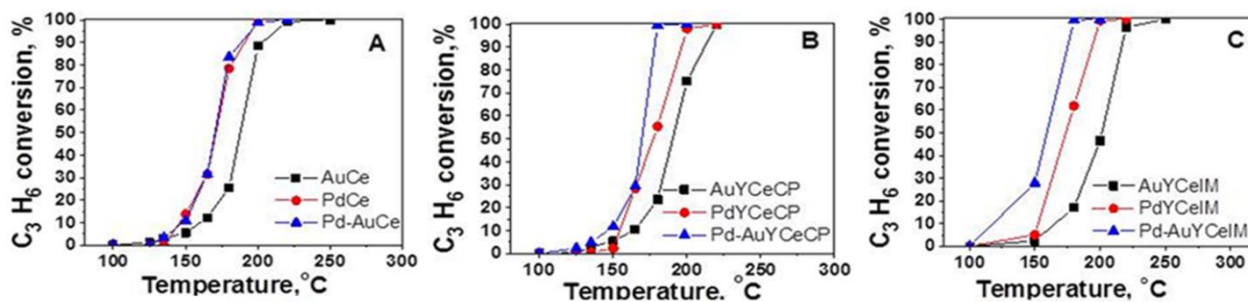

Fig. 1. Effect of monometallic (Au and Pd) and bimetallic Pd-Au loading on ceria (A) and Y-doped ceria supports (B and C) on propene conversion degree.

Sample characterization

Specific surface area (S_{BET}) values are given in table 1. All S_{BET} values are ranging around 100 ± 10 $m^2.g^{-1}$. No substantial differences between mono- and bimetallic catalysts were observed; slightly lower values were found in the case of CP supports.

Table 1. Specific surface area (S_{BET}), and hydrogen consumption of the studied catalysts

Catalyst	S_{BET} ($m^2.g^{-1}$)	HC ($mmol.g^{-1}$)	
		to 250 °C	to 800 °C
AuCe	102.4	0.5	0.5
PdCe	101.9	0.6	1.0
Pd-AuCe	108.5	0.6	1.2
AuYCeCP	90.0	0.7	0.7
PdYCeCP	99.6	0.7	1.4
Pd-AuYCeCP	90.0	0.7	1.1
AuYCeIM	107.2	0.6	0.6
PdYCeIM	105.0	0.7	1.2
Pd-AuYCeIM	105.5	0.5	1.1

X-ray diffractograms of all the studied catalysts showed typical reflections of face-centred cubic fluorite-type structure of CeO_2 (Fig. 2). A separate Y_2O_3 phase was not visible. Pd was not detectable in the Pd containing samples. For the gold containing samples a weak Au(111) peak was registered at $2\theta = 38.2°$ suggesting absence of alloying between Au and Pd. As expected ceria and gold average particle sizes were in accord with those obtained previously using the same synthesis procedure [4].
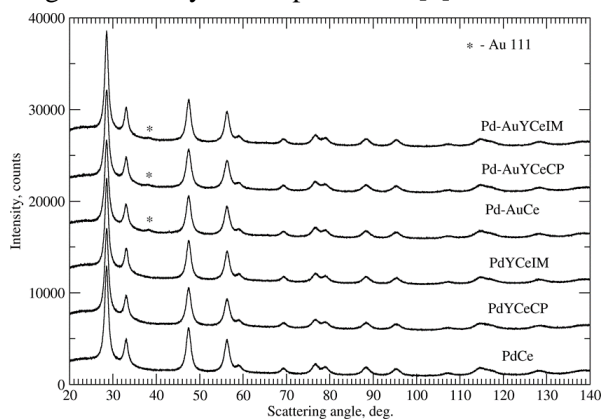

Fig. 2. XRD patterns of the studied Pd and Pd-Au catalysts.

The average particle size of bare ceria, Y-doped ceria prepared by IM, and Y-doped ceria prepared by CP was 5.7, 7.1, and 8.1 nm, respectively, whereas the average particle size of gold in Au and Pd-Au entities on bare ceria, Y-doped ceria prepared by IM, and Y-doped ceria prepared by CP was 4.1, 4.0, and 5.8 nm, correspondingly.

As reported before, based on HRTEM/HAADF analyses evaluated gold average particle size in AuCe, AuYCeIM, and AuYCeCP (valid for the Pd-Au samples as well) was 2.1, 2.6, and 3.2 nm, respectively [21]. Measurements of interplanar distances in selected HRTEM images of AuYCeIM catalyst evidenced the presence of Y_2O_3 crystals (not detectable by XRD) even in the case of 1 wt.% dopant content [21]. HRTEM measurements of Pd-containing samples are not reported. Due to similarity in lattice distances of PdO and CeO_2 it was difficult to distinguish unambiguously between ceria and PdO crystals by HRTEM [9].

Sample reducibility was evaluated by means of TPR measurements. The reduction of ceria proceeds in two steps: surface layer reduction at around 500 °C and bulk reduction above 800 °C [22]. It is known that the presence of noble metals is beneficial for the ceria reduction. Such effect has also been observed for gold when supported as nanoparticles. Many studies have confirmed the first observations of the research groups of Stephanopoulos [23] and Andreeva [24] that the role of nanogold is to lower substantially the temperature of the ceria surface layer reduction. Doping ceria with low concentration of Y^{3+} ions leads to random oxygen vacancies formation accompanied by enhanced reduction of Au/Y-doped ceria catalysts [4 and references therein].

TPR profiles of Au, Pd, and Pd-Au on ceria and Y-doped ceria supports are illustrated in figure 3. The TPR profiles of bare ceria supported mono- and bimetallic catalysts are compared in figure 3A. The T_{max} of the peak assigned to surface ceria reduction of AuCe (contribution of positively charged gold particles can be neglected) was 120 °C and the reduction process was finished up to 200 °C. Differently from samples with nanosized gold favouring ceria surface layer reduction [24], low intense broad peaks at higher temperatures could be seen in the presence of Pd. They were related to enhanced reduction of deeper ceria layers caused by palladium. The first TPR peaks in the profiles of Pd and Pd-Au catalysts should be assigned to the reduction of both ceria surface layers and oxidized palladium (revealed by XPS data on fresh catalysts). Compared with the TPR peak of AuCe, the peaks of Pd containing samples were narrower and located at lower temperatures, especially in the case of Pd-AuCe (T_{max} at 43 and 23 °C, respectively). The same tendency was observed on comparing the TPR profiles of Au, Pd, and Pd-Au on Y-doped ceria prepared by CP and IM (Figs. 3B and C). The lowest T_{max} at 20 °C was registered for Pd-AuYCeIM, however, a lower intensity of this peak was noticeable.

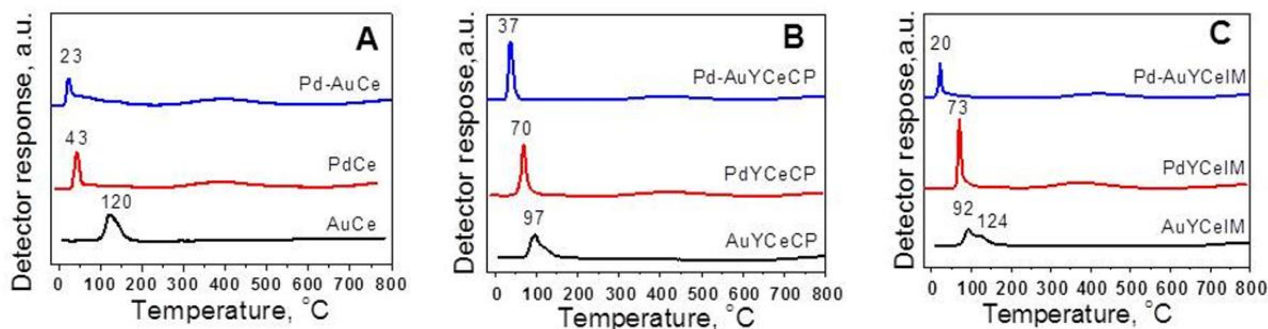Sample reduction behaviour was also evaluated based on calculated hydrogen consumption (HC).

Fig. 3. TPR profiles of the studied ceria- (A) and Y-doped ceria-supported (B and C) monometallic (Au and Pd) and bimetallic Pd-Au catalysts.

Table 1 presents experimental HC values during TPR up to 250 and 800 °C. In agreement with literature data, theoretical HC values for ceria surface layer reduction without changes in ceria bulk structure are limited to 17% [25] or 20% [26]. This means stoichiometric HC values of 0.49–0.58 and 0.49–0.57 mmol.g^{-1} for bare ceria and ceria doped with 1 wt.% Y_2O_3, respectively. Hydrogen consumption of AuCe was in the frame of the theoretical values for ceria surface layer reduction; a relatively higher HC in the case of CP method as compared to the IM mode was observed (even HC of the AuYCeCP catalysts exceeded 20% reduction).

For palladium containing catalysts, bearing in mind that the calculated HC value needed for the process $PdO + H_2 \rightarrow Pd^0$ is 0.08 mmol.g^{-1}, the HC for ceria surface layers reduction up to 250 °C was almost the same as for the matching gold catalysts. These close results justify the similarity of the CPO activities regardless of support nature. However, there existed differences in the oxidation activities determined by the type of loaded metal. The bimetallic catalysts exhibited the best performance irrespective of support composition. Sample activities followed an inverse trend of the reduction T_{max}: the C_3H_6 conversion varied with the supported metal catalysts as Pd-Au > Pd > Au. The TPR T_{max} of the catalysts followed the order T_{max} of Pd-Au catalysts < T_{max} of Pd catalysts < T_{max} of Au catalysts. Such an inverse relationship confirmed the important role of the oxygen being supplied by the support. Indeed the T_{max} of the TPR peak is indicative of the easiness with which the oxygen from ceria can be extracted.

A lower HC value obtained for Pd-AuYCeIM catalyst, which was mentioned above in relation to TPR peak intensity, is not in agreement with its highest CPO activity up to 180 °C, the latter being the temperature at which complete oxidation was also achieved over Pd-AuYCeCP. Tabakova *et al.* have observed a similar discrepancy for bimetallic catalyst of best catalytic performance in a study of Pd-Au deposited on Fe-modified ceria for complete

benzene oxidation [9]. Based on XPS evidence for metallic Pd particles at the onset of the low temperature TPR peak, the authors explained unexpectedly decreased reducibility by the formation of inactive Au_xPd_y alloy species in the hydrogen flow [9].

XPS data on fresh and used samples in CPO are summarized in table 2. All samples contained Ce^{4+} and a minor percentage of Ce^{3+} ions. A certain amount of Ce^{3+} in the fresh samples could be related to X-ray beam induced reduction during XPS analysis. For a fresh AuCe catalyst, the Au $4f_{7/2}$ binding energy (BE) was typical of metallic Au^0 [27]. Yttrium presence induced a slight increase of the BE especially for AuYCeCP catalyst sample, thus indicating occurrence of partly positively charged $Au^{\delta+}$. Charged $Au^{\delta+}$ particles were also registered in Pd-Au catalysts. Calculated atomic ratios showed that the Au catalysts exposed a gold-enriched surface of nominal Au/Ce of 0.03. After palladium deposition over gold, the Au/Ce ratio in the Pd-Au catalysts became closer to the nominal value. The monometallic palladium catalysts were characterized by Pd $3d_{5/2}$ BE of 337.1 eV that is typical of Pd^{2+} [28]. The bimetallic samples exhibited somewhat higher Pd 3d binding energy as compared to the monometallic ones, which may be attributed to a charge transfer from palladium to the more electronegative gold species. The Pd/Ce atomic ratio was always much higher than the nominal one (0.016) showing a strong surface segregation of palladium.

The binding energy of Y $3d_{5/2}$ at 157.6 ± 0.2 eV is typical of Y^{3+}. In the palladium containing samples, another Y $3d_{5/2}$ component at a lower energy (about 153.3 ± 0.3 eV) was registered. Such a component assigned to partially reduced yttrium ($Y^{(3-\delta)+}$) has been related to alloying effect in the presence of the noble metal [29]. Enrichment of the ceria surface by yttrium was evidenced by the Y/Ce atomic ratio values, which were larger than the analytical one (0.016).

XPS analysis of samples after reaction was carried out for the best performing bimetallic cata-

lysts. As shown in table 2, gold in the used samples was slightly more reduced as compared to the fresh catalysts. Palladium exhibited two components, one at high energy typical of oxidized palladium and the other due to reduced palladium.

Pd 3d and Y 3d spectra of the bimetallic fresh and spent catalysts are displayed in figures 4 and 5. It was interesting to observe that the largest amount of reduced palladium was found in the Pd-AuYCeIM catalyst as compared to the other two ca-
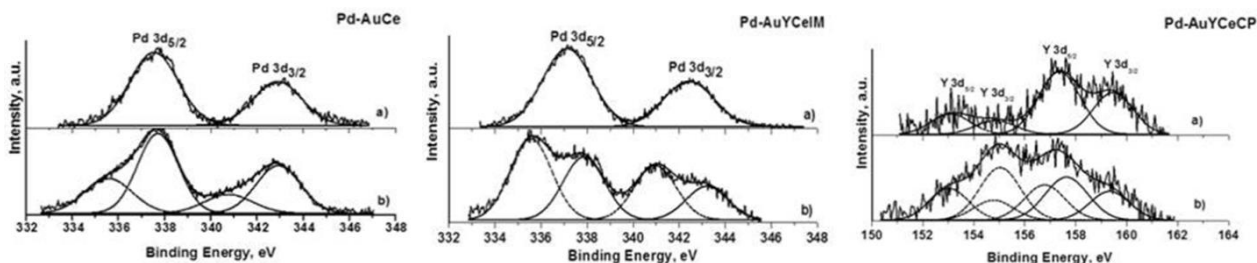

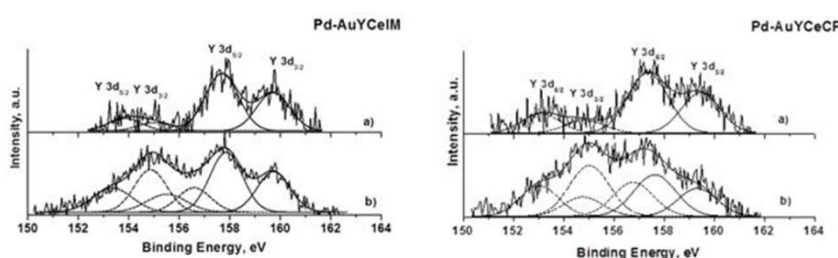Fig. 4. Pd 3d XP spectra of Pd-Au catalysts: a) fresh,
b) after CPO.

Fig. 5. Y 3d XP spectra of Pd-Au catalyst: a) fresh,
b) after CPO.

Table 2. XPS data in terms of binding energy and atomic ratio

Sample	Au 4f$_{7/2}$ eV	Pd 3d$_{5/2}$ eV	Y 3d$_{5/2}$ eV	O 1s eV	Pd/Ce (0.016)	Au/Ce (0.03)	Y/Ce (0.016)
AuCe	84.3			529.3 (57%) 531.6 (43%)		0.06	
AuYCeCP	85.1		157.7 (66%) 153.8 (34%)	529.5 (79%) 532.0 (21%)		0.06	0.09
AuYCeIM	84.5		157.4	529.4 (77%) 532.0 (23%)		0.06	0.15
PdCe*		337.1		529.6 (80%) 531.4 (20%)	0.14		
PdYCeCP*		337.2	157.5 (57%) 153.4 (43%)	529.6 (73%) 532.2 (27%)	0.49		0.25
PdYCeIM*		337.1	157.7 (81%) 153.2 (19%)	529.4 (78%) 531.7 (22%)	0.36		0.21
Pd-AuCe	84.5	337.6		529.5 (76%) 531.8 (24%)	0.25	0.03	
Pd-AuCe spent in CPO	84.2	337.7 (76%) 335.3 (24%)		529.3 (69%) 531.5 (20%) 533.2 (11%)	0.20	0.02	
Pd-AuYCeCP	84.8	337.5	157.4 (70%) 153.2 (30%)	529.4 (82%) 531.5 (18%)	0.32	0.04	0.04
Pd-AuYCeCP spent in CPO	84.2	337.8 (70%) 335.6 (30%)	157.6 (33%) 155.1 (37%) 153.1 (30%)	529.5 (60%) 531.8 (24%) 533.5 (16%)	0.27	0.02	0.15
Pd-AuYCeIM	84.8	337.2	157.6 (63%) 153.7 (37%)	529.3 (64%) 531.4 (36%)	0.33	0.04	0.07
Pd-AuYCeIM spent in CPO	84.2	337.9 (44%) 335.6 (56%)	157.8 (45%) 155.9 (33%) 153.2 (22%)	529.3 (60%) 531.7 (24%) 533.2 (16%)	0.19	0.04	0.17

talysts. On the contrary, in the bimetallic catalysts the Y 3d appeared more reduced for the CP Y-doped ceria with respect to the IM Y-doped ceria.

A Langmuir-Hinshelwood mechanism is generally claimed to operate in the catalytic oxidation of hydrocarbons over Pd-containing catalysts, although a redox Mars-van Krevelen mechanism cannot be excluded [30]. As it was already mentioned, Hosseini *et al.* [7] proposed Langmuir-Hinshelwood mechanism for the propene oxidation over bimetallic gold and palladium catalysts on titania. The present XPS observation of increased reduction after CPO over palladium, particularly with Pd-AuYCeIM sample, would further support the formation of a complex between the organic molecule and Pd-O species followed by reduction of palladium and oxidation of organic molecule. Enhanced reduction of palladium in this particular catalyst is attributed to the presence of gold and yttrium acting in synergy.

CONCLUSIONS

Mono- (Au, Pd) and bimetallic Pd-Au particles (Pd loaded over already deposited Au) in Y-modified (1 wt.% Y_2O_3) ceria supports exhibited a high catalytic activity in the reaction of CPO. Y dopant presence in the cerium oxide and synthesis method of mixed oxide support (impregnation or coprecipitation) did not significantly affect the catalytic performance.

These results are in agreement with catalyst reducibility evaluated from hydrogen consumption data during TPR. Moreover, CPO activity was affected by Au, Pd, or Pd-Au presence. Generally, the Pd catalysts were more active than matching Au catalysts. The bimetallic catalysts exhibited the best CPO activity. This function of activities correlated well with oxygen mobility through ceria surface layers. Modification of the oxidation state of gold, palladium, and yttrium after catalysis revealed a redox character of propene oxidation over the studied catalysts. A relatively low temperature of total propene oxidation of 180 °C, reached over Pd-Au catalysts on Y-doped ceria, and manifested good stability during longstanding catalytic performance make the bimetallic catalysts promising for reducing the energy requirements for VOCs abatement.

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МОНОМЕТАЛНИ (Au, Pd) И БИМЕТАЛНИ Pd-Au ЧАСТИЦИ, НАНЕСЕНИ ВЪРХУ Y-ДОТИРАНИ ЦЕРИЕВООКСИДНИ НОСИТЕЛИ ЗА ПЪЛНО ОКИСЛЕНИЕ НА ПРОПЕН

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(Резюме)

Монометални (Au, Pd) и биметални Pd-Au частици, нанесени на Y-дотирани цериевооксидни носители, са изследвани в моделната реакция на пълно окисление на пропен за пречистване на въздух от алифатни въглеводороди. Y-модифицираните цериевооксидни носители (1 тегл.% Y_2O_3) са синтезирани чрез импрегниране или съутаяване. Злато- (3 тегл.% Au) и паладий-съдържащите (1 тегл.% Pd) катализатори са получени по метода на отлагане чрез утаяване. Биметалните Pd-Au катализатори са синтезирани чрез добавяне на паладий към предварително нанесено злато. Получените катализатори са охарактеризирани с помощта на ниско-температурна адсорбция на азот (БЕТ), рентгенова дифракция, температурно-програмирана редукция и рентгенова фотоелектронна спектроскопия. Каталитичната активност не зависи съществено от наличието на Y-допант и метода на синтез на смесенооксиден носител. Пълното окисление на пропен в присъствие на златните катализатори е при 220 °C. Pd-съдържащите катализатори проявяват по-висока окислителна активност в сравнение със златните, показвайки 100% окисление на пропен при 200 °C. Най-ниската температура за пълното окисление на пропен, 180 °C, е постигната в присъствие на биметалните Pd-Au катализатори и проведеният продължителен каталитичен тест показва стабилна работа без загуба на каталитична активност.