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2016

Dostupný z <http://www.nusl.cz/ntk/nusl-254030>

Dílo je chráněno podle autorského zákona č. 121/2000 Sb.

Tento dokument byl stažen z Národního úložiště šedé literatury (NUŠL).

Datum stažení: 20.04.2024

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Total oxidation of ethanol over Au/Ce_{0.5}Zr_{0.5}O₂ catalysts

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Catalytic oxidation is a green and efficient way to handle man-made emissions of volatile organic compounds (VOC), which represent serious environmental and health issue. Despite high purchasing costs, supported noble metal catalysts are favoured over metal oxides due to their high activity and selectivity, excellent stability and superior resistance against poisoning.

Ceria attracted attention as VOC oxidation catalyst and support due to its unique property of storing and releasing oxygen. It can play the role of promoter and reducible support, provide lattice oxygen, increase the dispersion of supported metals and prevent their sintering or coke formation.

Gold supported on ceria has been reported as a good catalyst for benzene, toluene, ethanol, propanol, propene, formaldehyde and ethyl acetate oxidation. Recently, ceria-zirconia mixed oxides were proposed as VOC oxidation catalysts. It was shown that the partial substitution of cerium with zirconium leads to improved oxygen storage capacity, redox properties, thermal resistance, and better catalytic activity in oxidation reactions.

We have demonstrated that depending on the type of VOC, the introduction of gold can increase catalytic performance and/or selectivity of ceria-zirconia catalysts [1-3]. The aim of this work was to investigate in a detail the effect of Au loading on the catalytic performance of ceria-zirconia supported catalysts in the total oxidation of ethanol.

The Ce_{0.5}Zr_{0.5}O₂ support was prepared from zirconium n-propoxide and of cerium(III)nitrate hexahydrate. The calcined support was impregnated with aqueous solution of gold(III)acetate, dried at 120 °C and calcined at 300 °C. The Au content in the catalysts according to ICP-AES analysis was 0.20, 0.37, 1.55 and 3.35 wt. %, respectively. Catalytic light-off experiments of ethanol gas-phase oxidation were carried out at 20 m³·kg⁻¹·h⁻¹ space velocity with 0.8 g/m³ of ethanol in air.

The surface area S_{BET} was 45 m²/g for the CeZr catalyst and decreased to 25 m²/g for the 3.35Au/CeZr catalyst. With increasing gold loading, the Au particle size estimated from X-ray diffraction increased from 13 nm to 22 nm (Table 1). However, this decrease was not detrimental for the catalytic performance of gold catalysts (Table 1).

Table 1

Au particle size and catalytic performance of the catalysts in the total oxidation of ethanol

Catalyst	Au particle size* (nm)	Temperature of 50 % conversion (°C)
CeZr	-	191
0.20Au/CeZr	13	181
0.37Au/CeZr	17	182
1.55Au/CeZr	18	182
3.35Au/CeZr	22	161

* determined from X-ray diffractograms

Concluding, it was shown that the addition of gold can improve the catalytic performance of

ceria-zirconia mixed oxide in ethanol oxidation. The positive effect of gold addition can be ascribed to improved redox properties of ceria-zirconia support due to the presence of Au nanoparticles.

References

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