CO oxidation over Ce-Ru-O catalysts

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Introduction

The catalytic oxidation of CO at low temperature has attracted considerable attention because of its wide applications in exhaust abatement for CO_2 lasers, trace CO removal in enclosed atmospheres, automotive emission control, and CO preferential oxidation for proton exchange membrane fuel cells.

Recently it was found that stabilized in oxygen rich media Ru is highly active in CO oxidation [1]. On the other hand, ceria is famous because of its high oxygen/vacancy mobility which plays an important role in CO oxidation [2, 3]. Our aim was to produce stable Ce-Ru solid solution capable to convert CO to CO_2 near to room temperature.

Materials and methods

Ceria doped with ruthenium ($Ce_{0.95}Ru_{0.05}O_2$ and $Ce_{0.9}Ru_{0.1}O_2$) catalysts were prepared by sol-gel method modified by CIT technique [4]. Stoichiometric quantities of metal precursors were dissolved in isopropyl alcohol and water. Solutions were stirred for 4 h at 60°C, and then mixed with simultaneous addition of citric acid solution. At this step sol was formed. In order to prepare gel the mixture was evaporated. Obtained gel was calcined at 110°C for 12 h. Finally obtained fluffy solid was calcined at 600°C for 4 h with a temperature ramp of 5°C per min. The structural features of catalysts were characterized by X-ray diffraction (XRD). The surface area of prepared samples was measured by BET method. Their catalytic activity in CO oxidation was tested in flow micro reactor using (1.0%CO+0.5%O_2,He rest) gas mixture at residence time 75 ms. The contribution of different Ru oxide species was estimated by TPR with CO.

Results and Discussion

Prepared samples $Ce_{0.95}Ru_{0.05}O_2$ and $Ce_{0.9}Ru_{0.1}O_2$ are characterized with some changes of CeO_2 structure according to XRD analysis. In the case of $Ce_{0.95}Ru_{0.05}O_2$ 50% of ruthenium was stabilized as solid Ce-Ru solution or Ru oxide species supported on ceria according to TPR analysis. The increase of ruthenium concentration in $Ce_{0.9}Ru_{0.1}O_2$ does not change absolute value of "stabilized ruthenium" giving rise the contribution of bulk ruthenium oxide. Peculiarities of co-crystallization of ceria and ruthenium oxide were manifested also in dynamic changes of surface area for mixed oxides in comparison with the pure CeO_2. Presence of ruthenium precursor prevents formation of big ceria crystals and results in the increase of surface area for mixed oxides then for ceria only as it is shown in Table 1.

As can be seen on Figure 1 catalytic activity of mixed oxides is much higher than for ceria, $Ce_{0.9}Ru_{0.1}O_2$ being the most active one. $Ce_{0.95}Ru_{0.05}O_2$ is less active but more stable. Its activity practically does not change with a number of runs.

The changes in activity with number of runs for $Ce_{0.9}Ru_{0.1}O_2$ seem to be caused by redox transformation of catalyst in the course of the reaction due to the presence of extra quantity of bulk RuO_2 oxide. In contrast to bulk RuO_2 oxide Ce-Ru solid solution seems to be more stable.

Table 1. Surface area

Catalyst	$m^2 g^{-1}$
CeO_2	9
$Ce_{0.95}Ru_{0.05}O_2$	32
$Ce_{0.90}Ru_{0.10}O_2$	25

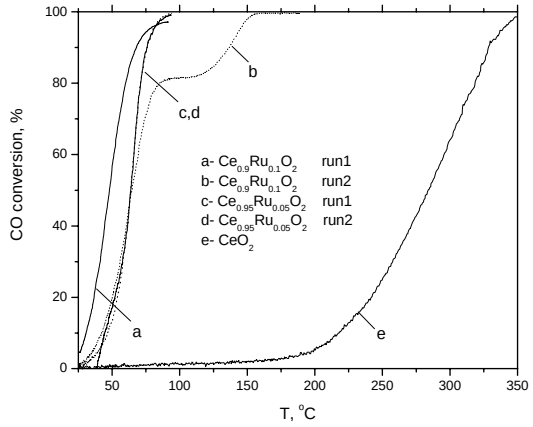

Figure 1. CO conversion vs temperature. Temperature ramp was 5°C per min.

Significance

The key-role of Ce-Ru-O solid solution in the CO oxidation revealed in the present work is important for the development of new effective catalysts for protection of environment.

References

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