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ABSTRACTS

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The Effect of Template Nature and the Composition of Double and Triple Oxide Catalysts Based on CeO_2 in CO Oxidation

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Mixed oxide catalysts on the base of ceria demonstrate promising properties in various catalytic reactions, including CO oxidation [1,2]. Additional improvement of low-temperature activity can be achieved by modification with the oxides of the metals with changeable oxidation state, such as CuOx [3] or MnOx [4]. The structure and texture of the catalyst have strong influence on catalytic action and can be influenced using different templates during synthesis. The aim of this work was to track the influence of the template nature on the catalytic action of CeZrOx (CZ, Ce:Zr=4) and CeSnOx, pristine and modified with MnO_x or CuO_x. Three types of templates were used: surfactant etyltrimetylammonium chloride (CTAB), polymer Pluronic 123 and biotemplate wood sawdust (SD). CZ [5], $Ce_{0.9}Sn_{0.1}O_2$ and $Cu/Ce_{0.9}Sn_{0.1}O_2$ [2] prepared without template were used for comparison. Modifiers (MnO_x or CuO_x) were co-precipitated Ce, Zr or Sn oxides; only Mn-CZ-IM catalyst was prepared by subsequent wet impregnation of CZ with Mn acetate solution. Calcination temperature of 500°C was used as providing more defect structure and better catalysts than higher values (e.g. 600°C). CO oxidation was performed in continuous-flow system with fixed-bed of catalyst and pulse feeding of stoichiometric mixture of reagents (2%CO, 1%O_2, balance He). Temperature was raised to 400°C with 50°C increments. The catalysts were investigated by XRD which confirmed fluorite structure of mixed CZ or cerium-tin oxides; temperature-programmed reduction (TPR) and XPS which revealed the presence and the ratios of Ce^{3+}/Ce^{4+}, Mn^{2+}/Mn^{3+} and Cu^{+}/Cu^{2+} in the corresponding systems and the conditions of their reduction. The texture was investigated using N_2 low-temperature adsorption-desorption and SEM (the relevant SEM images are shown in Fig.1, S_{BET} values in Table 1). Energy-dispersive spectroscopy (EDS) during SEM was used to confirm the uniform distribution of the elements on the surface of catalysts. The nominations of the catalysts, the S_{BET} values, Ce^{3+}/Ce^{4+} and $M^{(n-1)+}/M^{n+}$ ratios on the surface (where M=Mn, with n=3, or Cu, with n= 2), found by XPS, the content of M, CO conversion at 200 °C (X_{CO}), and the temperatures of 50% CO conversion (T_{50}) are presented in Table 1.

The comparison with co-precipitated analogs demonstrates that the use of all templates leads to significant increase of S_{BET}. However the addition of MnO_x decreases S_{BET} due to deterioration of complexation. Modification of CZ with Mn or Cu reduces Ce^{3+}/Ce^{4+} ratio. The highest Mn^{2+}/Mn^{3+} ratio was achieved in Mn-CZ-IM catalyst, in which manganese oxides were introduced by post-impregnation.