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CESIUM DOPED Co-Mn-Al MIXED OXIDES IN TOTAL METHANOL OXIDATION

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The more strict requirements for the VOC emission limits makes the finding of highly active and cheap catalysts, substituting conventional rather expensive noble metal catalysts, highly demanding. In this regard, mixed oxides of transition metals are most promising. In our previous study¹, we found that the Co-Mn-Al catalyst prepared from coprecipitated layered double hydroxide (LDH) precursor is very active in total oxidation of ethanol and its activity can be increased by incorporation of potassium.

In this study, we focused on both preparations of Co-Mn-Al precursors by mechanochemical treatment of corresponding nitrates and NH_4HCO_3 , and on modification of oxides by doping with cesium. The prepared precursors and catalysts were studied by TGA/DTA, XRD, UV-Vis, NH_3 -TPD and tested in total oxidation of methanol. The results obtained are summarized in Table 1.

Table 1 Properties of the prepared Co-Mn-Al catalysts and their catalytic activity in total oxidation of methanol

Catalyst	Method	Co+Mn wt. %	Cs wt. %	Cs source	T ₅₀ °C	T ₉₀ °C
110214 ZZ	CP ^a	70.4	0	—	193	215
100621 K	CALC ^b	61.3	0	—	211	234
KL111019C	MCh ^c	63.9	0	—	194	211
KL111019A	MCh ^c	63.9	2.9	NO_3^-	162	183
KL111020	MCh ^c	63.9	2.6	CO_3^{2-}	161	181
KL111019B	MCh ^c	63.9	3.4	CO_3^{2-}	170	192

^acoprecipitation, ^bcalcination of nitrates, ^cmechanochemical procedure, T₅₀, T₉₀ – the temperatures at which 50 and 90 % conversion of methanol was observed under reaction conditions: 0.1 g, 20 ml/min (4.5 % O_2 , 95 % helium, 0.5 % methanol), 12 000 ml g⁻¹ h⁻¹, the heating rate 10 °C min⁻¹.

It was confirmed by XRD measurements that in the patterns of the precursors prepared mechanochemically, a phase corresponding to hydrotalcite-like LDH was observed with characteristic 2 θ value at 11.5. Other phase (rhodochrosite MnCO_3) was also detected. After calcination at 500 °C, Co_3O_4 with spinel structure was found, as well as Mn_2O_3 and Mn_3O_4 phase.

The most active catalysts were Cs/Co-Mn-Al oxides

prepared mechanochemically (Fig. 1). The found T₅₀ temperatures were around 165 °C and the T₉₀ temperatures around 186 °C. The non-doped catalysts showed T₅₀ > 190 °C and T₉₀ > 210 °C. The non-modified catalyst prepared mechanochemically exhibited similar activity as the calcined coprecipitated LDH. The least active catalyst was that prepared by calcination of the metal nitrates.

Thus, mechanochemical preparation leads to precursors with LDH-like structure and therefore, the procedure is a good alternative to coprecipitation. Cesium in amount ~ 2.6 wt.% increases catalytic activity in methanol oxidation.

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References:

- ¹ Jiráťová, K.; Mikulová, J.; Klempa, J.; Grygar, T.; Bastl, Z.; Kovanda, F.: Appl. Catal. A 2009, 361, 106-116.

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