COMPARISON OF STRUCTURE OF HYDROTALCITE MATERIALS PREPARED BY DIFFERENT SYNTHESIS METHODS

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INTRODUCTION

Hydrotalcites, due to adjustable composition[1] are widely examined as a catalyst for processes such as dry reforming of methane[2],[3], carbon dioxide hydrogenation[4], selective catalytic reduction[5]. Size of an active center is of importance in catalytic processed, that's why reduction of elemental cell determines uniform distribution of the material, increases the active surface and prevents from sintering[6].

EXPERIMENTS

Samples containing Mg and Al in molar ratio 3:1 and Ni, Mg and Al in molar ratio 1:2:1 subsequently were prepared via three alternative methods, and later were characterised with XRD (Panalytical Empyrean diffractometer, equipped with Cu anode as a source of X-ray radiation, the patterns were recorded over the 2θ range 3 to 90° in steps of 0.013°).

Coprecipitation at low supersaturation

Preparation was carried according to methodology proposed by Cavani[1] with the constant pH in range 8.5-9.

Urea hydrolysis

Solution containing metal nitrates precursors was mixed with urea solution, in molar ratio of urea to to nitrate ions of 4:1, basing on the methodology by Zeng et al.[7].

Solution combustion synthesis

Solutions with nitrate precursors in chosen molar ratios were prepared with stoichiometric amount of urea, in accordance to form oxides, and 0.2 g Na_2CO_3 per 1 g of solid mixture. Such solution was heated to 50℃ for 1 hour under constant stirring. Then, solution was combusted at 450℃. Powder obtained from each combustion was cooled down and put to the 0.1M solution of Na_2CO_3 for 5 minutes, under dynamic stirring. After that time, samples were washed with distilled water and dried in 70℃ as it was suggested by Davila et al.[8].

RESULTS AND DISCUSSION

Different preparation methods of hydrotalcites do result in different size of the crystallites and presence of additional phases. Both size and phase composition may be influenced with the temperature of the process, pH and time. Pure hydrotalcite phase was obtained only with urea hydrolysis, also, this method resulted in the smallest size of crystallites.

Table 1 **Comparison of the composition and crystallite size of the prepared samples**

Method	Sample	XRD composition	Crystallite size of hydrotalcite [nm]
Coprecipitation at low supersaturation	Mg-Al	Hydrotalcite, Al_2O_3, $CaCO_3$	7.04
	Ni-Mg-Al	Hydrotalcite, Al_2O_3, $CaCO_3$	19.97
Urea hydrolysis	Mg-Al	Hydrotalcite	7.25
	Ni-Mg-Al	Hydrotalcite	15.68
Solution combustion synthesis	Mg-Al	Hydrotalcite, Al_2O_3,	15.11
	Ni-Mg-Al	Hydrotalcite, $Mg_{0.4}Ni_{0.6}O$	21.19

CONCLUSION

According to this research, the most precise method of hydrotalcite preparation is urea hydrolysis, which provides a highly crystalline, one-phase product with small size of the crystallites. Solution combustion synthesis leads to formation of relatively big crystallites and additional oxide phases. Coprecipitation gave satisfactory size of the crystallite in Mg-Al sample, however additional phases were present.

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