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Relations between synthesis and microstructural properties of copper/zinc hydroxycarbonates

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Abstract:

Cu/Zn Hydroxycarbonates obtained by co-precipitation of Cu^{2+} and Zn^{2+} with Na_2CO_3 have been investigated regarding phase formation and thermal decomposition in two series with varying Cu/Zn ratios prepared according to the decreasing pH and constant pH method. Hydrozincite, aurichalcite and (zincian)-malachite were found to form at differing Cu/Zn ratios for both series. For the constant pH preparation the Cu/Zn ratio in zincian-malachite was close to the nominal values whereas excess values were found for the decreasing pH samples. The degree of crystallinity as well as the thermal decomposition temperatures were lower for the constant pH series. All samples containing aurichalcite revealed an unexpected decomposition step at high temperatures evolving exclusively CO_2. The differences in composition and microstucture were traced back to the different pathways of solid formation for the two preparation methods. Substantial changes were observed during the post-precipitation processes of ageing and washing. The effects were studied in detail on samples with a cation ratio of Cu/Zn 70:30 mol %. Ageing of the precipitates in their own solutions is accompanied by a spontaneous crystallization of the initially amorphous solids. The decreasing pH sample develops from a hydroxy-rich material comprising basic copper nitrate (gerhardtite) as an intermediate. Only small changes in the chemistry of the samples were detected for the constant pH precipitation. The findings are summarised into a scheme of solid formation processes that explains the phenomenon of a "chemical memory" of the precipitates when they are converted into Cu/ZnO model catalysts.