

## SYNTHESIS OF MULTIMETALLIC LAYERED MATERIALS AND CHARACTERIZATION OF THE CALCINATION PRODUCTS

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The layered double hydroxides (LDHs), also known as hydrotalcite-like materials or as anionic (more properly speaking, anion exchanging) clays, are a large group of natural and synthetic materials readily produced when suitable mixtures of metal salts are exposed to base. These materials have the general formula  $[MII_{1-x}MIII_x(OH)_2] (A^{n-})_x/n mH_2O$ . Metal cations are located in coplanar octahedra  $[M(OH)_6]$  sharing edges and forming  $M(OH)_2$  layers with the brucite (cadmium iodide-like) structure. Partial substitution of the divalent cations by trivalent ones gives rise to a positive charge in the layers, balanced by anions ( $A^{n-}$ ) located between the hydroxylated layers, where water molecules also exist. The anions and the water molecules are hosted together in the interlayer gallery. The nature of the cations in the brucite-like sheets (which is not restricted to +2/+3 combinations) and the interlayer anions together with the coefficient  $x$  value may be varied in a broad range, giving rise to a large class of isostructural materials [1,2]. Decomposition of LDHs at moderate temperatures leads to mixed oxides displaying high specific surface areas, good interdispersion of the metal cations, and reactivity, which are of interest because of their catalytic applications [3].

In this study we used the co-precipitation method at constant pH for the synthesis of layered double hydroxides containing  $Zn^{2+}$ ,  $Ni^{2+}$  and  $Al^{3+}$  with cationic ratio 1-4 and carbonate, nitrate or citrate as charge balancing anions. The samples have been characterized by elemental chemical analysis, powder X-ray diffraction, and FT-IR spectroscopy. Their thermal stability has been assessed by thermogravimetric and differential thermal analyses (TG-DTA) and high temperature synchrotron powder X-ray diffraction (PXRD). Investigations on the high temperature phase transitions were carried out at the synchrotron beamline B2 at HASYLAB (DESY, Hamburg). The diffractometer

was equipped with capillary furnace (STOE) and the on-site readable image-plate detector OBI. Samples mounted into quartz capillaries of diameter 0.3 mm were heated and cooled at the temperature range from RT to 800°C. The wavelength was 0.65131 Å.

Thermal decomposition LDHs containing carbonate or nitrate in the interlayer starts at 200–300°C and proceeds through intermediate amorphous phases. The final decomposition product is a mixture of  $MIIIMIII_2O_4$  spinel and  $MIIO$  oxide because a  $MII/MIII$  ratio larger than 1 is required to form a stable hydrotalcite. The thermal stability of these materials is strongly related with several factors, such as both the nature of cations and the cationic ratio in the brucite-like sheets, the nature of the compensating anions and the crystallinity degree of LDH lattice.

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