Applied Clay Science 48 (2010) 542–546

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

Note

Incorporation of cerium ions by sonication in Ni–Mg–Al layered double hydroxides

Octávio R. Macedo Neto [a], Nielson F.P. Ribeiro [b], Carlos A.C. Perez [b], Martin Schmal [b], Mariana M.V.M. Souza [a,*]

[a] Escola de Química — Universidade Federal do Rio de Janeiro (UFRJ), Centro de Tecnologia, Bloco E, sala 206, CEP 21941-909, Rio de Janeiro, RJ, Brazil
[b] NUCAT/PEQ/COPPE/UFRJ, Centro de Tecnologia, Bloco G, sala 128, CEP 21945-970, Rio de Janeiro, RJ, Brazil

ARTICLE INFO

Article history:
Received 5 August 2009
Received in revised form 19 February 2010
Accepted 23 February 2010
Available online 1 March 2010

Keywords:
Layered double hydroxide
Hydrotalcite-like compounds
Mixed oxides
Coprecipitation
Sonication

ABSTRACT

The incorporation of Ce in Ni–Mg–Al layered double hydroxides (LDHs) was studied by coprecipitation and under sonication. Mixed oxides were obtained by calcination. X-ray diffraction (XRD) patterns of the as-synthesized samples showed formation of well-crystallized LDHs and CeO_2. Rietveld refinement was employed to calculate the content of cerium that was incorporated in the LDH structure. Sonication increased the incorporation of Ce in the structure. The specific surface areas of cerium-containing LDHs prepared by the conventional method were similar to that of free-Ce sample. Synthesis under sonication significantly increased the specific surface area. After calcination both specific surface areas and pore volumes increased compared to the corresponding LDH.

1. Introduction

Layered double hydroxides (LDHs), or hydrotalcite-like compounds, have many industrial applications as catalysts, catalyst precursors, adsorbents and ion exchangers (Cavani et al., 1991). These compounds may be applied in catalytic fields as such or after thermal decomposition. Heating induces dehydration, dehydroxylation and loss of compensating anions, forming mixed oxides with basic properties, high specific surface area, homogeneous dispersion of the metal ions and a better resistance to sintering than the corresponding supported catalysts (Tichit et al., 1995; Casenave et al., 2001).

The acid–base and/or redox properties of LDHs and their derived mixed oxides depend on the chemical composition, preparation methods and treatment conditions. Ni–Mg–Al mixed oxides derived from LDHs have been extensively studied as catalysts in different reactions, such as hydrogenation of acetonitrile (Lebedeva et al., 1999), partial oxidation of methane and light paraffins (Basile et al., 1998; Schulze et al., 2001), reforming of methane (Tsyganok et al., 2003; Fonseca and Assaf, 2005) and aqueous-phase reforming of ethanol (Cruz et al., 2008). Cerium oxide (CeO_2) is widely used as a promoter in various redox reactions due to its reducibility and high oxygen storage capacity, besides preventing the metal sintering and decreasing coke formation on nickel catalysts (Yao and Yao, 1984; Trimm, 1997).

The introduction of Ce ions in the hydroxide layer of LDHs is difficult because of its large ionic radius, when compared to Mg^{2+} and Al^{3+}. Das et al. (2006) incorporated cerium ions in Mg–Al LDH. Lucrédio et al. (2007) prepared Ni–Mg–Al LDHs and used anion exchange of the Ce-EDTA complex to incorporate cerium ions. Only a part of the cerium chelate was introduced in the interlayer space. Daza et al. (2008) also used the method with Ce-EDTA complexes, showing, however, that the complex was not intercalated.

Conventionally, LDHs are synthesized by coprecipitation from metal nitrates at a fixed pH under stirring, followed by a long aging and/or hydrothermal treatment to improve the crystallinity. The substitution of the conventional aging step by microwave irradiation has been described in the literature, with substantial reduction of the crystallization time (Fetter et al., 1997; Rivera et al., 2006). Kooli et al. (1997) described the use of ultrasound to intercalate vanadate ions in Mg–Al LDH. Climent et al. (2004) have performed an extensive study of the application of ultrasound irradiation during the coprecipitation step for LDH synthesis. Mg–Al LDHs prepared under sonication presented an increase in the specific surface area and the number of defect sites in the solid, leading to a higher basicity. However, to our knowledge, no one reported the use of ultrasound to incorporate large cations, such as cerium ions, into the hydroxide layer of LDHs.

In the present study, we investigated the preparation of LDHs containing Ni, Mg, Al and Ce in the hydroxide layer by conventional coprecipitation and under sonication.

2. Experimental

2.1. Synthesis

The LDHs were prepared by coprecipitation from aqueous solutions at room temperature, always containing 20 mass% of NiO. 200 mL of solution

* Corresponding author. Tel.: +55 21 25627598; fax: +55 21 25627596.
E-mail address: mmattos@eq.ufrj.br (M.M.V.M. Souza).

0169-1317/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.clay.2010.02.015