

Research paper

Rapid preparation of nano hexagonal-shaped hydrocalumite via one-pot mechanochemistry method



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ABSTRACT

Hexagonal-shaped hydrocalumite (Ca–Al layered double hydroxides) was successfully synthesized by one-step mechanochemistry method within only 30 min. The product was characterized by X-ray diffraction, Fourier Transform Infrared spectroscopy, field emission scanning electron microscopy, energy dispersive spectroscopy, elemental mapping analysis, transmission electron microscopy, X-ray fluorescence, and thermogravimetric analysis. The results showed that the formation of a well-defined hydrocalumite and the microstructural evolution were strongly dependent on the milling time. From XRD profiles, a well-crystallized hydrocalumite with higher peaks intensity was formed after 60 min of milling, while the sharpness and intensity of LDH-derived XRD peaks decreased as milling time increased. Microscopic analysis showed that the product had hexagonal layered structure with lateral dimension of 100 to 200 nm. In addition, the thermal decomposition of the product is reported in details. This study offers a low-cost approach to synthesis of hydrocalumite owing to simplicity and extremely short processing time.

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1. Introduction

Hydrocalumite (HC) are anionic clays with the chemical formula of $\text{Ca}_{1-x}\text{Al}_x(\text{OH})_2 \text{A}^{n-}_x$, where A^{n-} represents the charge balancing anions such as CO_3^{2-} , Cl^- , SO_4^{2-} , OH^- , $\text{Al}(\text{OH})_4^-$ or NO_3^- (Sampieri et al., 2011). HC also belong to the family of layered double hydroxides (LDH) and differ from Hydrotalcites in that their main layer has well-ordered Ca/Al distribution in the hydroxide layers due to the large cationic radius of Ca^{2+} in comparison to Mg^{2+} , while the water and anions are placed in the interlayer spaces (Zheng et al., 2015). Some anions are preferentially intercalated to LDH over others, thus it may be used for targeted anion removal in terms of environmental protection (Tóth et al., 2014). The carbonate ion owing to its high affinity is the most preferably intercalated anion and it is easily trapped in LDH (Sanderson et al., 2013). The presence of the carbonate in the interlayer region significantly affects the chemical reactivity of the LDH so that Yoon et al. (2014) reported calcined layered double hydroxide as an effective

absorbent for chloride ions in the cement matrix. Linares et al. (2016) also demonstrated that carbonated hydrocalumite can be used as a promising antacids using a synthetic gastric juice compared with commercial ones. In general, HC has other industrial and environmental applications like mopping up of oil from oil–water mixtures (Dutta, and Pramanik, 2013), the removal of environmental hazards in acid mine drainage (Li et al., 2008), and a mechanism for the disposal of radioactive wastes (Das et al., 2003).

In past decade, many different chemical methods have been applied for fabrication of anionic clays. These methods include co-precipitation at different pH for the removal of heavy metal (Kovanda et al., 2003), hydrolysis (Hibino and Ohya, 2009), sol-gel (Paredes et al., 2006), Microwave-assisted structure reconstruction which help the crystalline growth of Hydrotalcite (Benito et al., 2008), hydrothermal (Labajos et al., 1992), sonication (Climent et al., 2004), which have drawbacks in environmental pollution and energy consumption. Among various processes, mechanical alloying (MA) as a solvent-free process has been attracted special attention as an alternative method to prepare commercially nanostructured materials with proper structural characteristics due to its simplicity and versatility (Fahami et al. 2011), however the information regarding preparation of clay materials through this process is still limited. It has been reported some new kinds of LDH such as Li-Al (French et al., 2010), and Ca-Sn (Ferencz et al., 2014) LDH by this

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method. Recently, Ferencz et al. (2015) reported their synthesis of Ca–Al–CO₃–LDH, using calcium hydroxide and aluminum hydroxide as raw materials, by a two-step MA process with some impurities (15.6 wt.% Katoite) in the final product. Note that Ca–Al–LDH was initially formed as a metastable compound in cement and converted to cubic Katoite (Ca₃Al₂(OH)₁₂) as a stable calcium aluminate hydrate during concrete curing (Wang et al., 2015). Qu et al. (2016) was also reported two steps mechanochemically induced Ca–Al–CO₃–LDH. Their initial results demonstrated that the milling of hydroxides only gave Katoite but no LDH phase. Thus, they added calcium chloride and calcium carbonate to their mixture and found that the addition of a third phase to Ca and Al hydroxides allowed the easy formation of stable Ca–Al–LDH phases.

Although few studies have examined the synthesis of various HC and the way in which they interact with various anions, the synthesis and characterization of Ca–Al–LDH via one-step mechanochemistry method has not been studied so far. So the present research was aimed to prepare and characterize Ca–Al–LDH through XRD, FT-IR, FESEM, elemental mapping analysis, EDS, XRF, TEM, and DTGA techniques.

2. Materials and methods

Calcium chloride (99%, Sigma-Aldrich), Aluminum chloride (99%, ACROS), and Sodium hydroxide pellets (97%, ACROS), were purchased and used without further purification. To prepare Ca–Al–LDH, the desired amounts of CaCl₂, AlCl₃, and NaOH were milled at different milling times (30, 60, 180, and 300 min) in a high energy ball mill (8000 M Mixer/Mill) utilizing hardened chromium steel vials (vol. 65 ml) under air atmosphere. The weight ratio of ball-to-powder (BPR) was 10:1. The powders were washed three times with deionized water (20 ml) on a paper filter, and then were dried under oven at 60 °C for 1 h. The samples milled at different milling time were labeled as S1, S2, S3, and S4, respectively. The final composition was adjusted according to the following reaction (R1):



Composition of the reaction products between chemicals was determined using Bruker D8 Advance ECO using Cu–K_α radiation ($\lambda \approx 1.54 \text{ \AA}$). The samples were scanned from 5 to 70° 2 θ at a step size of 0.03°. The XRD profiles were compared to standards compiled by the Joint Committee on Powder Diffraction and Standards (JCPDS), which involved card #31-0245 for Hydrocalumite. Fourier transform infrared spectroscopy (FT-IR, Bruker Tensor 27, USA) was used to determine the functional groups of products. All spectra were recorded at ambient temperature in the range 4000–600 cm⁻¹. Morphological evaluation of Ca–Al–LDH was carried out using FE–SEM (FEI Helios Nanolab 400 SEM) and TEM (JEOL JEM 1200 EXII). EDS analysis attached to the FESEM was used to determine the elemental composition. Thermal analysis was executed with a TGA (TA Q50 Thermogravimetric Analyzer) during heating in an argon atmosphere at a rate of 10 °C/min from room temperature to 800 °C.

3. Results and discussions

3.1. XRD analysis

Fig. 1 shows the XRD profiles of samples as a function of milling time (30, 60, 180, and 300). Based on Fig. 1, the main product of 30 min milling was in excellent agreement with the standard HC. With increasing of milling time up to 60 min, a well-crystallized Ca–Al–LDH with higher peaks intensity of HC was formed. Ca–Al–LDH displayed peaks corresponding to (002), (004), (112), (020), and ($\bar{3}$ 16) crystal planes. This trend continued for the 180 min milled specimen with slight decrease in peaks intensity of HC. Further increase in the milling time up to 300 min led to further decrease of the dominant peaks particularly for

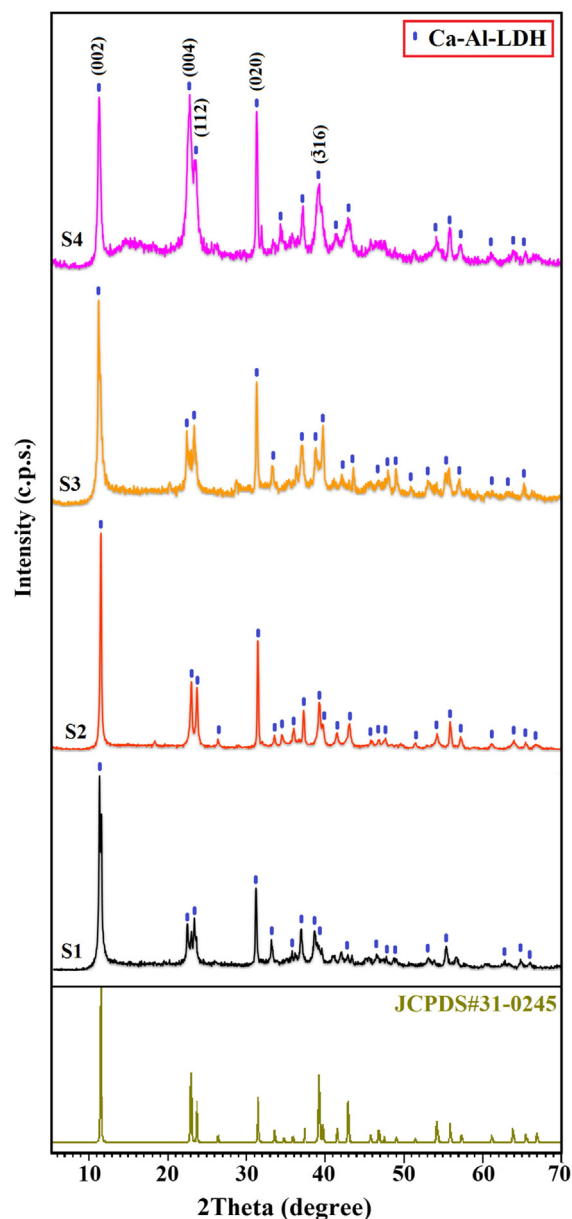


Fig. 1. XRD patterns of specimens (Ca–Al–LDH) milled at different milling time (S1 = 30, S2 = 60, S3 = 180, and S4 = 300).

(002) plane. As is evident from Fig. 2, with increasing milling time, the LDH-derived XRD peaks decreased in both sharpness as well as intensity, and shift toward larger d spacings (i.e. lower 2 θ). This effects could be

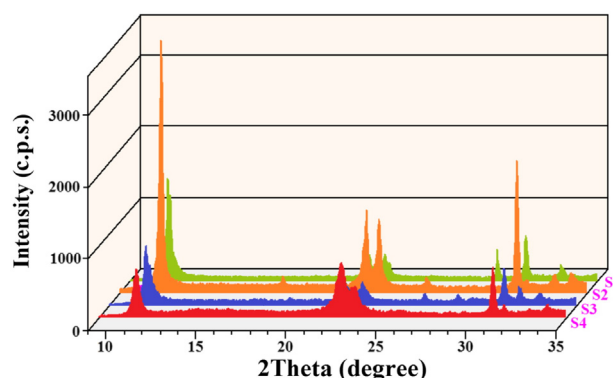


Fig. 2. The magnified 3D XRD profiles (2 θ = 5–35) of milled samples (S1, S2, S3, and S4).

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