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Recycling of lime mud and fly ash for fabrication of anorthite ceramic at low sintering temperature

Juan Qin, Chong Cui*, Xiaoyu Cui, Ahmad Hussain, Chuanmeng Yang, Shihai Yang

School of Materials Science and Engineering, Nanjing University of Science and Technology, Nanjing 210094, China

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Abstract

Lime mud is a kind of waste generated during causticization reaction of an alkali recycling process in paper industry. Lime mud and fly ash were reused as raw materials to fabricate anorthite ceramics through solid state reactions. Both sintering temperature and lime mud content influenced the crystalline phases in the prepared ceramics. Anorthite was the major phase in all samples (samples L36, L40, L50 and L60) and it was prominent in sample L36 (containing 36 wt% lime mud). The results also showed that anorthite ceramic can be synthesized at low sintering temperature (1100 °C). Gehlenite and wollastonite were formed in the samples possessing higher calcium (above 40 wt% lime mud) or at lower sintering temperatures. Bulk density, water absorption and compressive strength were measured. These ceramics were of light weight and had high water absorption. Recycling of lime mud and fly ash as raw materials of anorthite ceramic is a feasible approach to solve the solid wastes.

Keywords: Lime mud; Anorthite ceramic; Crystalline phase; Sintering temperature

1. Introduction

Lime mud is an industrial waste obtained during causticization reaction of an alkali recycling process in paper industry, as described in Eq. (1) [1]. The major chemical component of lime mud is calcium carbonate ($CaCO_3$). It was estimated that about 0.5 tons of lime mud can be produced from 1 ton of pulp [2,3]. China is the second largest paper producer country all over the world and about 50 million tons of papers were generated per year [4]. In 2011, the lime mud production of China had reached to 10 million tons and it is increasing consistently with the growing demand for paper [2].

$$Na_2CO_3 + CaO + H_2O = 2NaOH + CaCO_3 \quad (1)$$

Due to high alkalinity (pH 9.7–13.5) and the existence of alkali metal ions (e.g. Cr, Mn and Fe) [5–9], lime mud is classified as toxic industrial waste. A proper treatment is required for lime mud. Landfill, as an ordinary method to dispose solid wastes, may cause harm to human, animals, groundwater, and plants [10,11] and lead to a waste of recyclable resources [12–15]. Hence, landfill is unsuitable for the disposal of lime mud [13,14]. To protect environment, the paper industry has to place more emphasis on the cost-effective resourcization of lime mud.

Attentions are paid to enhance novel strategies and expand the available applications for better disposal of lime mud. Firstly, land application is one of the limited methods which is available to manage lime mud. As an alkaline material, lime mud can be used to control acid mine drainage and ameliorate acid soils [16]. Secondly, investigations of precipitation or immobilization of heavy metals have indicated that lime mud has a high removal efficiency during a waste water treatment process [2,17–19]. Thirdly, it was found that lime mud can be used as building materials (e.g. bricks, cements and concretes) [20,21]. However, high moisture content (about 39–60%) and the existence of silica in lime mud have created many difficulties in handling [22–24]. Therefore, more benign and efficient applications of lime mud should be put forward.

Anorthite ($CaAl_2Si_2O_8$) is a kind of plagioclase feldspar. Theoretically, its chemical composition is 43.2% SiO_2, 36.6% Al_2O_3, and 20.2% CaO on the basis of weight and its density is 2.76 g/cm^3. In heat exchangers, biomedicine, electronics and other

*Corresponding author.
E-mail address: cuichong@njust.edu.cn (C. Cui).

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industrial fields, anorthite is regarded as a crucial component of substrate materials [25–29]. However, anorthite cannot be mined naturally in mass; as a result it is hard to meet the demand of industry. Although lime mud and fly ash are industrial wastes, their chemical composition are relatively stable, as shown in Table 1 [30,31]. The stability of composition of lime mud and fly ash is enough to reuse them as raw materials for industrial applications. And taking their chemical composition into consideration, a possible effective approach to reuse these wastes is to produce anorthite ceramic. Sutcu et al. [32,33] designed a kind of porous anorthite ceramic from paper processing of residues and clay of various sources (i.e. enriched clay, commercial clay and fireclay) at 1200–1400 °C. Ke et al. [34] and Cheng et al. [35] reported the fabrication of anorthite-based porcelain by using ball clay, quartz, alumina, feldspar and three different sources of CaO (i.e. dolomite, wollastonite and calcite) sintered at 1200–1230 °C. Kurama and Ozel [36] fabricated an anorthite ceramic by using four different sources of CaO (i.e. $Ca(OH)_2$, $CaCO_3$, marble powder and gypsum mold waste) sintered at 1000–1300 °C. Reusing of lime mud and fly ash as raw materials of anorthite synthesis will not only protect the environment but also lead to a new low-cost synthesis technology of anorthite.

The purpose of this research was to fabricate anorthite ceramic by using lime mud and fly ash. Emphasis was put on the investigation of crystalline phases in solid state reactions. Physicochemical properties such as bulk density, water absorption, compressive strength and available lime content were characterized. These achieved results may contribute to the transformation of other solid wastes like lime mud into products with the highest possible market value.

2. Experimental procedure

2.1. Raw materials

Lime mud was collected from the alkali recycling process of a papermaking company in Xuzhou, China. Fly ash was obtained from a power plant in Nanjing, China. Major chemical compositions of materials were measured according to BS EN 196-2 [37], as presented in Tables 2 and 3. The moisture content of lime mud is 45.6%, which was calculated from Eq. (2). X-ray diffraction (XRD, Model D8 advance) with CuK_α radiation ($\lambda = 1.542$ Å) at 40 kV was used to analyze the crystalline phases of raw materials. Calcium carbonate ($CaCO_3$) is present as the main crystalline phase of lime mud, as shown in Fig. 1(a). Lime mud was used as a source of calcium and a pore-making material. Quartz (SiO_2), mullite ($Al_6Si_2O_{13}$) and glassy phase are present as the main crystalline phases of fly ash, which can be observed in Fig. 1(b).

$$\text{Moisture content} = 100\%(m - m_0)/m \quad (2)$$

Table 1
Change range of chemical composition of lime mud and fly ash (wt%).

Material	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	L.O.I.
Lime mud	3.4–11.0	0.5–1.4	0.2–1.2	44.4–52.0	0.6–3.4	31.5–43.5
Fly ash	33.9–59.7	16.5–35.4	1.5–19.7	0.8–10.4	0.7–1.9	1.2–33.6

Table 2
Chemical composition of raw materials (wt%).

Raw material	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	L.O.I.
Lime mud	7.01	0.94	0.47	45.79	3.39	37.16
Fly ash	53.47	30.48	4.73	2.94	0.95	4.26

Table 3
The nomenclature and composition of the investigated samples (wt%).

Sample no.	Raw material		Chemical composition					
	Lime mud	Fly ash	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	L.O.I.
L36	36	64	42.55	23.54	3.73	13.01	1.52	11.99
L40	40	60	41.07	22.60	3.59	14.37	1.60	13.04
L50	50	50	37.06	20.05	3.23	18.07	1.81	15.88
L60	60	40	32.55	17.18	2.81	22.23	2.05	19.07

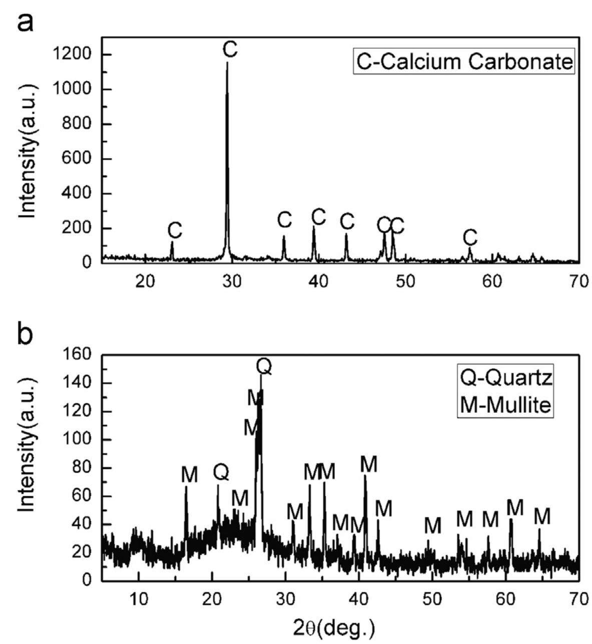

Fig. 1. XRD patterns of raw materials. (a) Lime mud and (b) fly ash.

where m and m_0 (in grams) are the weight of sample before and after drying, respectively.

2.2. Sample preparation

Phase diagram of $CaO–SiO_2–Al_2O_3$ represents a broad anorthite forming area in a silica-rich region [38]. Lime mud and fly ash were reused as raw materials of anorthite ceramics. The nomenclature and composition of lime mud–fly ash mixtures are presented in Table 3. Raw materials were mixed directly in a laboratory mortar mixer (Model JJ-5) for 10 min. The mixtures were pressed into 40 mm × 40 mm × 160 mm cuboids by a hydraulic universal tester (Model WE-300B) under 10 MPa. The green compacts were dried in air at room temperature for 24 h and