



Simple process for synthesis of layered sodium silicates using rice husk ash as silica source



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ABSTRACT

β , δ , and α phases of layered sodium silicates were synthesized using waste rice husk ash (RHA) as the raw material by a simple sintering process. X-ray diffraction (XRD) measurements revealed a phase transformation from β through δ to α as the reaction temperature and time of synthesis was increased. Raman studies confirmed the synthesized products have a layered SiO_4 tetrahedral structure. Further, a Si–O–Si (Na) band bond force constant sequence for the three phases was speculated ($\delta > \beta > \alpha$) that agrees with the ion binding sequence of the phases. Scanning electron microscopy (SEM) elucidated the typical micromorphology of the synthesized products, which was structurally similar to that of RHA. Optimal synthesis conditions produced layered sodium silicates that exhibited Ca^{2+} and Mg^{2+} binding capacities of 406 and 453 mg/g, respectively. This method can therefore be used to simply and cheaply convert agricultural waste into valuable products.

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1. Introduction

Layered sodium silicates have a polymeric layered crystalline structure with complicated polymorphism [1]. At least five phases have been acknowledged, though the common framework phases are δ , α , and β [2]. Their foundations are built from the condensation of silicate tetrahedra into a single sheet, with interlamination achieved through sodium cations. This makes them structurally similar to the natural mineral bentonite, and they therefore have the potential for application as a grinding aid, adsorbent, dispersant, or catalyst carrier [3]. Furthermore, their high sodium content gives them a pH buffering ability, strong absorption, and excellent ion binding capacity [4], providing a suitable alternative for phosphorus-based detergents that would be less polluting. Layered sodium silicates are therefore of great interest to both academia and industry.

Commercial layered sodium silicates are typically mixtures of δ (~90%), α (~10%), and β (~10%) phases [2] prepared using sodium silicate solutions as raw materials. These solutions are thermally spray dried to produce granules that are then sintered at high temperature under different conditions [4–6]. However, researchers have found that this conventional approach is not

suitable for producing a high content of single-phase product [2,5]. To address this, Falamaki incorporated an appropriate quantity of pure-phase seeds with a specific particle size to induce the direct formation of a particular phase and suppress the formation of other phases [7]. Kahlenberg et al. have also used the pretreatment of raw materials (slowly drying liquid sodium silicate at 75 °C for eight weeks), followed by sintering for 20 h to obtain pure-phase products [8]. However, the high cost of the raw materials, prolonged production cycle, and wide range of control variables in these processes greatly decreases the industrial value of the products produced, which has limited their application. Researchers have therefore concentrated on finding an economically viable approach for the synthesis of layered sodium silicates based on a low-cost source and simple process. Sun et al. used zirconium oxychloride waste effluent for the preparation of layered sodium silicates to reduce the production cost associated with the raw materials [9]. Yang et al. synthesized layered sodium silicates from sodium silicate solutions using the mirabilite method, which provides a new resource for their synthesis [10]. The Korea Research Institute has also applied for a patent on improved technology for layered sodium silicate production [11].

Rice husk (RH) is a common agricultural residue, with a global annual output of ~1.2 billion tons [12,13], but contains 10–21% silicon in a hydrated amorphous form. This amorphous silica is a desirable raw material for the preparation of silica and silicates,

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making RH a potential biomass resource [14–17]. However, most RH is currently discarded as waste because it lacks any direct value for commercial application. This not only wastes a valuable silica resource, but can also create environmental issues due to its extremely slow decomposition rate [18]. There is therefore an urgent need to find a way of using RH more effectively.

Incinerating RH under controlled conditions can enrich its amorphous silica content, with the rice husk ash (RHA) obtained being one of the most silica-rich raw materials [19–22]. In addition to being cheap and abundant, RHA has many other advantages such as high reactivity, large surface area, and a superfine size. The use of RHA as a silica source for the preparation of economical products [23–28] has been explored, but there are few reports pertaining to the preparation of layered sodium silicates from RHA.

In this paper, we report the synthesis of β -, δ -, and α -phase layered sodium silicates from RHA using a simple sintering process. The effect of changing the reaction temperature and time is investigated, and the structure and morphology of the silicates produced are characterized and their ion binding capacities are determined. This study is advantageous as it converts agricultural waste into valuable products using a low cost and simple process.

2. Experimental

2.1. Materials and reagents

RH was collected locally, from which RHA was prepared in-house by pyrolysis. Analytically pure NaOH (Kelong Chemical, Sichuan, Chengdu) was used in the process.

2.2. Preparation of layered sodium silicates

The RH was washed with distilled water to remove any dust or impurities. This was incinerated at 600 °C for 120 min to obtain RHA, which was subsequently mixed with a 1 M NaOH solution (molar ratio of Si/Na = 1) using a mechanical stirrer. The mixture was then poured into a ceramic crucible and sintered at 680–800 °C for 5–210 min. The samples were cooled to room temperature as the oven cooled naturally. The experimental

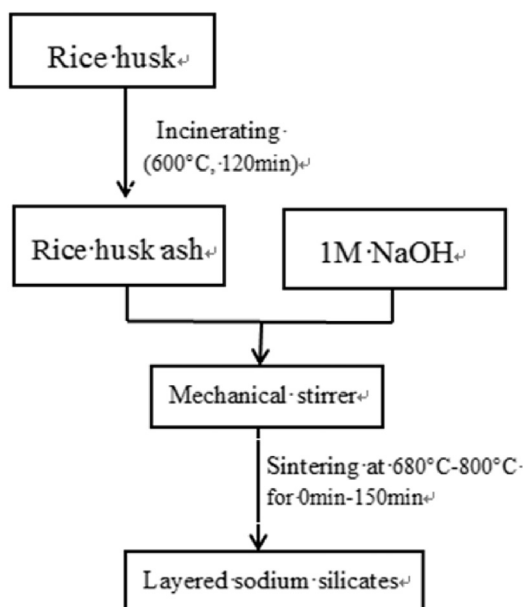
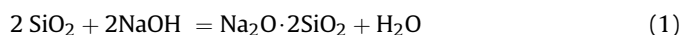


Fig. 1. Schematic of the experimental procedure used.

process is shown schematically in Fig. 1. The molar ratio of Si/Na in each sample was established using the following reaction:



2.3. Characterization

The chemical composition of the RHA was analyzed by X-ray fluorescence (XRF-1800). The crystal structures of the synthesized samples were determined by X-ray powder diffraction (XRD) (Rigaku DmaxIIIc diffractometer) with Ni-filtered Cu K α radiation. Raman spectra were obtained using a Renishaw inVia Raman instrument. Morphological characterization was performed by scanning electron microscopy (SEM, model: Quanta 250 FEG, USA). N₂ adsorption/desorption measurements were performed using a QUADRASORB SI surface area and pore size analyzer. Transmission electron microscopy (TEM) images were obtained with a JYTO11–1996 instrument. The Ca²⁺ and Mg²⁺ binding capacities were determined by ethylenediaminetetraacetic acid (EDTA) titration.

3. Results and discussion

The chemical composition of the RHA shown in Table 1 indicates that the major ingredient is SiO₂, along with small amounts of Al₂O₃, Fe₂O₃, CaO, MgO, TiO₂, K₂O, and Na₂O. The broad intense peak at $2\theta = 22^\circ$ in Fig. 2 indicates that the RHA has a high content of amorphous silica, with the absence of any sharp peaks confirming that the solid has no ordered crystalline structure. As the RHA clearly contains pure silica in an amorphous state with few impurities, it is a viable raw material for the preparation of layered sodium silicates.

The effects of reaction temperature and time on the synthesis of layered sodium silicate were investigated by XRD (Fig. 3). The phase contents of the synthesized samples were determined based on Lucas's study on quantitative phase analysis [2,5]. Based on Fig. 3a and Table 2, the reaction temperature for phase evolution can be divided into two sections. In the interval from 680 °C to 720 °C, a high content of β phase is obtained at 680 °C, but this decreases with an increase in δ phase up to 720 °C. In the interval from 720 °C to 800 °C the δ phase has an analogous evolution to the β phase, with the α phase tending to form at higher temperatures until its phase content reaches a maximum at the extinction of the δ phase. The effect of reaction time on the synthesis is shown in Fig. 3b and Table 3. Here, a similar phase transformation is observed, with an evolution from β to δ up to 120 min and stable α phase starting to be detected at 150 min.

According to the XRD patterns, it is reasonable to speculate that β -Na₂Si₂O₅ (JCPDS: 22-1123) can be obtained as an initial product at low temperature and with a short reaction time, while higher temperatures and longer times favor the formation of stable α -Na₂Si₂O₅ (JCPDS: 22-1397). The transitional δ -Na₂Si₂O₅ (JCPDS: 22-1396) can only be prepared within a narrow range of conditions, suggesting the following phase transformation sequence occurs during the process: $\beta \rightarrow \delta \rightarrow \alpha$ (Tables S1, S2, and S3). This is consistent with the phase evolution process reported for the synthesis of layered sodium silicates from sodium silicate solutions [6].

Table 1
Chemical composition of RHA (%).

SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MgO	CaO	TiO ₂	K ₂ O	Na ₂ O
98.64	0.38	0.238	0.218	0.249	0.03	0.125	0.12

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