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Synthesis of ordered mesoporous alumina with high thermal stability using aluminum nitrate as precursor



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ABSTRACT

By using the inexpensive aluminum nitrate as precursor, a highly ordered mesoporous alumina with high thermal stability has been synthesized successfully via an evaporation-induced self-assembly pathway associated with solvothermal pre-hydrolysis process. The resultant mesoporous alumina maintains the ordered hexagonal mesostructure, narrow pore-size distribution, high BET surface area and large pore volume even after thermal treatment at 900 °C.

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1. Introduction

Ordered mesoporous alumina (MA) materials with tunable structures, high surface areas and large pore volumes have potential applications as catalysts or catalyst supports [1]. A series of ordered MAs have been successfully synthesized through the sol-gel process [2] or by utilizing the nano-casting method [3]. Among these processes, the evaporation-induced self-assembly (EISA) pathway [2] has been demonstrated to be a simple and fast way to get ordered and thermally stable MA, however, the producing cost is an important concern because of the use of expensive and toxic aluminum alkoxides. Although considerable efforts have been devoted to reduce the cost of MA synthesis recently by using the inexpensive inorganic aluminum salts to replace aluminum alkoxides [1,4], unfortunately disordered mesostructures with poor thermal stability were fabricated in most cases. Therefore, there are still great interests in developing a novel, economic and environmentally benign approach to prepare highly ordered and thermally stable MA.

Herein, we present a feasible approach to synthesize ordered MA with high thermal stability by using the inexpensive aluminum nitrate as precursor. To favor the formation of ordered mesostructure, the key is to maximize the concentration of Al-OH species from the hydrolysis of aluminum nitrate to enhance their hydrogen bonding interaction with the surfactant molecules [5], which can be achieved by the solvothermal pre-hydrolysis (STPH) process. With this strategy, the ordered MA with 2D hexagonal mesostructure can be readily obtained. More importantly, the

synthesized MA exhibits a high thermal stability up to 900 °C, and possesses narrow pore-size distribution, high surface area and large pore volume.

2. Experimental section

Synthesis procedure: In a typical synthesis, 5.63 g of Al(NO₃)₃·9H₂O was dissolved in 15 mL of mixed solution of anhydrous ethanol and deionized water ($V_{\text{EtOH}}/V_{\text{water}}=4/1$). Then, the mixture was transferred to a beaker without cover for the STPH treatment at 80 °C for 5 h to promote the hydrolysis of Al³⁺ into Al-OH species. After that, the obtained light-orange alumina xerogel with a large number of Al-OH species was taken out and slowly added to 30 mL of anhydrous ethanol solution dissolved 1.8 g of P123 (EO₂₀PO₇₀EO₂₀, EO=ethylene oxide, PO=propylene oxide) and 0.6 g of citric acid (CA). After being vigorously stirred for 24 h at 30 °C, the resultant mixture was transferred to a dish and underwent solvent evaporation at 45 °C for 48 h and thermal treatment at 100 °C for 24 h, respectively. The final product was calcined at 400 °C for 5 h to remove the template and almost all of CA. The other MAs were prepared analogously by using AlCl₃·6H₂O and Al₂(SO₄)₃·18H₂O as precursors, respectively. The obtained samples were named as SE-Al(n), SE-Al(c), and SE-Al(s) (n, c, and s refer to Al(NO₃)₃·9H₂O, AlCl₃·6H₂O, and Al₂(SO₄)₃·18H₂O, respectively). For comparison, the MA prepared by the similar EISA method without the STPH process using Al(NO₃)₃·9H₂O as precursor was named E-Al(n). Notably, the amount of aluminum precursor used in each case was 15 mmol, and the solvent could be recycled and reused during the synthetic process by using the home-made drying oven.

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Thermal stability evaluation: The thermal stability was investigated by treating calcined samples in air at 750 and 900 °C for 1 h. Both temperature thermal treatment processes were correspondingly abbreviated as LT and HT.

Characterization: Powder X-ray diffraction patterns were recorded on a Shimadzu XRD-6000 diffractometer using Ni-filtered Cu K α (0.154 nm) radiation. Transmission electron microscopy (TEM) experiments were performed on a JEOL 2011 microscope operated at 200 kV. N₂ adsorption was conducted on a Quantachrome analyzer at -196 °C. Before measurements, the samples were degassed at 180 °C in vacuum for 10 h. The BET method was used to calculate the specific surface areas. The pore-size distributions were derived from the adsorption branches of the isotherms using the BJH method. The total pore volumes were calculated from the amount of nitrogen adsorbed at a relative pressure of 0.99.

3. Results and discussion

Fig. 1 depicts the XRD patterns of MAs prepared with different inorganic aluminum salts as precursors before and after the high-temperature treatment. It shows that SE-Al(n) exhibits two well-resolved Bragg peaks (Fig. 1A), according to the TEM observation of the well-ordered hexagonal arrays along [110] and [100]

orientations with uniform pore size and wall thickness (Fig. 2A), which can be attributed to $p6mm$ hexagonal symmetry. From the intense (100) peak, a d_{100} spacing of 9.81 nm is calculated (Table 1), corresponding to a unit cell parameter of 11.3 nm. However, for E-Al(n) prepared without the STPH process, only a broad (100) peak is observed (Fig. 1A), indicating that the STPH plays a key role to synthesize highly ordered mesostructure when $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ is used as precursor. We assume that the STPH is advantageous for the endothermic hydrolysis reaction of inorganic aluminum salts and thus facilitates the transformation of Al^{3+} into Al-OH species. With increasing the concentration of Al-OH species, the hydrogen bonding interaction between P123 and aluminum species will be significantly enhanced, which will promote the cooperative assembly process and further construct an ordered mesostructure of final SE-Al(n). In addition, CA can behave as a ligand and bond with Al-OH species in a bidentate or bridging fashion, which can effectively slow down the condensation reaction of Al-OH species and maintain more Al-OH species to enhance their hydrogen bonding interaction with P123.

On the other hand, compared with the XRD pattern of SE-Al(n), SE-Al(c) and SE-Al(s) exhibit relatively lower signal-to-noise ratio in their XRD patterns and broader full width at half-maximum (FWHM) as judged from the (100) peak, suggesting a less mesostructural ordering (Fig. 1A), which confirms the radius and charge of the counteranions will impose a great effect on the assembly

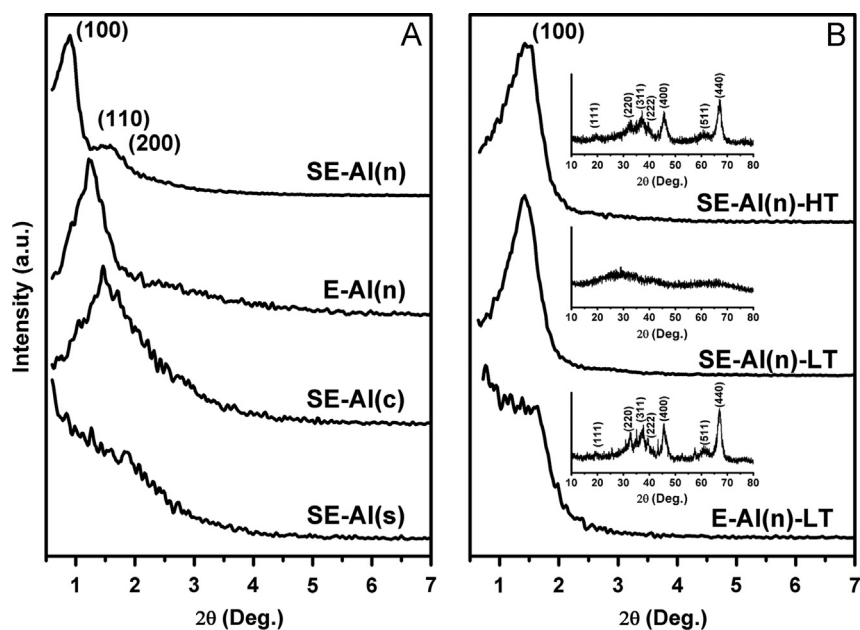


Fig. 1. XRD patterns of calcined (A) and high temperature treated (B) samples.

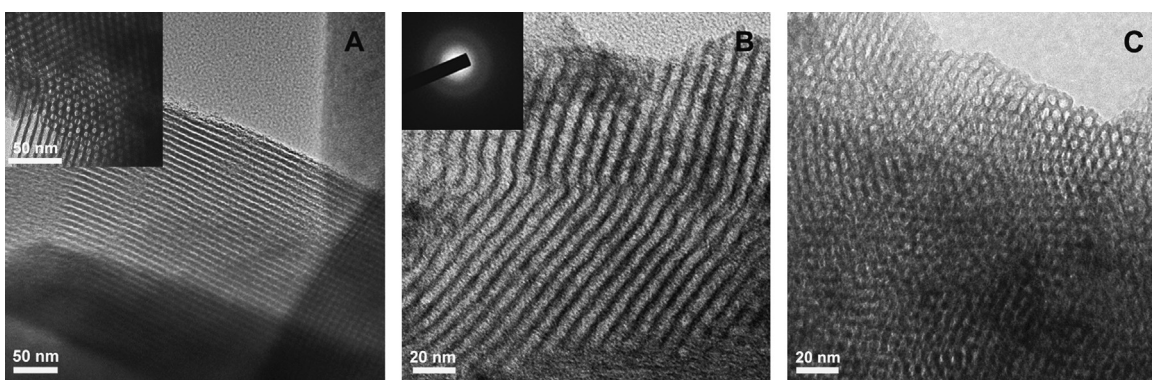


Fig. 2. TEM images of SE-Al(n) (A), SE-Al(n)-LT (B), and SE-Al-HT (C) (the inset in B is corresponding SAED pattern).

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