



# Synthesis of aluminum nitride nanoparticles by a facile urea glass route and influence of urea/metal molar ratio



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## ABSTRACT

Attention toward nanosized aluminum nitride (AlN) was rapidly increasing due to its physical and chemical characteristics. In this work, nanocrystalline AlN particles were prepared via a simple urea glass route. The effect of the urea/metal molar ratio on the crystal structure and morphology of nanocrystalline AlN particles was studied using X-ray powder diffraction (XRD), scanning electron microscope (SEM) and transmission electron microscope (TEM). The results revealed that the morphology and the crystal structure of AlN nanoparticles could be controlled by adjusting the urea/metal ratio. Furthermore, a mixture of Al<sub>2</sub>O<sub>3</sub> and *h*-AlN was detected at the urea/metal molar ratio of 4 due to the inadequate urea content. With increasing the molar ratio, the pure *h*-AlN was obtained. In addition, the nucleation and growth mechanisms of AlN nanocrystalline were proposed.

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## 1. Introduction

Aluminum nitride (AlN) has attracted much attention recently. It has been applied as an important ceramic substrate material due to its high thermal conductivity, low thermal expansion coefficient, low dielectric constant, good mechanical strength, thermal stability, lack of toxicity, stable crystal structure and relatively low cost [1–3]. These unique properties have made AlN an attractive material for high-tech industrial applications such as electrical packaging and heat sinks. Therefore, much attention has been given to the preparation of AlN. In general, AlN could be fabricated through a variety of methods, such as carbothermal nitridation [4–8], direct nitridation of aluminum powders [9,10], chemical vapor deposition [11] and plasma-base process, among which carbothermal nitridation and direct nitridation of aluminum powders have been the most widely used in the industrial production. However, both of these two methods have a variety of drawbacks, i.e., high-energy consumption, large size products, and low pure products of direct nitridation of aluminum powders and complicated production processes, high production temperatures, long production process of carbothermal nitridation.

Recently, it has been reported that a new approach, the urea glass route, can be used to synthesize the metal nitrides materials

such as TiN, VN, CrN, GaN and MoN [12–14]. In addition, compared with other methods, this route shows many advantages. Firstly, it is a simple method due to the simple and cheap raw reagent materials. Secondly, the growth temperature is lower than that of the low temperature combustion synthesis method which uses the similar starting materials [15,16]. Thirdly, no further purifications are necessary before or after the temperature treatment. Particularly, the products have nanosize. Although the preparation of nanosized AlN using the urea glass route had been reported by some researchers [17–19] but the focus was placed on metal sources, such as metal chloride and nitrate. However, no significant systematic studies have been conducted to investigate the effect of urea/metal molar ratio (*R*) on the synthesized AlN powders. It had been reported that the urea/metal molar ratio could be applied to tailor molecular precursors which might play an important role on the purity and crystal structure of the formed metal nitrides. Sardar et al. [18] demonstrated that the crystal structure of GaN transformed from hexagonal to cubic as increasing the urea content. Giordano et al. [12] reported that the pure Mo and W nitrides were able to be prepared by changing the urea/metal molar ratio suggesting the urea contents play an important role on the purity of the metal nitride products. Therefore, in this paper, a varying urea/metal molar ratio (*R*) was employed for the synthesis of the nanosized AlN via the simple urea glass route. Moreover, the effect of *R* on the phase composition, the morphology and microstructure of the synthesized AlN nanoparticle was investigated in detail. Furthermore, the nucleation and growth mechanisms of AlN nanoparticle were proposed.

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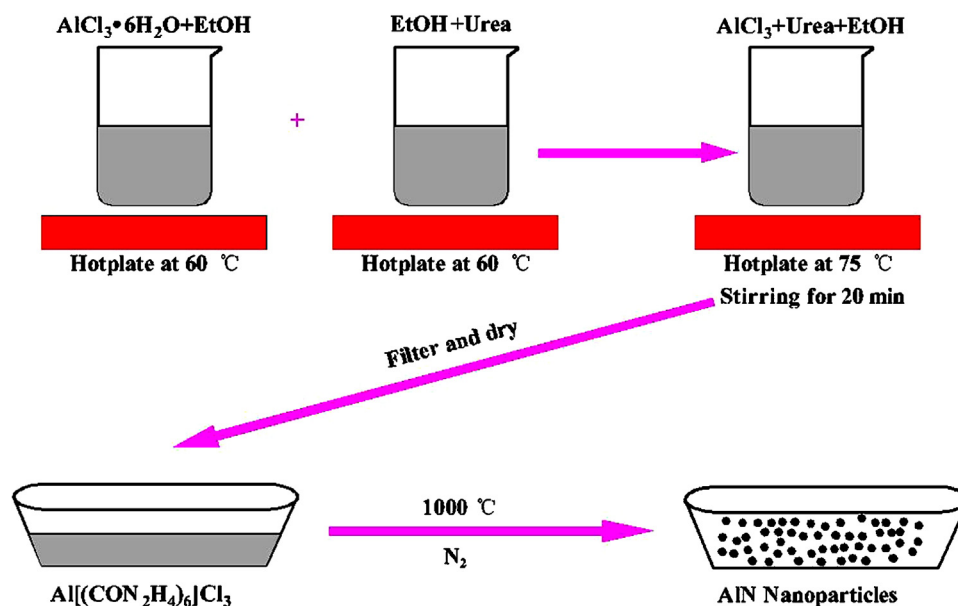


Fig. 1. Scheme for the AlN nanoparticles samples preparation route.

## 2. Materials and methods

### 2.1. Materials preparation

The aluminum chloride ( $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ , 97%), urea ( $\text{CON}_2\text{H}_4$ , 99.0%), anhydrous ethanol ( $\text{C}_2\text{H}_5\text{OH}$ , 99.5%) and nitrogen ( $\text{N}_2$ , 99.999%) were used as raw materials to prepare AlN nanoparticle. All starting materials were purchased from commercial sources and were used without further purification. The fabrication process of the AlN nanoparticles is shown in Fig. 1. The basic procedure employed for the synthesis of AlN was to heat compounds of aluminum chloride with urea at an appropriate temperature. Firstly, aluminum chloride and urea were dissolved in anhydrous ethanol to obtain a concentrated solution, respectively. Then, the aluminum chloride solution was added slowly into the urea solution at  $70^\circ\text{C}$ . The urea content of the urea solution was various to obtain the different urea/metal precursor molar ratio ( $R$ ). The  $R$  value was set at 4, 6 and 10 respectively and the resultant aluminum–urea chloride compound samples were named as R4, R6 and R10, respectively. The obtained aluminum–urea chloride compound was separated by filtration and dried at  $80^\circ\text{C}$  for 5 h. Finally, the aluminum–urea chloride compound was put into a tube furnace and heated under flowing  $\text{N}_2$  at  $1000^\circ\text{C}$  for 5 h to obtain AlN. The AlN nanoparticles fabricated from different aluminum–urea chloride compound samples were named as R4S, R6S and R10S, respectively.

### 2.2. Characterization

Thermal gravimetric analysis (TGA) of the aluminum–urea chloride compounds and urea were performed in air at a heating rate of  $10^\circ\text{C}/\text{min}$  with a Swiss Mettler TGA/DSC1. Simultaneous the Fourier transform infrared spectroscopy (FTIR) of the aluminum–urea compounds and urea were recorded on a Nicolet MAGNA-560 spectrophotometer. The AlN nanoparticles were characterized by X-ray diffraction (XRD) for phase identification using  $\text{Cu K}\alpha$ -radiation. XRD measurements were performed on a Bruker D8 Advanced X-ray diffractometer from  $20^\circ$  to  $80^\circ$  with a scan speed of  $4^\circ/\text{min}$ . Scanning electron microscope (SEM) examination of the AlN nanoparticle samples was carried out in FEI Nanosem 430. The samples were coated by sputtering an Au alloy prior to imaging. To provide further insight into AlN particles,

transmission electron microscope (TEM) investigations were performed. TEM samples were prepared by dispersing a small amount of solid powder in anhydrous ethanol and then using an ultrasonic bath for dispersing without any further dispersing agent. TEM images were obtained with a JEOL JEM100CXII operating at an accelerating voltage of 300 kV.

## 3. Results and discussion

### 3.1. FTIR spectra of aluminum–urea chloride complexes

Fig. 2 shows the FTIR spectra of the urea and aluminum–urea chloride complexes prepared with different  $R$ . The related peak positions and their assignments are listed in Table 1. It could be observed that the curve shapes of the R4, R6 and R10 samples were similar while they exhibited different trend in comparison with the curve of the urea spectrum. The asymmetric and symmetric stretching vibrations of  $\text{NH}_2$  groups in the urea were observed at  $3442\text{ cm}^{-1}$  and  $3396\text{ cm}^{-1}$  while both of two peaks in the R4, R6 and R10 samples shifted to a higher frequency (see Table 1). This revealed that an intermolecular linkage between urea molecules

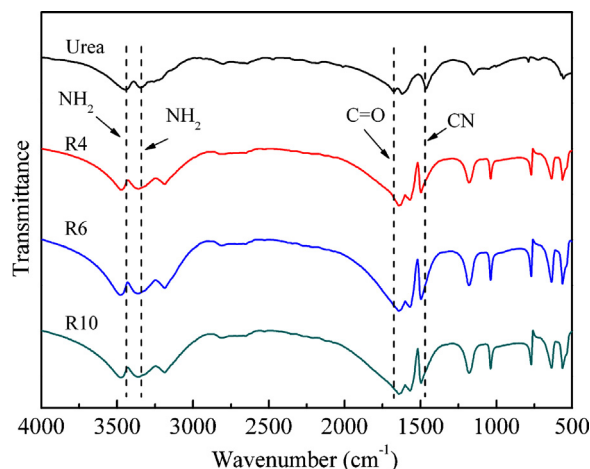


Fig. 2. The FTIR spectra results of urea and aluminum–urea chloride complexes.

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