



The influence of metal Mg on micro-morphology and crystallinity of spherical hexagonal boron nitride



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ABSTRACT

This search used the boric acid and borax as a source of boron, urea as a nitrogen source, Mg as metal catalyst, and thus prepared different micro-morphology and crystallinity hexagonal boron nitride powders under a flowing ammonia atmosphere at a nitrating temperature of 750 °C. The effect of Mg content on the crystallinity and micro-morphology of hexagonal boron nitride powders was studied, and the Mg action mechanism was explored. Without the added surfactant, the graphitization index (GI) was 6.87, and the diameter of the spherical h-BN was bigger. When the added Mg were 0.1 g, 0.3 g, 0.5 g and 0.7 g, the (GI) decreased to 6.04, 5.67, 4.62 and 4.84, respectively. When the Mg content was higher (0.9 g), GI value increased rapidly, and the crystallinity became bad. When the Mg content was 0.5 g, the dispersion of h-BN powders was at its optimum and refinement apparently, and the crystallinity at its highest.

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1. Introduction

LED light source is the fourth generation new type high efficiency and energy saving light source after the incandescent lamp, fluorescent and HID lamp [1], and the energy-saving achieves 80% under the same luminous efficiency. However, when an electric current passes through, if the heat of high-power, lightweight LED electronics does not disperse in time, the semiconductor PN section temperature will increase: the service life of LED will reduce. The Heat dissipation problem has become the bottleneck to the popularization and application of the high power LED new high efficient light source. To this end, the development of high thermal conductive plastics has become the research focus among the scientific and technical workers. High thermal conductive plastic has lighter quality, good insulation performance, more secure, one-time plastic injection molding, high production efficiency than aluminum heat dissipation material. If the electroplating process that often is used in aluminum product production is avoided, the serious pollution of metal of electroplating effluent on water and soil caused also be avoided, and more environmental protection, meanwhile the

cooling effect will close to that of the aluminum heat dissipation material. But if the thermal conductive plastics reach needed thermal conductivity effect, the high thermal conductivity filler is necessary. Hexagonal boron nitride (h-BN) has a low thermal expansion coefficient, high thermal conductivity [2–3], and the thermal conductivity of the hot pressing sintering products of h-BN as high as pure iron. It also is the largest thermal conductivity, the lightest ceramic material. At present, the high crystallinity and globular hexagonal boron nitride is most ideal thermal conductive plastic filler [4–5].

Hexagonal boron nitride is a covalent compound [6]. It has been synthesized as early as the 19th century, but it began to develop into a kind of widely used material until the second half of the 20th century. The h-BN crystal structure is similar to that of graphite. There is not spherical hexagonal boron nitride in the nature, and the h-BN synthesis needs to rely on artificial. So far, tubular, globular, cocoon-like, fibrous, arborescence-like, and rod-like h-BN powders have been successfully synthesised [7–9]. For these different morphologies of h-BN powders, the spherical forms had the greater filling ability compared to other morphologies of h-BN powders when used as thermally conductive plastics: the random orientation of spherical microcrystalline phases also reduces the anisotropy of h-BN [10–13]. In addition, the three-dimensional order of the atomic lattice of the h-BN powder (crystallinity) also has a great influence to the thermal conductivity of the thermal

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conductive plastic [14–15]. The higher the degree of crystallinity, the better thermal conductivity. Past, the research mainly focuses on the organization structure and mechanical properties of h-BN and the influence of metal Mg on the synthesis of boron nitride nanotubes. The research on the spherical h-BN crystallinity is rare, and the study on the influence of metal Mg on crystallinity report less. So, the main purpose of this study is the preparation of high crystallinity, spherical hexagonal boron nitride, and the influence mechanism of metal Mg on high crystallinity spherical hexagonal boron nitride is analysed clearly. The result has great theoretical reference value and establishes the theory foundation to the popularization and application of the fourth generation new type high efficiency and energy saving lighting technology

2. Experimental materials and methods

2.1. Experimental materials

The experiments used borax ($\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$) and boric acid (H_3BO_3) as sources of boron, urea ($\text{CO}(\text{NH}_2)_2$) as nitrogen source, Mg as metal reducing agent, acryl amide ($\text{C}_3\text{H}_5\text{ON}$) as monomer, *N,N*-methylenebisacrylamide ($\text{C}_7\text{H}_{10}\text{N}_2\text{O}_2$) as cross-linking agent and ammonium persulfate ($(\text{NH}_4)_2\text{S}_2\text{O}_8$) as initiator. The mole ratio of borax and boric acid was 1:1. The mole ratio of boron atoms and nitrogen atoms was 1:4, and the quality ratio of network agent between acryl amide and *N,N*-methylenebis-acrylamide and ammonium persulfate was 140:4:25. The experimental materials used were all of analytical grade, and the suppliers are all Shengyang Sinopharm Chemical Reagent Co., Ltd., China.

2.2. Sample preparation

First, a quality certain ratio of $\text{Na}_2\text{B}_4\text{O}_7 \cdot 10\text{H}_2\text{O}$, H_3BO_3 and $\text{CO}(\text{NH}_2)_2$ was poured into deionised water at a certain molar ratio, this was then mixed and stirred to form a transparent solution. Then $\text{C}_3\text{H}_5\text{ON}$ and $\text{C}_7\text{H}_{10}\text{N}_2\text{O}_2$ were added to this mixed solution, and stirring continued until completely dissolved. The resulting mixed solution was placed in a constant temperature water bath at between 60 and 70 °C, and stirred. Again the $(\text{NH}_4)_2\text{S}_2\text{O}_8$ was stirred into the mixed solution until the mixture became a gel suitable for use as a former body. After the precursor gel was put in a drying oven at 60 °C, dehydrated and ground, the h-BN precursor was finally obtained.

In order to avoid metal Mg was oxidized, h-BN precursors and magnesium powders were uniform mixed in vacuum glove box, and the mixture was placed in the tubular atmosphere furnace quickly, vacuuming. In a flowing ammonia atmosphere, the mixture was heated to 300 °C at a rate of 5 °C/min for 1 h. Then the mixture was heated to 750 °C at a rate of 10 °C/min for 3 h and cooled along with the furnace. After the product was cleaned by pickling, washing, alcohol washing, and residual carbon was removed at 600 °C in a rapid-heating furnace: h-BN powder was thus obtained.

2.3. Powder test and performance characterization

The material phase composition was analysed by PW3040/60 type X-ray diffraction, and the tests used a copper target with a scanning speed of 15°/min and an angle scanning range of $10^\circ \leq 2\theta \leq 90^\circ$. By comparing the results of those diffraction peaks revealed by XRD diffraction with the card database, the phase composition of the samples could then be determined.

The micro-morphology of the samples was observed by S4800 type scanning electron microscopy. Then anhydrous ethanol

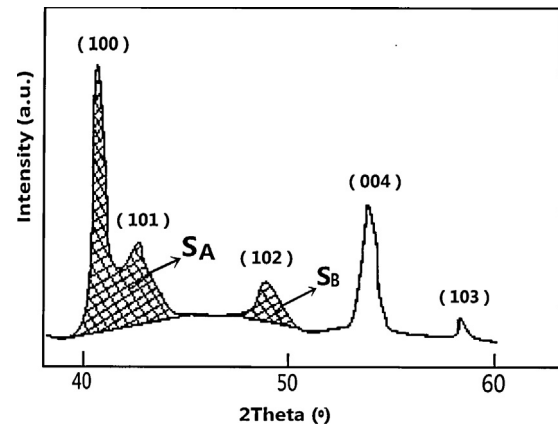


Fig. 1. The illustration of the defined areas used in determination of graphitization index.

was used as dispersion medium, and some samples were dispersed by ultrasound. Again the dispersed sample was dropped in the polishing copper column surface and scanned by scanning electron microscopy after sputter-coating in gold.

2.4. The characterization method of crystallinity

Because different preparation technology, the crystallinity of h-BN has a wide range change in 2-d to 3-d ordered structures. To characterise the three-dimensional order of h-BN, the crystallinity was represented by graphitization index (GI). With increased GI value, the three-dimensional order of the atomic lattice will decrease, and the crystallinity also decreases. Otherwise, the crystallinity will be better. The complete six-part structure will form when the GI value is 1.60. Generally speaking, the product has good crystallisation at $1.60 \leq \text{GI} \leq 5.00$ [16]. The GI value can be quantitative calculated through the (100), (101), and (102) crystal plane diffraction peak areas of the XRD diffraction spectrum as shown in Eq. (1.1).

$$\text{GI} = \frac{S_{(100)} + S_{(101)}}{S_{(102)}} \quad (1.1)$$

where $S_{(100)}$, $S_{(101)}$, and $S_{(102)}$ were respectively the (100), (101), and (102) crystal plane diffraction peak areas for h-BN: $S_A = S_{(100)} + S_{(101)}$, and $S_B = S_{(102)}$, as shown in Fig. 1.

This research used the grid method to simple calculate graphitization index: the XRD diffraction pattern of a sample was partially enlarged to maintain accuracy in the calculation, then the pre-prepared grid was overlain on the amplified XRD diffraction pattern of the product. Then the total number of grid squares contained in the (100), (101), and (102) crystal plane diffraction peaks were calculated, i.e., S_A was surrounded by (100) and (101) diffraction peak areas and S_B was surrounded by the

Table 1

The standard free energy of reactants and resultants under 750 °C.

The free energy GI (KJ mol ⁻¹)						
B ₂ O ₃	Mg	[B]	NH ₃	MgO	BN	H ₂
-1381.64	-49.04	-12.38	-265.40	-652.7	-282.92	-149.28

Table 2

The standard free energy of reactants and resultants.

The standard free energy GI (KJ mol ⁻¹)						
B ₂ O ₃	Mg	[B]	NH ₃	MgO	BN	H ₂
-1381.64	-49.04	-12.38	-265.40	-652.7	-282.92	-149.28

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