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Review article

Aluminium nitride cubic modifications synthesis methods and its features. Review

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ABSTRACT

This article presents a brief review of the research progress achieved in the field of aluminium nitride (AlN) metastable cubic modifications synthesis. It covers mainly the synthetic approaches, such as carbothermal reduction nitridation (CRN), chemical vapour deposition (CVD), solvent thermal synthesis, high pressure phase transition and thin-film technology (molecular beam epitaxy, atmospheric reactive plasma nitriding, magnetron sputtering, pulsed laser deposition). A classification of synthetic approaches has been proposed, which is based on the difference between the aluminium compounds involved in synthesis. The basics of cubic aluminium nitride stabilization and possible technological conditions for the synthesis of the metastable cubic phase have been formulated.

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1. Introduction

Aluminium nitride has the highest thermal conductivity among ceramic materials. Aluminium nitride synthesis technologies have been developed since the end of the 20th century owing to the progress in LED and computer production. The publication and

inventive activities in this field have been showing a stable positive dynamics since 1991 [1].

AlN in different forms, such as dispersed powder, single crystals and thin films, has found numerous applications in various industries.

The high inventive activity with respect to AlN application in electronic industry can be explained by the tendency to reduce the weight and size of electronic devices improving simultaneously their performance, for example, the expansive growth in the compact and high-performance tablets market [2–5]. The optoelec-

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tronic properties of aluminium nitride allow it to be used as LED [3,4,6] and PCBs [2], while its high thermal conductivity makes it possible to employ AlN in the production of brackets for LED technology, ceramic substrates [5,7–9] and thermal interfaces for electronic lasers [2].

In addition, research efforts are expended on searching of new ways of aluminium nitride application. Having a large electromechanical coupling factor and temperature stability, as well as a high acoustic velocity, AlN can be used in surface acoustic wave (SAW) devices, such as resonators [10,11], filters, sensors and actuators [12].

There are numerous of investigations examining the possibilities of using AlN properties for the production of sensors [13,14]. For example, highly c-axis oriented AlN films on a Si substrate were investigated as surface acoustic wave resonators for temperature sensing applications, because the resonator frequency depends linearly on temperature [15,16]. Erbium doped aluminium nitride nanoparticles can be also used as nanothermometers, since the photoluminescence peaks from Er^{3+} are related to temperature [17]. AlN films, in particular on flexible polymer substrates, can find application as highly sensitive pressure [18] and ME sensors [19].

Owing to a wide bandgap of AlN, it is used in the production of solid-state cells that are photosensitive to ultraviolet radiation [20–22]. AlN structures have a number of advantages: their sensitivity range resides in the UVC field, they perform parameters stability, provide the operation of devices at high temperatures and enhanced radiation.

AlN can be integrated into the micro electromechanical system (MEMS) technology. Although AlN has a lower piezoelectric coefficient compared to popular piezoelectric materials, such as ferroelectric lead-zirconate-titanate (PZT) or ZnO, it is preferred in many MEMS applications due to a lower dielectric loss tangent and a higher signal-to-noise ratio (SNR) [23–26].

Moreover, a large second order nonlinear susceptibility coefficient along the c-axis, high thermal conductivity, and a wide transparency window of AlN make it an excellent candidate for the generation of UV-C laser light [27].

Besides the electronic industry, aluminium nitride is widely used for the development of composite materials with a high electrical resistance [28], thermal conductivity [29–32] and mechanical strength [33].

Since the investigations in the field of hexagonal aluminium nitride synthesis technologies have reached the level of industrial scale production mastering numerous application areas, the challenge now is to synthesize aluminium nitride modifications with improved properties.

Currently, one of the main tendencies in this area is the synthesis and investigation of ternary systems with AlN: $\text{AlTiN}/\text{AlTiVN}$ [34] as a cutting material with high hardness; AlGaN quantum cells [35–37]; AlN/BN composites, which possess the properties of aluminium nitride and at the same time can be easily machined to produce odd-shaped parts [38].

Aluminium nitride nanoparticles can be used as components of nanohybrids with graphene. Graphene with its thermal conductivity of $5000 \text{ W/m}^2\text{K}$ has a high potential as a filler for improving the thermal conductivity of polymer composites, but its low dispersion in the polymer matrix requires the use of additional modifiers. AlN nanoparticles with a high thermal conductivity forming nanohybrids with graphene can be used as dispersing agents. It is interesting to note that graphene causes the stabilization of the AlN cubic modification [39].

AlN films find potential applications in light emitting diodes, laser diodes, solar cells, multi-junction transistors, detectors, sensors and Bragg reflectors because of their tunable band gap from

ultraviolet to infrared range [40,41]. These films can also be used as cladding layers on GaN based laser diode.

Ce doped wurtzite-type aluminium nitride can be used as high-efficiency electroluminescent applications over a wide wavelength range [42].

$(\text{SiC})_{1-x}(\text{AlN})_x$ pseudo-binary solid solutions are of interest as a class of wide bandgap semiconductors with band gap energies varying in the range from 3 to 6 eV at $0 \leq x \leq 1$ [43].

AlN is stable under normal conditions in the hexagonal structure of wurtzite w-AlN. However there are cubic phases of aluminium nitride with rock salt rs-AlN ($d = 4.045 \text{ \AA}$) and zinc-blend zb-AlN ($d = 4.38 \text{ \AA}$ and $d = 7.913 \text{ \AA}$) [44–46] structures. Aluminium nitride with a cubic microstructure has a higher thermal conductivity ($250\text{--}600 \text{ W/m}^2\text{K}$) as compared with hexagonal aluminium nitride, as well as electrical resistance (10^{16} Ohm/cm) and mechanical strength (hardness $40\text{--}50 \text{ GPa}$) [47].

Therefore, another challenge is the creation of cubic aluminium nitride samples. Synthesis of cubic aluminium nitride with advantageous properties compared to the hexagonal AlN is not only an applied, but also a fundamental study. Cubic aluminium nitride is a metastable phase and the problem of its stabilization requires a comprehensive assessment both from the theoretical and practical viewpoints. It is no less important to investigate the effect of the crystal structure on the thermal properties of refractory compounds.

The thermodynamic and thermal-conducting properties of AlN as a ceramic material are completely determined by phonons. The phonon intrinsic mean free path is an important physical characteristic, which plays a critical role in determining these properties. In work [48], the mean free path values in cubic and hexagonal aluminium have been determined theoretically. As expected, the results simulated within the Fermi's golden rule showed an excess in the phonon mean free path in the cubic crystal lattice compared to the hexagonal one. Moreover, this excess increases with temperature; so, at temperatures above ambient the phonon mean free path in the cubic phase is approximately four times higher than in the hexagonal phase. From the expression of thermal conductivity:

$$K = 1/3 C v \lambda$$

(λ – the phonon mean free path, v – the average velocity of phonons, C – the average specific heat capacity of phonons), it is determined that at temperatures above ambient temperature the thermal conductivity of cubic aluminium nitride is 2.7 times higher than in hexagonal AlN. There are two main reasons why the thermal conductivity of the cubic AlN would be higher than that of the hexagonal AlN. The first reason is a more simple crystal structure of the cubic phase with only three optical phonon branches. The hexagonal crystal structure consists of nine optical branches leading to a much stronger phonon–phonon interaction compared to the cubic phase. The second reason is that the average phonon speed in the cubic structure is higher than in the hexagonal structure.

For the bulk aluminium nitride, the wurtzite structure is thermodynamically stable under normal conditions; at high pressures ($15\text{--}20 \text{ GPa}$) the rock salt structure becomes more stable [49–56]. The properties of hexagonal aluminium nitride and their dependence on the particle shape and size, the methods of its production and applications are well known and have been described in monographs [57] and review articles [58,59]. Zinc-blend AlN is a metastable modification in the whole investigated pressures range, although it is noted that this phase can be formed by epitaxy at relatively low temperatures on substrates having the cubic symmetry. The lattice parameters of these structural modifications obtained by different authors vary considerably. Obviously, this is caused by

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