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Non-aqueous gelcasting of AlN ceramics using a low-toxicity monomer (DMAA) as gelling agent

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ABSTRACT

In this study, high-performance aluminum nitride (AlN) samples were prepared by gelcasting based on a low-toxicity monomer and organic solvents. We prepared a 50 vol% solid content AlN suspension with a solvent ratio of 3:1, 6 wt% N,N-dimethylacrylamide (DMAA) and 0.15 wt% Polyethyleneimine (PEI); after degassing, low viscosity and high stability were observed. It was proven that the AlN green bodies exhibited a bending strength of 18.68 MPa, which met the requirements for machining. An optimum debinding process was developed and the TG-DSC curves showed that the AlN green bodies were heated at 1 °C/min from 450 to 800 °C. The relative density, apparent porosity, bending strength, and thermal conductivity of the AlN sintered bodies prepared by non-aqueous gelcasting were 98.5%, 0.22%, 310 MPa, and 159 $W\,m^{-1}\,K^{-1}$ respectively, representing optimum conditions. The uniform miscrostructure of the sintered bodies was observed by SEM images.

1. Introduction

Aluminum nitride (AlN) ceramics have excellent properties, including high thermal conductivity (theoretical value: 320 $W\,m^{-1}\,K^{-1}$), low dielectric coefficients (8.7), and low thermal expansion coefficients consistent with silicon [1–4]. Due to these advantages, AlN has been considered as a package material for large-scale integrated circuits and semiconductor circuits because of the ideal heat dissipation. However, several conventional forming processes do not meet certain strict requirements such as shape, reliability, performance, and dimensional tolerances especially in the preparation of ceramic materials of sufficient size and with complex shapes. Gelcasting, a wet and near-net-shape forming technique, was invented by scientists at the Oak Ridge National Laboratory (ORNL), USA in the 1990s [5]. In this process, a concentrated suspension with good liquidity and low viscosity is plunged into a mold to form a gelled body by means of a polymerization reaction [6,7]. In recent years, multiple ceramic materials based on aqueous and non-aqueous gelcasting have been created, such as ZrO_2 [8], SiC [9], Si_3N_4 [10], YAG [11], ZTA [11], SiO_2 [12], Al_2O_3 [13-17], AlN [18-22], and other materials. Nevertheless, the industry has been reluctant to the forming technique because the neurotoxin monomer acrylamine (AM) that is used during the process threatens human health and the environment [11]. Therefore, researchers have developed low-toxicity or non-toxicity gel systems that have demonstrated excellent properties similar to the AM gelcasting system. It has been reported that the properties of AlN ceramics based on the polymerization of 1,6-Hexanediol diacrylate [19], Sorbitol polyglycidyl ether (SPEG) [20], Hydantion epoxy resin [21] and DMAA [18,23], were greatly improved.

It is well-known that AlN powder is sensitive to aqueous gelcasting. For example, Guo et al. prepared AlN ceramics using AlN powder modified to improve the anti-hydrolysis property with $Al(H_2PO_4)_3$ and H_3PO_4 as coating agents. This processing route is not only time consuming but also does not completely protect the AlN powder from hydrolysis [18,25]. Recently, Jiang et al. explored a non-aqueous gelcasting system using ethanol as a solvent [19–21]. However, the resultant samples were prone to cracking defects caused by the low boiling point and high volatility of the ethanol during drying [24], particularly in large-sized products. Therefore, there is still an open quest for finding a suitable non-aqueous gel system for the preparation of AlN ceramic slurries with high solid loading and low viscosity. Guo et al. [23] used a monomer (DMAA) and a mixture (polyethylene glycol and ethanol) for gelcasting of AlN ceramics and obtained a bending strength of the green bodies as high as 18.68 MPa.

In this study, the effects of the DMAA content and the solid content on the viscosity of AlN ceramic slurries and the properties of green bodies were studied. After sintering, the relative densities, apparent porosities, mechanical properties, thermal properties, and microstructures of the green and sintered bodies were investigated.

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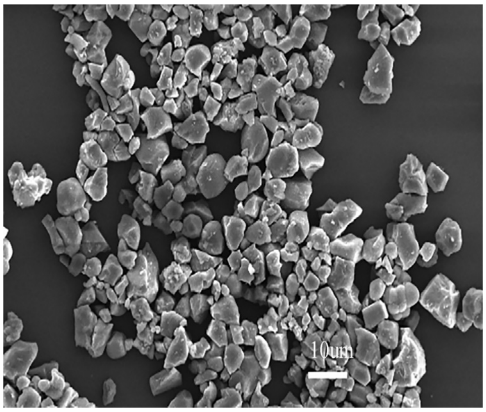

Fig. 1. SEM photograph of AlN powders.

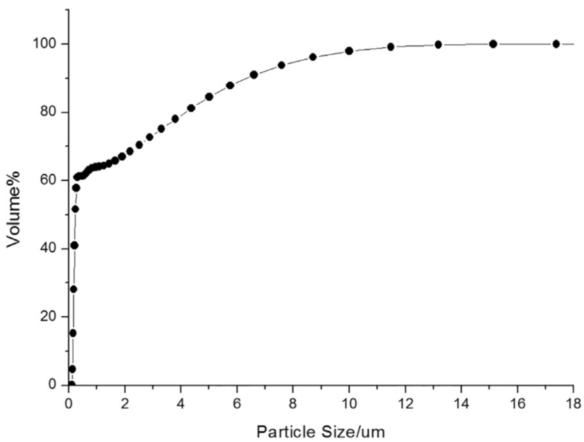

Fig. 2. Particle distribution of AlN powders.

Table 1
Main experimental supplies.

Experimental supplies	Purity	Functionality
AlN	99.9%	Powder
DMAA	99.4%	Monomer
MBAM	98%	Crosslinker
PEI	50%	Dispersant
BPO	CP	Initiator
Ethanol	AR	Solvent
N,N-dimethylacetamide	CP	Solvent
PEG-200	99.9%	Solvent
Yttrium oxide	99.9%	Sintering additive

AlN:Advanced Technology & Materials Co. Ltd., Beijing, China; DMAA:Kowa Co. Ltd., Japan; MBAM:Chemical Reagent Research Institute, Tianjing, China; PEI:Aladdin chemistry Co. Ltd., Shanghai, China; BPO:Lingfeng chemistry Co. Ltd., Shanghai, China; Ethanol:Kemiou Chemical reagent Co. Ltd., Tianjing, China; PEG-200:Guanghua Sci-Tech Co. Ltd., Guangdong, China.

2. Experimental part

2.1. Raw materials

Commercial AlN power was used as the raw material; the scanning electron microscopy (SEM) photographs and particle distribution patterns of the AlN powders are shown in Figs. 1 and 2, respectively. DMAA, N,N-methylenebisacrylamide (MBAM), and PEI (molecular weight: 70,000) were used as monomer, crosslinker and dispersant, respectively. Benzoyl peroxide (BPO, 1 g BPO was dissolved per 5 ml N,N–dimethylacetamide) was used as the initiator. Ethanol (low-viscosity organic solution) and polyethylene glycol (PEG, molecular weight:

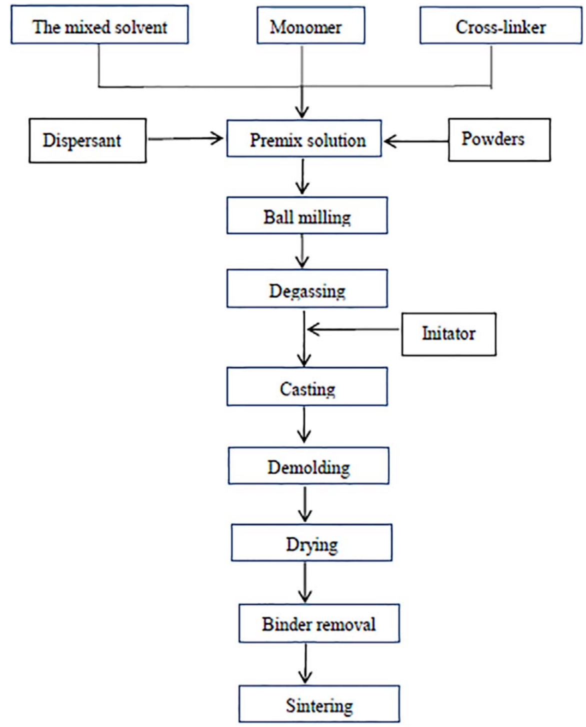

Fig. 3. Schematic of the gelcasting processing.

Table 2
Main instruments used in the experiment.

Laboratory instruments	Model	Manufacturers
Rotation rheometer	R/S CC25	Brookfield Corporation, USA
Scanning electron microscope (SEM)	JSM-5900	JEO, Tokyo, Japan
Universal testing machine	CM-62303	MTS System Corporation, China
Thermal constant analyzer	LFA447	Netzsch, selb, Germany

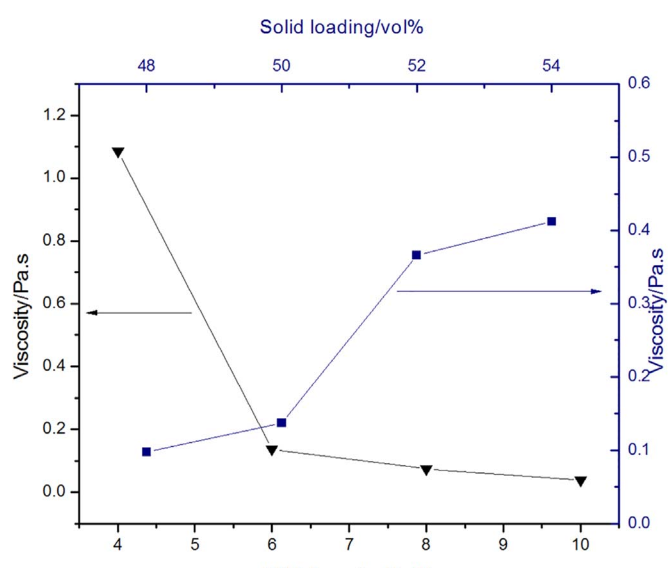

Fig. 4. Effect of the DMAA content and solid content on viscosity of the AlN slurries with a solvent ratio of 3:1 and 0.15 wt% PEI.