



ELSEVIER

Contents lists available at ScienceDirect

## Journal of the European Ceramic Society

journal homepage: [www.elsevier.com/locate/jeurceramsoc](http://www.elsevier.com/locate/jeurceramsoc)

Original article

## Method for fabricating microwave absorption ceramics with high thermal conductivity

Yongqin He<sup>a,b</sup>, Xiaoyun Li<sup>a</sup>, Jingxian Zhang<sup>c,\*</sup>, Xiaoguang Li<sup>c</sup>, Yusen Duan<sup>c</sup>, Mingming Huang<sup>c</sup>, Hainan Bai<sup>c</sup>, Dongliang Jiang<sup>c</sup>, Tai Qiu<sup>a</sup><sup>a</sup> College of Materials Science and Engineering, Nanjing Tech University, Nanjing 210009, China<sup>b</sup> Jiangsu Collaborative Innovation Center for Advanced Inorganic Function Composites, Nanjing Tech University, Nanjing 210009, China<sup>c</sup> State Key Laboratory of High Performance Ceramics and Superfine Microstructures, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050, China

## ARTICLE INFO

## Keywords:

AlN ceramics  
Phenolic resin  
Microwave absorption  
Thermal conductivity  
Carbon films

## ABSTRACT

In this work, a high-performance microwave absorption ceramic together with high thermal conductivity was proposed through the introduction of phenolic resin as the carbon source into AlN ceramic substrate. The phenolic resin was initially mixed with AlN powder and sintering additives in ethanol to form homogeneous slurries, followed by drying, pyrolysis, dry pressing and pressureless sintering to develop nano carbon in situ in AlN substrate. The well controlled microstructure with homogeneous distribution of nano sized carbon lead to the high thermal conductivity and excellent microwave absorption properties. by adjusting the phenolic resin content, the ceramic showed a minimal reflection coefficient of about  $-30$  dB with an effective bandwidth of about 2 GHz together with a high thermal conductivity of about 135.1 W/m K.

## 1. Introduction

Microwave absorption materials have attracted more and more attention due to their wide application in the stealth technology, telecommunication and vacuum electronic devices [1,2]. It is known that the electromagnetic energy will be turned into heat energy after entering the microwave absorption materials. The heat energy must be dissipated quickly, especially for the high power electronic devices. Otherwise, the high residual heat might affect the conventional operation of devices. Microwave absorption materials based on a beryllium oxide (BeO) matrix have traditionally been the candidate in the high power applications owing to their high thermal conductivities [2]. However, with more and more attention on health and environment, the applications of BeO have been restricted due to its toxicity. In recent years, aluminum nitride (AlN) has been found to be a promising candidate for the matrix of microwave absorption materials due to its high thermal conductivity, non-toxic, low dielectric constant, high electrical resistance and high chemical stability [3,4]. Lots of researches focused on the thermal and dielectric properties of the AlN-based ceramics have been reported, especially the AlN-SiC ceramics [5–8]. Although the intrinsic properties of SiC and the formation of AlN-SiC solid solution are beneficial to the microwave absorption properties, the thermal

conductivity was largely reduced due to the formation of solid solution. Carbon materials are also the candidate for the lossy phase and have attracted more and more attention. The mechanical, electrical and thermal properties of the ceramics consisted of AlN and various carbon materials have been investigated with the introduction of special nano-sized powder, such as carbon nanofiber, carbon nanotube, graphene etc. [9–11]. However, the thermal conductivity of the resulting ceramics was not high, because it is difficult to disperse carbon materials homogeneously in AlN substrate and usually agglomeration are formed, which lead to problems in sintering, microstructure control and also the thermal conductivity [10]. In addition, the microwave absorption properties of these systems were rarely investigated.

Lossy ceramics is a complicated system. Usually nano sized lossy phase is preferred because it will be more effective and the addition content can be reduced compared to big sized one for the same dielectric performance. This is also effective for keeping the high thermal conductivity of lossy ceramics because the addition of second phase will always show negative effect on the thermal conductivity. In addition, less lossy phase will also be beneficial for the densification. However, the dispersion of nano sized lossy phase in AlN substrate is quite difficult. In this paper, phenolic resin, a polymer was proposed as the precursor of nano carbon source for the preparation of the AlN based

\* Corresponding author at: State Key Laboratory of High Performance Ceramics and Superfine Microstructures, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050, China.

E-mail addresses: [15261806381@163.com](mailto:15261806381@163.com) (Y. He), [jxzhang@mail.sic.ac.cn](mailto:jxzhang@mail.sic.ac.cn) (J. Zhang).

<http://dx.doi.org/10.1016/j.jeurceramsoc.2017.09.042>

Received 16 February 2017; Received in revised form 25 September 2017; Accepted 27 September 2017  
0955-2219/ © 2017 Published by Elsevier Ltd.

lossy materials. To the best of our knowledge, the microwave absorption properties of AlN-phenolic resin system have not yet been reported.

This paper aimed at introducing an effective method for fabricating microwave absorption materials with high thermal conductivity. The effects of phenolic resin on the microstructure, thermal, dielectric and microwave absorption properties of the composites were investigated.

## 2. Experimental procedure

AlN powder (Grade F, Tokuyama Corp., Tokyo) and phenolic resin (QiNan Adhesive Material, Shanghai, China) with a carbon yield of about 60 wt.% after pyrolysis were used as the starting materials. 3 wt.%  $Y_2O_3$  (analytical reagents) was used as sintering additive. The composites containing  $x$  wt.% phenolic resin (named as SPR $x$  ( $x = 0, 1.5, 3, 4.5, 6, 6.5$ )) were ball-milled for 4 h with ethanol and zirconia balls in a polypropylene jar. After drying, the mixtures were sieved (100 mesh), uniaxially pressed in a steel die at 20 MPa and then isostatically pressed at 200 MPa. The powder compacts were heated to 900 °C for 3 h for the pyrolysis of phenolic resin and then pressureless-sintered at 1800 °C for 2 h in a flowing nitrogen atmosphere.

The bulk densities of the samples were measured using Archimedes' method. The crystalline phases were analyzed by X-ray diffraction (XRD, D8 Advance, Bruker, Germany) with  $Cu K_{\alpha}$  radiation. The fracture surface of the samples was observed by a scanning electron microscopy (SEM, S-4800, Hitachi, Japan) equipped with energy dispersive spectroscopy (EDS). The carbon content was measured by a carbon sulfur analyzer (CS2000, Eltra GmbH, Germany). Thermal diffusivity was measured by a laser-flash apparatus (LFA427 Nanoflash, Netzsch Instruments Co. Ltd., Selb, Germany) at room temperature. The thermal conductivity was calculated using the heat capacity of pure AlN (0.75 J/g K) [12] and the density of the samples measured in this paper. The complex permittivities of the samples were measured by a network analyzer (VNA, N5225A, Agilent).

## 3. Results and discussions

### 3.1. Densities and phase compositions

Table 1 shows the bulk density and relative density of the samples. It can be seen that the phenolic resin had no remarkable influence on the densification process when the content of it was less than 4.5%. However, with further increase of phenolic resin content to 6%, the relative density decreased from 98.4% to 95.4%. This may be caused by the residual carbon which can inhibit the rearrangement of AlN particles. The gaseous product produced by the pyrolysis of phenolic resin can also restrain the densification process.

The XRD patterns are shown in Fig. 1. It showed that the main phase of all the samples was AlN. The secondary phases were affected by the content of phenolic resin due to the carbothermal reduction as shown in the following:



In the SPR0, the  $Al_2O_3$  layer on the surface of AlN particles was sufficient to react with the  $Y_2O_3$  additive to form the  $Al_5Y_3O_{12}$  (33-0040) phase. With the addition of phenolic resin, the  $Al_2O_3$  layer was

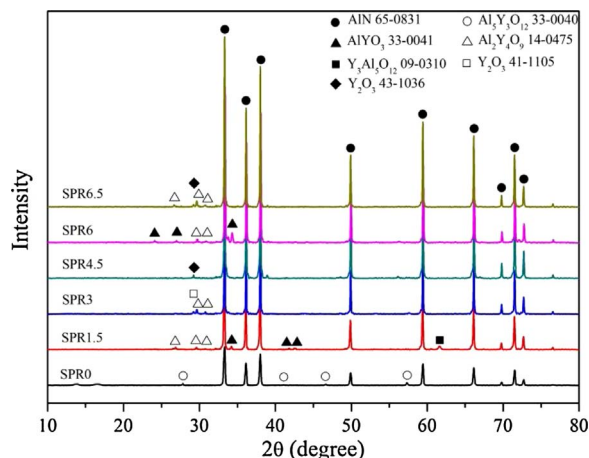


Fig. 1. XRD patterns of the samples with different phenolic resin content sintered at 1800 °C for 2 h in  $N_2$ .

removed by the carbothermal reduction gradually. The secondary phases were transformed to the  $AlYO_3$  (33-0041) phase and the  $Al_2Y_4O_9$  (14-0475) phase in the SPR1.5, the  $Al_2Y_4O_9$  (14-0475) phase and the  $Y_2O_3$  (41-1105) phase in the SPR3 and the  $Y_2O_3$  (43-1036) phase in the SPR4.5. In addition, the change of the secondary phase from the SPR4.5 to the SPR6 may be caused by the pores which can hinder the contact between the  $Al_2O_3$  layer and the residual carbon.

### 3.2. Dielectric properties

The dielectric properties are normally characterized by the relative complex permittivity ( $\epsilon = \epsilon' - j\epsilon''$ ). The real part of permittivity ( $\epsilon'$ ) represents the polarization relaxation, and the imaginary part of permittivity ( $\epsilon''$ ) represents the ability of the dielectric loss. Fig. 2(a) shows the real part of permittivity of the samples. It can be seen that the real part of permittivity showed small changes when the phenolic resin content increased from 0 wt.% to 4.5 wt.% and was close to that of the monolithic AlN whose average value was 8.2 in this study. However, when the phenolic resin increased from 4.5 wt.% to 6 wt.%, the real part of permittivity increased from 8.19 to 15.82, and further increased to 18.93 at 10 GHz when the phenolic resin content was 6.5 wt.%. The imaginary part of permittivity of the samples was shown in Fig. 2(b). Obviously, the variation of the imaginary part of permittivity was consistent with the real part of permittivity. The average imaginary part of permittivity of the samples SPR0-SPR4.5 was below 0.2, which showed limited dielectric loss. When the phenolic resin content increased to 6 wt.%, the average imaginary part of permittivity of SPR6 increased to 4.76. With further addition of phenolic resin, the average imaginary part of permittivity increased to 34.41, which predicted good dielectric loss. Fig. 2(c) shows the dielectric loss of the samples. No obvious dielectric loss was observed when the phenolic resin content increased from 0 wt.% to 4.5 wt.%. However, when the phenolic resin content reached to 6 wt.% and 6.5 wt.%, the average dielectric loss of SPR6 and SPR6.5 jumped up to 0.30 and 1.76. This dielectric properties is comparable to that reported for typical AlN-SiC, AlN-Mo etc. system, reported in literatures [2,13,14]. This might be due to the high microwave absorbing properties of in situ formed nano sized C as shown later.

This interesting phenomenon is related to the microstructure of the samples. When phenolic resin content is below 4.5 wt.%, samples presented typical morphology of AlN ceramics, Fig. 3(a). The grey part was AlN grains and the white part located at the triple junctions and distributed along grain boundaries was the secondary phase, and no obvious pores can be observed in the samples. However, when the phenolic resin content increased to 6 wt.% and above, irregular shaped films located at the triple junctions and filled in the pores were

Table 1

Bulk density and Relative density of the samples.

Sample	Bulk density ( $g/cm^3$ )	Relative density(%)
SPR0	$3.27 \pm 0.02$	$98.8 \pm 0.6$
SPR1.5	$3.26 \pm 0.02$	$98.4 \pm 0.6$
SPR3	$3.21 \pm 0.01$	$97.8 \pm 0.3$
SPR4.5	$3.25 \pm 0.03$	$98.4 \pm 0.9$
SPR6	$3.14 \pm 0.02$	$95.4 \pm 0.6$
SPR6.5	$3.12 \pm 0.01$	$94.6 \pm 0.3$

Download English Version:

<https://daneshyari.com/en/article/7898670>

Download Persian Version:

<https://daneshyari.com/article/7898670>

[Daneshyari.com](https://daneshyari.com)