



Review Article

Development of high-thermal-conductivity silicon nitride ceramics

You Zhou*, Hideki Hyuga, Dai Kusano¹, Yu-ichi Yoshizawa, Tatsuki Ohji, Kiyoshi Hirao

National Institute of Advanced Industrial Science and Technology (AIST), Nagoya 463-8560, Japan

ARTICLE INFO

Article history:

Received 13 January 2015

Received in revised form 23 March 2015

Accepted 23 March 2015

Available online 7 April 2015

Keywords:

Silicon nitride

Thermal conductivity

Reaction-bonding

Nitridation

ABSTRACT

Silicon nitride (Si_3N_4) with high thermal conductivity has emerged as one of the most promising substrate materials for the next-generation power devices. This paper gives an overview on recent developments in preparing high-thermal-conductivity Si_3N_4 by a sintering of reaction-bonded silicon nitride (SRBSN) method. Due to the reduction of lattice oxygen content, the SRBSN ceramics could attain substantially higher thermal conductivities than the Si_3N_4 ceramics prepared by the conventional gas-pressure sintering of silicon nitride (SSN) method. Thermal conductivity could further be improved through increasing the β/α phase ratio during nitridation and enhancing grain growth during post-sintering. Studies on fracture resistance behaviors of the SRBSN ceramics revealed that they possessed high fracture toughness and exhibited obvious *R*-curve behaviors. Using the SRBSN method, a Si_3N_4 with a record-high thermal conductivity of $177 \text{ Wm}^{-1}\text{K}^{-1}$ and a fracture toughness of $11.2 \text{ MPa m}^{1/2}$ was developed. Studies on the influences of two typical metallic impurity elements, Fe and Al, on thermal conductivities of the SRBSN ceramics revealed that the tolerable content limits for the two impurities were different. While 1 wt% of impurity Fe hardly degraded thermal conductivity, only 0.01 wt% of Al caused large decrease in thermal conductivity.

© 2015 The Ceramic Society of Japan and the Korean Ceramic Society. Production and hosting by Elsevier B.V. All rights reserved.

Contents

1. Introduction	221
2. Thermal conductivity of silicon nitride	222
3. High-thermal-conductivity silicon nitride via the SRBSN route	222
4. Fracture Toughness of High-Thermal-Conductivity Silicon Nitride Ceramics	225
5. Effects of metallic impurity elements on thermal conductivity of SRBSN ceramics	226
5.1. Effects of impurity iron	226
5.2. Effects of impurity aluminum	226
6. Summary	227
Acknowledgements	229
References	229

1. Introduction

Energy and environment-related problems are serious social issues. In order to save energy as well as to reduce the emission of carbon dioxide, energy sources tend to shift from fossil

fuel to electric power, hence highly efficient use of electric power becomes more and more important. Power electronic devices are key technologies for this purpose, and they have been widely used for a variety of applications such as industrial robots, hybrid motor vehicles, and advanced electric trains [1]. Driven by the demand for more efficient control and conversion of electric power, power device technology is advancing toward higher voltage, larger current, greater power density, and this trend is poised to be accelerated with the replacement of Si by the wide-band gap semiconductors (SiC and GaN) in the near future [2,3]. However, the high power will induce large thermal stresses in the devices, which poses great challenges for the assembly of the devices and the

* Corresponding author. Tel.: +81 527367102.

E-mail address: you.zhou@aist.go.jp (Y. Zhou).

¹ Japan Fine Ceramics Co., Ltd., Sendai, 981-3203, Japan.

Peer review under responsibility of The Ceramic Society of Japan and the Korean Ceramic Society.

packaging materials, especially the brittle ceramic substrates that provide functions of electrical insulation and heat dissipation. Therefore, both good mechanical reliability and high thermal conductivity are required for the ceramic substrates used for the high-power electronic devices.

So far, AlN has been used as a major ceramic substrate material for power devices because it exhibits high thermal conductivity over $200 \text{ Wm}^{-1} \text{ K}^{-1}$. However, mechanical properties of AlN are not sufficient (generally, bending strength of 300–400 MPa, fracture toughness of 3–4 $\text{MPa m}^{1/2}$), which results in low reliability of the substrates. The electronic industry is eager to seek alternative high-thermal-conductivity substrate materials with good mechanical properties, therefore attention is turned to Si_3N_4 ceramics.

Over the last decades, Si_3N_4 ceramics have been investigated as high temperature structural materials, and now Si_3N_4 with bending strength over 1 GPa can be prepared by choosing appropriate sintering additives and fully developing a bimodal microstructure composed of interlocked rod-like grains [4]. However, thermal conductivity of Si_3N_4 ceramics is usually rather low ($<70 \text{ Wm}^{-1} \text{ K}^{-1}$). If thermal conductivity of Si_3N_4 can be improved and at the mean time its good mechanical properties can be maintained, Si_3N_4 would undoubtedly be an attractive substrate material for high-power electronic device applications. Such an expectation has fascinated many researchers to improve thermal conductivities of Si_3N_4 ceramics by a variety of means in recent years. In this paper, previous studies on thermal conductivity property of Si_3N_4 are briefly reviewed. Following that, some recent achievements of our research group on fabricating high-thermal-conductivity Si_3N_4 ceramics via a reaction-bonding and post-sintering method are introduced.

2. Thermal conductivity of silicon nitride

Si_3N_4 mainly exists in two hexagonal polymorphs, namely α - and β - Si_3N_4 , which are generally regarded as low and high temperature crystal forms, respectively [5]. Industrial synthesis routes mainly lead to α - Si_3N_4 , which converts to the more stable β - Si_3N_4 phase during high temperature sintering. Si_3N_4 is a highly covalent compound, and it transports heat primarily by phonons at room temperature and below. In 1995, Haggerty and Lightfoot [6] predicted that the intrinsic thermal conductivity of Si_3N_4 might be $200\text{--}320 \text{ Wm}^{-1} \text{ K}^{-1}$ at room temperature. Later, Watari et al. [7] estimated that the upper limit of the intrinsic thermal conductivity of β - Si_3N_4 could be $400 \text{ Wm}^{-1} \text{ K}^{-1}$. Moreover, Hirotsuki et al. [8] estimated the theoretical thermal conductivity of single crystal α - and β - Si_3N_4 , using the molecular dynamics method in conjunction with the Green–Kubo formulation, as a function of temperature. In their calculation, the estimated thermal conductivities of α - and β - Si_3N_4 , along the a -axis and c -axis at room temperature were approximately 105 and $225 \text{ Wm}^{-1} \text{ K}^{-1}$, and 170 and $450 \text{ Wm}^{-1} \text{ K}^{-1}$, respectively. However, thermal conductivities of polycrystalline Si_3N_4 ceramics are much lower than the intrinsic values of the single crystal due to the following reasons.

Owing to its strong covalency and low diffusivity, sinterability of Si_3N_4 is poor. Si_3N_4 is generally densified by liquid-phase-sintering mechanisms, where some oxide sintering aids are added and they react with Si_3N_4 as well as the silica phase on the surface of Si_3N_4 particles to form a liquid phase which promotes densification through rearrangement and solution-reprecipitation mechanisms during sintering. After sintering, the liquid phase converts to glassy or partially crystallized phases (oxynitrides) in the sintered material. They may exist as isolated secondary phases at the triple point junctions surrounded by three grains, or as a continuous thin film (around 1 nm thickness) on the boundaries between two adjacent grains. Because thermal conductivities of these oxynitride

secondary phases are quite low (less than $5 \text{ Wm}^{-1} \text{ K}^{-1}$), their existence in the microstructure causes reduction of the thermal conductivity of the sintered material. Compared with the triple-junction phases, the detrimental effect of the grain boundary phases is greater due to its continuity. Kitayama et al. [9] assessed the effects of the grain boundary phases on the overall thermal conductivities of Si_3N_4 ceramics by using a modified Wieners' model, and their calculation indicated that the detrimental effect of the grain boundary phases could be alleviated when the grain sizes of the β - Si_3N_4 grains were larger than several micrometers. Thus, promoting grain growth is an effective way of improving thermal conductivity. However, it should be aware that microstructural coarsening often results in lower mechanical strength of the material.

Besides the secondary and grain boundary phases which reside outside Si_3N_4 grains, there exist a variety of imperfections called lattice defects (impurity atoms, vacancies, dislocations, stacking faults, etc.) within the Si_3N_4 grains. Because Si_3N_4 transports heat primarily by phonon (lattice vibration), lattice defects in Si_3N_4 crystals can induce phonon scattering, thereby reducing thermal conductivity. It has been reported that solution of oxygen into Si_3N_4 crystals generates vacancies at the Si sites in Si_3N_4 lattice [10,11]. Si vacancies can scatter phonons and lead to lower thermal conductivity. In order to improve thermal conductivity, it is essential to lower the content of oxygen dissolved in Si_3N_4 lattice (i.e., lattice oxygen content).

Therefore, choosing a Si_3N_4 powder with low impurity oxygen content as a starting material is decisively important for preparing Si_3N_4 ceramics with high thermal conductivity. Sintering additives, which are indispensable for achieving densification of Si_3N_4 , also play an important role in reducing lattice oxygen content of the sintered Si_3N_4 . Using sintering additives with high oxygen affinity (e.g., rare earth oxides) [7,12], or choosing sintering additive compositions which lead to high nitrogen/oxygen ratios of the liquid phase during sintering were effective ways of reducing lattice oxygen content of the sintered Si_3N_4 . For example, Hayashi et al. [13] reported that while a Si_3N_4 ceramic with a thermal conductivity of $120 \text{ Wm}^{-1} \text{ K}^{-1}$ was prepared by using a Yb_2O_3 -MgO sintering additive, the thermal conductivity could be increased to $140 \text{ Wm}^{-1} \text{ K}^{-1}$ by replacing MgO with MgSiN_2 in the sintering additive so that nitrogen/oxygen ratio of the liquid phase was increased.

However, for Si_3N_4 starting powders, even the top grade high-purity commercial powder contains more than 1 wt% of oxygen residing both on the surface and within the lattice, which imposes an upper limit on the attainable thermal conductivity of Si_3N_4 ceramics. In order to raise the upper limit to a higher level, a starting powder containing less oxygen should be used. Motivated by such an idea, our research group proposed a strategy of preparing high thermal conductivity Si_3N_4 via a route of sintering of reaction-bonded silicon nitride (SRBSN), which is a well-known process of fabricating Si_3N_4 ceramics from an Si starting powder instead of a Si_3N_4 powder [14–17], in consideration of the fact that Si powders containing much less oxygen and metallic impurities than Si_3N_4 powders are commercially available thanks to the advancement of modern semiconductor industry. Our experimental results have verified the effectiveness of the SRBSN strategy, by which both higher thermal conductivity and higher mechanical strength were achieved, compared to the conventional sintering of silicon nitride powder (SSN) route [18–28].

3. High-thermal-conductivity silicon nitride via the SRBSN route

The sintering of reaction-bonded silicon nitride (SRBSN) method consists of two processing steps. Firstly, an Si compact composed of Si powder and sintering additives is heated to a temperature

Download English Version:

<https://daneshyari.com/en/article/1473258>

Download Persian Version:

<https://daneshyari.com/article/1473258>

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