



PHYSICAL CHEMISTRY 2006

Proceedings

*of the 8th International Conference
on Fundamental and Applied Aspects of
Physical Chemistry*

September 26-29,
Belgrade, Serbia

ISBN 86-82139-26-X
Title: Physical Chemistry 2006. (Proceedings)
Editors Prof. dr A. Antić-Jovanović
Published by: The Society of Physical Chemists of Serbia, Studentski trg 12-16, P.O.Box 137, 11001 Belgrade, Serbia
Publisher: Society of Physical Chemists of Serbia
For publisher: Prof. dr S. Anić, president of the Society of Physical Chemists of Serbia
Printed by: "Jovan" Printing and Published Comp;
250 Copies; Number of Pages: x + 442; Format B5;
Printing finished in September 2006.
Text and Layout: Aleksandar Nikolić
250 – copy printing

SYNTHESIS OF CRYSTALLINE SILICON OXYNITRIDE COMPOSITES

A. Egelja, J. Dukić, S. Bošković, A. Radosavljević-Mihajlović and B. Matović

Vinča Institute of Nuclear Sciences, Material Science Lab., Belgrade, Serbia

Abstract

Silicon oxynitride / silicon nitride ($\text{Si}_2\text{N}_2\text{O}/\text{Si}_3\text{N}_4$) ceramics have been prepared from Si_3N_4 powder and amorphous silica (SiO_2) by hot pressing at different temperature. It was found that material sintered at lower temperature exhibit fine composite structure composed of equiaxed $\alpha\text{-Si}_3\text{N}_4$ grains and $\text{Si}_2\text{N}_2\text{O}$ crystals. At higher temperature the growing of $\text{Si}_2\text{N}_2\text{O}$ particles as well as phase transformation from $\alpha\text{-Si}_3\text{N}_4$ to $\beta\text{-Si}_3\text{N}_4$ phase take place.

Introduction

The ceramic silicon oxynitride ($\text{Si}_2\text{N}_2\text{O}$) has for the most part been considered as an inconvenient and undesirable by-product sometimes formed during the production of Si_3N_4 . However, in recent years the applications of $\text{Si}_2\text{N}_2\text{O}$ as a structural ceramic has begun to be considered owing to the excellent high temperature properties and chemical inertness of the material [1]. The refractory behavior of the $\text{Si}_2\text{N}_2\text{O}$ is explained by much better stability in oxidizing environments at high temperature than the Si_3N_4 material [2].

A convenient method for the formation of the $\text{Si}_2\text{N}_2\text{O}$ has yet to be established and it is for this reason that researchers have examined many alternative routes for its production. In this work $\text{Si}_2\text{N}_2\text{O}$ materials have been prepared by direct reaction between the two components:



It is well known that reaction (1) is very sluggish and sintering additives like Al_2O_3 and Y_2O_3 have to be added in order to enhance the reaction rate by forming a liquid phase [3]. As a silica source, usually is used crystalline quartz, however we have used amorphous silica powder.

The intention of this paper is to study the sintering reaction between Si_3N_4 and very reactive amorphous silica without using additive.

Experimental

Starting material for this investigation was a mixture of SiO_2 and Si_3N_4 (UBE) with the quantitative ratio 1:1. This mixture was treated in the vibratory mill for 2 h in the presence of liquid ethanol. The green bodies were heated by hot pressing in the graphite mould with the applied pressure of 25 MPa at 1400°C/4 h for the first sample and 1750 °C/1 h for the second one. The reaction products were analyzed by XRD using diffractometer with $\text{CuK}\alpha$ as the target., while mechanical characterization included

Vickers hardness and toughness measurements. Density was determined by standard Archimedes method. Micrographs were obtained with a Philips scanning electron microscope (SEM).

Results and discussion

Fig. 1. shows the evolution of the crystalline phases formed as a function of the reaction temperature. At low temperature (1400 °C) there are two phases: $\text{Si}_2\text{N}_2\text{O}$ and $\alpha\text{-Si}_3\text{N}_4$ with 30 wt% and 70 wt% respectively. This indicates that $\text{Si}_2\text{N}_2\text{O}$ is obtained by direct reaction and without intermediate stages. At higher sintering temperature (1750 °C) the phase transformation from $\alpha\text{-Si}_3\text{N}_4$ to $\beta\text{-Si}_3\text{N}_4$ phase has already taken place. The amount of $\text{Si}_2\text{N}_2\text{O}$ slightly increases whilst the intensity of $\alpha\text{-Si}_3\text{N}_4$ is significantly reduced.

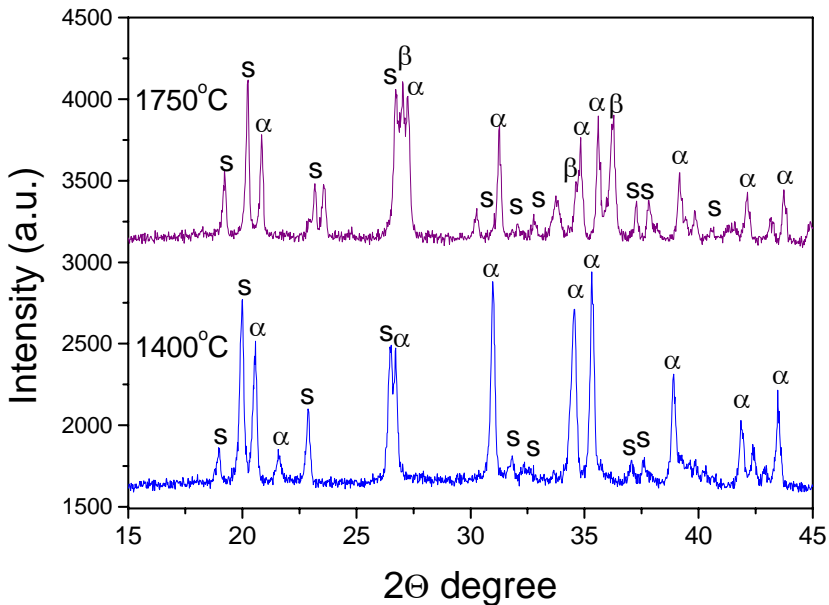


Fig. 1. XRD patterns of samples hot-pressed at 1400 °C and 1750 °C.
S- $\text{Si}_2\text{N}_2\text{O}$, $\alpha\text{-Si}_3\text{N}_4$, $\beta\text{-Si}_3\text{N}_4$.

The microstructure of ceramics sintered is shown in Fig. 2. Material sintered at 1400 °C exhibits fine and equiaxed $\alpha\text{-Si}_3\text{N}_4$ grains and very well formed crystals of silicon oxynitride having polyhedral form, which is combination of forms in trigonal system. With increasing sintering temperature the size of the silicon oxynitride particles increased. In the other side, morphology of the growing silicon nitride particles is changed significantly from equiaxed to elongated due to phase transformation from $\alpha\text{-Si}_3\text{N}_4$ to $\beta\text{-Si}_3\text{N}_4$ phase.

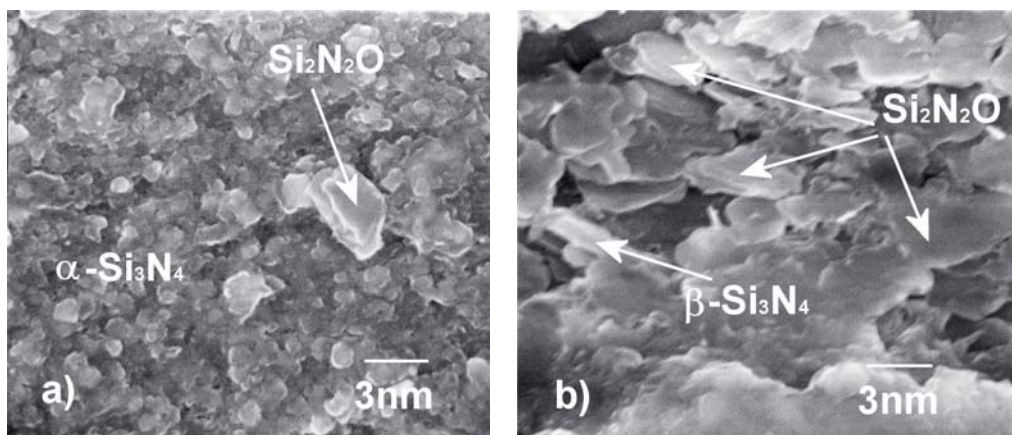


Fig. 2. SEM image of polished and chemical-etched cross section of hot-pressed $\text{Si}_2\text{N}_2\text{O}$ composites produced from the mixture of $\text{Si}_3\text{N}_4/\text{SiO}_2$ at 1400 °C (a) and 1750 °C (b), respectively. The arrows show $\text{Si}_2\text{N}_2\text{O}$ plate crystals, $\beta\text{-Si}_3\text{N}_4$ elongated grains and $\alpha\text{-Si}_3\text{N}_4$ rounded grains.

Conclusion

On the basis of the experimental results, it can be concluded that at low temperature the reaction products are mixture of different oxides and $\text{Si}_2\text{N}_2\text{O}$ with morphology that exhibit well-defined crystal geometry. Increasing temperature, the amount of $\text{Si}_2\text{N}_2\text{O}$ slightly increases whilst the phase transformation from $\alpha\text{-Si}_3\text{N}_4$ to $\beta\text{-Si}_3\text{N}_4$ phase takes place.

Acknowledgement

We acknowledge the support of the project No. 142016 by the MSEP of Serbia.

References

- [1] M. Washburn, *Ceram. Bul.*, 1967, **7**, 667-671.
- [2] D. Park, H. Choi, B. Han, H. Kim, D. Lim, *J. Mater. Res.*, 2002, **9**, 2275-2280.
- [3] H. Emoto, M. Mitomo, C. Wang, *J. Eur. Ceram. Soc.*, 1998, **18**, 527-533.