*Key Engineering Materials Vols. 189-191 (2001) pp 516-521*

*doi:10.4028/www.scientific.net/KEM.189-191.516*

# Study of SiC-SiAlON Composite Formation by Nitridation of SiC-Al62.5Si Mixture

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**Keywords:** Mechanical Alloying, Nitridation Composite, SiC-SiAlON Composite

**Abstract.** SiC-SiAlON composite formation was achieved by means of nitridation in a furnace with graphite heating element. SiC and Al62.5Si alloy powders with broad grain size distribution were used as raw materials. After mixing, the powders were compacted into cylindrical green bodies of 25mm diameter and 10 mm height by uniaxial pressing with subsequent cold isostatic pressing. Green densities of about 2.35 g/cm$^3$ were achieved. Nitridation was carried out in stagnant nitrogen atmosphere under normal pressure (0.1 MPa). Heating rate was 20 $^0$C/min up to 1000 $^0$C, and 10 $^0$C/min up to 1400 $^0$C. Soaking time at 1400 $^0$C was 8 hours. The composite obtained was characterized by scanning electron microscopy, energy dispersive spectroscopy, and X-ray diffraction.

## Introduction

SiC-SiAlON ceramic composites have a high potential for application at high temperatures due to their inherently good mechanical, chemical and thermal properties [1-3]. Manufacturing of such composites requires reliable control of parameters such as temperature, atmosphere, time of nitridation and also of initial composition. These parameters have strong influence on the composite formation.

The understanding of the nitridation mechanism of aluminum-silicon metallic alloy is of great importance for further development of SiC-SiAlON refractory composites. The SiAlON solid solution would be formed in a mixture with SiC due to the reaction between Al-62.5 Si alloy and nitrogen ($N_2$) during heat treatment at high temperatures in a graphite resistance furnace.

Various processing routes were used to obtain SiC-SiAlON composite [ 3 ]. However, direct nitridation of aluminum-silicon metallic alloy as a processing route for β-SiAlON bonded SiC formation has not been reported.

The purpose of this investigation is to develop the SiC-SiAlON refractory composite using the direct nitridation of Al-62.5Si (wt% ) in a mixture with SiC powders with broad particle size distribution, and to determine the composition of β-SiAlON phase formed, which may be described as SiC-β-$Si_{6-z}Al_zO_zN_{8-z}$, where $0 \leq z \leq 4.2$.

$Si_{6-z}Al_zO_zN_{8-z}$ is the general formula for the β-SiAlON solid solutions series extended along the line with a constant metal-to-nonmetal atomic ratio, ( Si, Al ):( O, N ). Such solid solutions have a crystal structure similar to β-$Si_3N_4$, as reported by several authors [ 4-6 ].

**Experimental Procedure**

Commercial powder of α-SiC (CASIL S.A.) and Al-62.5Si (wt.%) metallic alloy were prepared by mechanical alloying technique. Powders of Al (BELGO BRASILEIRA S.A.) and Si (LIASA) were used for metallic alloy preparation. The alloy obtained by high energy milling was characterized for granulometric composition on a Micromeritecs Sedigraph 5100 and by scanning electron microscopy ( SEM ) performed on Philips XL30 scanning electron microscope. The resulting phase composition was investigated by X-ray diffraction (XRD) on a Rigaku DMAX 2000 diffractomer. The overall composition of the prepared starting powder mixture was formulated as 25 ( wt%) Al62.5Si and 75 (wt%) α-SiC with broad particle size distribution ( 21-336 μm ). The powders of Al-62.5Si alloy and of α-SiC were mixed for 30 min together with phenolic resin in a ball mill. The powder mixture was subsequently uniaxially and cold isostatically pressed at 150 MPa and 200 MPa, respectively. The green density was calculated using geometrical method. The green bodies were then heated in a furnace with a graphite-heating element under nitrogen atmosphere ( normal pressure of 0.1 MPa ). Heating rate up to 1000 $^0$C was 20 $^0$C/min and 10$^0$C/min up to 1400 $^0$C. Soaking time at 1400 $^0$C was 8 h. Weight gain ( $w_l$) was determined after heat treatment. Final density ( $\rho_f$ ) was measured on a gas multivolume picnometer Micromeritics 1305. Porosity ( $\phi$ ) was calculated according to [7]

$$\phi\,(\%) = [\,w_l/\rho_f\,] \times 100 / V_c \qquad (1)$$

Where :

| | | | |
|---|---|---|---|
| $\phi$ | = | porosity | ( % ) |
| $w_l$ | = | weight | ( g ) |
| $\rho_f$ | = | final density | ( g/cm$^3$ ) |
| $V_c$ | = | volume | ( cm$^3$ ) |

The microstructure was investigated by scanning electron microscopy ( SEM Philips XL30 ) on polished surfaces. The energy dispersive spectroscopy ( EDS ) analysis performed on EDAX DX-AUTO was used for chemical composition determination. X-ray diffraction ( Rigaku DMAX 2000 diffractometer ) was used for phase composition characterization.

**Results and Discussion**

The particle size distribution of Al-62.5Si alloy determined by sedimentation analysis is presented in Fig.1. The results show that the Al-62.5Si alloy powder obtained by high energy ball milling has mean particles size of 3μm. As can be seen from Fig.2, the powder consists of very small uniform particles with apparent agglomerates.

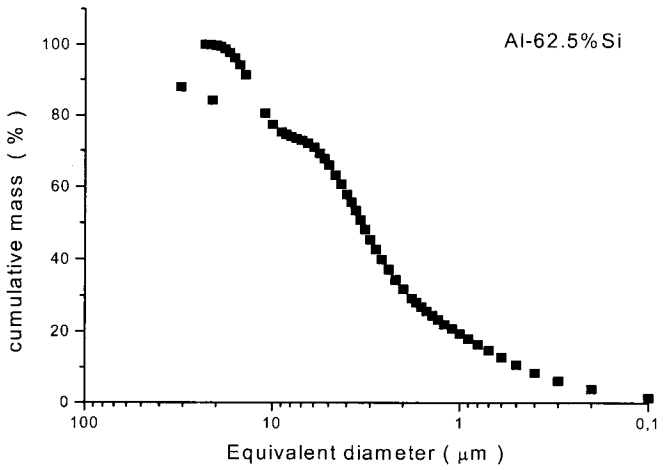


Fig. 1 – Particle size distribution of Al-62.5Si alloy powder produced by high energy ball milling for 15h.

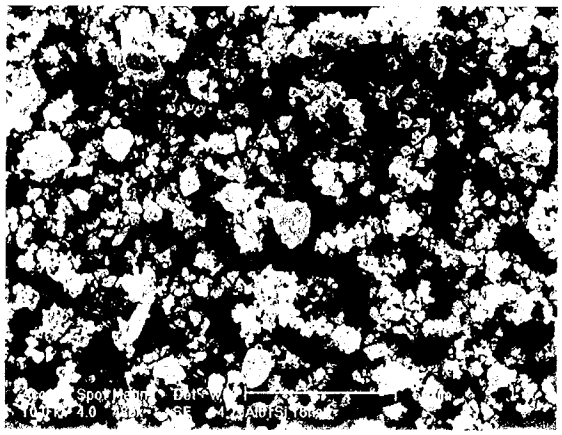

Fig.2 - SEM micrograph of Al-62.5Si alloy powder produced by high energy ball milling for 15h

The Al-62.5Si alloy powder milled for 15 h was analyzed by X ray diffraction. The results are presented in Fig.3, where the presence of silicon and aluminum can be clearly seen.

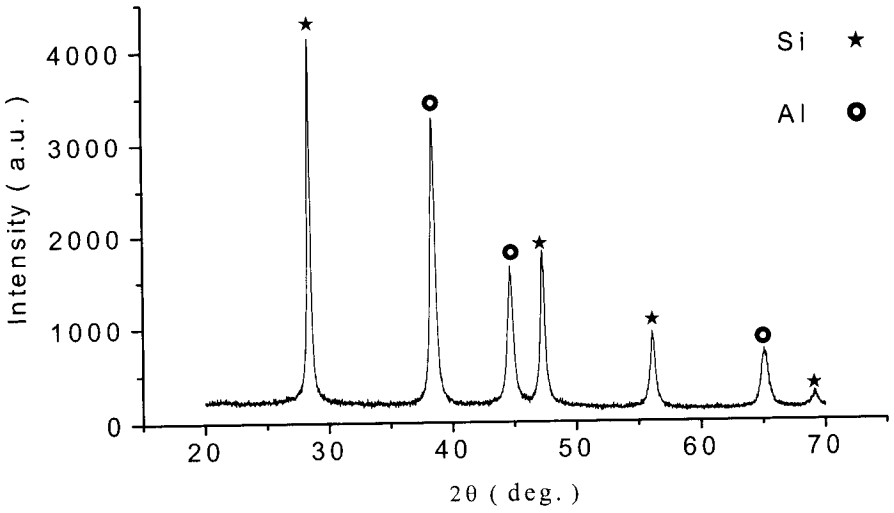


Fig.3 X-ray diffraction pattern for the Al-62.5Si alloy milled for 15h

The average green densities were determined to be 2.35 g/cm$^3$. Average final densities were up to 68% of theoretical. The final porosity of the material after nitration at 1400°C according to microstructural analysis was up to 15%.

The micrograph of the composite polished surface obtained by SEM is shown in Fig.4. The microstuctural analysis revealed the presence of a continuous β-SiAlON phase at the SiC grain boundaries, that appears as a light colored structural component. The SiC grains are embedded in a β-SiAlON continuous matrix, thus forming a ceramic composite. The corresponding EDS spectrum presented in Fig.5 was taken from the region between SiC grains. The presence of aluminum and silicon was detected. It can be noted that the Al content is higher than that of Si. This result confirms the presence of β-SiAlON phase in the light-colored regions.

Fig.4 – SEM micrograph of SiC-SiAlON polished surface

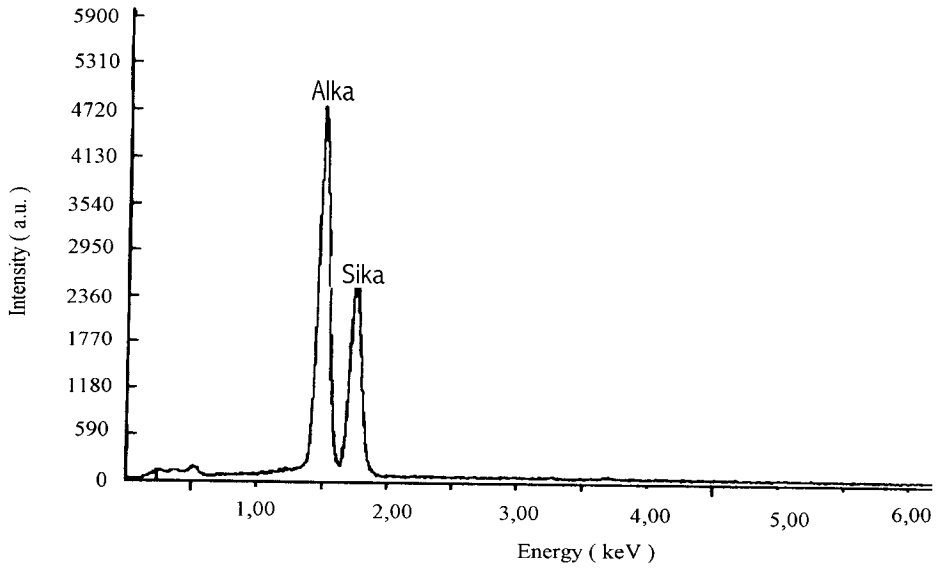


Fig.5 – EDS spectrum taken from region between SiC grain in Fig.4

The X ray diffractogram of the SiC-β-SiAlON composite synthesized at 1400°C is presented in Fig. 6. The crystalline phases present in the material were identified as 2H-SiC and β-SiAlON.

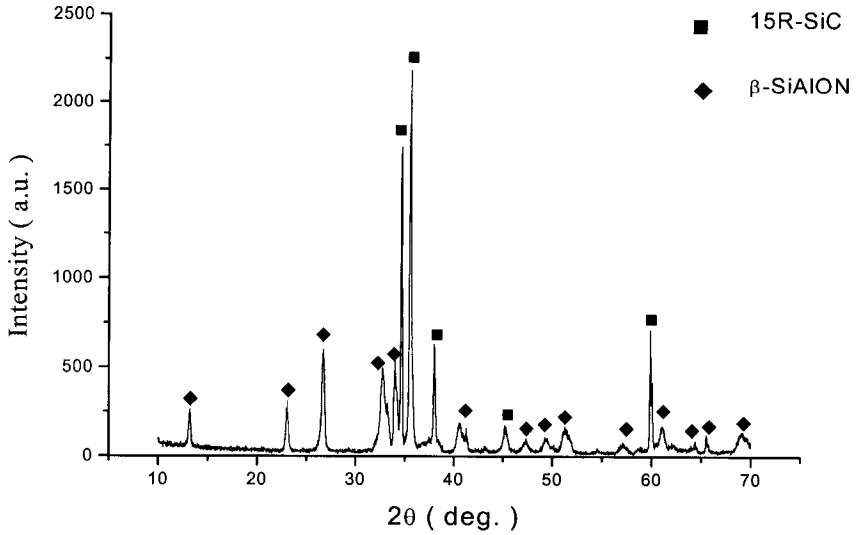


Fig. 6 - X ray diffraction patterns of SiC-SiAlON composite synthesized at $1400^0$C for 8h

X-ray data were used to determine the solid solution substitution coefficient z from the $\beta$-$Si_{6-z}Al_zO_zN_{8-z}$ formula. The values of interplanar distances (d values) for $\beta$-$Si_3N_4$ from JCPDS File 36-

1333 were compared with the experimental values calculated from the experimental data for 2θ. The lattice parameter refinement by least-square refinement software of Benoit [7], which is a PC version of the well-kown Appelman and Evans software [8], produced the values of the lattice parameters a = 7.671 and c = 2.966

Based on the existing published data for the unit cell dimension of β′-$Si_{6-z}Al_zO_zN_{8-z}$ as a function of solid solution [9,10] composition and considering the calculated lattice parameters of the β-SiAlON phase formed in our composite, z value was found as z = 2.2 Consequently, the composition of the composite was determined as SiC-β-$Si_{3.8}Al_{2.2}O_{2.2}N_{5.8}$

## Conclusions

The possibility of a SiC-β-SiAlON ceramic composite synthesis by means of high-temperature nitridation of a powder mixture of SiC and Al62.5Si alloy was shown. The synthesized composite was stable up to 1400$^0$C in nitrogen. β-SiAlON phase formation was confirmed by SEM and XRD characterization. The investigated processing route can be successfully used for advanced refractory materials manufacturing.

## Acknowledgements

The authors thank CNPq for financial support. They also wish to thank Dr. Vassyli A. Izhevskyi for helpful discussions.

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**Advanced Powder Technology II**
doi:10.4028/www.scientific.net/KEM.189-191

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doi:10.4028/www.scientific.net/KEM.189-191.516