LOW COST IN-SITU FORMATION OF AIN - β - SiC

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Abstract

AlN-β-SiC composite powders have been prepared by in-situ high temperature reaction process. By appropriate mixing of Charred Rice Husk (CRH) and aluminium powder, samples containing 30-70% AlN (balance SiC) have been produced. Nitridation has been done by high purity N_2 gas at temperatures ranging from 1400-1700°C. Amorphous SiO_2 and carbon present in CRH have been utilized for the formation of β-SiC.

At about 1550°C the reaction products are only β-SiC and AlN, whereas, below 1550°C other phases, like α-Al_2O_3, Al, Si are the reaction products. It has also been observed that above 1600°C SiC starts going into the solid solution of AlN. The final products have been identified and characterized by XRD, SEM and EDX analysis.

Introduction

Aluminium nitride (AlN) is an excellent ceramic material. Its high electrical resistivity (>10^3 ohm-cm) and high thermal conductivity (>200 W/m K^{-1}) [1] make it uniquely suitable for electronic applications. Thermal expansivity of AlN (≈4.3 x 10^{-6}/K at RT) is quite similar to Si and it has a good wettability character with Si [2] and has a low dielectric constant (K = 8.9 at 1 MHz at RT)[1]. All these unique properties make AlN as an important material for substrate hybrid, power circuits etc. When high pressure is applied, AlN undergoes a brittle - to - ductile transition and is able to absorb high level of energy [3]. As a result AlN finds application as armour materials [4]. Since AlN does not react with molten Al and mostly unaffected by molten ferrous alloys - it has been considered as high performance refractory materials in these area [1,5]. Because of high thermal conductivity AlN has a large market in high-heat conductive plastic package application [6].

However, the widespread use of AlN has been restricted because of its modest mechanical properties [7], like poor oxidation characteristics and also production of finely divided AlN is expensive [5]. Hence, to make use of this potential materials in the field of structural applications improvement of strength and toughness are needed at a reasonable cost of production.

At present SiC has been considered to be most popular and effective material for improving mechanical properties of different composite materials [8]. SiC has high elastic modulus, excellent wear behavior, density very close to AlN and quite high thermal conductivity.

Inspite of above mentioned excellent properties of both AlN and SiC, until recent years, very little attention has been given on the development of AlN-SiC composites. These unique covalent ceramics are able to form a wide range of solid solutions. The range depends on raw materials and processing temperatures [9]. Above 2100°C a nearly composite range of solid solutions are possible [9]. It has also been reported that this solid solution formation is quite slow [10].

In this work attempt has been to develop an insitu process to produce simultaneously AlN and SiC. Since one of the raw materials is a waste product, i.e., Rice Husk, the overall cost of product will be low. Depending on the amount of raw materials one can produce products like 70 AlN - 30 SiC and 30 AlN - 70 SiC.

Experimental Procedure

Rice husk was chosen for the formation of SiC [11] and aluminium powder (-150 mesh) for AlN. At first rice husk was cleaned with 1N HCl, 0.5N NaOH and distilled water. Cleaned rice husk was dried at about 100°C for 6-8 h. Volatile matters like moisture, rice-bran oil etc. were removed from rice husk by heating at about 400°C in inert atmosphere, the heated product is termed as CRH.

Definite proportions of CRH and aluminium powder were mixed thoroughly in a ball-mill. A stainless steel container and steel balls were used for milling. The milled mixture was mixed with 5% (wt/vol) of 5% PVA solution and compacted under 2 Ton pressure. The 20 mm diameter tablets were then subjected to heat treatment at various temperatures ranging from 1400 to 1700°C in nitrogen flowing atmosphere in a graphite resistance furnace (ASTRO 2000, USA). Excess carbon of the reacted product was removed by heating at about 700°C in oxygen atmosphere.

The final products were examined by Scanning Electron microscope (SEM), EDS (JEOL 800, Japan, Kevex, USA) and XRD (SIMENS D500, Germany).

Results and Discussion

In this study, it has been possible to produce samples like 70% AlN-30% β-SiC, as well as 30% AlN-70% β-SiC. Since the overall nature of reactions are same in all cases, for discussion the results of 70AlN-30 β-SiC are only dealt with here.

XRD analyses show that the formation of both AlN and β-SiC start around 1300°C. Formation of AlN accelerates at about 1400°C, but along with β-SiC and AlN, α-Al_2O_3, Si and unreacted Al are also found in the reaction product, Fig.1. The formation of these products can be attributed to the following reactions :

$$SiO_2 + C \longrightarrow SiC + 2CO$$
$$3SiO_2 + 4Al \longrightarrow 3Si + 2Al_2O_3$$
$$Al_2O_3 + C + N_2 \longrightarrow 2AlN + 3CO$$
$$2Al + N_2 \longrightarrow 2AlN$$
$$Si + C \longrightarrow SiC$$

All the unreacted Al reacts with N_2 at 1475°C to form AlN, but still some α-Al_2O_3 and Si remain in the final product till the temperature reaches 1550°C. XRD result of the product obtained at 1550°C shows the presence of optimum temperature to get only β-SiC and AlN, Fig.3. Again above 1550°C the cubic β-SiC starts changing to hexagonal α-SiC, Fig.4 (at 1620°C). It is interesting to note that at about 1700°C α-SiC which had been formed before this temperature, went into the solid solution in AlN [12]. As a result, XRD analysis of the product formed at 1710°C shows only AlN and β-SiC, Fig.5. This clearly shows that only hexagonal α-SiC goes into the solid solution in hexagonal AlN. Therefore, along with AlN only the unchanged β-SiC can be detected in XRD results. At about 1700°C α-SiC starts forming and dissolves into the solid solution : apparently this temperature (1700°C) is lower than the reported one [13]. The reasons may be SiO_2 in CRH is in amorphous condition, the reactants, i.e. SiO_2, C and Al are in intimate contact with each other and the particle sizes of products i.e. AlN and β-SiC are very fine, some cases they are in submicron sizes, Fig.6.

The reaction between SiC and AlN has also been confirmed by SEM studies. During the formation of β-SiC a good amount of β-SiC whiskers are also formed along with particulates, Fig.6. As the reaction temperature increases beyond 1620°C, the β-SiC whiskers start disappearing - indicating transformation of β to α-form and then solid solution formation with AlN, Fig.7. But the overall percentages of Al and Si remain more-or-less same with what it was added in the starting mixture, Fig.8.

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TABLE - 1
Reaction products obtained at various temperatures

Temp.(°C)	AlN	β-SiC	α-SiC	α-Al_2O_3	Si	Al
1315	minor	minor	nil	minor	minor	major
1405	minor	minor	nil	minor	minor	major
1475	major	minor	nil	minor	minor	nil
1550	major	major	nil	nil	nil	nil
1620	major	major	major	nil	nil	nil
1710	major	major	nil	nil	nil	nil

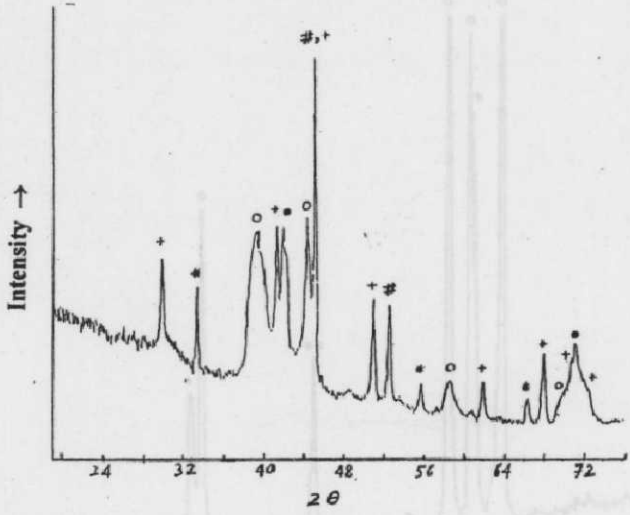

Fig. 1 - XRD Analysis of AlN-SiC prepared at 1404 °C
o : AlN, + : α-Al_2O_3, ● : β-SiC, # : Al, ★ : Si

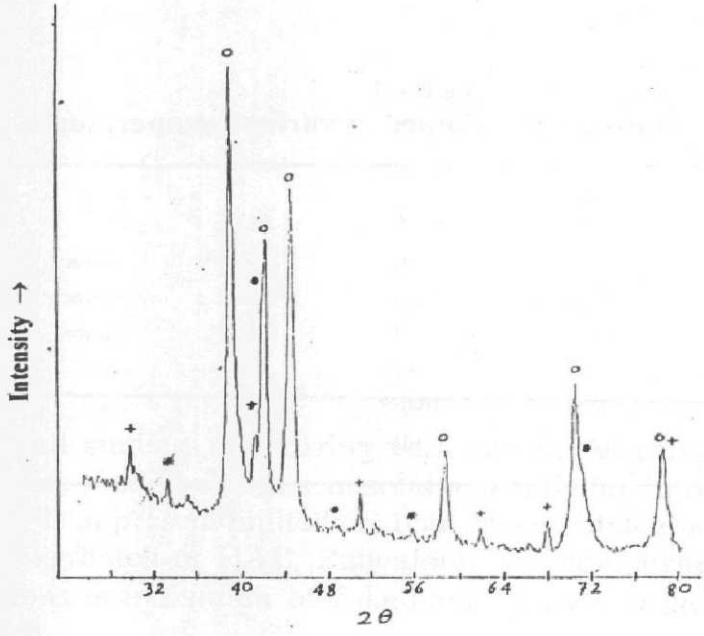

Fig. 2 - XRD Analysis of AlN-SiC prepared at 1475 °C
o : AlN, + : α-Al_2O_3, ● : β-SiC, ★ : Si

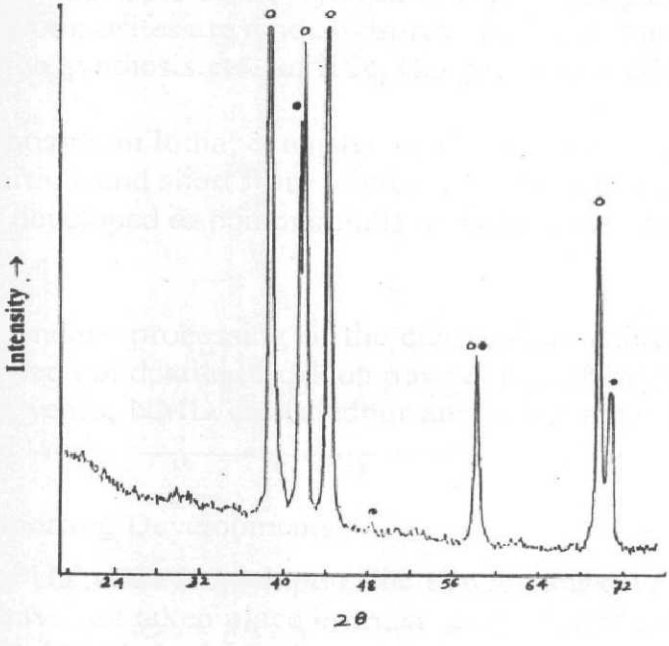

Fig. 3 - XRD Analysis of AlN-SiC prepared at 1550 °C
o : AlN, ● : β-SiC.

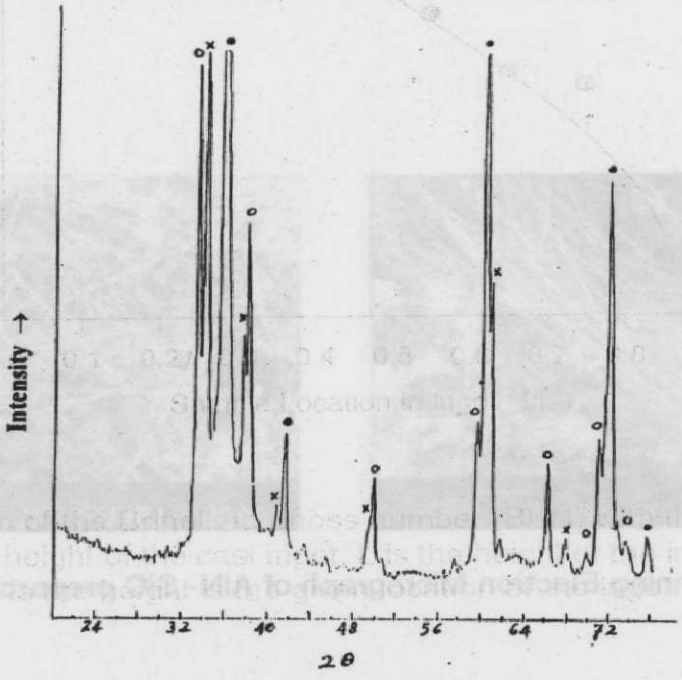

Fig. 4 - XRD Analysis of AIN-SiC prepared at 1620 °C
o : AIN, ● : β-SiC, x : α–SiC

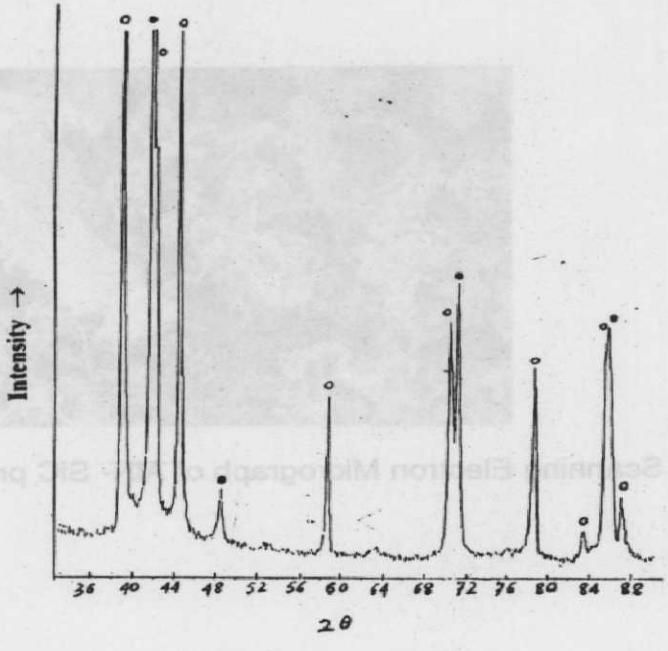

Fig. 5 - XRD Analysis of AIN-SiC prepared at 1710 °C
o : AIN, ● : β-SiC.

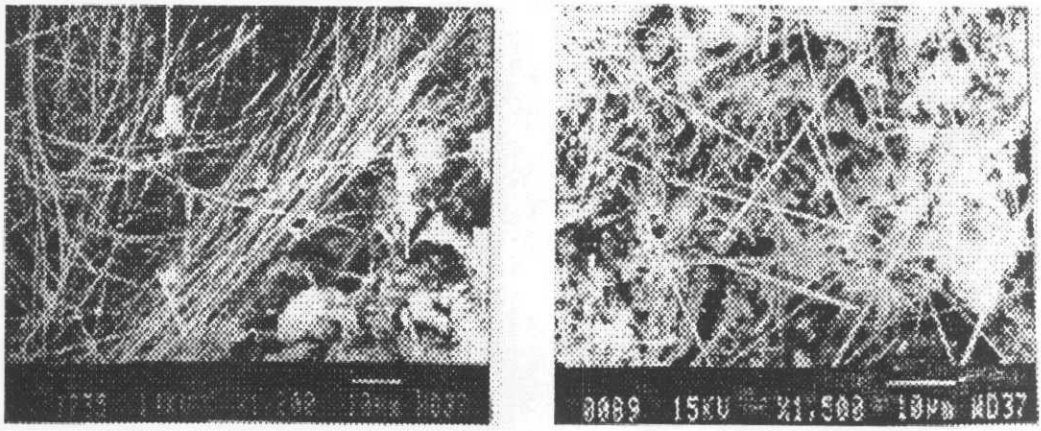

Fig. 6 - Scanning Electron Micrograph of AlN- SiC prepared at 1550° C

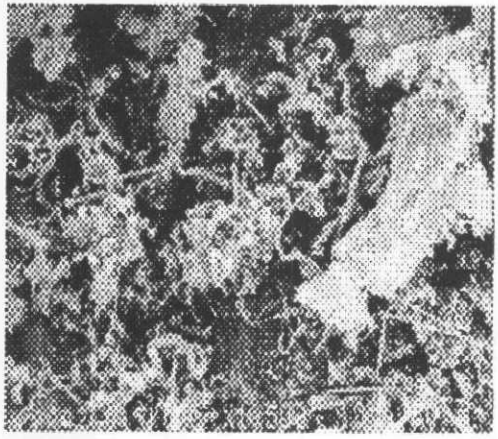

Fig. 7 - Scanning Electron Micrograph of AlN- SiC prepared at 1710° C

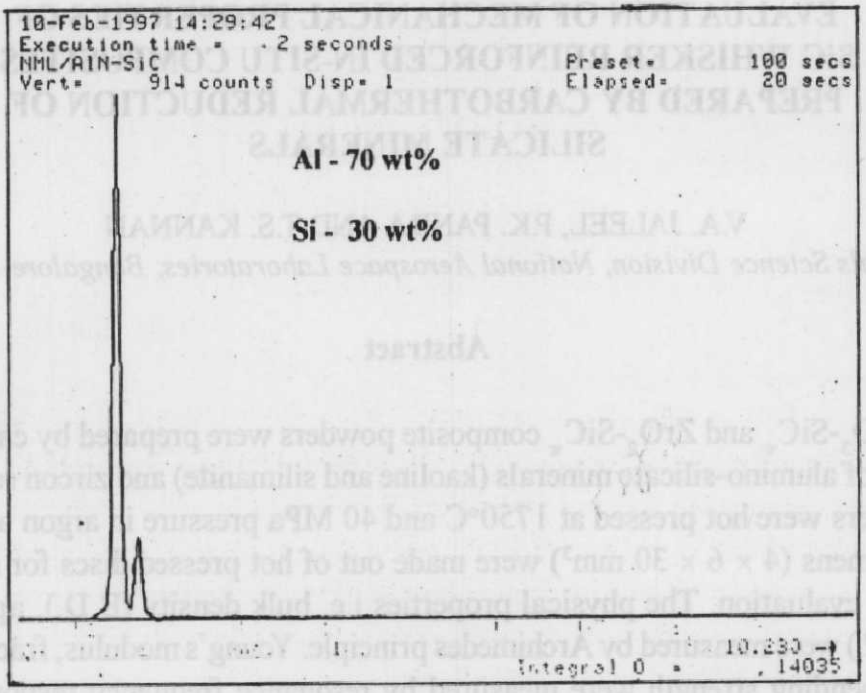

Fig. 8. EDX Analysis of AlN-SiC