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Synthesis of Si_2N_2O-SiC Composites from Desert Sand†

RADWAN Mohamed* and MIYAMOTO Yoshinari**

Abstract

Si_2N_2O-SiC composite powders in various compositions were synthesized by the nitriding combustion of the mixtures of natural desert sand, reclaimed silicon and graphite. Qualitative and quantitative analysis of the composites' phases were made from the x-ray diffraction patterns. Their sintering behavior was observed by densification at 1600 °C for 2 minutes using the new spark plasma sintering technique.

KEY WORDS: (Combustion synthesis) (SHS) (Si_2N_2O) (SiC) (Composite) (Sand)

1. Introduction

Desert sand is a unique natural raw material because it is abundant, cheap and pure in the earth's crust. This fact attracted us to investigate the synthesis of useful materials that can result from its combustion in nitrogen gas. The combustion synthesis[1] method is an advanced process used successfully for preparation of a variety of refractories and ceramics such as borides, nitrides, carbides, and silicides[2-4]. It depends on the strong exothermic heat formation from the chemical reaction which propagates spontaneously and rapidly through the reactants converting them into a product. Simple equipment, high-energy efficiency, and high synthesis rate are its main advantages.

From natural sand, we prepared powders of Si_2N_2O-SiC and Si_2N_2O-Si_3N_4 composites besides the monolithic Si_2N_2O. This paper reports the results of the combustion synthesis of the Si_2N_2O-SiC composite powders.

2. Experimental Procedure

Untreated desert sand (99.7 % purity) from Egypt, reclaimed silicon (94 % purity) from Toho Zinc Co. Ltd., Japan, and graphite (99.7 % purity) were used as the raw materials. The starting compositions were determined by the following chemical equation:

$$(1\text{-}Y)[3(1\text{-}X)/2Si + (1\text{-}X)/2SiO_2 + XSi_2N_2O + (1\text{-}X)N_2] + Y(Si + C) = (1\text{-}Y)Si_2N_2O + YSiC$$

The mixing ratios X and Y used and the nominal product compositions (in molar ratios) are given in **Table 1**. The starting mixtures preparation and the combustion synthesis were the same as described in our previous report[5].

Qualitative and quantitative phase determination in the synthesized powders was performed by X-ray powder diffraction analysis with Cu-$K\alpha$ radiation.

The sintering experiments were carried out at constant conditions (1600 °C sintering temperature in 400 °C/min heating rate and 2 minutes holding time under 30 MPa sintering pressure) using Dr. Sinter® Model 1050 SPS apparatus (Sumitomo Coal Mining Company, Ltd., Japan). The additive of CaO-Al_2O_3 (5 wt%) was used as a sintering-aid.

Table 1 Mixing ratios of starting reactants and nominal composition of the products.

Y	*X*	*Calculated composition*	*Si_2ON_2 (wt%)*	*SiC (wt%)*
0.25	0.075	$3Si_2ON_2 + SiC$	88.2	11.8
0.33	0.075	$2Si_2ON_2 + SiC$	83.3	16.7
0.5	0.075	$Si_2ON_2 + SiC$	71.4	28.6
0.67	0.075	$Si_2ON_2 + 2SiC$	55.6	44.4

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3. Results and Discussion

The change in the formation enthalpy of the synthesis reaction at various Y ratios is shown in **Fig. 1**. Increasing the SiC content in the composite products tends to decrease the exothermicity of the starting reactants. This trend was reflected on their corresponding temperature profiles, **Fig. 2**, which were measured during the combustion by using a thermocouple placed halfway in the burden. The combustion temperature was decreased from 1868 °C to 1600 °C when the SiC content was increased from 19.7 wt% to 57.6 wt%. But, starting mixtures with Y greater than 0.67 failed to sustain reaction in the self-propagation mode. Among the reasons are the low exothermicity of these mixtures and the high volume of graphite in the reactants which may prevent good contacts between Si particles and N_2. The thermodynamic calculations suggest that in such mixtures, the principal reaction is the nitriding of Si and the heat released by this reaction can promote further reactions of Si_2N_2O and SiC formation.

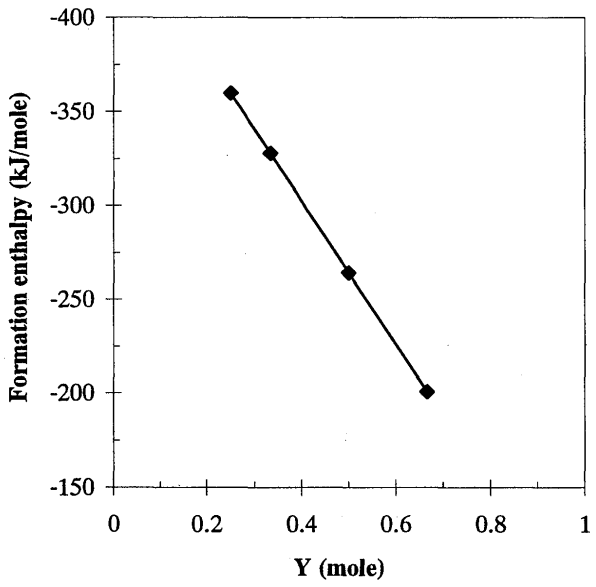

Fig. 1 Formation enthalpy of Si_2N_2O-SiC synthesis reactions at different mixing ratios.

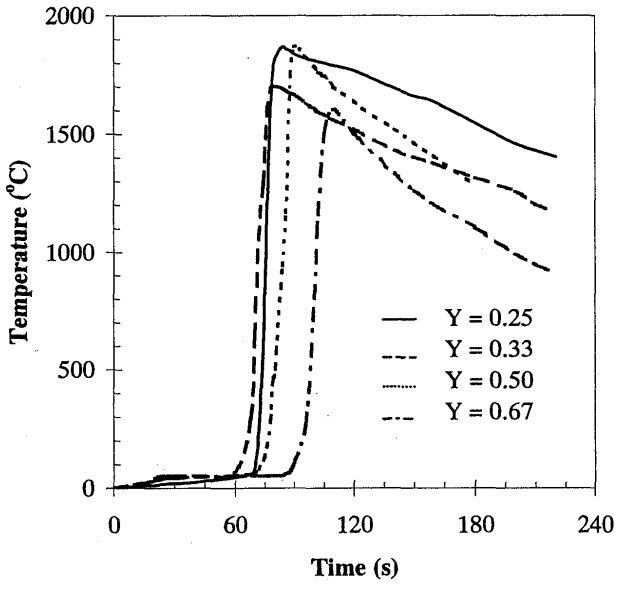

Fig. 2 Temperature profiles of Si_2N_2O-SiC composite synthesis reactions.

Figure 3 shows the change in the Si_2N_2O and SiC contents in the synthesized composite powders determined from their x-ray diffraction patterns. From the XRD analyses of synthetic Si_2N_2O-SiC composites with known compositions, we found that the amount of each phase could be calculated from the intensities of their main peaks. **Figure 3** shows that composites with SiC contents up to 57.6 wt% were synthesized. The measured contents differ from the calculated ones which may be due to the vaporization of Si_2N_2O during the high-temperature combustion reaction. Since these reactions were reproducible, it is possible to control the composition of the resultant composite.

The results of measuring densities, hardness, and fracture toughness of the sintered powders are shown in **Figs. 4 and 5**. The increase of SiC content in the composite hindered achieving the full densification. This is because of the covalent nature of the hard SiC and usually SiC is densified at much higher temperatures. However, the Vickers hardness increased significantly as can be predicted. Since these results are still preliminary, we expect to improve the densities and both the hardness and toughness of the sintered composites in the near future.

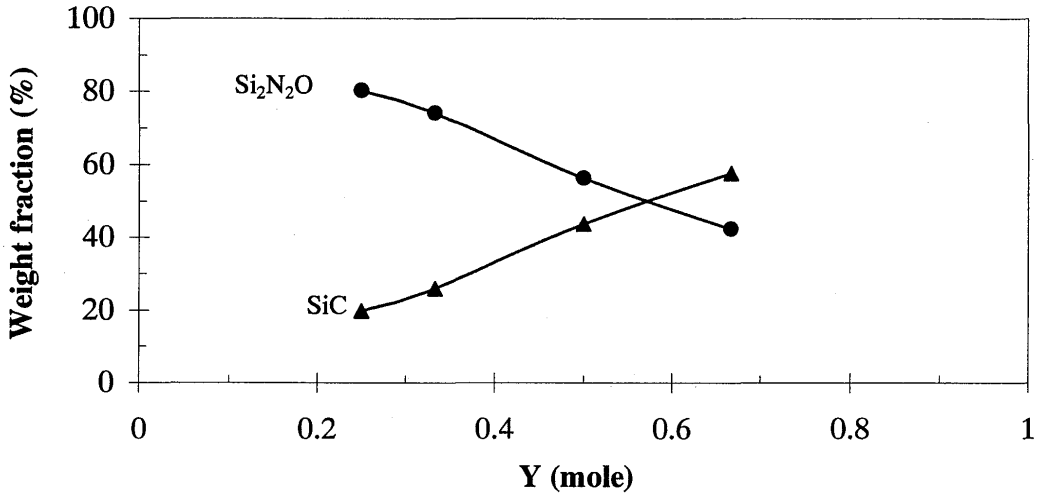

Fig. 3 Change of composition in the synthesized Si_2N_2O-SiC composites.

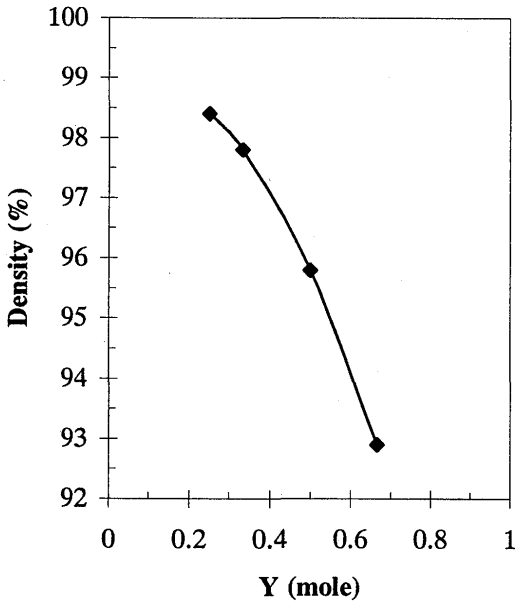

Fig. 4 Change of the density of the sintered Si_2N_2O-SiC composites.

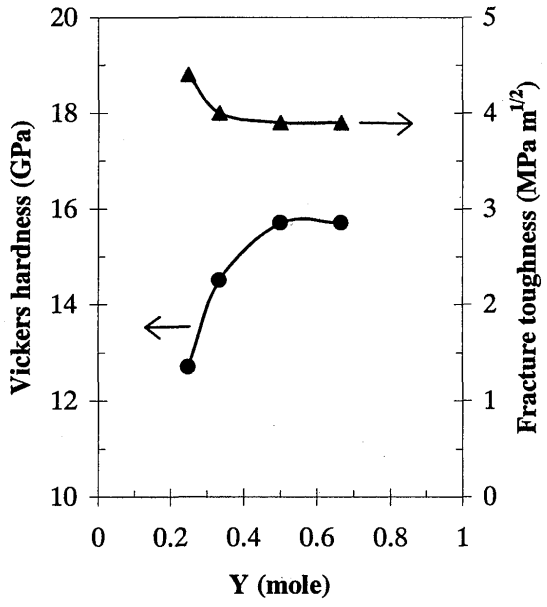

Fig. 5 Vickers hardness and fracture toughness of the sintered Si_2N_2O-SiC composites.

4. Summary

Desert sand is cheap, natural, abundant ore in high purity form and can be considered as a valuable starting material for the synthesis of many Si-based ceramics using the nitriding combustion synthesis method. This research reports the results of nitriding combustion of desert sand to Si_2N_2O-SiC composite powders. Composites with SiC contents up to 57.6 wt% were prepared.

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