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Short communication

Carbide matrix composites by fast MW reaction-sintering in air of B_4C–SiC–Al mixtures

Adrian Goldstein *, Roman Ruginets, Liana Geifman

Israel Ceramic and Silicate Institute, Technion City, 32000 Haifa, Israel

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Abstract

The behavior of B_4C/SiC/Al mixtures during MW heating **in air** was studied. It was determined that B_4C/SiC/Al mixtures generate well-densified specimens. The fired specimens are made from a B_4C matrix, the porosity of which is filled with products of the reactions of Al with B_4C and the gases of air. Phases detected included Al_2OC, $Al_{27}O_{39}N$, $Al_{10}N_8O_3$, $Al_3B_{48}C$ and AlN. The SiC high aspect ratio grains are protected in this environment from oxidation, being able to act as a toughening filler.

Keywords: A. Microwave processing; Sintering; C. Mechanical properties; D. Carbides

1. Introduction

The processing of ceramics in microwave (MW) powered furnaces was determined to have specific and significant benefits. Sintering temperatures and time could be reduced, microstructures refined and new types of processing, not feasible in conventional furnaces, were made possible through the use of MW systems [1–8]. For instance mixtures of B_4C/Al have been converted after fast MW heating in air, into dense boron carbide matrix composites [8]. Such composites exhibited a combination of mechanical properties quite similar to monolithic B_4C. Boron carbide ceramics have important engineering applications in domains like nuclear reactor control rods, lightweight armor, etc. However, the fabrication of well-densified parts requires expensive processing including hot pressing at temperatures >2000 °C [9], while the composites mentioned above can be obtained by rapid sintering in air under 2000 °C.

In continuation of the work described previously [8], the behavior of B_4C/SiC/Al mixtures when exposed to the action of MW fields (air atmosphere) was studied here.

2. Experimental

The boron carbide powder used was the HP grade (H.C. Starck, Gosslar, Germany), which has an average particle size of 3.5 μm. A high aspect ratio, coarse (−400 mesh) SiC_g (Carborundum, Buffalo, USA) was also used, together with the 405 grade Al powder of Alcan-Toyo-Europe (Paris, France). The Al powder had a purity of 99.75% and an average particle size of 5.5 μm. The examined B_4C–SiC–Al mixtures included 20–45% SiC and 10–35% Al, the remainder being B_4C.

Green parts of various shape and size (20–60 mm) were formed by mild unidirectional pressing followed by isostatic cold pressing (250–300 MPa).

The MW processing was performed with the aid of a system working at 2.45 GHz, based on a large (L = 1.22 m), multimode applicator equipped with mode stirrers (model 101 of MMT, Knoxville, TN, USA). The atmosphere was air. The specimens were placed into a refractory heating chamber, which provided thermal insulation so that the temperature did not exceed 700 °C on its external surface. The components of the heating chamber were made of pure alumina. The specimens were buried in a mixture of bubble alumina and coarse yttria grog.

Various MW power–time profiles were examined. Peak power levels ≤2.5 kW were required, and firing run lengths were in the 50–60 min domain, with 25–30 min at peak temperature (cooling stage, with power off, not included). The qualitative phase composition was derived from XRD patterns (model APD

* Corresponding author. Tel.: +972 4 8222107; fax: +972 4 8325339.
E-mail address: goldaad@actcom.net.il (A. Goldstein).

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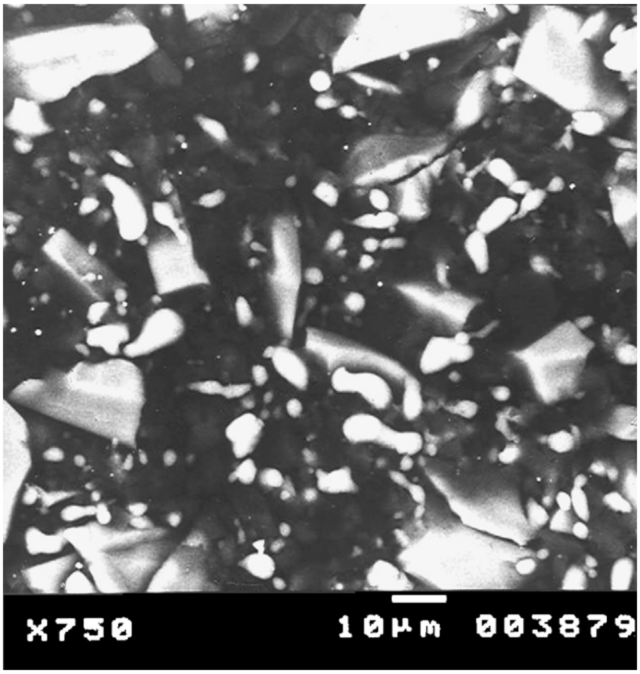

Fig. 1. Green, B_4C/35% SiC/35% Al specimen (after CIP). White spots = Al particles; large, edged particles = SiC; black matrix = B_4C. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

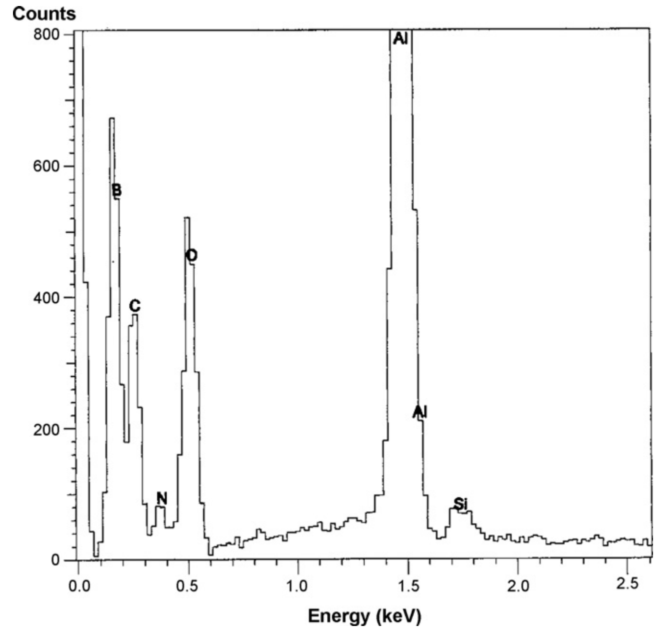

Fig. 2. Elemental composition of part derived from a B_4C/35% SiC/35% Al by MW sintering (air). EMPA (EDS).

2000 of IS, Riva del Garda, Italy), while the microstructure, including element maps, was examined by electron microscopy (models SM 5000 and 8600 Superprobe of JEOL, Tokyo, Japan). Because a quantitative determination of the complex composites phase composition could not be obtained, calculation of theoretical densities was not possible. The Vickers hardness (HV) was measured under 1 kg load with a DVK 15 indenter (Matsuzawa, Seiki, Tokyo, Japan). The toughness was determined with the aid of the K_{IC} coefficient, which was derived from indentation cracks data (approach allowing only rough estimations), according to the method of Antsis et al. [10]

3. Results and discussion

In Fig. 1, the microstructure of a green specimen (B_4C/35% SiC/35% Al composition) is shown. Such specimens (loads of 30–40 g) were transformed into dense parts when suitable heating schedules were used. The maximal temperatures attained were in the 1800–1850 °C range. The chemical analysis of a fired specimen (Fig. 2) shows that it includes B, C, Si, Al, N and O. The microstructure of the composite and the pattern of the elements surface distribution are illustrated in Fig. 3. The white features of Fig. 3 (BSE image) are SiC grains, while in the grayish regions surrounding the SiC, a superposition of Al, O, N and some C is observed. These regions are a mixture of phases resulting from the reactions of Al with B_4C and the air. A typical pattern for the case of an initial B_4C/35% SiC/35% Al composition is shown in Fig. 4. The main Al containing phases identified were Al_2OC, $Al_{27}O_{39}N$, $Al_{10}N_8O_3$, AlN and $Al_3B_{48}C$. The XRD patterns vary to some extent from run to run and as a function of the B_4C/SiC/Al ratio. Wet chemical analysis shows that only small amounts of the initial Al content were lost by vaporization.

The above results suggest that the sequence of events following MW irradiation of the B_4C/SiC/Al powder compacts includes the partial sintering of the B_4C. The monolithic B_4C regions thus obtained (variable morphology, tens to hundreds of microns in size) appear as black spots in Fig. 2. The space between the B_4C domains is filled with the products of the reactions of Al with the B_4C and the air. The SiC contributes to the heating, but does not participate in chemical reactions. Oxygen is consumed in reactions with Al and B_4C, so that SiC degradation is prevented. Oxygen capturing by Al is efficient enough to massively retard the reaction of the gas with B_4C. As a result, part of the initial B_4C remains as such despite being exposed for a relatively long time (at high temperatures) to air.

The mechanical properties of the composites – which included enough Al ($\geq$20%) to allow densification to a level at which the water absorption was $\leq$3.5% – were quite attractive.

Table 1
The density and the mechanical properties of sintered carbide specimens

Material	Bulk density (g/cm^3)	Water absorption (%)	HV 1 (GPa)	K_{IC} (MPa m$^{1/2}$)
Composite from B_4C/35% Si/35% Al (MW sintering)	2.87	1.2	19–25	3.2–3.6
Composite from B_4C/35% Al (MW sintering)	2.73	0.9	19–25	2.9–3.2
Monolithic B_4C (hot pressing)	2.49	$<$0.2	28–33	2.4–2.8

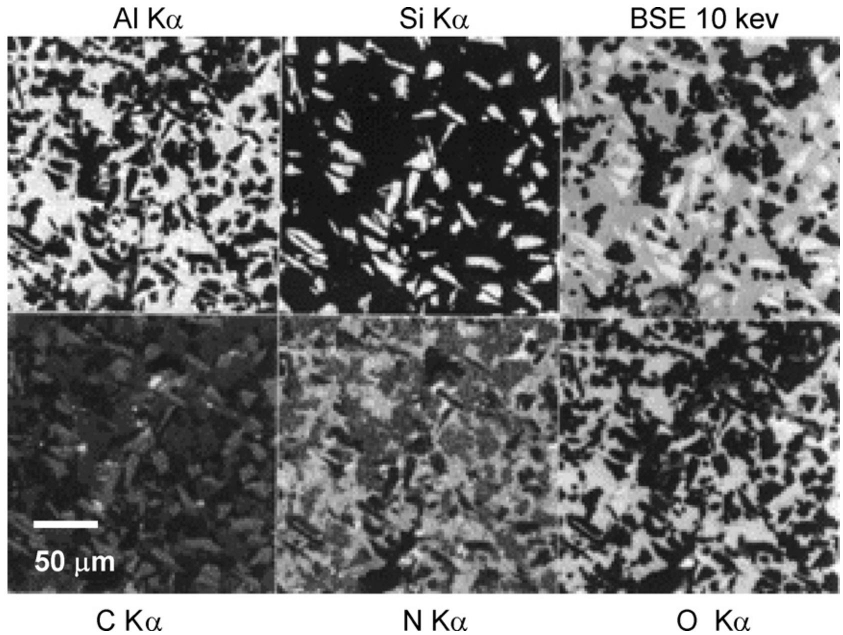

Fig. 3. Microstructure and element (Si, Al, C, N, O) distribution maps of composite derived from B_4C/35% SiC/35% Al; BSE image = microstructure; EMPA = element maps.

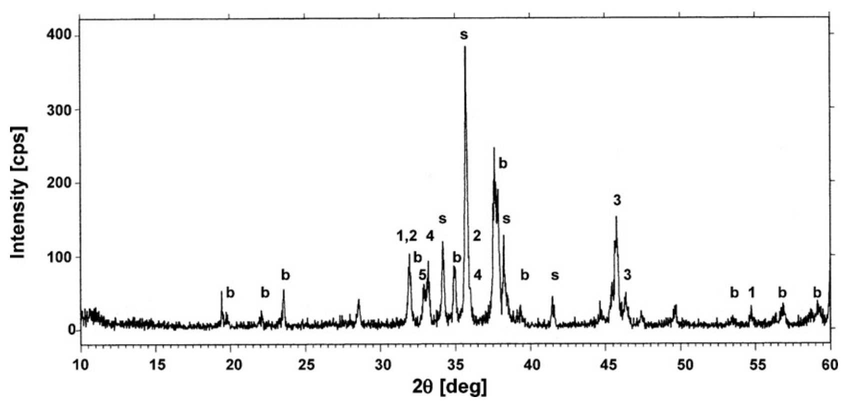

Fig. 4. XRD pattern of dense part obtained by MW sintering of B_4C/35% SiC/35% Al powder compact. $b = B_4C$; s = SiC; 1 = Al_2OC; 2 = $Al_3B_{48}C_3$; 3 = $Al_{27}O_{39}N$; 4 = AlN; 5 = $Al_{10}N_8O_3$.

In Table 1 the characteristics are shown for a composite derived from the optimal composition (B_4C/35% SiC/35% Al). For comparison purposes, the properties of a composite obtained by the MW processing of a B_4C/35% Al mixture and those of a monolithic B_4C (fabricated by hot pressing at 2220 °C) are also given. The presence of uniformly dispersed large SiC grains has a beneficial effect on toughness. At the macroscopic level, the B_4C/SiC/Al derived composites showed a significantly reduced warpage propensity when compared to parts derived from B_4C/Al mixtures.

4. Conclusions

The MW heating, in air, of B_4C/SiC/Al mixtures, with a suitable composition, is conducive to the obtainment of quite dense composites (water absorption $\leq$3.5%). Sintering of the B_4C skeleton is only partial and reaction products fill the porosity. In this environment, SiC is not attacked by oxygen, and is able to contribute to matrix toughness increase. The new composites exhibit low specific gravity and high hardness; such a combination of properties is attractive for some applications like lightweight ceramic armor.

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