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$HoAlO_3$: A novel liquid aid for low temperature densification of SiC ceramics

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ABSTRACT

In this communication, a novel liquid sintering aid consisting of Ho-Al-O elements was analyzed, using the complementary techniques of electron back-scattered diffraction (EBSD) and transmission electron microscopy (TEM). This liquid forming additive was demonstrated to be $HoAlO_3$, showing an exact stoichiometric ratio; it had good wettability with the adjacent SiC grains.

1. Introduction

Carbide-based materials are promising candidates for several technological applications [1–3]. SiC/SiC ceramic matrix composites (CMCs) attracted increasing attention in recent years, as they can be used for both structural and functional applications; examples include aerojet engines, stationary gas turbines and nuclear fusion reactors [4].

Compared with other fabrication methods such as chemical vapor infiltration (CVI) and liquid silicon infiltration (LSI), polymer impregnation and pyrolysis (PIP) is a cost-effective technology, which has significant advantages in fabricating high-performance SiC_f/SiC composites [5]. The process is normally performed several times until the porosity decreases down to a suitable value. However, the presence of large pores among the fiber bundles can inhibit the final densification. To avoid this, selective fillers are introduced into the matrix to achieve enhanced densification [6]. The use of filler and matrix with similar characteristics can ensure the compatibility of the composite.

Dense SiC fillers are always preferable. Since monolithic SiC is quite difficult to sinter, researchers worldwide are looking for suitable additives to densify SiC ceramics at a sufficiently low temperature, and meanwhile, to prevent fibers suffering from damage. Various liquid phase assistants have been tested until now [7–9]. In our previous research, a novel Al_2O_3-Ho_2O_3 sintering aid was developed for densifying SiC ceramics at a temperature as low as 1700 °C [10]. The liquid system formed a Ho-Al-O based phase during the high temperature firing. However, rare reference or data are available for this compound. Ho-Al-O could form substances with stoichiometric ratios of 4:12:9, 1:1:3 and 3:5:12 respectively, each possessing a unique crystalline structure. In our earlier study, the high-temperature liquid phase was identified to be $HoAlO_3$ according to the XRD measurements. However, investigations are still required for further understanding this novel liquid phase.

The purpose of this paper is to perform a more detailed study of this novel $HoAlO_3$ agent, using advanced and complementary characterization techniques, such as electron back-scattered diffraction (EBSD) and transmission electron microscopy (TEM).

2. Material and methods

Sintered SiC-Al_2O_3-Ho_2O_3 pellets with a tailored composition of 92.5 wt% SiC plus 7.5 wt% Al_2O_3-Ho_2O_3 (mole ratio of Al_2O_3 and

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Ho_2O_3=1:1) were fabricated via spark plasma sintering (SPS) at 1700 °C for 1 h. Details of the powder processing, as well as sintering procedures, have been reported elsewhere [10]. The sample was named S-7.5–60. The sintered bulks were polished to a 1-μm finish using diamond abrasives. Scanning electron microscopy (SEM; Magellan 400, FEI, Eindhoven, the Netherlands) equipped with EBSD, together with TEM (JEM-2100F, JEOL, Tokyo, Japan) techniques were adopted for the microstructural analyses of the materials. The features of SiC grain sizes were determined by measuring at least 80 grains from the SEM micrograph. The nano-indentation hardness, as well as modulus of the $HoAlO_3$ phase, were determined using a Nano Indenter (G200, Agilent, US), with a load and an indentation depth of 500 mN and 10,000 nm, respectively.

3. Results and discussion

Representative SEM micrographs of the sintered S-7.5–60 bulks are displayed in Fig. 1-a and 1-b. The relative density was as high as 98.6%, which was a very good value for a low-temperature-sintered SiC sample. SiC grains with diameters ranging from 0.1 to 2.0 μm could be observed. The size distribution of SiC grains is shown in Fig. 1-c (calculated based on Fig. 1-a), with the mean particle size being 0.8 μm. It is clear that the bright phase was distributed homogenously between adjacent SiC grains; this confirmed the good wetting behavior between the SiC matrix and the secondary phase. As shown by Fig. 1-d (patterned bright phase in Fig. 1-b after EBSD mapping), the Ho-Al-O substance was identified to be $HoAlO_3$. At high temperature, the two oxide species can react to form the mixed compound according to the following equation:

$$Ho_2O_3 + Al_2O_3 = 2HoAlO_3 \quad (1)$$

It is highly likely that some energy would be released during this reaction to boost the densification progress. Yet no data are available for $HoAlO_3$; because of this, further study is needed.

A representative TEM image is displayed in Fig. 2-a. The morphology of the SiC matrix differed from rod-like to equiaxial-like, and the black region represented the secondary liquid phase. During the 60 min dwell at high temperature, the morphology of SiC grains were developed, with their size increasing from nano- into submicron- scale, due to the intensified action of included liquid-phase. This image shows the advantage of using $HoAlO_3$ in the sintering of the nano-SiC raw powder; in fact, the microstructural features of the standard submicron SiC raw powders normally show heavily enlarged grain morphology [11].

In our previous study, the mechanical properties were a major concern. However, the sintering mechanism should be studied as well. As shown in Fig. 2-b, a liquid pocket with a width of about 0.1 μm was detected. For liquid-solid binary material system, the dihedral angle plays a critical role in determining the interfacial behavior and, hence, the homogeneity and the properties of the final material; generally, a lower the dihedral angle corresponds to a better wetting behavior, and more effective diffusion of material substances. The relationship between the dihedral angle ϕ and

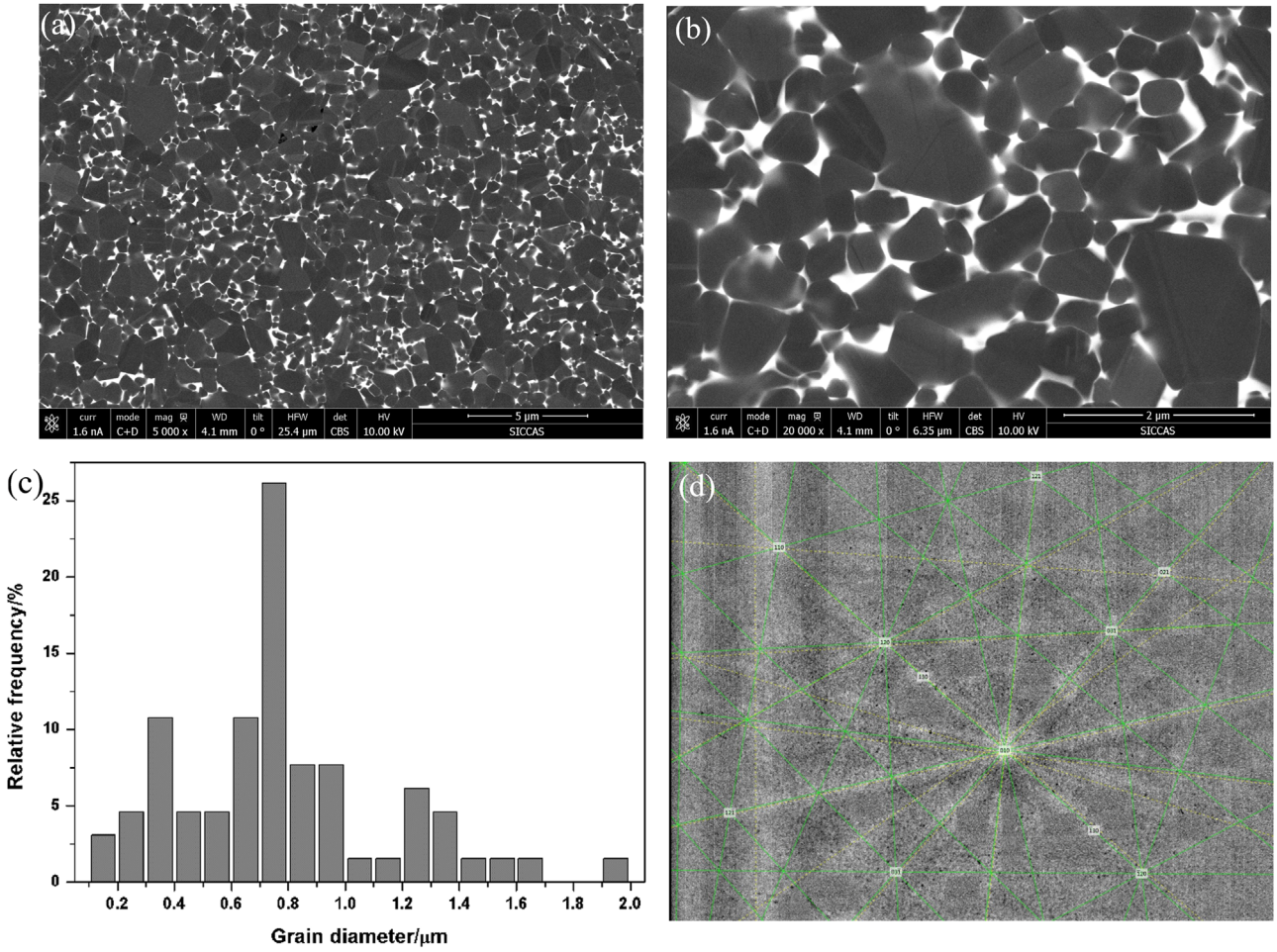

Fig. 1. Representative SEM micrographs of the sintered S-7.5–60 ceramics. (a) Low (a) and high (b) magnifications; (c) size distribution of SiC grains; (d) EBSD analysis of the secondary Ho-Al-O phase.