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Wettability of silicon carbide ceramic by Al_2O_3/Dy_2O_3 and Al_2O_3/Yb_2O_3 systems

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Abstract: Wettability is an important phenomenon in the liquid phase sintering of silicon carbide (SiC) ceramics. This work involved a study of the wetting of SiC ceramics by two oxide systems, Al_2O_3/Dy_2O_3 and Al_2O_3/Yb_2O_3, which have so far not been studied for application in the sintering of SiC ceramics. Five mixtures of each system were prepared, with different compositions close to their respective eutectic ones. Samples of the mixtures were pressed into cylindrical specimens, which were placed on a SiC plate and subjected to temperatures above their melting points using a graphite resistance furnace. The behavior of the melted mixtures on the SiC plate was observed by means of an imaging system using a CCD camera and the sessile drop method was employed to determine the contact angle, the parameter that measures the degree of wettability. The results of variation in the contact angle as a function of temperature were plotted in graphic form which showed that the curves displayed a fast decline and good spreading. All the samples of the two systems presented final contact angles of 40º to 10º indicating their good wetting on SiC in the argon atmosphere. The melted/solidified area and interface between SiC and melted/solidified phase were evaluated by scanning electron microscopy (SEM) and their crystalline phases were identified by X-ray diffraction (DRX). The DRX analysis showed that Al_2O_3 and RE_2O_3 reacted and formed the $Dy_3Al_5O_{12}$ (DyAg) and $Yb_3Al_5O_{12}$ (YbAg) phases. The results indicated that the two systems had a promising potential as additives for the sintering of SiC ceramics.

Keywords: liquid-phase sintering; silicon carbide; rare earth oxide; wettability; contact angle

Silicon carbide ceramic materials are widely used due to their advantageous properties, such as relatively low density, high hardness, high stability and thermal conductivity. These properties render SiC highly resistant to wear, thermal shock, and chemical attack[1–6]. Silicon-carbon bonds are highly covalent; hence, SiC has a low self-diffusion coefficient. This property, allied to the fact that SiC possesses two crystalline phases, β-SiC, which is stable up to 2000 ºC, and α-SiC, which is stable up to 2300 ºC, limits the production of high density SiC ceramics via solid-state sintering, which occurs at temperatures ranging from 2050 to 2200 ºC[2,7–9]. Therefore, liquid-phase sintering (LPS) of SiC is preferable, since it gives this ceramic material high density and better final properties via control of its microstructure, and LPS is faster[2,3,8].

Additives are used as liquid phase formers in LPS, in which the main property of the liquid must be good wettability, i.e., it must present a low contact angle[4–6,8,10].

Several metal oxides are used as liquid-forming additives in SiC sintering[2–5,10,11]. Negita[12] studied the interaction of several oxides with SiC and concluded that the most stable ones are rare earth oxides. Therefore, additives chosen for this study were Dy_2O_3 and Yb_2O_3, particularly in view of the fact that they have not yet been studied for this purpose. In the literatures[13–15], there are many phase diagrams for many oxides systems.

The behavior of the liquid on the solid surface is evaluated by the contact angle, θ, as indicated in Fig. 1. The contact angle is a function of the interfacial energies of the solid-liquid (γ^{SL}), liquid-vapor (γ^{LV}), solid-vapor (γ^{SV}) systems, and ΔG is the variation in Gibbs free energy, as shown in the following equations[4,8,10].

$$\gamma^{SV}=\gamma^{SL}+\gamma^{LV}\cos\theta \quad (1)$$

$$\Delta G=-\gamma^{LV}(1+\cos\theta) \quad (2)$$

The technique most commonly used to measure the contact angle involves measuring the profile of a drop of liquid deposited on a solid surface. This technique is called the sessile drop method[5,8]. As can be seen, therefore, the phenomenon of wettability is of utmost importance in liquid-phase sintering.

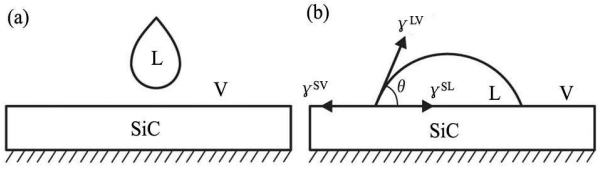

Fig. 1 Representation of wetting
(a) Before adhesion; (b) After adhesion[4]

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The objective of this work was to evaluate the wetting behavior of SiC by the Al_2O_3/Yb_2O_3 and Al_2O_3/Dy_2O_3 systems with different compositions as a function of temperature, using the sessile drop method.

1 Experimental

Wetting tests were performed on solid-state sintered SiC plates with 98.9% density and dimensions of approximately 10 mm×10 mm×4 mm, which were sanded and polished with diamond suspensions of up to 3 μm to minimize their surface roughness.

The compositions of the oxide mixtures were calculated based on the phase diagram of the Al_2O_3/Yb_2O_3 system depicted in Fig. 2, and the eutectic composition indicated by the arrow in the diagram was taken as reference. No phase diagram was found for the Al_2O_3/Dy_2O_3 system. Therefore, the phase diagram of the Al_2O_3/Yb_2O_3 system was used as a basis to solve this problem, since their rare earths are very similar[4,5,13–15].

The quantities of Al_2O_3 (99.99% purity), Dy_2O_3 (99.9% purity) and Yb_2O_3 (99.9% purity) required to compose the eutectic and quasi-eutectic mixtures were calculated. Compositions of the two oxide systems above (5% and 10%) and below (5% and 10%) the eutectic point were studied. The composition and nomenclature (codes) of the different mixtures are described in Table 1.

The oxides were weighed on laboratory scales with a precision of 0.01 g, milled in a planetary mill for 20 min using isopropyl alcohol as a medium (99.9% purity), and oven-dried at 110 ºC for 24 h. The mixtures were then die-pressed into cylindrical 3 mm diameter ×3 mm high samples. These dimensions and shapes are in accordance with the DIN 51730 standard[16].

For the wetting test, the cylindrical samples were placed on a SiC plate. The sample-plate sets were then placed in an electric graphite resistance furnace (ASTRO)

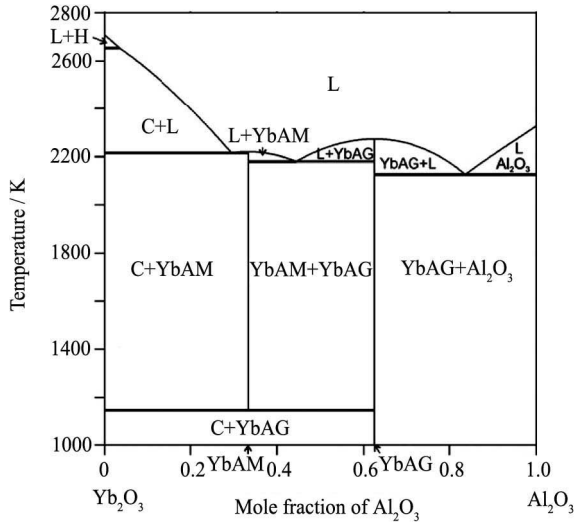

Fig. 2 Phase diagram of the Yb_2O_3/Al_2O_3 system (the arrow indicates the used eutectic point, which was taken as reference in this study[13])

Table 1 Mass composition of the powder mixtures prepared for the Al_2O_3/ Yb_2O_3 and Al_2O_3/Dy_2O_3 systems*

Codes of the samples	Composition/wt.%			Remarks
	Al_2O_3	Yb_2O_3	Dy_2O_3	
EY	56.79	43.21	0.0	Eutectic Al_2O_3-Yb_2O_3
EY5B	49.89	50.11	0.0	5% B
EY10B	43.96	56.04	0.0	10% B
EY5A	64.91	35.09	0.0	5% A
EY10A	74.59	24.41	0,0	10% A
ED	58.10	0.0	41.90	Quasi-eutectic Al_2O_3-Dy_2O_3
ED5B	51.14	0.0	48.73	5% B
ED10B	45.34	0.0	54.66	10% B
ED5A	66.15	0.0	33.85	5% A
ED10A	75.61	0.0	24.39	10% A

* A indicates "above" and B "below" the eutectic composition

and heated in an argon atmosphere up to the samples' melting temperature, applying a heating rate of 20 ºC/min. The samples' behavior as a function of temperature was observed using an image capturing system consisting of a CCD camera (photographic system).

Using ImageJ software, the wetting angle was determined from the captured images, with two measurements for each side of the meniscus. The melting temperature was determined for all the compositions, as recommended by the DIN 51730 standard[16].

Upon completion of the tests, the samples were sectioned longitudinally and cold embedded in resin. The samples were then sanded and polished with diamond paste up to 1 μm to analyze the interface between the melted mixture and SiC by scanning electron microscopy (SEM, LEO 1450VP).

The crystalline phases were evaluated based on X-ray diffraction with Cu Kα radiation in the range of 10º to 90º.

Graphs were created based on the results of melting temperatures and contact angles, using the OriginPro software program.

2 Results and discussion

The evolution of the contact angle with increasing temperature was analyzed based on the experiments and the captured images. Fig. 3 illustrates the behavior of the sample of the Al_2O_3/Dy_2O_3 system in the ED mixture, while Fig. 4 shows the same phenomenon for the EY mixture. These representations were chosen randomly among all the additives under study.

As can be seen in Figs. 3 and 4, the samples of all the compositions begin to change their shape as a function of temperature. According to the DIN 51730 standard, the temperatures of 1825 °C, Fig. 3(c), and of 1850 °C, Fig. 4(d), are considered melting temperatures.

The variation in contact angle differed in each composition. Fig. 5 illustrates the behavior of the contact angle