Available online at www.sciencedirect.com
ScienceDirect
Journal of the European Ceramic Society 26 (2006) 3881–3886
www.elsevier.com/locate/jeurceramsoc

Wetting behaviour of Y_2O_3/AlN additive on SiC ceramics

R.M. Balestra *, S. Ribeiro, S.P. Taguchi, F.V. Motta, C. Bormio-Nunes

Department of Materials Engineering (DEMAR), Faculty of Chemical Engineering of Lorena (FAENQUIL), Polo Urbo Industrial, Gleba AI6, CP 116, 12600-970 Lorena, SP, Brazil

Received 2 May 2005; received in revised form 27 November 2005; accepted 16 December 2005
Available online 20 February 2006

Abstract

The wetting of SiC plate by Y_2O_3/AlN additive was analysed using the sessile drop method. The wetting behaviour was observed by image capture system using a CCD camera during the heating, in argon atmosphere. The contact angle was measured as a function of temperature and time. After the wetting test the SiC plus additive samples were cut in order to observe the thickness plate cross section. The additive area and the interface between SiC and additive were analysed using scanning electron microscopy (SEM) and energy dispersive spectrometry (EDS). The wetting of SiC by Y_2O_3/AlN is influenced by the presence of a solid phase in some of the additive drops that depends mainly on the additive composition and consequently on the temperature. The measured contact angles were below 7°, reaching 0° for Y_2O_3/AlN additive tested at the eutectic composition, indicating a very good wettability of Y_2O_3/AlN on the SiC.

Keywords: Wettability; Interfaces; SiC; Y_2O_3; AlN

1. Introduction

It is well known that the bonds between silicon and carbon atoms in SiC (silicon carbide) are very strong, as a consequence, SiC has a low self-diffusion coefficient. This property limits the production of high density SiC ceramics by solid phase sintering. In order to get a high density ceramic, some metallic oxides (additives) can be added to SiC. The objective is to form a liquid phase during the sintering. The liquid phase sintering (LPS) results in a material with an homogeneous microstructure and therefore suitable to achieve good mechanical properties such as a high toughness.[1–6]

The important variable of the LPS process to obtain SiC ceramic is the wetting of the solid phase (SiC) by the liquid phase (additive). The wetting behaviour is measured through the contact angle (θ) between the solid SiC and the liquid drop formed by the additive. The solid–liquid systems may be of two types, i.e. non-reactive and reactive. In non-reactive systems, the contact angle θ is expressed as a function of the surface energies solid–liquid (γ^{SL}), liquid–vapour (γ^{LV}), solid–vapour (γ^{SV}), as well as the adhesion work (W_a). The mutual dependence of surface energies, adhesion work and contact angle is described by Young and Yong-Dupré equations,[7–16] that are given by Eqs. (1) and (2):

$$\gamma^{SV} = \gamma^{SL} + \gamma^{LV}\cos\theta \tag{1}$$

$$W_a = \gamma^{LV}(1 + \cos\theta) \tag{2}$$

When a reaction occurs at the interface, the free energy change per unit area per unit time also enhances wetting. In this case, the Young equation should be corrected for this driving force.[7,10] The smallest contact angle in reactive system is given by Eq. (3),

$$\cos\theta_{min} = \cos\theta_0 - \frac{\Delta\gamma_r}{\gamma_{LV}} - \frac{\Delta G_r}{\gamma_{LV}} \tag{3}$$

where θ_0 is the contact angle of the liquid on the substrate in the absence of any reaction, ΔG_r is the Gibbs free energy of the chemical reaction, γ_r is the change in the interfacial energies brought about by the chemical reaction.

One possible source of reaction is related to the oxygen partial pressure in the furnace. The reaction of oxygen with the substrate of SiC can form a thin film of SiO_2. However, at temperatures higher than 1475 K, in vacuum, the SiO_2 film evaporates according to the reaction of Eq. (4).[10]

$$2\langle SiO_2\rangle_s + \langle SiC\rangle_s \rightarrow 3(SiO)_g \uparrow + (CO)_g \uparrow \tag{4}$$

* Corresponding author. Tel.: +55 012 3159 9954; fax: +55 012 3153 3006.
E-mail address: roselibalestra@yahoo.com.br (R.M. Balestra).

0955-2219/$ – see front matter © 2006 Elsevier Ltd. All rights reserved.
doi:10.1016/j.jeurceramsoc.2005.12.022

Under these conditions a pure SiC surface is formed and it is more realistic for this system than the measurement at lower temperatures.

The reaction of the oxygen with the additives is very important in metal/ceramic oxides. Usually a thick oxide film forms on the sessile drop and oxidation is evident because the surface of the liquid is not smooth at melting.[17,18] However, in oxide additive the oxygen is already part of the composition and an appreciable effect on interfacial energies are not expected. The furnace atmosphere can also modify the viscosities of the melt. The melt viscosity can change with composition of the melt and with temperature, as shown in the literature.[10,14–16,19]

The usual experimental method used to measure the contact angle is the sessile drop method. In this method, a photography system register the additive sample for increasing temperature in determined time intervals pictures of the drop shape evolution, just before and after the drop starts to melt.[12–14] The contact angle is measured in each picture of a sequence, using a computational program.

Due to the importance of the wettability on the liquid phase sintering, it is the objective of this work to evaluate the wettability of the Y_2O_3/AlN additive on the SiC as a function of the temperature, time and compositions using the sessile drop method.

2. Experimental

The substrates were SiC plates of density 98.9%, sintered via solid state obtained from Wacker – Chemie GmbH, Germany. The substrates had dimensions 10 mm × 10 mm × 4 mm, were ground and polished using diamond suspension down to 1 μm to minimise the surface roughness. SiC plate surfaces were cleaned by acetone and deionised water.

GRADE C powders of Y_2O_3 and AlN from Hermann C. Starck – HCST were used to produce the additives for three different compositions. One of the selected compositions is that of the eutectic point (YN2) and the other two are just at the left (YN1) and at the right side (YN3) of the eutectic one. These compositions are indicated in the Y_2O_3–AlN phase diagram showed in Fig. 1. In order to get the additives samples, Y_2O_3 and AlN powders were mixed using isopropyl alcohol in an attrition milling for 1 h at 1000 rpm. The slurry was dried at 80 °C in a vacuum rotating drier. Green bodies spheres of 4 mm diameter were pressed at 90 MPa using a Monostatic 50 Powder Press – Simac. The molar and mass concentrations of Y_2O_3 and AlN of each additive are presented in Table 1.

Table 1
Compositions of the additives

Additive code	Composition			
	mol%		wt.%	
	Y_2O_3	AlN	Y_2O_3	AlN
YN1	50	50	84.64	15.36
YN2	57	43	87.96	12.04
YN3	60	40	89.21	10.79

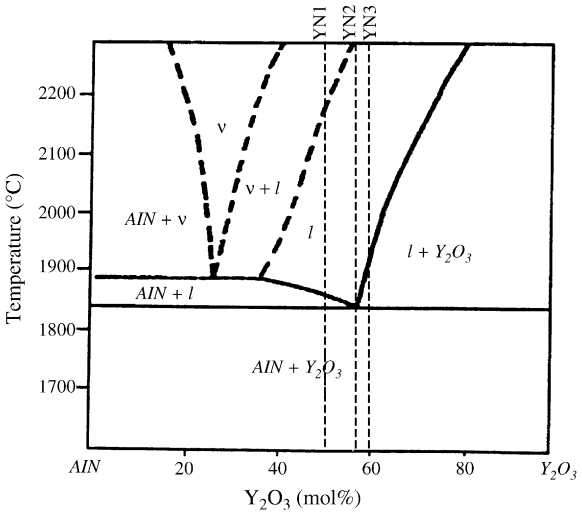

Fig. 1. Phase diagram of the Y_2O_3/AlN system.[3]

The additive spheres were put on the SiC plate and the set was heated up in a graphite furnace of Thermal Technology Inc. (ASTRO) at 10 °C/min, under a pressure of 0.1 MPa of analytical grade argon. Before starting the test, the furnace camera was flushed out with argon to remove the oxygen. In a first experiment, an imaging system capture with a CCD camera (photography system) registers in determined time intervals, pictures of the drop shape evolution for increasing temperature. This test was performed for YN1, YN2 and YN3 additives. While, the experiment at constant temperature (T = 1850 °C) was carried out only for the additive YN2. During the tests, the pictures are taken when visible changes in the spreading of the additive liquid on SiC can be noticed. The contact angle is measured in each picture of a sequence, using a QWin Leica software.

After wetting experiments, images of the interfaces between SiC and additives were taken by SEM/BSE (scanning electron microscopy/back-scattered electrons). The elements present in each phase of the additive region were identified via energy dispersive X-ray analysis (EDS). The EDS measurements and SEM images were carried out in a LEO-Zeiss 1450VP microscope, at 20 kV accelerating voltage.

3. Results and discussion

Fig. 2 shows three rows of images for additive drop shape evolution. Four representative images are shown in each row for YN1 (first row from the top), YN2 (second) and YN3 (third) additives. These images were registered during wetting tests for variable temperature. One test for isothermal conditions (T = 1850 °C) was made for the YN2 additive and the corresponding row of images is shown in Fig. 3. From these images is observable that the surface of the drops is very smooth, indicating that no reaction is occurring between the additive and the furnace atmosphere.

The experimental data of the contact angle θ as a function of temperature T is plotted in Fig. 4 together with the fitting curves for each additive. Experimental results of θ versus T were fitted for temperatures $T \geq T_0$. T_0 corresponds to the eutetic temper-