

Pressure infiltration of Al and Al-12 wt% Si alloy into compacts of SiC and oxidized SiC particles

J. T. TIAN

Departamento de Química Inorgánica, Universidad de Alicante, Apartado 99, E-03080, Alicante, Spain
E-mail: *jttian@yahoo.com;tian@ua.es*

J. M. MOLINA

Departamento de Física Aplicada, Universidad de Alicante, Apartado 99, E-03080, Alicante, Spain

J. NARCISO

Departamento de Química Inorgánica, Universidad de Alicante, Apartado 99, E-03080, Alicante, Spain

C. GARCÍA-CORDOVILLA

Centro de Investigación y Desarrollo, Flat Rolling Products, Alcoa Europe, Apartado 25, E-03080, Alicante, Spain

E. LOUIS

Departamento de Física Aplicada, Universidad de Alicante, Apartado 99, E-03080, Alicante, Spain; Unidad Asociada de the Consejo Superior de Investigaciones Científicas, Universidad de Alicante, Apartado 99, E-03080, Alicante, Spain

Oxidizing SiC particles is a common practice in the fabrication of SiC/Al composites through direct mixing of the liquid metal and the ceramic particles. This practice is addressed to reduce the reaction between the two materials. In a previous work it was concluded that oxidizing SiC particles scarcely affected the threshold pressure for infiltration of pure Al into compacts made of those particles. In this work we present a detailed study of this problem for infiltration of pure Al and the eutectic Al-Si alloy. Our results indicate that oxidizing of SiC particles does not affect significantly either the threshold pressure or the infiltration kinetics. This conclusion is equally valid for both pure Al and the eutectic alloy. © 2005 Springer Science + Business Media, Inc.

1. Introduction

A common practice in the fabrication of silicon carbide/aluminum (SiC/Al) composites through compositing (direct mixing and stirring of the semi-solid metal and the ceramic particles) is the oxidation of the SiC particles prior to mixing of the two materials. The formed SiO₂ layer is expected to have two effects: protection of SiC from Al attack and modification of wetting at the SiC/Al interface. In a previous work it was reported that oxidation of SiC particles can produce at most a decrease of around 10% on the threshold pressure for infiltration of pure Al, though this effect was ascribed to changes in particle morphology induced by oxidation [1]. On the other hand, Laurent *et al.* [2], using the sessile drop technique, concluded that the SiO₂ layer did not improve low temperature wetting at the SiC/Al interface. Moreover, wetting kinetics was slowed down and even blocked due to the presence of SiO₂, giving an apparent contact angle as high as 150° at 973 K.

In this work we investigate pressure infiltration of pure Al and the Al-12 wt% Si alloy (eutectic composi-

tion, hereafter referred as Al-12Si) into compacts made of as-received and oxidized SiC particles. The Al-Si alloy was chosen because Si additions are rather common in the commercial fabrication of SiC/Al composites. Both the infiltration kinetics and the threshold pressure were investigated. Besides, pressureless infiltration of an organic fluid into the SiC and oxidized SiC compacts were also performed at room temperature to determine the intrinsic permeability of the compacts. Our results indicate that, in agreement with previous studies [1, 2], oxidation of SiC particles scarcely affects pressure infiltration performance of both pure Al and the eutectic Al-Si alloy.

2. Experimental procedures

2.1. Materials

Commercially pure aluminum and silicon (99.98 wt%) were used to obtain the Al-Si eutectic alloy. The alloy was fabricated in the same crucible used to carry out the infiltration experiments (see below). α -SiC particles of purity better than 98.5 wt% were used in all infiltration

TABLE I Characteristics of the as-received and oxidized SiC particles used in this work

Particles	D	Span	S	λ	ρ	V _p
SiC	17.1	0.78	0.36	3.0	3.21	0.56
SiC _{ox}	17.0	0.87	0.35	2.6	3.05	0.53

The table reports data for: the average diameter *D* (in μm), the span of the particle size distribution, defined as $[D(90)-D(10)]/D(50)$ where $D(x)$ is the diameter (also in μm) below which *x*% of the particles are found, the specific surface area of the particles *S* (m²/g) as measured by the BET technique, the geometric factor λ as derived from Equation 1, the particle density (in g/cm³) and the particle volume fraction *V_p* of the compacts made of those particles.

experiments. In order to reduce the span of the size distribution (see Table I) the as-received SiC particulate was cleaned in a stirred ethanol bath discarding the above floating fraction. After three times cleaning, the particulate was dried at 110°C for 2 h.

2.2. Oxidation and characterization of SiC particles

Oxidation of SiC particulate was carried out under static air atmosphere at a temperature of 1000°C for a batch time of 24 h. After oxidation, the agglomerated particles were stirred in ethanol and then dried at 110°C for 2 h. Unoxidized and oxidized SiC (hereafter referred to as SiC and SiC_{ox}, respectively) particles were then characterized by means of several techniques. The most relevant data are reported in Table I. Particle average diameter and size distribution were determined by means of laser scattering. The density of SiC_{ox} particles was determined from: (i) the weight change of particles before and after oxidation, assuming densities of 3.21 and 2.2 g/cm³ for SiC and the amorphous SiO₂ layer, and (ii) using a small bottle, which allows to measure slight change in the volume of liquid in the bottle and thus gives an error in the density measurements of less than 1%. The density obtained by means of these two methods was very similar (the average is reported in Table I). The specific surface area of the particles was measured by means of the Brunauer, Emmett and Teller (BET) technique, which is known to give the results relevant to pressure infiltration of liquid aluminum (see Refs. [3–5] for details). The specific surface area of round particles can be written as:

$$S = \frac{6\lambda}{\rho D} \tag{1}$$

where ρ and *D* are the density and average diameter of the particles, and λ—a geometric factor that accounts for deviations from sphericity and surface roughness. Inserting the experimental results for *S*, *D* and ρ in Equation 1 we obtained the geometric factor. The results are reported in Table I.

The SiC and SiC_{ox} particles compacts were made by addition of small amounts of particles into quartz tubes (~5 mm of inner diameter) and combining strokes of a weight until a height of approximately 35 mm was obtained (see Ref. [1] for details). The particle volume

fractions in the compacts, determined as described in Ref. [1], are given in Table I.

Comparing the results for SiC and SiC_{ox} particles, the following points are worth of comment. An estimation of the increase in particle diameter promoted by oxidation gives ~0.5 μm, a change too small to be easily detected. In fact, our results indicate that no significant change is produced by oxidation either in the particle diameter or in the span of the size distribution. However, the specific surface area is appreciably smaller in the oxidized particles suggesting that oxidation smoothes the particle surface. This is not compatible with the smaller particle volume fraction of the compacts made of oxidized particles.

2.3. Pressure infiltration

Pressure infiltration of liquid Al and Al-12Si alloy into SiC and SiC_{ox} compacts was carried out in a pressurized chamber. As the equipment and the infiltration procedures are described in detail in Ref. [6], here we only comment on the most relevant aspects. The compacts were preheated for 3 min by holding just above the melt the quartz tubes containing the compact. The tubes were then introduced into the melt and the pressure chamber closed as fast as possible. Gas pressure was applied with nitrogen for 120 s. All infiltrations were carried out at 700°C. After infiltration, the tube containing the infiltrated compact was taken out and air-cooled, and the infiltrated height were measured. The organic fluid used for pressureless infiltration of SiC and SiC_{ox} compacts was polyethyleneglycol-200 (PEG, purity of 99 wt%). After the infiltration was initiated, the infiltration height and time were recorded. At least three compacts were infiltrated for each particulate.

3. Results and discussion

3.1. Threshold pressure for infiltration

As shown in Fig. 1 the experimental results for infiltration in all systems investigated here follow Darcy’s

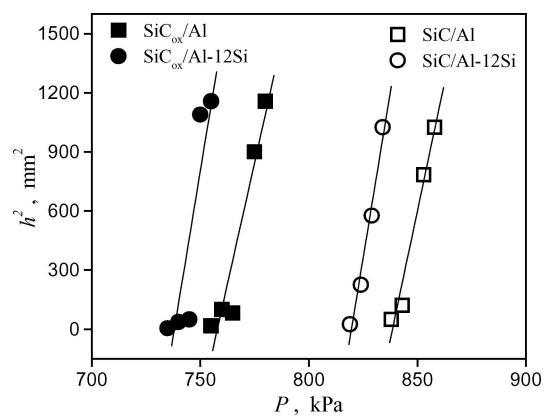


Figure 1 Plots of the square of the infiltrated height *h*² (in mm²) as a function of applied pressure *P* (in kPa) for SiC and SiC_{ox} particle compacts infiltrated with Al and Al-12Si alloy. The straight lines fitted to the data are: $h^2 = 52.3(P-839)$ ($R = 0.99$) for SiC/Al (hollow squares); $h^2 = 67.0(P-820)$ ($R = 0.99$) for SiC/Al-12Si (hollow circles); $h^2 = 49.2(P-758)$ ($R = 0.95$) for SiC_{ox}/Al (solid squares); $h^2 = 67.1(P-738)$ ($R = 0.89$) for SiC_{ox}/Al-12Si (solid circles), where *R* is the correlation coefficient.

law [3]:

$$h^2 = \frac{2kt}{\mu(1 - V_p)}(P - P_0) \quad (2)$$

where h is the infiltration height, k —the intrinsic permeability of the compact, t —the infiltration time, μ —the viscosity of the melt, V_p —the particle volume fraction, P —the applied pressure, and P_0 —the threshold pressure (also called capillary pressure). A fitting of the results for the square of the infiltration height h^2 versus P (at fixed infiltration time t) by means of a straight line, allows deriving the threshold pressure P_0 for infiltration and the intrinsic permeability k of the compact.

The experimental results for the threshold pressure are reported in Table II. A rather sharp decrease is noted apparently due to oxidation of SiC particles. However, as shown in Table I, this decrease could be related to the lower particle volume fraction of compacts made of SiC_{ox} particles. The threshold pressure P_0 in Equation 2 is related to the contact angle θ and the particle average diameter D through the so-called capillary law or Laplace equation [3],

$$P_0 = -6\lambda\gamma_{lv} \cos \theta \frac{V_p}{(1 - V_p)D} \quad (3)$$

where γ_{lv} is the liquid-vapor surface tension and λ is the geometric factor in Equation 1. Then, introducing all parameters given in Tables I and II, the experimental data for the threshold pressure, and the results for the geometric factor λ derived in the preceding section, we obtained the contact angles reported in Table II. The results clearly indicate that oxidation of SiC does not increase the wetting at the SiC/Al interface; in fact, just the opposite occurs, in agreement with the sessile drop results reported in Ref. [2].

An additional interesting point concerns the effect of Si on the threshold pressure. Si addition slightly decreases the surface tension of aluminium (see Table II). Thus, one may expect that Si additions should promote a decrease in the threshold pressure [7]. This is so for both oxidized and unoxidized particles (see Table II).

TABLE II Results of the fittings of the experimental data for the square of the infiltrated height h^2 versus applied pressure P (see Fig. 1) obtained from infiltration of Al and Al-12Si into compacts of as-received and oxidized SiC particles, with $h^2 = s(P - P_0)$

System	s	P_0	γ_{lv}	θ	a
SiC/Al	52.3	839	861	131°	4.3×10^{-4}
SiC/Al-12Si	67.0	820	836	132°	3.7×10^{-4}
SiC _{ox} /Al	49.2	758	861	137°	4.4×10^{-4}
SiC _{ox} /Al-12Si	67.1	738	836	137°	4.0×10^{-4}

The slope s is given in mm²/kPa and P and P_0 in kPa. The surface tension of the liquid metal γ_{lv} (in mN/m) is also given [8, 9]. The contact angle θ was derived from Equation 3 and the constant a from Equation 4. In carrying out the latter calculation we took 1.15 mPa·s and 0.77 mPa·s for the viscosity of Al and Al-12Si, respectively, at the infiltration temperature [10].

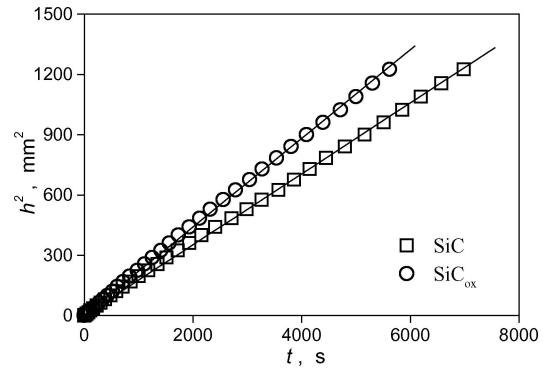


Figure 2 Experimental results for the square of the infiltrated height h^2 (in mm²) versus time t (in second) obtained from pressureless infiltration at room temperature of PEG into SiC and SiC_{ox} particle compacts. The slopes of the fitted straight lines are reported in Table III.

3.2. Infiltration kinetics and permeability of the compacts

The results of Table II indicate that infiltration kinetics is increased to some extent by Si additions, while particle oxidation has no observable effect. The PEG experiments were aimed to evaluate the intrinsic permeability of the compacts. Fig. 2 and Table III show the results derived from pressureless infiltration of PEG into SiC and SiC_{ox} compacts. As depicted in Fig. 2 the data for the square of the infiltrated height versus infiltration time can be very accurately fitted by means of straight lines, in full agreement with Darcy law. Fig. 2 and Table III indicate that the SiC_{ox} compacts have higher permeability than the SiC compacts. In order to quantify this result we used the empirical law that relates the compact permeability to the particle diameter [3],

$$k = aD^2 \quad (4)$$

where a is a constant that characterizes the permeability of the compact. Introducing all data in Tables I–III we derived the results for the constant a reported in Table III. We note that oxidation substantially increases the permeability of the compact. We have also derived the constant a from the infiltration experiments for Al and Al-12Si. The results reported in Table II are all very similar and significantly differ from those derived from PEG infiltration. The fact that infiltration of Al and Al-12Si alloy takes usually place under unsaturated conditions does not explain these discrepancies. Better data are required to attain definitive conclusions.

TABLE III Slopes of the straight lines (in mm²/s) fitted to the experimental results for the square of the infiltrated height h^2 versus time t obtained from infiltrations of PEG into SiC and SiC_{ox} particle compacts

System	Slope	θ	a
SiC/PEG	0.176	37.5°	1.6×10^{-4}
SiC _{ox} /PEG	0.221	24°	2.3×10^{-4}

The contact angle at the SiC/PEG interface, as measured by means of the sessile drop technique, and the constant a in Equation 4 are also given. The viscosity and surface tension of PEG used to derive a are 60 mPa·s and 42.5 mN/m, respectively.

4. Concluding remarks

The results presented here agree with previous data in the sense that oxidation of SiC particles does not improve infiltration performance in the SiC/Al system. A similar result was obtained for the eutectic Al-Si alloy. In fact the contact angle derived from the infiltration experiments increased upon oxidation of SiC particles, in agreement with sessile drop measurements of the contact angle [2]. The decrease in threshold pressure promoted by Si additions can be understood in terms of the decrease that Si induces in the surface tension of pure Al. Infiltration with a wetting organic fluid allowed to conclude that oxidation significantly increases the permeability of the compacts. Unfortunately the results derived from Al and Al-Si infiltration disagree with that result, probably due to its insufficient accuracy.

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