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Decreasing the Infiltration Threshold Pressure of Al-12wt%Si into Alumina Particle Compacts by Sn or Pb Layers

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Abstract

Pressure infiltration of liquid aluminum alloys into ceramic preforms is a widely used method to fabricate aluminum based composites. Aiming to improve infiltration performance, a novel method is proposed that consists of placing a thin film of a low melting point metal having a surface tension substantially smaller than that of the alloy. In particular the effects of a thin layer of tin or lead on infiltration of Al-12wt%Si into alumina compacts are investigated. The threshold pressure is significantly reduced with respect to that for the alloy, although it is slightly higher than that for pure tin or lead. The performance and characteristics of the method are discussed in some detail.

1. Introduction

During the last decades, gas pressure infiltration of liquid metals into preforms of ceramic or graphite particles has been put forward as a very efficient process for the fabrication of Metal Matrix Composites (MMC) [1-4]. At present, this method is one of the best suited to the fabrication of composites containing high volume fractions of reinforcement, having, in addition, the capability of near net-net shape production. The most important limitation of all liquid-phase processes being used to produce MMC resides upon the compatibility of the reinforcement and the metal matrix [1]. This problem is particularly important in the case of aluminum matrix composites due to oxide layer that usually covers the aluminum surface. This layer increases the threshold pressure for infiltration, slows down the process, and increases porosity. Several procedures addressed to disrupt or weaken the oxide layer and, consequently, improve infiltration performance, have been reported in the literature. Among them we mention: reducing the oxygen partial pressure [2], increasing the infiltration temperature [5] and adding chemicals that partially dissolve the oxide layer [4]. Methods that focus on improving wettability by coating the reinforcing particulate with a metal [6] are, unfortunately, less economical and non-advisable.

Aiming to improve infiltration performance, a novel method is proposed in this work. The method consists of placing a thin film of a low-melting-point low-surface-tension metal (Sn or Pb) at the compact end through which infiltration takes place. In this way, during the process the particle compact is firstly infiltrated by the pure metal, which is swept by the aluminium alloy. Some preliminary results on infiltration of graphite by aluminum alloys can be found in Ref. [7]. Here, the efficiency of the method in the infiltration of compacts of alumina particles by Al-12wt%Si is discussed in detail. In order to help in the interpretation of the results, infiltration of pure metals was also carried out. The effects on the threshold pressure and on the infiltration kinetics are investigated.

In addition, other aspects of the method proposed here are evaluated on samples infiltrated under vacuum and solidified directionally and substantially larger than those used in capillary studies. In particular it was checked that a large portion of the composites was free of tin or lead, those metals being mostly concentrated near the infiltration front.

2. Materials and experimental procedures

Alumina particles AA18 were used in the infiltration experiments. The particle size distribution was determined by means of laser light scattering, obtaining a mean diameter of 22.3 μm and a span of 0.81. Applying the conventional Brunauer, Emmett and Teller (BET) technique to nitrogen adsorption isotherms measured in an Autosorb 6B system from Quantachrome Instruments, a particle specific surface area of 0.2 m^2/g was obtained. The aspect ratio of the alumina particles, defined as the ratio of the maximum and the minimum Feret diameter, was measured obtaining a mean value of 1.51. Al-12wt%Si (hereafter referred to as Al-12Si) alloy and pure tin and lead (99.9%), supplied by Leichtmetallkompetenzzentrum Ranshofen GmbH and Panreac, respectively, were used. Some useful data for the alloy and the pure metals are reported in Table 1.

Particles were packed into quartz tubes of 4.5 mm inner diameter and 20 cm in length, by alternating vibrations and strokes of a 35 g piston dropped from a height of approximately 10 cm. At each packing step, approximately 0.05 g of particulate were added, after which 2 seconds of vibrations and 2 strokes were applied. The process was repeated until the compact reached a height of approximately 5 cm. The bottom side of the compact was plugged with alumina paper of 6% of theoretical density, to avoid depacking and skim off the oxide layer. In order to determine the particle volume fraction V_p , the quartz tube was weighed before and after the powder was packed inside it, and its outer diameter measured at five points along its length. Using these data, and the length of the packed powder, a particle volume fraction V_p of 0.57 ± 0.01 was obtained. The density of the particles used in this determination is 3.96 g/cm^3 . In some samples a thin layer (3 mm thick) of either tin or lead was placed between the particle compact and the alumina paper.

The infiltration apparatus used to determine the threshold pressure and the infiltration kinetics was described in detail elsewhere [2,3]. The Al-12Si alloy and the pure metals were melted in alumina crucibles, degassed and cleaned by bubbling argon gas during at least 30 min. The quartz tube containing the packed powder was attached at the top of the pressure chamber and preheated by holding it above the melt for no more than 300 s. Just before immersion, the metal surface was thoroughly cleaned. Infiltration was carried out at 913 K, during which the tube with the preform is maintained in air atmosphere. Pressure was raised up to a chosen value by introducing nitrogen at a rate of 50-60 kPa/s. After a fixed period of time at constant pressure (40 s in this work), the chamber was vented at 30-70 kPa/s. Then the sample was taken out of the melt and air-cooled. No de-wetting was observed. Once the sample was solidified and cooled, the infiltration height was measured with a precision gauge.

The major source of errors in the present experiments is related to particle packing. Other direct errors related to the measurement of the infiltrated height, applied pressure, quartz tube radius, etc., are much smaller. In the present experiments great care was taken in preparing the preforms. The maximum relative error in V_p derived from the many preforms that have been prepared to carry out the present experiments, is around $\pm 2\%$.

In order to investigate other aspects of the process larger pieces were fabricated in a pressure chamber that allowed: a) melting and infiltrating under vacuum/inert atmospheres, and, b) directional solidification of the infiltrated sample. The latter was essential in reducing significantly the porosity of the composite. The ceramic particles were packed in a quartz crucible 100 mm high and an inner diameter of 17 mm. Packing was carried out following the procedure described above. Once packing was completed, a layer 3 mm thick of tin or lead and an alloy rod 75 mm in length were placed on top of the ceramic compact. Once placed the crucible into the chamber, a vacuum of 50 Pa was attained by means of a rotary pump, followed by the introduction of nitrogen up to a pressure of 0.1 MPa. The cycle vacuum/pressure was repeated several times. Melting at 650°C was done under a 100 Pa nitrogen atmosphere. Subsequently, metal infiltration was induced by raising pressure with nitrogen gas up to 400-600 kPa and maintaining it for at least 2 min. It should be noted the relevant applied pressure in this system is around one atmosphere higher than in the capillary experiments described above, as in the latter the pressure in the perform voids is one atmosphere whereas in the former is only 100 Pa.

3. Results and discussion

3.1 Threshold pressure for infiltration

Fig. 1 depicts the experimental results for the square of the infiltrated height h versus applied pressure P at a fixed infiltration time t (time at constant pressure) for all systems investigated in this work. The results can be satisfactorily fitted by means of Darcy's law [2]:

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

where P_0 is the threshold pressure, V_p is the particle volume fraction in the preform, μ is the viscosity of the alloy and k is the preform permeability. By fitting straight lines to the experimental results for h^2 versus t one can derive the threshold pressure P_0 . In addition, the preform permeability k can be derived from the slope of the straight line, provided that the viscosity of the liquid is known (see below). Data scattering produced by different error sources, such as the packing process or the measurement of the infiltration height, is discussed in [4].

The results reported in Table 2 clearly indicate that infiltration with pure tin and lead requires a threshold pressure that is almost half of that for Al-12Si alloy. This is mainly a consequence of the lower surface tension of the two pure metals, as can be easily deduced from the capillary or Laplace law,

$$P_0 = - 6 \lambda \gamma_w \cos \theta \frac{V_p}{(1 - V_p)D} \quad (2)$$

where γ_w is the liquid-vapor surface tension, D is the average particle diameter, θ the contact angle at the particle/liquid interface and λ a geometric factor that accounts for particle morphology. Using Eq. (2) one may check that in both metals the decrease in surface tension with respect to Al-12Si accounts for around 85% of the drop in threshold pressure. The remaining 15% is surely due to a reduction in the contact angle at the ceramic/liquid interface [12]. This can be quantified by combining Eq. (2) for

each metal with that for the Al-12Si alloy (note that we are always dealing with the same ceramic perform),

$$\frac{\cos \theta^{Al-12Si}}{\cos \theta^{metal}} = \frac{\gamma_{lv}^{metal}}{\gamma_{lv}^{Al-12Si}} \left(\frac{P_o^{Al-12Si}}{P_o^{metal}} \right) \quad (3)$$

From this equation one can easily derive the contact angle at the alumina/tin(or lead) interface. The results reported in Table 2 for the two metals are compatible with those derived from sessile drop measurements [10] (note, however, that the temperatures at which infiltration was carried out in this work are higher than those at which the contact angle was measured).

In the case of Al-12Si infiltration with a layer of either tin or lead, the threshold pressures are in between those for the pure metals and the alloy (see Table 2). The reduction with respect to the case without layer is in both metals around 30%. This important improvement suggests that the present method may have some technological relevance; however, its efficiency requires further evaluation (see below). Now the question is why the capillary pressure has increased with respect to the pure metals. The probable origin of this drawback is diffusion of Al or Si (or both) to the Sn (or Pb)/alumina interface during raise of pressure that may affect to both surface tension and wetting. In addition one may ask why infiltration, once initiated, proceeds at pressures well below threshold for Al-12Si. To understand this point we note that now the alloy displaces Sn (or Pb) at the ceramic surface, instead of air, as in the case without layer. In addition we note that it is likely that the oxide layer that usually covers aluminium alloy is, in this case, substantially reduced.

3.2 Infiltration kinetics

The slopes of the straight lines fitted to the experimental results for the square of the infiltrated height versus applied pressure characterize the infiltration kinetics (see Eq. (1)). The results are given in the caption of Fig. 1. The higher infiltration kinetics in Al-12Si as compared to the pure metals is mostly due to its lower viscosity (see Table 1). This is most clearly seen by calculating the compact permeability k by means of Eq. (1). The results are reported in Table 2. The almost coincidence of the three results can be rationalized by factorizing the permeability in two terms [1],

$$k = k_r \cdot k_s \quad (4)$$

The saturated permeability k_s , is the permeability for fully saturated flow and is characteristic of the preform only, no matter the nature of the infiltrating fluid. The relative permeability k_r , instead, depends on the degree of saturation, which is in turn related to the contact angle at the ceramic/liquid interface. Then, the rather similar contact angles in the three cases (see Table 1) may explain the also similar compact permeability derived from the infiltration kinetics.

In analyzing the results for infiltration of Al-12Si with a layer of tin or lead we have derived the compact permeability by introducing the viscosity of either the pure metal or the alloy. The results are also reported in Table 2. Although the results for Pb are reasonable and may enter into what can be considered as experimental uncertainty, we have no explanation for the rather high permeability

derived in the case of Sn. Anyhow we may safely conclude that the procedure proposed here, being effective in decreasing the threshold pressure, does not significantly affect the infiltration kinetics.

3.3 Microstructure of the composites

Mixing of the pure metal and the alloy that can occur along the infiltration process may deteriorate the properties of the composite. It will thus be desirable that mixing occurs only in a small region close to the infiltration front. This is actually what the optical micrographs of Figure 2 indicate. The bulk of composites infiltrated at the higher pressure (600 kPa) are almost free from lead or tin. For lower infiltration pressures small amounts of tin or lead were detected in the bulk of the compacts, mostly filling small voids. These results have been quantified by means of EDAX (see Fig. 3). Despite of the fact that EDAX results can only be considered as relative, the results of Fig. 3 reinforce our conclusions. An alternative way to quantify the amount of tin or lead is to analyze the melting peak of tin or lead in a DSC curve. Our results indicate that the amount of tin in the bulk of composites fabricated at the higher pressures was below 0.2% in volume.

4. Concluding Remark

Aiming to improve the performance of infiltration of aluminum alloys into ceramic preforms, a novel method has been proposed. The method consists of placing a thin film of a low melting point low surface tension metal at the compact end through which infiltration occurs. Results for infiltration of Al-12wt%Si into alumina particles performs with layers of either tin or lead indicate that a 30% reduction of the threshold pressure can easily be attained without deteriorating the infiltration kinetics. In addition it has been shown that the bulk of the composites have negligible amounts of tin or lead, being those metals mostly located near the infiltration front. The present results support and reinforce previous and preliminary results obtained on graphite performs [7] and suggest that the method may have a wide applicability.

Acknowledgements

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FIGURE CAPTIONS

Figure 1. Square of the infiltrated height h^2 versus applied pressure P for alumina particle compacts infiltrated with Sn, Pb, the Al-12Si alloy, and the Al-12Si alloy with a thin layer of either Pb or Sn placed at the infiltration front (see text). The fitted straight lines are: $h^2=27.7(P-309)$ –pure Sn-, $h^2=18.8(P-261)$ -pure Pb-, $h^2=39.2(P-600)$ -Al-12Si-, $h^2=55.6(P-408)$ –Al-12Si/Sn-, and $h^2=21.3(P-415)$ -Al-12Si/Pb-, with h in mm and P in kPa.

Figure 2. Optical micrographs of samples produced by infiltration of Al-12Si into an alumina particle preform at 600 kPa with a thin layer of Sn placed at the infiltration front. Samples were taken from a region away (a) and near (b) the infiltration front.

Figure 3. Percentage of Sn versus the distance to the Al-12Si end, as obtained by means of EDAX, in the same composite samples of Fig. 2. Infiltration was carried out at the pressures given in the Figure. Measurements were carried out on samples 17 mm in diameter and 40 mm in length infiltrated under vacuum and solidified directionally (see text).

TABLES

Table 1. Surface tension γ_v and viscosity of tin, lead and Al-12wt%Si alloy, at the infiltration temperature T_i . The melting point is also given.

	Melting point (K)	T_i (K)	Viscosity (mPa s)	γ_v (mN/m)
Sn	505	913	1.10 ^[2]	525 ^[10]
Pb	601	833	1.65 ^[8]	436 ^[10]
Al-12Si	848	913	0.79 ^[4]	855 ^[9]

Table 2. Threshold pressure and compact permeability derived from infiltrations of several metallic materials into alumina compacts at a temperature T_i and an infiltration time of 40 s. The contact angles at the Alumina/Sn and Alumina /Pb interfaces were derived from Eq. (3).

	T_i (K)	P_0 (kPa)	k (10^{-14} m ²)	α (this work)	θ
Sn	913	309	16.4	116°	116-135 ^{d[10]}
Pb	833	261	16.7	117°	117 ^{d[10]}
Al-12Si	913	600	16.7	-	122 ^{o[11]}
Al-12Si+Sn layer	913	408	23.6-32.9	-	-
Al-12Si+Pb layer	913	415	9.0-18.9	-	-

Fig.1

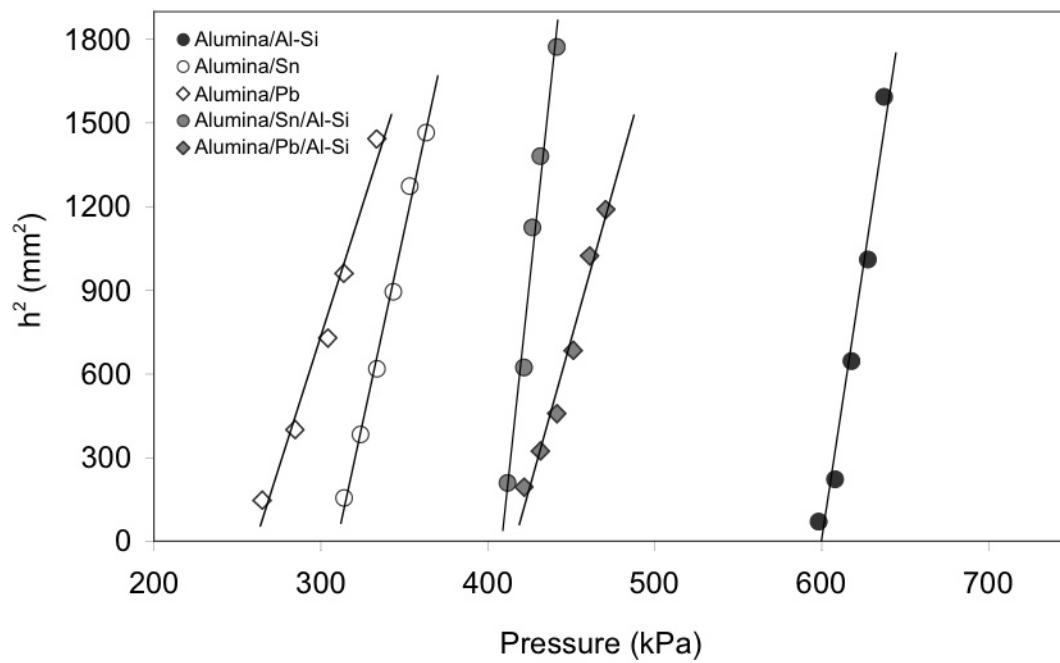
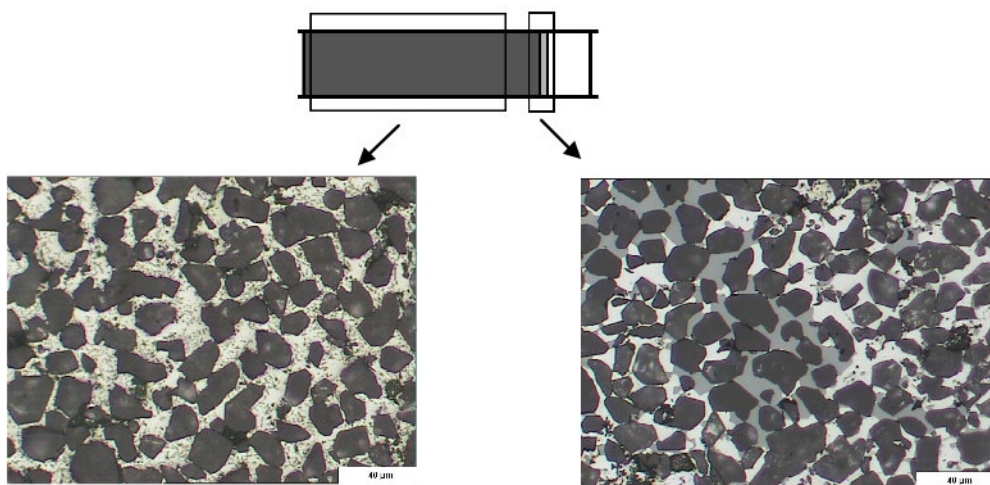
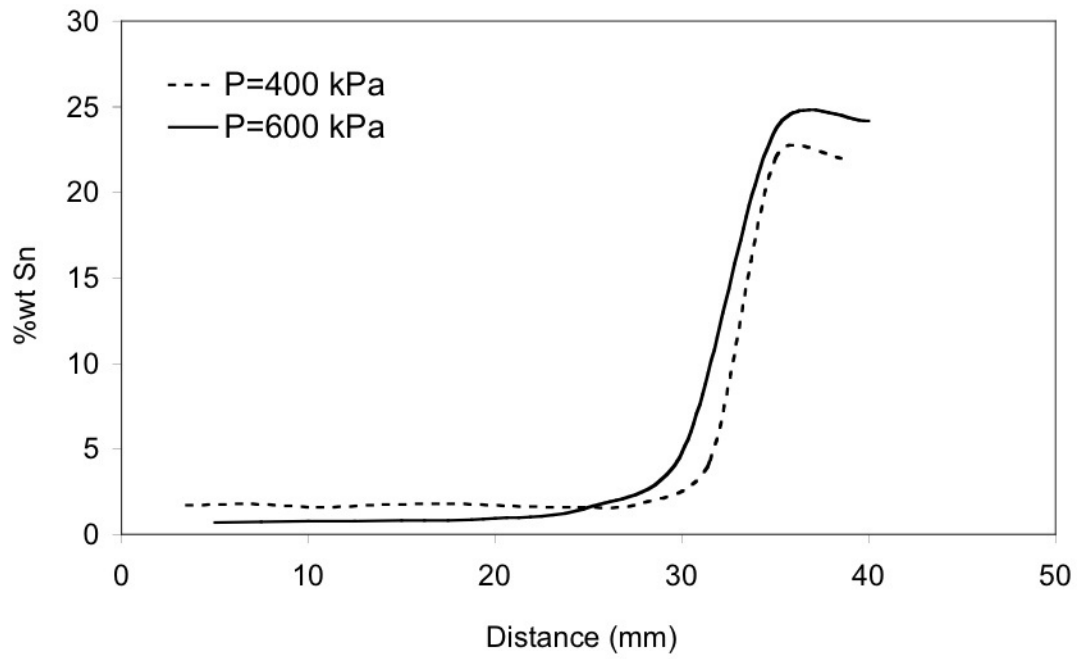


Fig.2



ACCEPTED M.

Fig.3



ACCEPTED M.