Effect of particle size and Mg content on the processing parameters of Al-Si-Mg/SiC_p composites processed by pressureless infiltration

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Recibido el 8 de marzo de 2007; aceptado el 4 de mayo de 2007

The effect of the following processing parameters, namely SiC particle size (20 and 75μm) and Mg content (3 and 6 wt.%), on residual porosity and on the degree of infiltration for SiC_p preforms by Al-Si-Mg alloys were investigated and quantified (here the subscript p means particles). The contribution of each of these parameters to the infiltration mechanism was determined employing an analysis of variance (ANOVA), and the effect of the levels of each parameter was examined using a surface response analysis. ANOVA results show that particle size is the parameter that most significantly affects the degree of infiltration and residual porosity. A surface response analysis shows that the degree of infiltration increases with an increase in magnesium content. The increasing rate in the degree of infiltration is higher for 20 μm than for 75 μm of SiC, whereas residual porosity diminishes with decreasing particle size; these two effects occur for both Mg concentrations in the alloy. Accordingly, the optimum parameters for a maximum degree of infiltration as well as for a minimum residual porosity are 20 μm SiC powders and 6 wt.% Mg in the alloy.

Keywords: Degree of infiltration; processing parameters; surface response analysis; Al-Si-Mg Alloy.

Se investigaron y cuantificaron los efectos de los siguientes parámetros de procesamiento, es decir el tamaño de partícula de SiC (20 y 75 μm) y el contenido de Mg (3 y 6% en peso), sobre el grado de infiltración para preformas de SiC_p con aleaciones de Al-Si-Mg y sobre la porosidad residual (aquí el subíndice p significa partículas). La contribución de cada uno de estos parámetros al mecanismo de infiltración se determinó empleando análisis de varianza (ANOVA), y el efecto de los niveles de cada parámetro se estudió usando análisis de respuesta de superficie. Los resultados de ANOVA muestran que el tamaño de partícula es el parámetro que más significativamente repercute sobre el grado de infiltración y la porosidad residual. Los análisis de respuesta de superficie muestran que el grado de infiltración aumenta al incrementar el contenido de magnesio. La razón de aumento en el grado de infiltración es mayor para 20 μm que para 75 μm de SiC, mientras que la porosidad residual decrece con la disminución del tamaño de partícula; estos dos efectos ocurren para ambas concentraciones de Mg en la aleación. Por consiguiente, los parámetros óptimos para el grado máximo de infiltración así como para la porosidad residual mínima son 20 μm para el tamaño de partícula de SiC y 6% en peso de Mg en la aleación.

Descriptores: Grado de infiltración; parámetros de procesamiento; análisis de respuesta de superficie; aleación de Al-Si-Mg.

PACS: 81.20.-n; 81.05.Mh; 81.05.Ni

1. Introduction

In the last two decades, silicon carbide-reinforced aluminum matrix composites have received a great deal of attention because they may exhibit greater strength and a lower thermal expansion coefficient than their individual components, from room temperature to elevated temperatures [1,2]. In general, these high volume fraction metal-matrix composites are being considered for wear resistance, structural, and electronic packaging applications. In addition, components such as cylinder liners, brake components, piston components, and ballistic armor plates may be produced by these ceramic preforms. Infiltration of molten metals into porous ceramic preforms is a technique suitable for processing composites. The infiltration can be classified into three categories based on the source of driving force: external pressure assisted, vacuum driven, and driven or pressureless capillarity; of there, pressureless infiltration is very attractive due to its cost effectiveness and near-net-shape capability [1]. However, in order for these materials to achieve their full potential, the Al-Si-Mg/SiC_p interface must be carefully tailored so that adhesion between the SiC and the aluminum alloy matrix is strong [2]. The production of Al/SiC_p composites requires the consideration of several important factors such as characteristics of the raw materials and process parameters. Two of the major problems frequently encountered in processing by the pressureless infiltration method are the presence of considerable levels of residual porosity and the development of unwanted reaction products, for example Al_4C_3. Residual porosity is related to an inadequate wetting of the silicon carbide by the molten aluminum, whereas unwanted phases are developed from the dissolution of the SiC reinforcement by the liquid aluminum [3-5]. Both problems can be overcome by adequately controlling the processing parameters such as alloy chemistry, temperature, atmosphere, preform porosity, particle size, etc. [6]. It has been reported that Mg in the system plays an important role on the wetting of ceramics by metals [7-9], and that the development of unwanted phases may be prevented, or at least retarded, either by additions of

silicon to the melt or by the presence of an oxide layer (SiO_2) on the SiC reinforcements [10-12]. It is always desirable to attain, simultaneously, the highest degree of infiltration and the lowest residual porosity in order to obtain the largest usable material volumes for technological applications. In this work, the effect of SiC particle size and the Mg level on the degree of infiltration of SiC_p preforms by aluminum alloys and residual porosity were investigated and quantified.

2. Experimental

The effect of the following processing parameters on the degree of infiltration of Al/SiC_p composites and residual porosity were investigated: SiC particle size and Mg content in the aluminum alloy. These parameters were studied for two particle sizes, 20 and 75 μm, and for two Mg doping concentrations, 3 and 6 wt.%. We chose these particular values because they are the optimized parameters for pressureless infiltration in the processing of Al-Si-Mg/SiC_p composites reported in the literature [13]. A full factorial experiment design of the 2^2 type was used. Factorial design allows us to determine the effect of a given factor at various levels on one or more response variables [14]. The contribution of each of the aforementioned parameters and their interactions on the variability of the degree of infiltration and residual porosity were examined using analysis of variance, and the effect of each of the levels was determined using surface response analysis. In Table I, a standard 2^2 factorial design indicating the established parameters and levels is shown. Silicon carbide preforms 40 and 60% porosity were prepared with 75 and 20 μm particle size powders, respectively. These percentages for the porosity were selected because, for processing on the degree of infiltration of silicon carbide preforms by Al-Si-Mg alloys, they represented the optimum values [6,13,15]. Both kinds of preforms were prepared by mixing thoroughly a predetermined amount of the SiC powders with 5 wt.% dextrin and distilled water. The SiC amounts are 7.68 and 11.52 g for 60 and 40% porosity, respectively. The dextrin was used as a medium binding with 5% wt. because this value is the content necessary for obtaining a mechanical consistency of the preform and, in turn, is the minimum value that does not present any problem when it evaporates. In addition, the distilled water helped to achieve a better mixing. The mixing was then compacted in a steel die to produce 3 cm $\times$ 4 cm $\times$ 0.5 cm slabs. The preforms were dried at 120°C in a forced air drier for two hours, and then cured at 225°C for two more hours. These last experimental conditions were previously studied, and it was found they are the best values for the infiltration process [15]. A preform was placed on top of a plate of the aluminum alloy (about 40 g) inside a ceramic container that was previously coated with boron nitride (BN). Here BN prevents not only the degradation of the mold but prevents the molten aluminum alloy from infiltrating into the porous mold. The chemical compositions of the alloys are shown in Table II, where the component percentages were determined by the spark emission technique with a resolution of about 0.010 nm. Infiltration trials were performed in a horizontal tube furnace with a 6.5 cm diameter alumina tube provided with end-cap fittings to control the process atmosphere. The preforms were heated in ultra high purity argon at a rate of 15°C/min up to 1150°C. At this temperature, in order to enhance the wetting of the SiC particles by the liquid aluminum, the atmosphere was changed to ultra-high-purity nitrogen and the system was held isothermally for 60 min. In previous studies [6,9], these values were found to be the best conditions to have adequate infiltrations. After cooling to room temperature in nitrogen atmosphere, the composites obtained were removed from the furnace to measure their densities. It is worth mentioning that, during heating, any residual binder was volatilized and removed from the system by the argon flow, whereas the presence of nitrogen in the atmosphere served, in addition to minimizing the oxidation of the substrate, to reintroduce the magnesium back into the aluminum melt [see below Eqs. (3) and (4)]. Specimens were sectioned and polished using standard metallurgical procedures, and microstructure characterization was performed using optical microscopy, scanning electron microscopy (SEM), and X-ray diffraction (XRD). The density of the composites was measured using the Archimedes principle. The degree of infiltration (%) in the composites was determined, knowing the value of the theoretical weight as well as the actual weight of the composites. Moreover, assuming full infiltration, the percentage residual porosity $\%P$ was calculated using the following formula:

$$\%P = \left(1 - \frac{\rho_C}{\rho_T}\right) \times 100, \tag{1}$$

where ρ_C is the measured density of the composite, and ρ_T is the theoretical density of the composite calculated using the law of mixtures.

TABLE I. Standard 2^2 factorial design showing the parameters investigated.

Trial	Mg (wt. %)	SiC Particle size (μm)
1	3	20
2	6	20
3	3	75
4	6	75

TABLE II. Chemical compositions (wt. %) of the alloys used in the experiment.

Alloy	Si	Mg	Total other elements*	Al
1	10.23	2.98	0.10	86.68
2	9.82	6.02	0.11	84.04

* Fe, Cu, Mn, Cr, Ni, Zn, B, Bi, Ca, Co, Li, Na, Pb, Sn, Sr, Ti, and V.

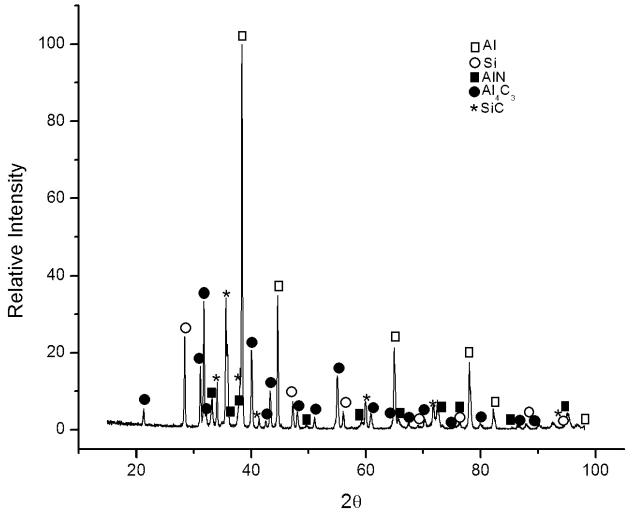

FIGURE 1. XRD pattern corresponding to composite obtained from trial 2.

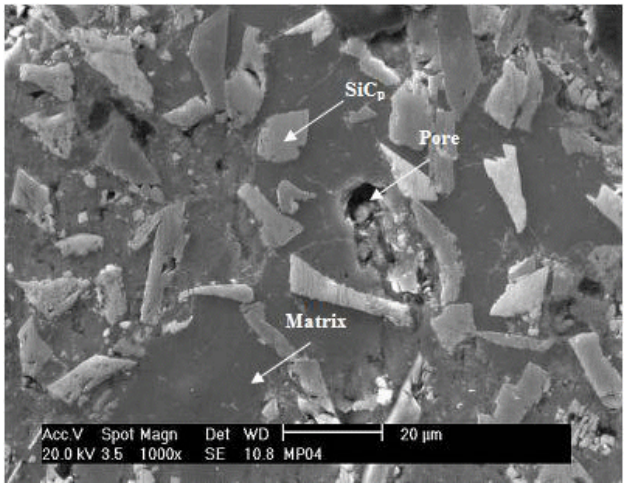

FIGURE 2. Scanning electron photomicrograph of the composite for trial 2.

3. Results and discussion

As mentioned above, specimens were sectioned and polished using standard metallurgical procedures. Once the samples had been optically checked, by means of optical microscopy, and it was ensured that they had been adequately polished, we proceeded to microstructurally characterize them using scanning electron microscopy (SEM) and x-ray diffraction (XRD). Figure 1 shows the XRD pattern only for the specimen of trial 2, which reveals the formation of Al_4C_3 and AlN phases. All of the trials present the same phases, but specimen 2 was selected in Fig. 1 because it exhibited the highest degree of infiltration (97.33%) and the lowest residual porosity (4.10%). It should be noted that the area of the composite analyzed by XRD corresponds to the face that was in direct contact with the aluminum alloy. Since this side of the preform is in fact the region that was in contact with the liquid aluminum for the longest time period, it is the most susceptible area for developing Al_4C_3 phase by partial dissolution of the SiC particles [9]. The chemical reaction is:

$$3SiC(s) + 4Al(l) \rightarrow Al_4C_3(s) + 3Si(s). \quad (2)$$

On the other hand, the presence of AlN is attributed to a series of chemical processes that occur between Mg and nitrogen in the atmosphere, where the formation of Mg_3N_2 is a prerequisite for AlN creation [10] in accordance with the following chemical reactions:

$$3Mg + N_2 \rightarrow Mg_3N_2 + \Delta T, \quad (3)$$

$$Mg_3N_2 + 2Al \rightarrow 2AlN + 3Mg. \quad (4)$$

All of the XRD patterns also show the same AlN formation in the four composites. In addition, we observed that the Mg_3N_2 phase was not formed in any sample, and therefore we assume that this phase was completely consumed in the formation of the AlN phase. If an undesirable Al_4C_3 phase is formed due to the dissolution of SiC$_p$ during the infiltration processing, it may cause a decrease in the degree of infiltration, and thereby increase the residual porosity. On the other hand, it can in no way be supposed that the AlN phase modifies the infiltration and residual porosity, because it is not present before the infiltration occurs. The aluminum nitride phase appears in subsequent stages according to whether the reaction is between nitrogen gas and liquid aluminum, or only gas phases in; in this last reaction type AlN phase generally is formed inside porous or composite porous preform. A photomicrograph showing a typical microstructure in specimen 2 is illustrated in Fig. 2. As can be appreciated in Fig. 2, SiC$_p$ particles exhibit several grey tones, including light grey, due to the different crystallographic orientations. For the same reason, it is often a great challenge to achieve a good color contrast between matrix and particles during the electron microscopy analyses. Nonetheless, due to the region's morphology and the confirmation of its composition by means of

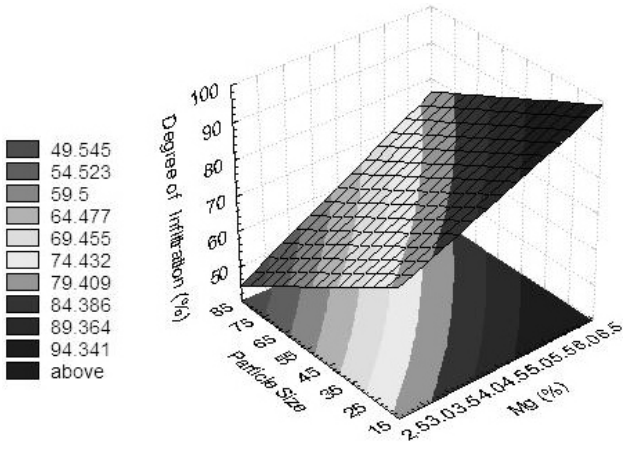

FIGURE 3. Surface response plot for the interaction between SiC particle size and Mg content in the aluminum alloy on the degree of infiltration.

TABLE III. ANOVA results for degree of infiltration.

Source	Sum Squares	Degrees of Freedom	Mean Square	Distribution F_o	Statistical Significance P
(A) Mg content in Al alloy*	840.295	1	840.2950	99.2724	0.000570*
(B) Particle size *	908.872	1	908.872	107.3740	0.000490*
AB	11.353	1	11.353	1.3412	0.311260
Error	33.858	4	8.4645		
Total	1794.377	7			

1% Significance

TABLE IV. ANOVA results for percentage residual porosity.

Source	Sum Squares	Degrees of Freedom	Mean Square	Distribution F_o	Statistical Significance P
(A) Mg content in Al alloy *	0.51511	1	0.51511	2.1357	0.217705
(B) Particle size*	59.45951	1	59.45951	246.5282	0.000096*
AB	21.81301	1	21.81301	90.4401	0.000682*
Error	0.96475	4	0.24119		
Total	82.75239	7			

1% Significance

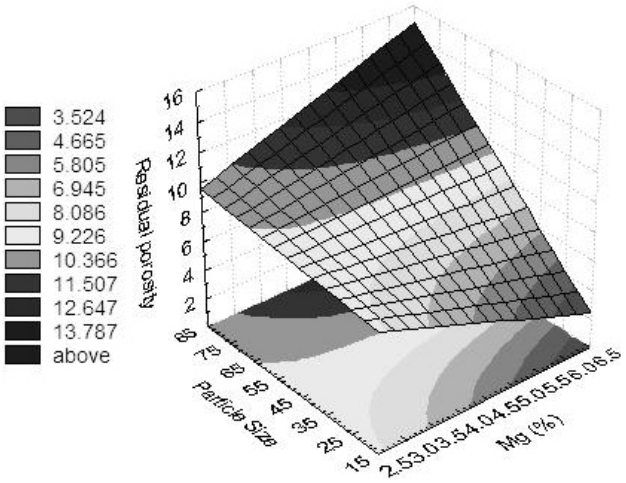

FIGURE 4. Surface response plot for the interaction between SiC particle size and Mg content in the aluminum alloy on the residual porosity.

energy dispersive x-rays, it is easy to identify the Al-matrix-embedded SiC particles. Results from the analysis of variance are shown in Tables III and IV. According to Table III, the parameters that most significantly affect the degree of infiltration within 1% significance are the SiC particle size and Mg content in the alloy. The effect of particle size can be fairly well explained in terms of the capillary pressure [6] as follows: particle size is expected to influence the residual porosity according to the following equation [16]:

$$\Delta\wp = \frac{2\gamma}{r}\cos\theta, \tag{5}$$

where $\Delta\wp$ is the capillary pressure difference, γ is the liquid surface tension, r is the capillary radius, and θ is the wetting angle. The smaller the capillary radius, the higher the pressure difference. Regardless of the Mg level in the alloy, the smaller the particle size, the lower the porosity. Therefore, a higher infiltration degree and a lower residual porosity are expected. To determine the effect of the levels of each factor on the degree of infiltration of SiC_p preforms, a surface response plot was constructed. According to Fig. 3, the highest degree of infiltration can be obtained by using an alloy with 6 wt.% Mg and a 20 μm particle size of SiC. On the other hand, the analysis of variance results show that particle size is the parameter that most significantly affects the residual porosity, followed by the interaction between SiC particle size and Mg content in the alloy. As can be seen from Fig. 4, surface response analysis reveals that the lowest residual porosity is obtained with 20 μm particle size and 6 wt.% Mg. Accordingly, the optimum parameters for a maximum degree of infiltration and minimum residual porosity in the composites are a 20 μm particle size of SiC and 6 wt.% Mg in the aluminum alloy.

4. Conclusions

The effect of the SiC particle size and Mg concentration in the aluminum alloy on the degree of infiltration and the residual porosity of SiC_p preforms by Al-Si-Mg alloys were investigated. Results from ANOVA are in excellent agreement with those obtained by surface response analysis. ANOVA results show that particle size is the parameter that most strongly affects the degree of infiltration, followed by the magnesium content in the alloy. The surface response analysis shows that the degree of infiltration increases not only with an increase in magnesium content, but also as the SiC particle size diminishes. A maximum degree of infiltration can be achieved

by using a 20 μm particle size and a 6 wt.% Mg aluminum alloy. ANOVA results also show that particle size is the parameter that most significantly affects the residual porosity, followed by the interaction between magnesium content in the alloy. The surface response analysis reveals that residual porosity diminishes with decreasing particle size and with an increase in magnesium content. Minimum residual porosity can be achieved by using 20 μm particle size and a 6 wt.% Mg aluminum alloy.

Acknowledgements

The authors gratefully acknowledge financial support from the National Council of Science and Technology (CONACyT) in México and Microabrasivos de México for supplying the SiC powders.

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