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Infiltration-Diffusional-Solidification (IDS): A P/M+Liquid Metal Infiltration Route for Processing Metal Matrix Composites

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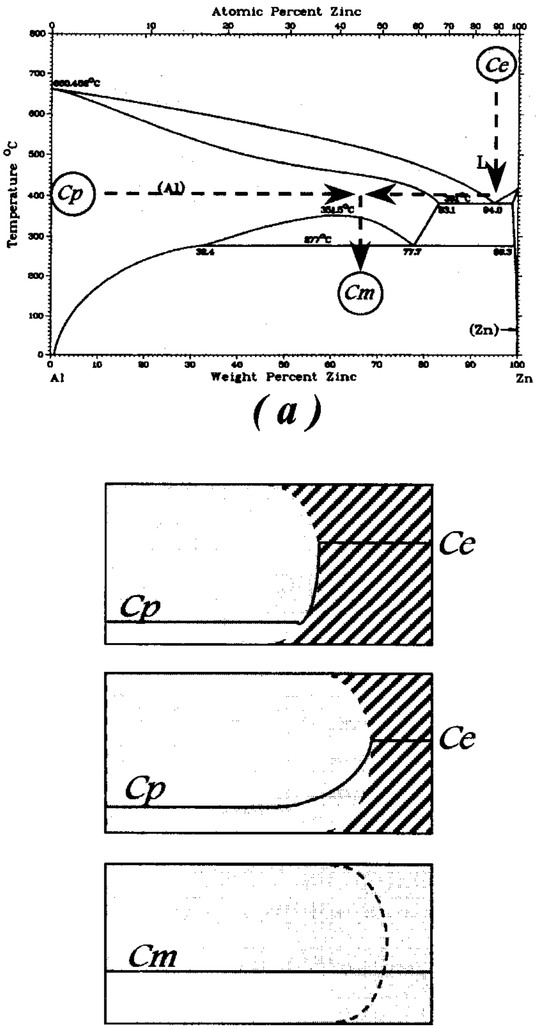
Abstract. A process for the production of MMCs, based on the infiltration by a low melting temperature eutectic alloy of porous preforms containing a mixture of reinforcing ceramic particles and pure metal powder is described.. The infiltration is done at temperatures higher than the eutectic temperature, but below the fusion temperature of the pure metal. After infiltration with the low melting temperature liquid, a diffusional solidification process occurs, where the solidification is controlled by the partition of the alloying elements between the solid and the melt. Preforms prepared with mixtures of Al powders and SiCp, were cold pressed to different pressures so as to obtain from 25% to 10% volume fractions of pores. The compression curves obtained are described. Infiltration experiments using eutectic Al-Cu alloys (33%weight Cu) and Zamak 5 (Zn-4% Al- 1%Cu) were run on the preforms, varying the infiltration pressure and the cooling rate after infiltration. Infiltrated samples were microstructurally characterized by optical and scanning electron microscopy. The final structures showed that the IDS concept is feasible, producing MMC's with homogeneous matrixes and regular distributions of SiCp.

Introduction

The unique properties that can be obtained by the use of Metal Matrix Materials (MMCs) widened the range of possible applications of metallic alloys. MMCs have mainly been used for structural applications, where high modulus, high specific strength, and low densities are desirable. Other desirable properties that can only be achieved by composite materials are increased wear resistance and very low or controlled thermal expansion. Among all MMCs, aluminum matrix reinforced with SiC particles (SiCp) composites, in particular, attract much attention due to the low cost of processing and cheap raw materials. The main processing routes for these MMCs involve either liquid metals processing, usually mechanical mixtures of ceramic particles and liquid metals, or powder metallurgy with consolidation by sintering or hot mechanical work. Other processes are the Osprey process (or spray co-deposition) and liquid metal infiltration of a ceramic preform. The low wettability of SiC based ceramics by molten Al requires the aid of external pressure to promote the infiltration of preforms. A mechanical device can achieve this external pressure as in squeeze casting or trough the action of inert gas pressurisation, as in the PIC process [1, 2].

The degree of homogeneity of the SiC particles dispersion is an important microstructural parameter for Al/SiCp MMCs, directly related to the mechanical obtained properties [3]. Chemical composition of the matrix, volume fraction and size distribution of the reinforcing particles and absence of undesirable phases formation at the interface between matrix and reinforcement are also key parameters for performance [3].

Pressure infiltration of porous ceramic preforms by molten metals is a cost competitive, near net shape route for fabricating MMCs. It is easy to prepare low volume fraction preforms of continuous fibers, producing low volume fractions ceramic particle preforms are nearly impossible. The minimal volume fractions attainable with particulate preforms are in the range of 45 to 60 percent in volume [4], due to the packing characteristics of equiaxed particles. MMCs produced by liquid metal infiltration normally have a large volume fraction of particulate reinforcements.



(a)

(b)

Fig. 1 Phase diagram for the Al-Zn system; Ce=composition of infiltrating liquid, Cp=composition of the pure metal preform and Cm=final average composition

E. M. Klier at MIT [1, 5] suggested a way to overcome this limitation, by a process where the infiltration of a packed bed of particles is followed by dispersion into more liquid metal using a semi-solid slurry mixer (the "Norton process"). A similar process is also used by Lanxide.

This work proposes an alternate route for the production of particulate reinforced MMCs with any volume fraction from 0 to circa 55%. The process is based on the infiltration by a low melting temperature eutectic alloy of a porous preform made with a mixture of reinforcing ceramic particles and a pure metal powder. The infiltration is done at temperatures above the eutectic and below the melting temperature of the pure metal. Once infiltrated with the eutectic, the solidification proceeds with a planar front and it is controlled by the partition of the alloy elements between the solid particles and the melt. A similar process was proposed by Langford [6] in the 70's for casting and welding steels and it is known in the literature as Diffusional Solidification (DS) [10] or sometimes isothermal (or adiabatic) solidification. In DS casting, a high alloy content liquid is brought in contact with a low alloy or pure material, and the liquid solidifies by rejecting solute to the surrounding low alloy solid metal. In his original paper, Langford used a mould filled with uniform sized steel shot, heated the mould to a temperature below the solidus of the steel shot and then infiltrated it with liquid cast iron, using an inert gas to pressurize the liquid. Figure 1 shows a schematic description of the mechanism of DS casting. After infiltration, the sample behavior is very similar to transient liquid sintering. The liquid gradually disappears by a diffusion controlled solidification process. The proposed name for this route for processing MMC's is Infiltration-Diffusional-Solidification or IDS. In our experiments the solidification occurs as much due to the diffusion as due to heat transfer, as the furnace is turned of during and after infiltration.

The benefits of this route are the rapid solidification, lower processing temperatures, near net shape possibilities and fundamentally, the possibility of producing MMCs with a low volume fraction of particulate reinforcements by infiltration with a molten metal.

In comparison with structures obtained by conventional solidification of metal containing dispersed particles, which tend to segregate interdendritically, in the ISD process particles are essentially homogeneously dispersed.

1. Producing a MMC with a given volume fraction of reinforcement and matrix composition by IDS

In order to derive equations that describe a MMC produced by IDS process, relating the volume fraction of reinforcement and the chemical composition of the matrix with the fraction of the theoretical density of the preform (f_{td}) , ceramic reinforcement/pure metal ratio k, composition of the infiltrating liquid Ce and densities of the reinforcement particles, pure metal particles and infiltrating liquid, it is assumed that there is no volume change of the preform during the infiltration and solidification, so the volume fraction of the reinforcement V_r in the preform is the same as in the final composite. Thus, V_r is defined by the level of compaction of the preform obtained by a mixture of pure metal powder and reinforcement particle. Defining V_r' as the volume fraction of reinforcement in the preform supposing it 100% dense, then the fraction of theoretical density of the preform f_{td} is given by equation (1),

$$V_r / V_r' = f_{td} \qquad (1)$$

and V_r' is given by equation (2)

$$V_r' = m_r*d_p / (m_p*d_r + m_r*d_p) \qquad (2)$$

where, m and d are mass and density, respectively, and subscripts $_r$ and $_p$ indicate reinforcement and pure metal powder, respectively. V_r can thus be written as:

$$V_r = k*d_p*f_{td} / (d_r + k*d_p) \qquad (3)$$

where k is the mass ratio of reinforcement powder and pure metal powder ($k = m_r / m_p$).

Supposing there is no mutual solubility nor reaction between the matrix and the reinforcement, the solute content of the homogenised matrix C_m of the MMC produced by IDS process is defined by the dilution of the solute content of eutectic alloy C_e into the pure metal powder of the preform:

$$C_m = M_e * C_e / M_m \qquad (4)$$

but

$$M_m = M_e + M_p \qquad (5)$$

and generically:

$$M_i = V_i * d_i / d_c \qquad (6)$$

where M and V indicate mass fraction and volume fraction, respectively, and subscript $_m$, $_e$, $_c$ and $_i$ indicate matrix, eutectic alloy, composite and generic constituent, respectively.

Assuming that all porosity of the preform is filled by the infiltrating alloy, and overlooking the contraction during solidification:

$$V_e = V_{pore} = 1 - f_{td} \qquad (7)$$

$$V_p = 1 - V_r - V_{pore} \qquad (8)$$

where V_{pore} is the volume fraction of pores in the preform

Thus, Cm can be written as:

$$C_m = (1 - f_{td})*d_e*C_e / [(1-f_{td})*d_e + (f_{td} - V_r)*d_p] \qquad (9)$$

In order to design a desired MMC with a given reinforcement volume fraction and a given matrix composition by IDS process, it is necessary to input the mass ratio of reinforcement and pure metal in the and volume fraction of porosity in the preform and the composition of the alloy. The description of the above makes reference to the infiltration by a molten eutectic alloy, but any alloy or even a pure metal can be used in the IDS process , provided that it's melting temperature is bellow the melting temperature of the metal powder in the preform.

Figs. 2 and 3 are plots containing the IDS parameters as described by Equations (3) and (9) for systems involving Al powder and SiC particle preforms, infiltrated with eutectic Al-33%Cu or Zn-5%Al alloys, respectively, supposing densities of 2.7 g/cm^3 for the Al, 3.2 g/cm^3 for the SiC, 3.6 g/cm^3 for Al-33%Cu and 6.7 g/cm^3 for Zn-4%Al-1%Cu. For composites produced from preforms compacted at pressures from 100 to 350 MPa, volume fractions of SiC varying from 0.22 to 0.28 are expected, as well as matrix compositions from 14 to 5 wt% Cu when infiltrated with Al-33%Cu or 54 to 24 wt% Zn when infiltrated with the 95wt%Zn alloy.

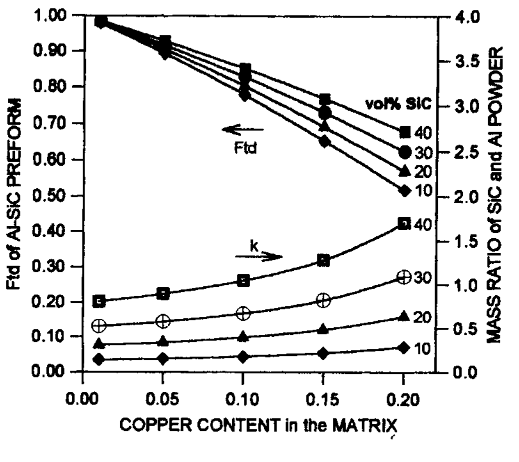


Fig. 2: IDS parameters for infiltration with Al-33%Cu, from equations (3) and (9).

As an example, in order to produce a composite with 20 vol.%SiC$_p$ dispersed in matrix of Al-20% solute, in the case of Al-Cu matrix it would be necessary a preform with 57% ftd containing a mass ratio of SiC particle / Al powder of 0.64 (Fig. 2), while in the case of Al-Zn matrix, the preform must have 92% ftd and 0.33 k (Fig. 3).

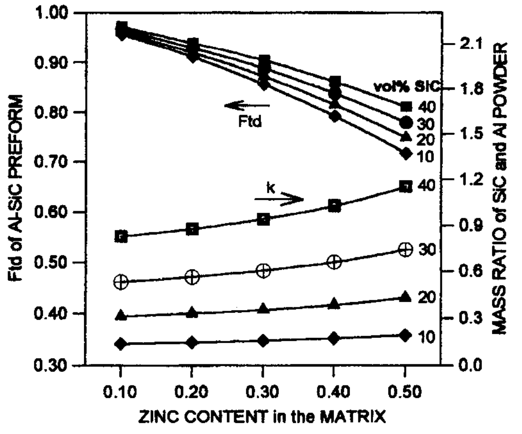


Fig. 3: IDS parameters for infiltration with Zn-5%Al, from equations (3) and (9).

Experimental Procedure

Commercial purity Al powders (Alcoa-Brazil, type 123, 51μm average size) were mixed for one hour with SiC particles (Carborundum "Black SiC powder", 320 class, with 42μm average size), using a ceramic reinforcement / pure metal ratio k of 34 wt% SiC / 66 wt% Al or 0.52. The compressibility curves for this powder mixture were determined for uniaxial pressing at room temperature using a cylindrical die with 1 cm^2 section, producing preforms with f_{td} from 0.7 to 0.9.

Preforms with f_{td} of 0.72; 0.74; 0.88 and 0.91 were used for infiltration. Preforms were loaded into cylindrical quartz glass crucibles, separated from the infiltrating alloy by a drilled graphite spacer. The assemble was introduced into an inert gas pressure infiltration casting device ("PIC" machine, built at EPUSP [1, 5]), which was evacuated to 10^{-3} mm Hg, heated to the desired temperature and then pressurized with N_2 at 4.5 MPa so as to achieve the infiltration. After the infiltration the samples were slowly cooled in the furnace.

Experiments were run with two different near eutectic compositions: binary Al-CuAl$_2$ alloy (Al-33 wt%Cu) and commercial zinc - aluminum AC41A or Zamak 5 alloy (Zn-4 wt%Al-1 wt%Cu). The infiltration with the Al-Cu eutectic, which melts at ~548 °C, were run at 590 °C while the infiltration with Zamak 5, which melts between 380 and 386 °C, were run at 470 °C.

The samples were cut, mounted, mechanically polished using diamond slurry and final polished colloidal silica, in some cases chemically etched, and analyzed by optical and scanning electron microscopy.

Results and Discussion

Fig. 4 shows a SEM image (backscattered electrons) of a composite produced from a 0.88 ftd preform. The eutectic alloy filled almost all the pores along the preform; a small amount of microporosity is left, originated from solidification shrinkage and the difficulty of penetration on tight space (crevices) between SiC particles. The SiC (dark grey phase) is homogeneously dispersed in the matrix, with no SiC clusters formation. The white phase seen in SEM was identified as CuAl2 (θ phase). The θ phase outlines the former Al particle boundaries, around and between the SiC particles. No reaction products were observed between the molten metal and the SiC. Similar structures were observed on the composite obtained from a 0.74 ftd preform. The main observed difference was the higher amount of θ phase found. In none of the samples the eutectic morphology assumed its usual "regular eutectic" lamellar morphology. This was interpreted as an evidence for the occurrence of diffusional solidification at first, until the temperature drops below the eutectic temperature.

When the final matrix composition falls inside the eutectic range, a divorced eutectic growth mechanism occurs, where the eutectic Al phase solidifies over the Al particles of the preform, while massive CuAlz phase nucleates heterogeneously at SiC particle surfaces. Figs. 5 show the solid state precipitation of 9 phase as needles or platelets in the central region of former Al grains, confirming the diffusion of Cu to the core of the former Al powder particles. A few long blade-like plates present in the microstructure were identified by microanalysis as Cu2FeAh. Solution heat treatments at 510 °C for 12 hours were enough to dissolve completely the A phase precipitates within the Al. With 100 h at 510 °C the volume fraction of 6 between the former Al particles diminished, approaching the equilibrium condition. Figure 6 shows an optical micrograph of the border of the sample. Three different regions can be identified: a region showing the Al-CuAl2 eutectic structure, a region showing the infiltrated composite and an intermediate region, all around the former preform region, with an average thickness of 1 mm, presenting a larger amount of A phase, both massive and lamellar, a slightly bigger volume fraction of SiC and larger and more frequent pores than in the inner region of the preform. The explanation proposed for this intermediate region is that during infiltration, the Al particles at the outer regions of the preform are partially dissolved by the liquid, so the preform collapses partially inward. The increased amount of pores is due to solidification shrinkage of the larger amount of liquid. The presence of a transition layer with a larger amount of reinforcement can eventually be exploited as a wear resistant layer outside a tough core, using a "property gradient material" approach.

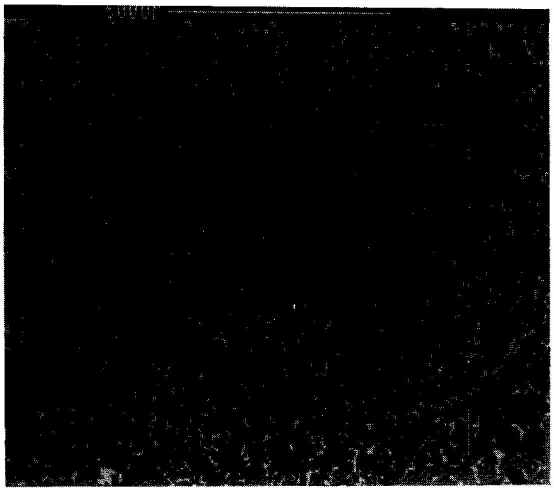


Fig.4 SEM image (backscattered electrons) of a 0.88 ftd preform infiltrated with Al-33%Cu. The white phase is $CuAl_2$ (θ phase).

Fig. 5 Solid state precipitation of θ phase as needles or platelets in the central region of former Al grains; SEM, backscattered electrons.

Fig. 6 Optical micrograph of the border of the sample, showing an Al-$CuAl_2$ eutectic structure region, the infiltrated composite and an intermediate region containing a larger amount of Cu-Al_2 θ phase.

Fig. 7 Detail of the intermediate region showing the eutectic matrix with embedded SiC particles

Figure 8 shows a similar area of a a 9% porosity (0.91 ftp) preform infiltrated with Zamak 5. The infiltration of a 28% porosity (0.72 ftp) preform by Zamak 5 at 470°C was not successful: the structure became very heterogeneous due to a partial dissolution of the Al powder particles. Infiltration at lower temperatures can probably prevent this.

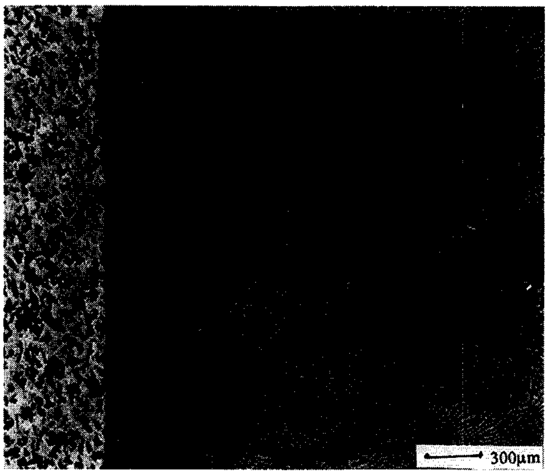


Fig 8 Micrograph of a 9% porosity (0.91 ftd) preform infiltrated with Zamak 5, showing a Zn-Al lamellar eutectic layer, external to the preform, a dilution layer containing SiC particles and a fully dense composite layer. Villela's etchant.

Fig.9 SEM image of the core of a 0.91ftd preform infiltrated with Zamak 5, etched with Villela's reactive, showing that the matrix composition is near the Al-Zn eutectoid (~23wt%Zn). Expected Vsic and Cm are 0.28 and 24 % Zn, respectively.

Conclusions

Metal matrix composites with Al-Cu and Al-Zn matrixes reinforced with particulate SiC were produced. The Infiltration-Diffusional-Solidification (IDS) process is a feasible process, allowing the production of MMC's with homogeneous matrixes and regular distributions of SiCp, without clustering, provided that the reinforcement particles have near the same size as the pure metal powder. The obtained structures rival the ones obtained by powder metallurgy, a more costly and not near-net-shape process.

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