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Metal matrix composites: production by the stir casting method

J. Hashim, L. Looney*, M.S.J. Hashmi

School of Mechanical and Manufacturing Engineering, Dublin City University, Dublin 9, Ireland

Abstract

Combining high specific strength with good corrosion resistance, metal matrix composites (MMCs) are materials that are attractive for a large range of engineering applications. Given the factors of reinforcement type, form, and quantity, which can be varied, in addition to matrix characteristics, the composites have a huge potential for being tailored for particular applications. One factor that, to date, has restricted the widespread use of MMCs has been their relatively high cost. This is mostly related to the expensive processing techniques used currently to produce high quality composites. In this paper, the relatively low cost stir casting technique is evaluated for use in the production of silicon carbide/aluminium alloy MMCs. The technical difficulties associated with attaining a uniform distribution of reinforcement, good wettability between substances, and a low porosity material are presented and discussed. © 1999 Elsevier Science S.A. All rights reserved.

Keywords: Metal matrix composites; Composites; Composites processing; Stir casting

1. Introduction

The aim involved in designing metal matrix composite materials is to combine the desirable attributes of metals and ceramics. The addition of high strength, high modulus refractory particles to a ductile metal matrix produces a material whose mechanical properties are intermediate between the matrix alloy and the ceramic reinforcement. Metals have a useful combination of properties such as high strength, ductility and high temperature resistance, but sometimes have low stiffness, whereas ceramics are stiff and strong, though brittle. Aluminium and silicon carbide, for example, have very different mechanical properties: Young's moduli of 70 and 400 GPa, coefficients of thermal expansion of 24×10^{-6} and $4 \times 10^{-6}/^{\circ}\text{C}$, and yield strengths of 35 and 600 MPa, respectively. By combining these materials, e.g. A6061/SiC/17p (T6 condition), an MMC with a Young's modulus of 96.6 GPa and a yield strength of 510 MPa can be produced [1]. By carefully controlling the relative amount and distribution of the ingredients of a composite as well as the processing conditions, these properties can be further improved.

Among the variety of manufacturing processes available for discontinuous metal matrix composites, stir casting is generally accepted as a particularly promising route, cur-

rently practised commercially. Its advantages lie in its simplicity, flexibility and applicability to large quantity production. It is also attractive because, in principle, it allows a conventional metal processing route to be used, and hence minimizes the final cost of the product. This liquid metallurgy technique is the most economical of all the available routes for metal matrix composite production [2], and allows very large sized components to be fabricated. According to Skibo et al. [3], the cost of preparing composites material using a casting method is about one-third to half that of competitive methods, and for high volume production, it is projected that the cost will fall to one-tenth. Table 1 shows a comparative evaluation of the different processes commonly used for discontinuously reinforced metal matrix composites (DRMMC) production.

In general, the solidification synthesis of metal matrix composites involves producing a melt of the selected matrix material followed by the introduction of a reinforcement material into the melt, obtaining a suitable dispersion. The next step is the solidification of the melt containing suspended dispersoids under selected conditions to obtain the desired distribution of the dispersed phase in the cast matrix. In preparing metal matrix composites by the stir casting method, there are several factors that need considerable attention, including

1. The difficulty of achieving a uniform distribution of the reinforcement material;

*Corresponding author. Fax: +353-01-704-5345

E-mail address: lisaaloney@dcu.ie (L. Looney)

Table 1
A comparative evaluation of the different techniques used for DRMMC fabrication [2]

Method	Range of shape and size	Metal yield	Range of volume fraction	Damage to reinforcement	Cost
Liquid metallurgy (stir casting)	wide range of shapes; larger size; up to 500 kg	very high, >90%	up to 0.3	no damage	least expensive
Squeeze casting	limited by preform shape; up to 2 cm height	low	up to 0.45	severe damage	moderately expensive
Powder metallurgy	wide range; restricted size	high		reinforcement fracture	expensive
Spray casting	limited shape; large size	medium	0.3–0.7	–	expensive
Lanxide technique	limited by pre-form shape; restricted size	–	–	–	expensive

2. wettability between the two main substances;
3. porosity in the cast metal matrix composites; and
4. chemical reactions between the reinforcement material and the matrix alloy.

In order to achieve the optimum properties of the metal matrix composite, the distribution of the reinforcement material in the matrix alloy must be uniform, and the wettability or bonding between these substances should be optimised. The porosity levels need to be minimised, and chemical reactions between the reinforcement materials and the matrix alloy must be avoided. In this paper, issues relating to the first three of these factors will be presented and discussed.

2. The distribution of reinforcement materials

One of the problems encountered in metal matrix composite processing is the settling of the reinforcement particles during melt holding or during casting. This arises as a result of density differences between the reinforcement particles and the matrix alloy melt. The reinforcement distribution is influenced during several stages including (a) distribution in the liquid as a result of mixing, (b) distribution in the liquid after mixing, but before solidification, and (c) redistribution as a result of solidification.

The mechanical stirrer used (usually during melt preparation or holding) during stirring, the melt temperature, and the type, amount and nature of the particles are some of the main factors to be considered when investigating these phenomena. Proper dispersion of the particles in a matrix is also affected by pouring rate, pouring temperature and gating systems [4].

The method of the introduction of particles into the matrix melt is one of the most important aspects of the casting process. It helps in dispersing the reinforcement materials in the melt. There are a number of techniques [5,6] for introducing and mixing the particles including

1. Injection of the particles entrained in an inert carrier gas into the melt with the help of an injection gun, wherein the particles are mixed into the melt as the bubbles rise through the melt;
2. addition of particles into the molten stream as the mould is filled;
3. pushing particles into the melt through the use of reciprocating rods;
4. spray casting of droplets of atomised molten metal along with particles onto a substrate;
5. dispersion of fine particles in the melt by centrifugal action;
6. pre-infiltrating a packed bed of particles to form pellets of a master alloy, and redispersing and diluting into a melt, followed by slow hand or mechanical stirring;
7. injection of particles into the melt while the melt is irradiated continuously with high intensity ultrasound; and
8. zero gravity processing which involves utilising a synergism of ultra-high vacuum and elevated temperature for a prolonged period of time.

The vortex method is one of the better known approaches used to create and maintain a good distribution of the reinforcement material in the matrix alloy. In this method, after the matrix material is melted, it is stirred vigorously to form a vortex at the surface of the melt, and the reinforcement material is then introduced at the side of the vortex. The stirring is continued for a few minutes before the slurry is cast. Harnby et al. [7] studied different designs of mechanical stirrers, as shown in Fig. 1. Among them, the turbine stirrer is quite popular. During stir casting for the synthesis of composites, stirring helps in two ways: (a) transferring particles into the liquid metal, and (b) maintaining the particles in a state of suspension.

Several of the methods listed have disadvantages and limitations. The development of the vortex during stirring is observed to be helpful for transferring the particles into the matrix melt as the pressure difference between the inner and the outer surface of the melt sucks the particles into the liquid [8]. However, air bubbles and all the other impurities on the surface of the melt are also sucked into the liquid by the same mechanism, resulting in high porosity and inclusions in the cast product. The formation of pores as a result of the vortex method is discussed in greater detail in Section 4. A vigorously stirred melt will entrap gas which proves to be extremely difficult to remove as the viscosity

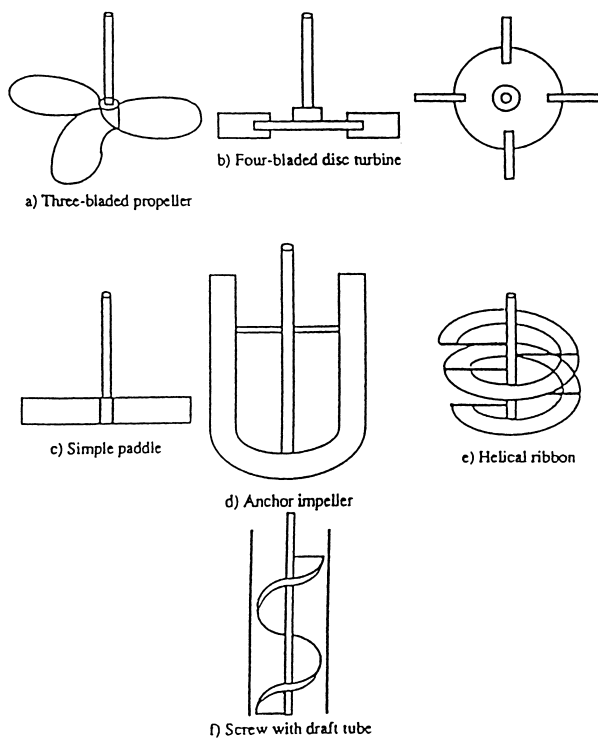


Fig. 1. Mechanical stirrer designs [7].

of the slurry increases. Gas injection of particles introduces a quantity of gas into the melt. Some of the methods, such as the ultrasonic, are very expensive and difficult to scale to production level. Zero gravity processing is a very complicated method, and is difficult to characterize. On account of centrifugal action, the distribution of the particles varies from the inner to the outer part of the billet because of the difference in centrifugal force [9]. Introducing the reinforcement particles from air to the stirred molten matrix will sometimes entrap the particles with other impurities, such as metal oxides and slag which are formed on the surface of the melt. While pouring is done, air envelopes are also formed between the particles, altering the interface properties between the particles and the melt, and retarding the wettability between them. In the case where the particles added are not at the same temperature as the slurry, the temperature, and consequently, the viscosity of the slurry will change vary rapidly.

The particle distribution in cast composites may become inhomogeneous even when a homogeneous state of suspension is maintained in the slurry. During the solidification of a liquid matrix alloy containing dispersed second phase particles, the particles in the melt can migrate towards or away from the freezing front, and a particle near the freezing front will either be rejected or engulfed. These two phenomena lead to the redistribution of the particles during solidification. This means that the solidification cell size, and hence the solidification rate influence the distribution of the reinforcement particles in the final ingot. Fine dendrite arm spacing (DAS) produces a more uniform distribution of the

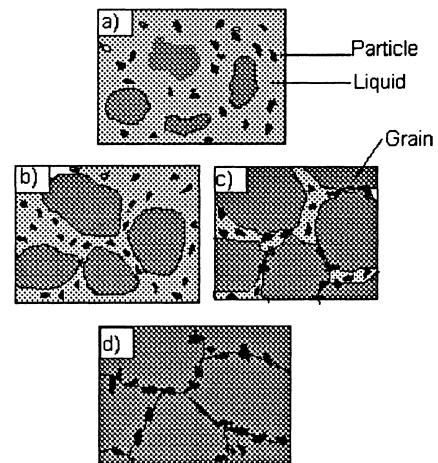


Fig. 2. Schematic representation of the particle distribution during solidification [11]: (a) grain growing freely in the liquid; (b) particle/grain interaction occurs; (c) some particles are pushed by the interface, while some others are engulfed; (d) the final distribution of particle in the matrix material.

particles, whereas larger DAS leads to particle clustering [10]. Rapidly solidified structures, therefore, give a better distribution of the particles due to finer dendrite size as well as due to a limited settling of the particles resulting from the reduced time during which the composites are in a molten state. A schematic representation of the distribution of the particles during solidification [11] is shown in Fig. 2. Secondary fabrication processes, such as extrusion, can modify the distribution of the particles, but complete declustering cannot be achieved even at the highest extrusion ratio [12].

A successful casting process must be able to produce a composite in which the particles are uniformly dispersed throughout the matrix. The thoroughness of the agitation is determined by many factors, such as the shape of the agitator, its speed, and its placement relative to the melt surface and the wall of the crucible. It is suggested that both the matrix and the reinforcement materials be pre-heated at a certain temperature before being mixed to release all the moisture and trapped air between the particles. The stirrer must be designed such that it avoids the agitation of the melt surface, and the formation of vortex must be avoided or minimised. The stirring speed should not be too high, but should be continuous for a few minutes before the material is poured into a mould through the bottom of the crucible. Bottom pouring is necessary in order to avoid impurities on the surface of the melt being cast into the mould.

3. Wettability between reinforcement material and matrix alloy

Wettability is another significant problem when producing cast metal matrix composites. Wettability can be defined as the ability of a liquid to spread on a solid surface. It also describes the extent of intimate contact between a

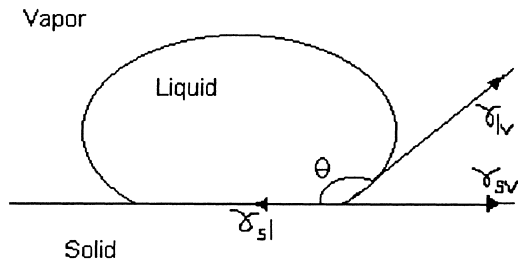


Fig. 3. Schematic diagram showing the contact angle that describes wettability [14].

liquid and a solid. Successful incorporation of solid ceramic particles into casting requires that the melt should wet the solid ceramic phase. The problem of the wetting of the ceramic by molten metal is one of surface chemistry and surface tension. The chemistry of the particle surface, including any contamination, or oxidation, the melt surface and oxide layer must be considered. The basic means used to improve wetting [13,14] are (a) increasing the surface energies of the solid, (b) decreasing the surface tension of the liquid matrix alloy, and (c) decreasing the solid–liquid interfacial energy at the particles–matrix interface. The bonding force between the liquid and solid phases can be expressed in terms of contact angle referred to in the Young–Dupre equation [15]. The magnitude of the contact angles (θ) in this equation, as shown in Fig. 3, describes the wettability, i.e. (a) $\theta = 0^\circ$, perfect wettability, (b) $\theta = 180^\circ$, no wetting, and (c) $0^\circ < \theta < 180^\circ$, partial wetting.

Several approaches have been taken to promote the wetting of the reinforcement particles with a molten matrix alloy [16], including the coating of the particles, the addition of alloying elements to the molten matrix alloy, the treatment of the particles, and ultrasonic irradiation of the melt. In general, the surface of non-metallic particles is not wetted by the metallic metal, regardless of the cleaning techniques carried out. Wetting has been achieved by coating with a wettable metal. Metal coating on ceramic particles increases the overall surface energy of the solid, and improves wetting by enhancing the contacting interface to metal–metal instead of metal–ceramic. Nickel and copper are well wetted by many alloys, and have been used for a number of low melting alloys. In general, these coatings are applied for three purposes viz. to protect the reinforcement from damage in handling, to improve wetting, and to improve dispensability before addition to the matrix. The type of coating, in terms of wettability, can be divided into coating which reacts with the matrix, and coating which reacts with the oxide layer of the metal.

The addition of certain alloying elements can modify the matrix metal alloy by producing a transient layer between the particles and the liquid matrix. This transient layer has a low wetting angle, decreases the surface tension of the liquid, and surrounds the particles with a structure that is similar to both the particle and the matrix alloy. The composites produced by liquid metallurgy techniques show

excellent bonding between the ceramic and the metal when reactive elements, such as Mg, Ca, Ti, or Zr are added to induce wettability [17]. The addition of Mg to molten aluminium to promote the wetting of alumina is particularly successful [18], and it has also been used widely as an addition agent to promote the wetting of different ceramic particles, such as silicon carbide and mica.

Heat treatment of the particles before dispersion into the melt aids their transfer by causing desorption of adsorbed gases from the particle surface. Heating silicon carbide particles to 900°C , for example, assists in removing surface impurities and in the desorption of gases, and alters the surface composition by forming an oxide layer on the surface [19]. The addition of pre-heated alumina particles in Al–Mg melt has been found to improve the wetting of alumina [20]. A clean surface provides a better opportunity for melt–particles interaction, and thus, enhances wetting. Ultrasonic techniques, various etching techniques, and heating in a suitable atmosphere can be used to clean the particle surface. Ultrasonic vibration has been applied to molten aluminium in order to improve the wettability of alumina particles [21].

There are many other methods which have been developed to improve wettability. These include (a) various processes of fibre treatment by molten sodium for the infiltration of carbon or alumina fibre by aluminium [22], (b) the TiB process involving the deposition of Ti–B on carbon fibre before infiltration in an oxygen free atmosphere by aluminium or magnesium [23], (c) pre-treatment of silicon carbide by dehydrated sodium tetraborate for infiltration by molten aluminium [24], (d) pre-treatment of carbon by tetraisopropyltitanate for infiltration by molten aluminium or magnesium [25], (e) pre-treatment of B_4C by one of the various alcohol or other organic solvents for infiltration by molten aluminium at elevated temperature [26], (f) dispersion of solid magnesium nitride between carbon fibres for infiltration by magnesium [27], (g) the LanxideTM process using magnesium alloy additive, nitrogen containing oxygen free atmosphere and non-disclosed temperature for infiltration by aluminium with several types of reinforcements [28,29], (h) the use of nitrogen in aiding the wettability of silicon carbide and alumina particles by magnesium [30], and (i) the addition of sodium tetraborate to aid the wetting of alumina particles by aluminium alloys [31].

A mechanical force can usually be used to overcome surface tension to improve wettability. However, in the experimental work of Zhou et al. [32], poor wettability could not be solved by mechanical stirring. They also proposed that it is necessary to break the gas layer surrounding the particles in order to achieve good wettability. When gas layers are broken and the particles are wetted, the particles will tend to sink to the bottom rather than float on the surface. Improvements in the wettability between the reinforcement and the molten alloy have been achieved using various metallic coatings on the reinforcement,

however, the interaction of a coating with the liquid metal during infiltration or stirring, and the influence of this interaction on the solidification microstructure and the mechanical properties are not well understood. Some of the coating techniques are also complicated and expensive. In the usual compocasting technique, long contact time is also necessary for the promotion of bonding between the ceramic phase and the matrix [33].

The attainment of complete wetting becomes more difficult to achieve as the particle size decreases. This is due to the increase in the surface energy required for the metal surface to deform to a small radius as the particles begin to penetrate through it. The smaller particles are also more difficult to disperse because of their inherently greater surface area. The second aspect of the problem is that finely divided powder shows an increasing tendency to agglomerate or clump together as the particle size decreases.

4. Porosity in cast metal matrix composites

The volume fraction of porosity, and its size and distribution in a cast metal matrix composite play an important role in controlling the material's mechanical properties. This kind of a composite defect can be detrimental also to the corrosion resistance of the casting. Porosity levels must, therefore, be kept to a minimum. Porosity cannot be fully avoided during the casting process, but it can, however, be controlled. In general, porosity arises from three causes: (a) gas entrapment during mixing, (b) hydrogen evolution, and (c) shrinkage during solidification. According to Ghosh and Ray [34], the process parameters of holding times, stirring speed, and the size and position of the impeller will influence the development of porosity. Their experimental work showed that there is a decrease in the porosity level with an increase in the holding temperature. It has been recommended that a turbine stirrer should be placed so as to have 35% liquid below and 65% liquid above [35]. According to Lloyd [36] and Samuel [37], structural defects such as porosity, particle cluster, oxide inclusions, and interfacial reaction are found to arise from unsatisfactory casting technology. It was observed that the amount of gas porosity in casting depends more on the volume fraction of inclusions than on the amount of dissolved hydrogen [38]. Composite casting will have a higher volume fraction of suspended non-metal solid than even the dirtiest conventional aluminium casting, so the potential for the nucleation of gas bubbles is enormous. It has been observed that the porosity in cast composites increases almost linearly with particle content.

The porosity of a composite results primarily from air bubbles entering the slurry either independently or as an air envelope to the reinforcement particles [39]. Most of the gas absorbed on the surface of the particles is H_2O , and it increases with decrease in the size of the particle. In the experimental work of Miwa et al. [40], it was found that the evolution process of H_2O gas relying on temperature is

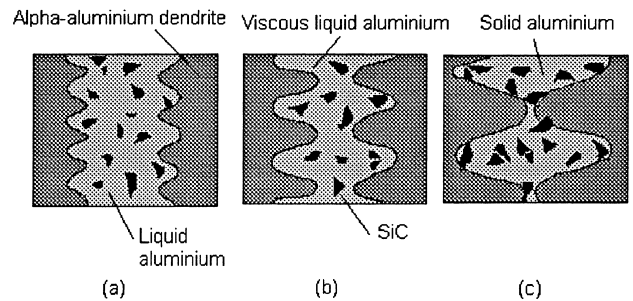


Fig. 4. The different stages of particle pushing and pore formation in silicon carbide reinforced composites during solidification [9].

mostly finished at temperatures between 200 and 600°C. Therefore, it is suggested that most of the H_2O gas absorbed on the surface of the particles can be liberated by treatment with isothermal heating at 600°C. The air trapped in the cluster of particles also contributes to the porosity.

Oxygen and hydrogen are both sources of difficulty in light alloy foundry. The affinity of aluminium for oxygen leads to a reduction of the surrounding water vapor and the formation of hydrogen, which is readily dissolved in liquid aluminium. There is a substantial drop in solubility as the metal solidifies, but because of a large energy barrier involved in the nucleation of bubbles, hydrogen usually stays in supersaturated solid solution after solidification. The nucleation and growth of pores during the solidification of A356/SiC particle reinforced composites, as shown schematically in Fig. 4 [9]. When solidification starts, a network of alpha-aluminium dendrites is developed (Fig. 4(a)). As solidification progresses, the silicon carbides that already exist in the melt are rejected in front of the advancing alpha-aluminium dendrite network (Fig. 4(b)). At this stage, there is an accumulation of hydrogen gas in a pocket of interdendritic liquid due to the decrease in the solubility accompanying solidification. When the temperature reaches the eutectic temperature, the growth of pores is limited by their abilities to expand in the remaining melt (Fig. 4(c)).

In casting metal matrix composites, there are several sources of gas. The occurrence of porosity can be attributed variously to the amount of hydrogen gas present in the melt, the oxide film on the surface of the melt, that can be drawn into it at any stage of stirring, and the gas being drawn into the melt by certain stirring methods. Vigorously stirred melt or vortex tends to entrap gas and draw it into the melt. It has been found that the presence of a vortex inhibits wetting. An experiment performed to determine the extent of the incorporation of gas in the molten mixture [2] is shown schematically in Fig. 5. Line A represents the surface of the melt before stirring, line B is the shape of the surface of the melt when rapid stirring forms a vortex, and line C is the surface level when stirring is stopped. Line C is significantly higher than line A, the difference being due to gas that was drawn into the melt by the vortex, and entrapped during the mixing process. Introducing the reinforcement particles by injection through an inert carrier gas, and several degassing techni-

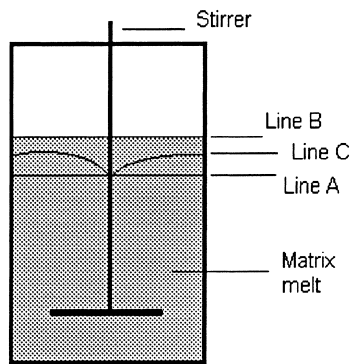


Fig. 5. The effect of vortex formation on the surface of a matrix melt [2].

ques will also increase the gas level in the melt. The pouring distance from the crucible and the mould should be as short as possible.

There are several strategies that have been used to minimise porosity, such as (a) compocasting in vacuum, (b) extensive inert gas bubbling through the melt, (c) casting under pressure, or (d) compressing, and extruding, or (e) rolling the materials after casting to close the pores. Degassing liquid aluminium alloy is a usual step in the casting procedure. When reinforcement materials are incorporated into a melt in air, the molten compound must be treated to remove the dissolved gas. Although various outgassing treatments are available (based on nitrogen gas, chlorine and/or vacuum treatment), it is difficult to reach a very low hydrogen content corresponding to the saturation of solid aluminium alloys. In this context, Girot et al. [8] have developed a procedure for gas removal. In this process, the usual cleaning, deoxidising and refining treatments are applied before degassing. Degassing is carried out in a vacuum chamber. At the end of the degassing step, the formation of bubbles is enhanced by an injection of nitrogen gas. The application of vacuum to the molten mixture of metal and particles during the mixing step can reduce the atmospheric gases available for introduction into the melt, and also tends to draw dissolved, entrapped and adsorbed gases out of the melt during mixing. Solidification shrinkage arises as a result of incorrect mould temperature and incorrect gating systems. It has been observed that increasing the mould temperature will improve the soundness of the casting, as shown by a decrease in the porosity level [41].

5. Conclusions

Processing variables such as holding temperature, stirring speed, size of the impeller, and the position of the impeller in the melt are among the important factors to be considered in the production of cast metal matrix composites as these have an impact on mechanical properties. These are determined by the reinforcement content, its distribution, the level of the intimate contact of the wetting with the matrix materials, and also the porosity content. Therefore, by controlling the

processing conditions as well as the relative amount of the reinforcement material, it is possible to obtain a composite with a broad range of mechanical properties. The method is potentially very cost effective, but widespread adoption is dependent on a satisfactory resolution of the technical difficulties presented.

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