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Dispersion of ceramic powders into particulate superplastic Al–Li alloy

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A new method for dispersing homogeneously fine ceramic powders into a particulate alloy matrix has been developed. The Al–Li matrix alloy, with controlled crystal grain size and shape, shows superplastic behaviour under certain conditions. The homogeneous dispersion of SiC in the matrix alloy was achieved by introducing the fine particles of filler along the grain boundaries within the alloy particles. This new compounding method yields a composite powder (i.e. each particle itself is a composite) rather than a mixture of different components.

1. Introduction

Aluminium has been widely used as a means of decreasing the density of structural materials. However, it is still necessary to improve the heat resistance, wear resistance and mechanical strength of the composites. For this purpose, in recent years a great effort has been directed to the production of ceramic–metal compounds [1–3] using three general compounding methods: (i) dry mixing of the raw powdered materials, (ii) incorporation of ceramic whiskers into the metal in the melt state (casting), and (iii) the mechanical alloying (MA) method. With the first two methods it is difficult to avoid component segregation arising from differences in the physical properties of the materials. For this reason, the MA technique is attracting increasing interest [4, 5]. There is a tendency towards using finer powders to obtain more homogeneous composites and to augment the surface of contact between the particles of the different components. However, fine powders possess low flowability and tend to form agglomerates; consequently, the homogeneous dispersion of the particles is difficult to achieve.

In this research, particulate Al–Li alloy (the matrix) has been compounded with SiC fine powder (filler). The alloy particles are formed of crystals whose size and shape can be accurately controlled. The alloy particles show superplastic behaviour under specific conditions. Using a high-speed, high-shear mill, the filler particles were progressively introduced into the alloy structure through the grain boundaries, resulting in a rather homogeneous composite. It is believed that this new compounding method, involving superplasticity phenomena, will become an useful technique for the design of new composites.

2. Experimental procedure

2.1. Materials

In these experiments, aluminium alloy powder was used as the matrix material and silicon carbide powder as the filler. Their physical properties are shown in Table I. The superplastic aluminium alloy (2090) atomized powder consists of spheres formed of spherical crystals of about 10 μm diameter. By superplasticity is meant the ability of the material to undergo a very large elongation without fracture under certain conditions (high temperature, low strain rate and low deformation stress). Fig. 1a shows the external aspect of the spherical alloy particles. Their internal structure can be seen in Fig. 1b, which shows the more or less constant crystal size. Fig. 1c shows the irregularly shaped SiC particles used as the filler.

2.2. Equipment and method

A high-speed, high-shear type mill [6] (Angmill, Hosokawa Micron) was used to prepare the composite powder. Measured amounts of aluminium alloy powder and SiC powder were introduced into the vessel after a preliminary mixing. The volume of mixture used in each experiment was about 10% of the volume of the mixer chamber. The compounding process was carried out in an inert atmosphere and under different operating conditions. The transformation of the particles throughout the process was assessed by SEM.

3. Results and discussion

3.1. Change in particle size of the aluminium alloy

Prior to the compounding experiments, a preliminary

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TABLE 1 Physical properties of the materials

Material	Diameter (μm)	Density ($\mathrm{kg\,m^{-3}}$)	Angle of repose (deg)
Al–Li alloy 2090[a]	100–210	2565	46
Silicon carbide	0.5–8.0	3234	53

[a] Components of the alloy (wt%): Al 94.98, Li 2.2, Cu 2.7, Zr 0.12.

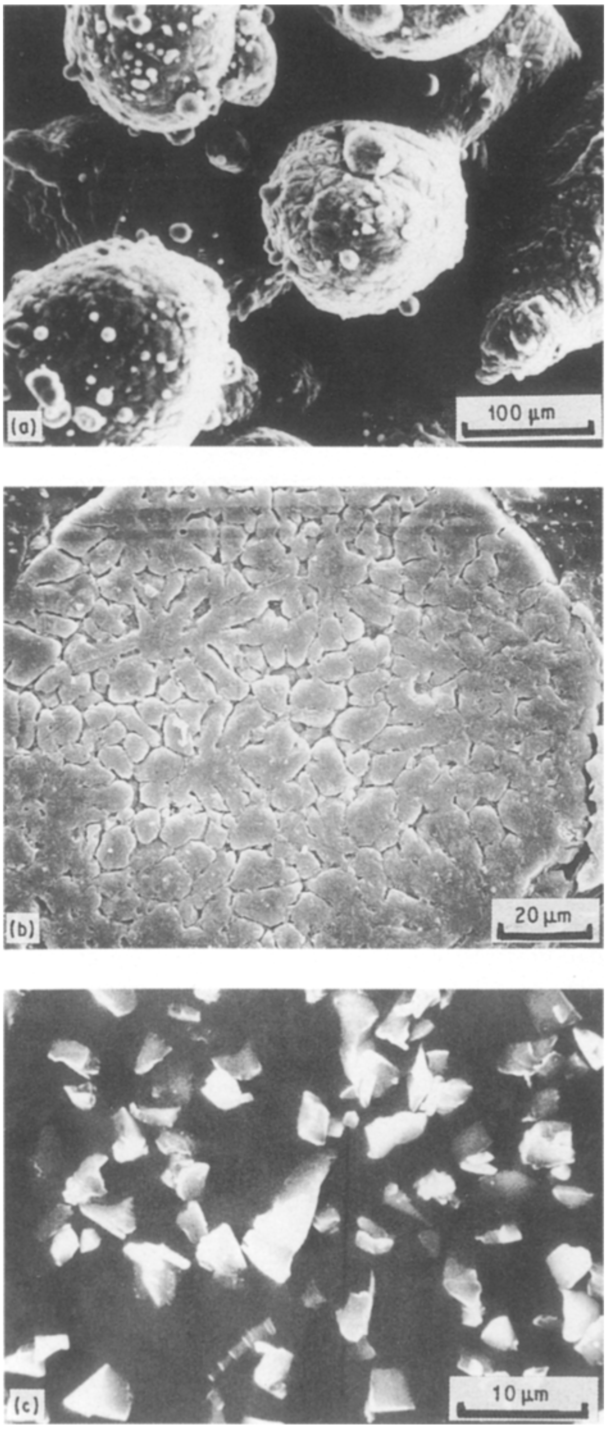

Figure 1 SEM photographs of the raw materials: (a) alloy particles, (b) cross-sectional view of the alloy particles, (c) SiC particles.

study on the effect of a high-speed, high-shear treatment of the aluminium alloy particles was carried out. Fig. 2 shows the temporal variation of the particle size distribution of a sample of aluminium alloy powder (average size 150 μm) treated in the Angmill at a rotation speed of 25 r.p.s. The shape of the original distribution does not change appreciably, although the appearance of a fraction of fines can be noticed. It can also be noticed that there is a very slight displacement in the mean size of the coarse fraction, which suggests that this variation in the particle size distribution is not caused by grinding or wear of the original particles. The raw material consists of large particles with small ones adhering to their surface (see Fig. 1a), and these small particles are progressively detached from the former as the mixing chamber rotates. Later on, some of the detached small particles adhere again to the large ones and the fraction of fines decreases.

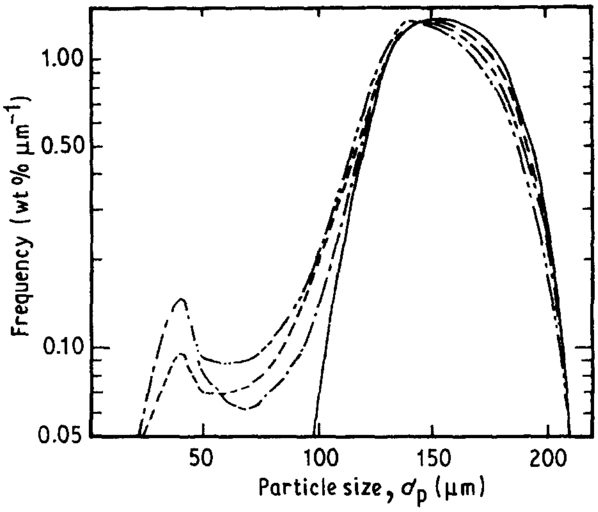

Figure 2 Changes in the alloy particle size distribution with time during treatment at 25 r.p.s. in the high-speed/high-shear mill: (——) 0s, (—·—) 600s, (–··–) 1800s, (– – –) 3600s.

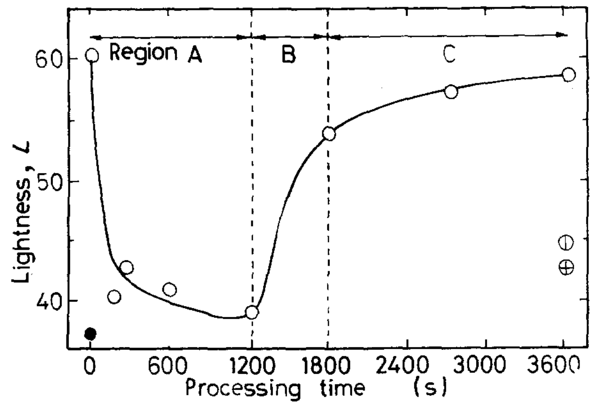

Figure 3 Temporal variation of the colour of a 94:6 alloy:SiC mixture: (●) SiC powder, (○) mixture; (⦶) ball mill 2 weeks, (⊕) ball mill 4 weeks.

3.2. Temporal variation of colour and temperature during mixing

Fig. 3 shows the temporal variation of the colour of a 94:6 aluminium alloy:SiC mixture. The mixture at time $t = 0$ is that obtained through “soft” mixing in a rotary-type mixer, and this is the mixture fed to the high-speed, high-shear mill. As seen in the figure, the mixture is progressively becoming black until a minimum value of lightness (maximum blackness) is reached. After passing through the minimum, the mixture slowly turns into its original grey colour. This figure is very useful for an analysis of the change in the surface condition of the powder throughout the compounding process. The initially existing agglomerates of SiC are gradually being broken up and adhere to the surface of the alloy particles. As a result, the surface area of black SiC particles exposed to light increases and the lightness of the mixture decreases. This first stage corresponds to the first portion (region A) of the lightness–time curve shown in the figure. The shearing and compression forces acting upon the powder cause fixation of the adhered fine particles of SiC on the alloy particles. Eventually, these fines penetrates into the alloy particles through the grain boundaries, thus decreasing the number of black fines exposed to light (region B). Friction between alloy particles provokes a further smoothing of their surface, and the penetration of fines into the body of the larger particles continues until the disappearance of SiC from the external surface of the alloy particles is almost completed (region C).

In the same figure, the values of lightness of the 94:6 mixtures obtained after 2 and 4 weeks of mixing in a ball-mill have also been represented. In this case, the lightness of the mixture decreased monotonically and failed to reach a minimum value even after a very long mixing run. It can thus be concluded that the compounding process being described is strongly influenced by the type of equipment used and that the complex set of mechanical forces acting upon the powder in the high-speed, high-shear mill used in this work permits the compounding of materials in a way not realizable with conventional equipment.

The temporal variation of the temperature of the mixture at a rotation speed of 25 r.p.s. is shown by the solid line in Fig. 4. The temperature rise takes place immediately after the mixer is set in motion and attains a steady value at about 600 s. Later, at about 1300 s, there is a sudden increase in the temperature of the mixture, which passes through a maximum, then decreasing gradually to attain a steady temperature. As the degree of coverage of the alloy particles by SiC increases (region A in the lightness–time curve), so does the friction coefficient and, accordingly, the temperature rises. The temperature peak in Fig. 4 corresponds to region B of Fig. 3, that is, that at which the fine particles of SiC begin to enter into the body of the aluminium alloy particles. Later, the gradual disappearance of fines from the surface of the alloy particles results in the smoothing of the latter and the temperature decreases (this stage corresponds to region C in Fig. 3). Three samples of powder, one from each one of the three regions, were taken and the friction coefficient measured (tilted plate method). In region B the friction coefficient μ_0 showed a maximum value, in accordance with the maximum observed in the temperature–time curve. Next, the composite powder obtained (i.e. that corresponding to the end of region C) was cooled down to room temperature and treated again at 25 r.p.s.; its temperature–time curve has also been represented in Fig. 4. In this case, the temperature increased monotonically and no maximum point was observed. Furthermore, the curve obtained was almost coincident with that of aluminium alloy alone, also plotted in the figure. This results from the fact that, once the SiC particles have penetrated into the alloy particles, these behave (as far as surface frictional phenomena is concerned) as if they were particles of the pure alloy.

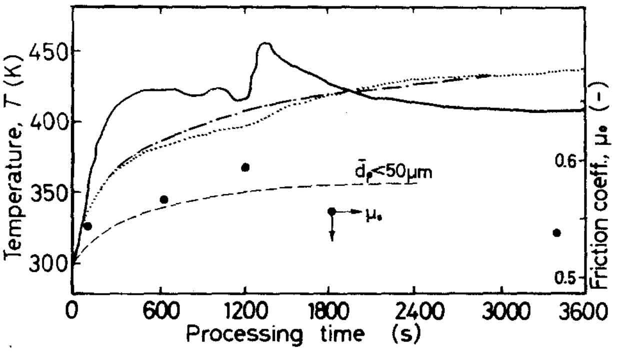

Figure 4 Temporal variation of the temperature of the mixture: (——) Al–Li/SiC powder mixture, (–·–) composite powder after treatment, (···) original alloy powder. Rotary speed 25 r.p.s.

When smaller alloy particles (< 50 μm) were used, the temperature–time curve (dashed line in Fig. 4) increased monotonically failing to show any peak. Similarly, its lightness (not plotted) decreased also monotonically and the mixture did not turn into the original metallic colour even after very long mixing times.

3.3. Composite particle surface and internal structure

Fig. 5 shows SEM pictures of the surface and the internal structure of the composite particles. In order to observe the internal structure, Keller liquid was used to etch the grain boundaries. The samples of these photographs are those corresponding to region A, very close to the minimum of the lightness–time curve. In Fig. 5a it can be noticed how the fine particles of SiC are homogeneously attached to the surface of the alloy particles. We could not observe the presence of either free fines or agglomerates. In Fig. 5b it can be seen that the compounding process in the high-speed, high-shear mill did not noticeably change the size or shape of the constituent crystals of the alloy particles. In the same figure the layer of SiC covering the alloy particles can also be seen, and the presence of SiC within the internal structure of the alloy is not observed.

Fig. 6a and b are SEM photographs of the surface and the internal structure of the composite corresponding to region C, that is, after the peak of the temperature–time curve has passed and the temperature has attained a steady value. From Fig. 6a it is concluded that the compounding treatment did not alter the size of the alloy particles but did slightly modify their shape, this becoming similar to an ellipsoid of revolution. The surface of the alloy particles appears very smooth and no particles of SiC can be observed on it. In Fig. 6b it is seen that the shape and

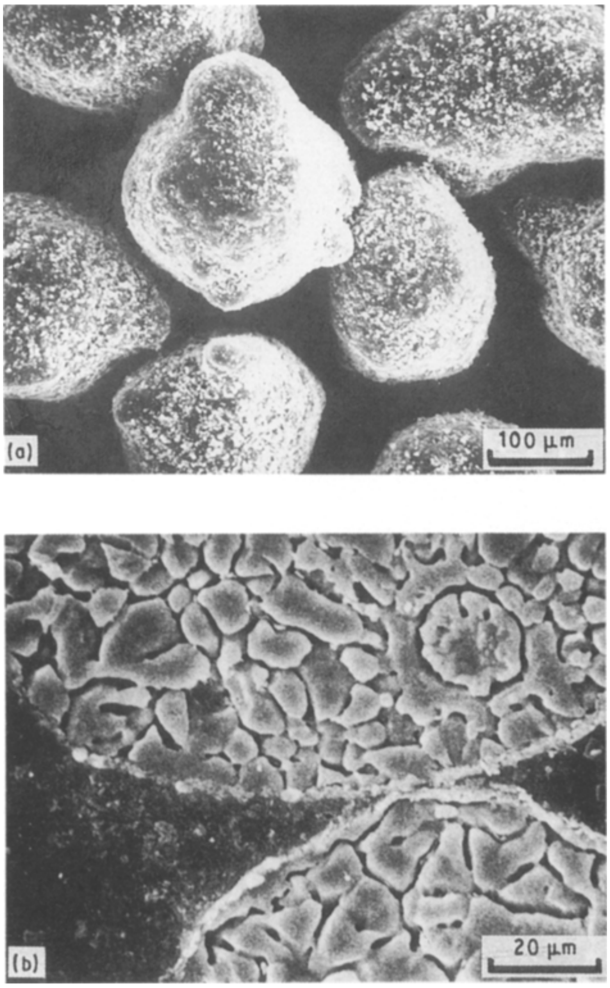

Figure 5 SEM photographs of composite particles corresponding to region A in Fig. 3: (a) external aspect, (b) cross-section.

size of the alloy crystals are the same as before and that the SiC particles are placed along the grain boundaries. Fig. 6c is the image obtained by X-ray microanalysis, showing the distribution of Si within the alloy particle. Fig. 6c is confirmation of the fact that the material disposed along the grain boundaries (Fig. 6b) is indeed SiC. Furthermore, it is noticed that the distribution of SiC inside the alloy particles is almost homogeneous, that is, there are SiC particles in contact with practically all of the crystals present in the alloy particle. The alloy particles show no cracks or voids in their internal structure.

3.4. Compounding process model

The material used as the matrix in the compounding process described in this work shows superplastic behaviour under certain conditions. It is necessary to create these conditions within the mill in order to have the material behaving as superplastic. Under the external forces applied, the alloy crystals can be elongated without fracture (returning later to the original state) and their relative positions change. In this manner, the fine particles of SiC adhering originally to the surface of the "external" crystals can occupy successively more "internal" positions within the alloy particles. The whole process has been sketched in Fig. 7 and has been subdivided into several stages as follows.

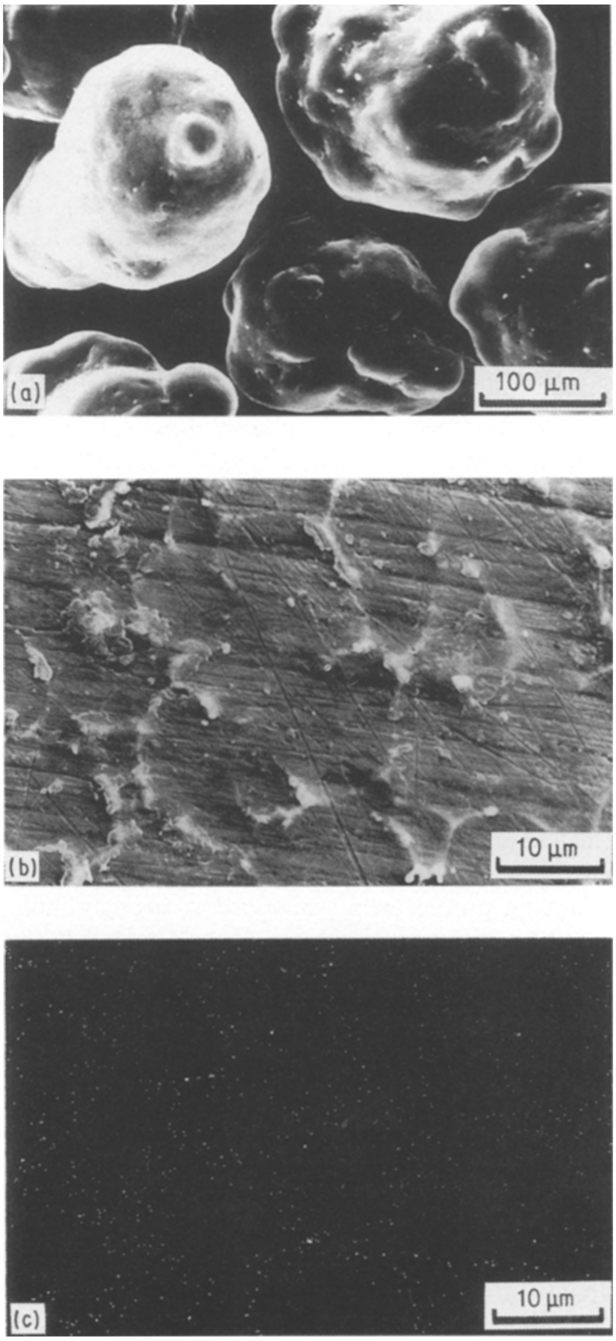

Figure 6 Composite particles corresponding to region C in Fig. 3: (a) SEM photograph of the external aspect, (b) SEM photograph of the cross-section, (c) X-ray image of the cross-section for SiC in the matrix alloy (white spots).

1. The fines attach to the surface of the alloy particles forming an ordered mixture [7, 8].
2. As the temperature increases due to interparticle friction, at some point the alloy particles show superplastic behaviour for very short periods of time.
3. When the superplastic behaviour becomes more pronounced, the crystal grain boundaries loosen and permit the partial introduction of fines.
4. For very short periods of time, the whole alloy particle becomes superplastic and the crystal surface slippery; the slide of crystals over each other and their relocation in new positions permit the gradual passage of SiC towards the centre of the alloy particle.

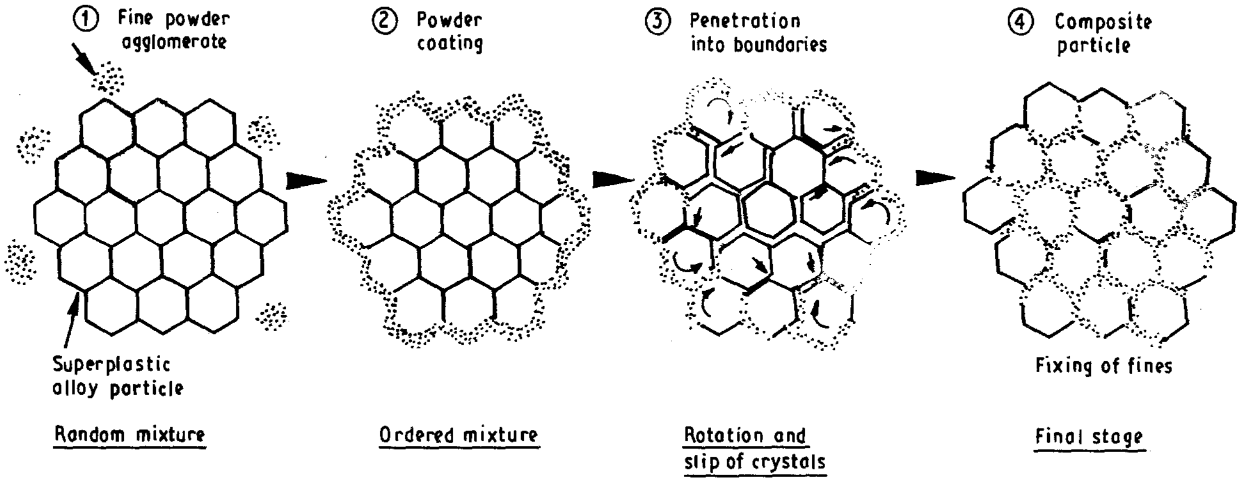

Figure 7 Compounding process model.

5. The mechanism commented on in stage 4, along with the mechanical external force communicated to the powder, results in the complete dispersion of the ceramic material within the matrix particle.

As mentioned before, when the alloy particles are small (even though the size of the constituent crystals is the same as for larger particles) no special peak appeared in the temperature–time curve and the fine SiC particles could not enter into the larger ones. One reason for this effect is that the external force is poorly transmitted through a bed of fine particles and, therefore, the frictional heating is not enough in this case to provoke the superplastic behaviour of the alloy. Besides this frictional effect, the existence of superplastic behaviour also depends on the number of crystals forming the particle.

4. Conclusions

A new compounding method, using superplasticity phenomena, has been developed to achieve the homogeneous dispersion of the filler within the matrix component. The main advantages of this technique may be summarized in two points: (i) there is no need to use a very fine matrix powder in order to obtain a composite of high degree of homogeneity and, therefore, a subsequent granulation of the product is not required; and (ii) composite powders (as opposed to composite mixtures) are obtained. This second point is of fundamental importance in the preparation of particles of special properties to be used in the formulation of new alloys and materials.

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