JOURNAL OF MATERIALS SCIENCE **38** (2003) 2761 – 2765

On the stability with temperature of the dislocation structure in 6061Al-15vol%SiC composites as studied by hardness and differential scanning calorimetry

A. BORREGO, G. GONZÁLEZ-DONCEL
Department of Physical Metallurgy, CENIM, C.S.I.C., Av. de Gregorio del Amo, 8, 28040, Madrid, Spain
E-mail: ggd@cenim.csic.es

The relative difference between the average dislocation density of 6061Al-15vol%SiC composites investigated by two independent methods (namely hardness, H, and differential scanning calorimetry, DSC), is discussed. They are interpreted in terms of the "actual" temperature at which the measurements are made with each technique. The present analysis accounts for the high microstructural stability of these composites at elevated temperatures. © *2003 Kluwer Academic Publishers*

Introduction

There is general agreement in that the high density of dislocations, ρ, in metal matrix composites, MMCs, e.g. Al with SiC whiskers, platelets or quasi-equiaxed particles, is one of the most important microstructural features in explaining their improved yield and tensile strength with respect the unreinforced alloys [1]. The increase in the dislocation density arises during the cooling stages in the processing of the materials as a consequence of the large difference between the thermal expansion coefficient of the metallic matrix and the ceramic reinforcement (≈6:1 in the Al/SiC system). Most of the work aimed at rigorous determination of ρ in MMCs has been conducted by transmission electron microscopy, TEM [1–6]. The determination of the dislocation density by TEM, however, involves a number of difficulties mainly due to the large number of micrographs required for reliable average values of ρ since the dislocations are not uniformly distributed throughout the matrix but mostly concentrated at metal-matrix interfaces. Accordingly, values ranging from 10^{12} to 10^{14} m^{-2} are commonly reported [1–6]. It is, therefore, worth further investigating other techniques which, although using indirect procedures, also provide useful information that can be correlated with the dislocation structure and stability as well as with ρ values. The measurements derived from such analysis could be, then, correlated with TEM data.

The effect of dislocations on several physical properties such as mechanical, electrical and thermal, has been used to address, qualitatively, the increase of dislocation density in MMCs, particularly when precipitation hardenable aluminium alloys are used as matrices [2, 7, 8]. This is because of the importance of diffusional processes, in particular dislocation pipe diffusion, on the evolution of macroscopic materials properties. On this basis, two independent methods to estimate average values of ρ in MMCs have been developed recently [9–12]. Specifically, the dislocation density in three 6061Al-15vol%SiC$_w$ has been estimated by the present authors taking into account the effect of dislocation pipe diffusion on the accelerated ageing process (or the reduction in τ_p, the time required to reach the peak hardness at a given ageing temperature) of hardenable aluminium alloys [9, 10] and on the formation of the metastable β'' phase peak of the matrix alloy [11, 12]. The accelerated ageing process has been followed by Vickers hardness measurements and the formation of the β'' phase peak has been studied by differential scanning calorimetry, DSC, experiments. The precipitation sequence (GP zones, β'', β', and stable β (Mg_2Si) phase) of 6061Al is well documented elsewhere [2, 13, 14]. It is the purpose of the present work to throw further ideas on the dislocation structure and materials microstructure stability by comparing the normalised dislocation density of these 6061Al matrix composites determined by these two methods to that of the base alloy. The present analysis reinforces the idea of that these composites possesses a high microstructural stability at elevated temperature.

Materials and experimental procedure

The materials used in this investigation were three 6061Al-15vol%SiC$_w$ composites consolidated by extrusion of un-canned powder compacts at three temperatures ranging between 300° and 500°C. The increase in extrusion temperature resulted in a higher directionality and less break-up of the SiC whiskers during the consolidation process. The composites show

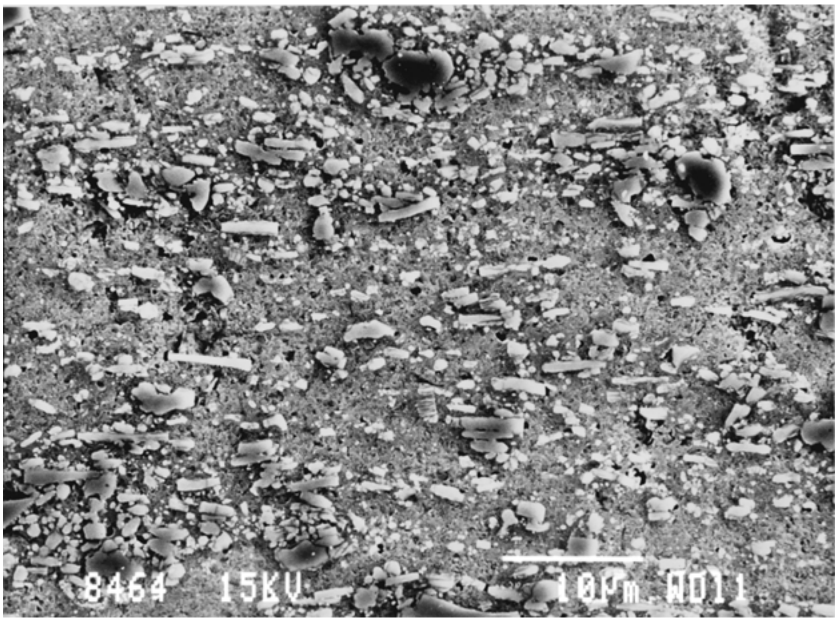

Figure 1 SEM micrograph of the MMC extruded at 498°C. A homogeneous distribution of whiskers can be observed. The extrusion direction is the horizontal.

a homogeneous distribution of whiskers in the Al matrix, Fig. 1. For comparison, unreinforced 6061Al powder was also consolidated by the same procedure. Table I presents the extrusion temperature of the different materials. A detailed description of the powder metallurgy, PM, route employed to prepare the materials is given elsewhere [9]. The precipitation of the composite matrices was followed, from the supersaturated solid solution, by Vickers hardness after annealing at 419 K (isothermal method) and DSC at four heating rates (non isothermal method). Further details about these two methods and the analytical procedure to determine dislocation densities are given in [9–12]. The microstructure of all materials was also studied by scanning electron microscopy, SEM.

Results and discussion

The two independent methods, hardness evolution and DSC, to determine dislocation density in 6061Al matrix composites are described in detail in references [9–12]. As follows is a summary of the procedure employed in each case.

Hardness measurements, H

This method (isothermal) is based on the fact that the increase in dislocation density of the MMCs with respect the unreinforced alloy is manifested on the accelerated ageing behaviour rather than on the increase in peak hardness upon heat treatment at a given temperature. On one hand, the increase in hardness is usually affected by several strengthening mechanisms; these include: the increase in the initial ρ, internal stresses, different grain/subgrain structure (Petch-Hall), particle strengthening (Orowan), precipitation/solid solution strengthening, and/or initial work hardening (dislocation multiplication) [15]. On the other hand, however, the accelerated ageing phenomenon can be, in some cases, attributed only to the increase in ρ [10]. In such a case, a direct correlation between the measurable decrease in τ_p and ρ is rigorous. This correlation is possible if instantaneous nucleation takes place at dislocations and if the growth of precipitates is driven by dislocation pipe diffusion. As will be shown, both two circumstances are satisfied in the present case.

TABLE I Extrusion temperature, time to reach peak hardness, τ_p, (in hours) and effective activation energy, $Q_{\text{eff}}^{\text{Si,Al}}$, for Si diffusion in Al of all materials

Material	Code	Extr. Temp. (°C)	τ_p (h)	$Q_{\text{eff}}^{\text{Si,Al}}$(kJ/mol)
6061Al	C8	525	56.2	120
	C32	300	26.1	94
6061Al/SiC	C34	359	24.5	95
	C38	498	14.5	88

At the ageing temperature of 419 K, which is above that of silicon cluster nucleation (≈350 K) [16], the high density of dislocations would provide short-circuit paths for heterogeneous nucleation and fast growth of metastable precipitates resulting in accelerated ageing of the matrix [17]. Therefore, the metastable precipitates are mainly nucleated at dislocation cores. Instantaneous nucleation is, then, assumed since heterogeneous nucleation at dislocations is virtually instantaneous [18]. For the above reason the precipitation rate, usually defined as the inverse of the time to reach the peak hardness, τ_p^{-1}, can be assumed to be proportional to the effective diffusion coefficient, D_{eff}, [19],

$$\frac{1}{\tau_p} = K \cdot D_{\text{eff}} \approx K \cdot (D_l + 10^{-18}\rho \cdot D_p)$$

with K a microstructure related constant and D_l and D_p the lattice and pipe diffusivity, respectively. At the ageing temperature of 419 K $D_l \ll 10^{-18}\rho D_p$; i.e., lattice diffusivity is negligible compared to pipe diffusivity.

TABLE II Normalised dislocation density calculated from hardness and DSC experiments for the different materials. Subindex denotes extrusion temperature

Material	$\rho/\rho_{6061\mathrm{Al}}$ (H)	$\rho/\rho_{6061\mathrm{Al}}$ (DSC)
6061Al	1.0	1.0
6061Al/SiC$_{300}$	2.1	11.5
6061Al/SiC$_{359}$	2.3	10.0
6061Al/SiC$_{498}$	3.9	25.5

This, allows one to assume that the ratio of dislocation density is the same as the ratio of precipitation rate, τ_p^{-1}. Hence, the τ_p ratio of the MMCs to that of the monolithic alloy yields the ratio of the dislocation densities of these materials [10].

Table I summarises τ_p for the composites and for the base alloy. The normalised dislocation densities of the materials, ρ_H, are presented in Table II. It should be noted that these values correspond to the dislocation densities at the ageing temperature (419 K). It is assumed that at this temperature no rearrangement of dislocations occurs [20]. Therefore, they can be taken as "low temperature" values of the dislocation density. It is worth noting that these normalised ρ values are close to those reported in the literature for 6061Al matrix composites measured by TEM. For example, Vogelsang *et al.* [4] have reported values of 5.0 and 2.5 for PM 6061Al-SiC$_w$ composites with 5 and 20 vol% reinforcement respectively. Arsenault *et al.* [1] found values of 2.8, 5.5, and 6, 8 in PM 6061Al-SiC$_w$ with 1, 5 and 20 vol% reinforcement, respectively. Finally, Bartels *et al.* [6] accounted for normalised ρ values for *in situ* 6061Al-TiB$_2$ composites of 2.3 and 8.0 for 3.4 and 6.8 vol% reinforcement, respectively.

DSC measurements

This method (non isothermal) takes advantage of the fact that the precipitation reaction of the metastable β'' phase in the 6061Al matrix is, basically, only influenced by the dislocations. This is because the growth of the axially symmetrical β'' precipitates is a diffusion controlled process of the Si atoms (Si is the slowest specie diffusing in aluminium) where dislocation pipe diffusion is found to be important. Since nucleation occurs in the early stages of the DSC experiment, i.e. at low temperature, homogeneous precipitation must occurs now in the aluminium matrix during the DSC experiments [11, 21]. Specifically, nucleation takes place mainly in vacancy clusters. A high density of vacancy clusters, formed during solution treatment, is retained after quenching prior to DSC experiments. Then, the nucleation rate, $I(t)$, decreases rapidly with time due to saturation of nucleation sites during the early stages of the transformation. The following expression is usually proposed for the nucleation rate [22]:

$$I\,(t) \approx C'' \cdot t^q$$

where $q < 0$ and C'' is a time constant with an Arrhenius type temperature dependence. Experimental evidence suggests that the time dependence of $I(t)$ is an intermediate case between instantaneous nucleation (all nuclei are present at $t = 0$; $I(t) = 0$) and continuous nucleation (nucleation rate is constant with time; $I(t) = \text{cte.}$) [8].

On the other hand, the effective activation energy for diffusion of silicon in the aluminium matrix, $Q_{\mathrm{eff}}^{\mathrm{Si,Al}}$, has been related to E, the apparent activation energy, and to the Johnson-Mehl-Avrami, JMA, exponent, n, through the relationship [11]:

$$\frac{3}{2}Q_{\mathrm{eff}}^{\mathrm{Si,Al}} + Q_n = nE$$

where Q_n is the activation energy for nucleation. Kovács *et al.* [16] have observed that the nucleation process in Al—Mg$_2$Si takes place during a short period of time, even at temperatures below 273 K. On the other hand, however, growth of the β'' precipitates does not occur in a reasonable time during isothermal annealing below 373 K [23]. All this suggests a negligible value of Q_n with respect $Q_{\mathrm{eff}}^{\mathrm{Si,Al}}$ and, hence:

$$\frac{3}{2}Q_{\mathrm{eff}}^{\mathrm{Si,Al}} \approx nE$$

Therefore, the growth of the axially symmetrical metastable precipitates is a diffusion controlled process of the Si atoms (Si is the slowest specie diffusing in Al) where dislocation pipe diffusion is found to be important. Table I lists $Q_{\mathrm{eff}}^{\mathrm{Si,Al}}$ values obtained for all materials. Then, ρ for each material can be readily calculated from the following equation [11]:

$$\rho \approx A_o \frac{D_{\mathrm{l}}^{\mathrm{Si,Al}}\left(Q_{\mathrm{l}}^{\mathrm{Si,Al}} - Q_{\mathrm{eff}}^{\mathrm{Si,Al}}\right)}{D_{\mathrm{p}}^{\mathrm{Si,Al}}\left(Q_{\mathrm{eff}}^{\mathrm{Si,Al}} - Q_{\mathrm{p}}^{\mathrm{Si,Al}}\right)} \tag{1}$$

where A_o ($\sim 10^{18}$ m^{-2}) represents the inverse of the dislocation core section in aluminium, $D_{\mathrm{l}}^{\mathrm{Si,Al}}$ and $D_{\mathrm{p}}^{\mathrm{Si,Al}}$ are the atomic lattice and pipe diffusion coefficients of Si in Al, respectively, and $Q_{\mathrm{l}}^{\mathrm{Si,Al}}$ and $Q_{\mathrm{p}}^{\mathrm{Si,Al}}$ are the associated activation energies. Since the dislocation densities obtained from Equation 1 have been calculated for the β'' peak ($\sim$530 K) they can be taken as "high temperature" values of the dislocation density. The ratios of ρ for the MMCs to that for the unreinforced alloy are listed in Table II. As can be seen, the normalised dislocation density of the MMCs obtained from DSC is one order of magnitude higher than that calculated from hardness experiments. Whereas good agreement is found at low temperature, no data of dislocation densities at high temperatures are presently available in the literature.

It should be noted that the precipitation stage of the materials at the peak ageing condition is such that the dislocation/precipitate interaction is the highest; in particular, higher than the interaction in the precipitation stage given by β'' exothermic peak of the DSC experiments. Therefore, ρ of MMCs normalised to that of the base alloy allows to eliminate the effect of the dislocation/precipitate interaction and only accounts for the effect of the reinforcing particles on the dislocation structure. It is assumed that the precipitation stage is different for each technique but, in each case, similar for all materials.

Correlation between hardness and DSC data

From the values of the normalised dislocation densities estimated by these two methods, the parameter P, defined as the ratio:

$$P = \frac{\rho_{\mathrm{DSC}}}{\rho_{\mathrm{H}}}$$

can be introduced. Since these two measurements of ρ can be correlated to those obtained at different temperatures, this parameter would represent the stability of the dislocation structure of the materials or their ability to retain the microstructure from recovery processes. Furthermore, since ρ_{DSC} and ρ_{H} are normalised values to those of the unreinforced alloy, P would account for the stability of the composites relative to that of the unreinforced alloy. Then, all data points through a straight line (without intercept), in a diagram of ρ_{DSC} vs. ρ_{H}, denote materials with the same stability of the dislocation structure stability, given by the slope P, Fig. 2. Specifically, the line defined by $P = 1$ indicates the stability line of base alloys. Points below this line ($P < 1$) would represent composite materials with lower stability than the monolithic alloy, whereas points above this line ($P > 1$) would represent materials with higher stability; i.e. the higher the slope, the higher the stability.

In this framework, the normalised dislocation density of the materials listed in Table II has been plotted in Fig. 2. As expected, the composites data fall well above the base line giving support to the idea of the higher microstructural stability of these materials. It is important to note that the data for the composites are close to a given stability line, $P \approx 6$ accounting for the similar stability of the composites. From this plot, it is also seen that the materials consolidated at low extrusion temperatures, C32 and C34 (300° and 359°C, respectively) have a slightly lower stability than the composite extruded at high temperature, C38 (498°C). This result is consistent with previous studies which show that the higher the extrusion temperature, the higher the directionality and the length, as well as a more homogeneous distribution, of the reinforcing whiskers in the aluminium matrix [10]. Therefore, it is likely that after the extrusion process at high temperature, the whiskers rearrange to form better barriers for dislocation motion/annihilation leading, hence, to a more stable microstructure of the composite than that developed at lower extrusion temperatures, in agreement with the present study.

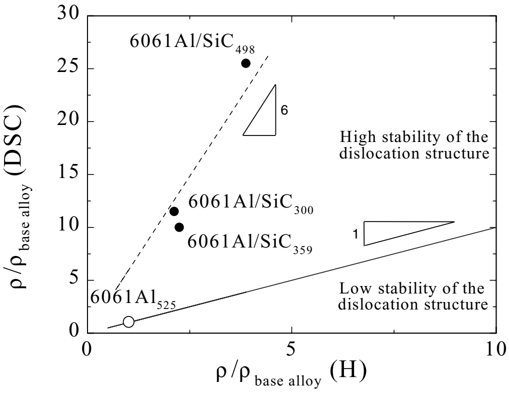

Figure 2 Dislocation density of MMCs normalised to that of 6061Al base alloy calculated from DSC experiments versus dislocation density calculated from hardness measurements.

Concluding remarks

As it has been shown, the dislocation density in 6061Al-15vol%SiC$_{\mathrm{w}}$ composites obtained by two methods, hardness and DSC, can be correlated to the dislocations present in the materials at different temperatures. For each technique, the dislocation structure is determined by the stage of the metastable microstructure (specifically the degree of dislocation/precipitate interaction) which is strongly dependent upon the thermal history. Comparison of these measurements, by the introduction of a microstructural stability parameter, P, throw light on the high temperature stability of these composites. The stability map proposed can be generalised to other discontinuously reinforced MMCs and can help to explain the high temperature behaviour (e.g. creep) of these structural materials, not yet well understood [24, 25]. Finally, the conclusions derived from this analysis agree with the fact that discontinuously reinforced MMCs with different aluminium matrices maintain their mechanical properties to higher temperatures than the unreinforced alloys, as it is evidenced from tensile testing at different temperatures [26, 27].

Acknowledgements

Work supported by CICYT (Spain) under projects MAT94-0887 and MAT97-1059-C02. One of the authors (A.B.) also thanks Volkswagen-Audi/CSIC financial support through a research grant.

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Received 13 December 2001
and accepted 26 March 2003