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Matrix grain refinement in Al-TiAl composites by severe plastic deformation: influence of particle size and processing route.

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Abstract

The microstructure and mechanical behaviour of Al-based composites reinforced with TiAl intermetallic particles has been examined in the as-extruded state and after processing by equal channel angular pressing (ECAP). The latter produces a grain size reduction in the aluminium matrix to values of 500 nm, using route A, and 750 nm, using route C. The ECAP produces up to a 75% increase in the yield stress of the composites, being more rapid when route A is used. The strengthening effect by ECAP is much larger than that obtained by increasing the volume fraction of reinforcement particles from 25% to 50% in these composites.

Keywords: Equal-channel angular pressing; grain refinement; metal-intermetallic composites; mechanical strength

1. Introduction

Processing by severe plastic deformation, for example by equal channel angular pressing (ECAP), is one of the areas of major research activity today, since it is possible to produce bulk materials with fine microstructures and interesting mechanical behaviour [1-2]. The materials produced typically show fine, submicron grains with a high retained dislocation density, with refinement or dissolution of precipitate and dispersoid particles present often occurring simultaneously. The materials obtained, with their fine-scale, heavily-worked microstructures, typically show high strength, in combination with good ductility, and hence are of considerable interest for many possible engineering applications. Much of the work reported has been carried out on single-phase materials, with a large amount of effort concentrated on the Al-Mg system where grain sizes of about 100-500 nm have been obtained [3-6]. Although some of the commercial alloys studied contained relatively coarse dispersoid or inclusion particles [7-8], only a few reports have specifically examined the role played by

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particles in the evolution of microstructure during severe deformation [9-10]. In the case of Al-based composites, ECAP offers the possibility of hardening the matrix as well as helping modify the distribution of reinforcement particles if these are sheared during deformation. Significant hardening of composites has indeed been observed in an Al-6061 based composite reinforced with alumina particles [11-13].

Recent studies of Al-based composites reinforced with TiAl intermetallic particles have confirmed the possibility of obtaining materials with a homogeneous distribution of reinforcement particles without any interface reactions by the direct extrusion of powders [14]. Although the strength of these composites was not very high at room temperature, it remained rather stable up to temperatures of 200ºC. A recent investigation [15] on the effect of processing such composites by ECAP to relatively low strain levels (2.8) confirmed that it was possible to improve significantly their room temperature strength by such microstructural refinement. The present study examines the effect of ECAP to larger strains on microstructural refinement of the aluminium matrix, and compares the mechanical strength achieved by the severely deformed composites to that produced in as-extruded materials with increased volume fraction or decreased size of TiAl reinforcement.

2. Experimental details

The Al-based composites were prepared from gas atomised aluminium powders of purity 99.5% and intermetallic powders of composition Ti-49at% Al. Both types of powders had similar sizes, 100±25 μm. To produce smaller sized reinforcements, the TiAl atomised powders were mechanically milled for 5 h, at 600 r.p.m., using a Pulverisette 7 planetary ball-mill, with a ball to powder weight ratio of 3. The intermetallic particle size was reduced to 20±25 μm by this procedure. Mixtures of aluminium powders were prepared with 25 and 50% by volume of TiAl particles in both the atomised and the milled state. The powder mixtures were blended to a homogeneous particle distribution using the same mill without balls, at a rotation speed of 400 r.p.m for 10 min. The powder mixtures were then encapsulated in an aluminium container and extruded at 450ºC with an extrusion ratio 10:1. Bars obtained were studied as reference materials. In addition, the mixtures containing 25% TiAl powders were extruded, also at 450ºC, with a 5:1 ratio to larger diameter bars for severe plastic deformation by equal channel angular pressing. ECAP was performed, at room temperature, in a 20 mm diameter, circular section die, with a tool angle of 120º and very small radius of curvature at the outer line of intersection of the two channels, producing a true strain per pass of 0.7 [1,2,4]. Materials were processed using both route A, without sample rotation between passes, and route C, with 180º rotation between passes [1,16].

Optical and scanning electron microscopy (SEM) was used to examine general microstructures, and specifically to study the particle size and distribution, and matrix grain size as a function of processing route and number of ECAP passes. The SEM observations were performed on electropolished samples with a JEOL 6500F microscope using back-scattered electron mode to obtain images from the matrix grains. Transmission electron microscopy (TEM) observations were carried out using a JEOL 2010FX instrument to examine dislocation densities and distributions, as well as to obtain more information on the types of boundaries present. For both SEM and TEM studies, electropolished samples were prepared using a mixture of 20% nitric acid in methanol at -30ºC and 20 V. Microstructural parameters were quantified by image analysis of SEM photographs, measuring at least five hundred grains and three hundred particles for each state examined. Particle and grain size were both determined as the equivalent diameter value, that is the diameter of the circle having the same area as the given particle/grain. This parameter was chosen since both particles and grains were close to equiaxed/circular in morphology. The size distribution of particles and grains was obtained from such analyses, as has been reported previously, see refs. [14,15]. While experimental measurements showed little error (estimated as 1%) and good reproducibility, there was a wide variation in the measured sizes, with smallest measured particles/grains for a given state being only 10-20% of the average value, and largest measured particles/grains being 2-3 times the average value. As shown elsewhere [14,15] the distribution of both grain size and TiAl particle size for any state may be described by an approximately Gaussian size distribution about the average value, with a standard deviation of size of about 20% of that average.

Mechanical properties were evaluated by testing samples in tension and compression at temperatures between room temperature and 350 ºC using a universal testing machine at a strain rate of $5x10^{-4}$ s^{-1}. A heating rate of 20 K/min was used for high temperature tests and samples were allowed to soak at temperature for 15 min before testing. The tensile samples had gauges of diameter 3 mm and length 20 mm, and the samples for compression were cylinders of diameter 3 mm and height 5mm. Three tensile or compression samples were tested for each condition, showing extremely good reproducibility of strength levels – always with data dispersion less than 5 MPa, or about ± 2%-5%, depending on the material. Yield stress was determined in each case as the stress where there was a deviation of 0.2% strain from the elastic line. This was determined from the stress-strain curve deduced from the applied load and crosshead displacement measured during testing. In general tensile testing was carried out on the as-extruded composites, both those prepared from as-atomised TiAl powders and those from milled powders, at room temperature and at high temperatures, while compression tests were conducted on material after ECAP where there was generally less material available for the preparation of many tensile samples. However, at certain conditions, specifically for samples processed by ECAP to 4-5 passes and to 8 passes, as well as a few other conditions, both tensile tests and compression tests were carried out, at room temperature and at high temperatures, to compare stresses at the onset of plasticity. Within the data scatter, i.e. dispersion of 5 MPa, or ± 2%-5%, the compression testing and tensile testing gave identical results, and hence data are presented indiscriminately in the results section. Finally, in addition to the tensile and compression tests, Vickers microhardness was measured on both the aluminium matrix and the intermetallic reinforcement using 50 g and 500 g loads respectively. Hardness values were determined as the average of 10 measurements, with again data showing scatter of a few percent about the determined average value.

3. Results

It has been shown previously [14] that the conventional high temperature extrusion of the different powder mixtures produced composite materials with full density and good cohesion between the aluminium matrix and the reinforcement particles. The amount of strain that could be imposed during subsequent ECAP processing of these composites containing 25% TiAl depended strongly on the initial reinforcement size. Thus, materials reinforced with large, atomised TiAl particles could withstand four ECAP passes (total strain 2.8) using route A and eight ECAP passes (total strain 5.6) using route C before showing cracking. Materials containing the finer, milled TiAl particles could only withstand two ECAP passes using route A and five passes using route C before similar cracking.

Microstructural observations have shown no differences in size and distribution of the reinforcement particles between the conventionally extruded and the ECAP processed materials. Thus, the severe plastic deformation did not affect the intermetallic particles, which neither cracked nor fractured. Both before and after the severe plastic deformation the average size of the atomised and milled TiAl particles was 100 and 20 μm respectively.

Figure 1 shows examples of microstructures observed in the SEM using backscattered electrons to produce atomic number contrast of the reinforcement particles and crystallographic contrast of the ultrafine grains in the aluminium matrix. Figs. 1a and 1b show such particles and matrix grains in a composite containing atomised particles after six ECAP passes. Fig. 1b shows the curvature of microstructure, in bands where grain refinement has occurred, produced by the plastic flow of the ductile matrix between the hard intermetallic particles during the severe deformation. Fig. 1c shows the distribution of the milled intermetallic particles in a composite subjected to three ECAP passes. Fig. 1d shows details, in the same sample, of the finer grain structure produced by ECAP at the interface between the aluminium matrix and the reinforcement particle (bright). Figure 2 shows typical grain distributions in the aluminium matrix, illustrating micrographs from which grain sizes and distributions were measured.

The evolution of grain size with number of ECAP passes for the two different routes are shown in Figure 3a. The initial grain size corresponds to that of the conventionally extruded materials used as a starting reference (2.5μm for the material reinforced with atomised intermetallic particles). The decrease in grain size from this large initial value depends on the ECAP route. Route A produces a rapid initial grain size reduction, to 610 nm after only two ECAP passes, with a near-saturation thereafter, to 520 nm after four passes. In contrast, route C produces a much slower grain refinement, to about 1μm after two passes, reaching only 750 nm after eight passes. An even slower reduction of matrix grain size is observed during ECAP deformation of material reinforced with milled particles, from the initial grain size of about 1.2 μm in as-extruded material to 740 nm after five passes.

Figure 3b shows the evolution of hardness of the aluminium matrix as a function of the number of ECAP passes. The hardness increase is faster for the material processed using route A, where a finer grain size is achieved after two and four passes. Materials deformed by route C reached higher hardnesses, however, even though the grain sizes achieved were somewhat larger than for route A. This indicates that grain size is not the only parameter affecting matrix strength during severe plastic deformation.

TEM studies on the different materials after ECAP have confirmed that the boundaries produced are mixtures of low, medium and high angle boundaries, and that material deformed to a given number of passes using route A exhibit a higher fraction of grain boundaries of somewhat higher misorientations than material deformed using route C. Figure 4 shows examples of typical boundary microstructures after four ECAP passes using route A (Fig. 4a) or route C (Fig. 4b). For the sample deformed using route A, the grain boundaries frequently show a fringe contrast characteristic of high angle boundaries whereas the sample produced using route C shows also some lower angle boundaries, still imaged as dislocations walls. It is not the intention of the present report to quantify further this assertion, for example presenting results of the spread of diffraction spots or Kikuchi line shifts in TEM diffraction, as has been presented elsewhere for different ECAP processed materials[4,10]. However, a quick assessment of a few tens of boundaries, examining Kikuchi lines on each side of a boundary, showed an average boundary misorientation of somewhat above 10º for the material of Fig. 4a deformed 4 passes using route A, and somewhat below 10º for material deformed instead using route C (Fig. 4b). Earlier reports comparing materials processed by route A and C [3,16] appear to come to similar conclusions, of route A leading to slightly faster increase of boundary misorientation, although the difference is not always large and may depend on alloy composition [3] and strain level considered [16].

Yield stresses of composites processed by ECAP in various ways and at various temperatures are shown in Figures 5 to 7. Fig. 5 compares the yield stress at room temperature as it varies with the number of passes for the same composite (reinforced with atomised TiAl particles) processed by the two different routes. For route A, the yield stress increases rapidly after the first ECAP passes but that it saturates and then slightly decreases after four passes. For route C, the increase in strength is less rapid and reaches a maximum after four passes, decreasing slightly between four and eight passes. The maximum yield stress achieved, independent of the route used, is about 75 % higher than that of extruded materials. This increase in strength is accompanied by a reduction in tensile ductility from 12% in the extruded material to about 2-3% for the materials processed by ECAP. This significant fall in ductility occurs after the first 2-3 passes, corresponding roughly with the increase to a maximum of the yield stress. While there was no metallographic evidence of internal cracking or of debonding at the matrix-reinforcement interface during ECAP, the large reduction of ductility after many ECAP passes raises doubts about the engineering utility of such heavily deformed composites, and may suggest that only materials strengthened to near the maximum yield stress by ECAP may be of further interest.

Figure 6 shows the evolution of the high temperature yield stress with the number of ECAP passes for materials processed using route C. Fig. 6a shows data for composites reinforced with atomised particles and Fig. 6b compares these materials with composites containing milled particles. The general trend of yield stress with number of passes is similar at all temperatures and as seen in Fig. 5, namely an initial increase in yield stress for the first few passes, to a maximum, and then a slow fall in yield stress with additional passes. The exception to this generalisation is the behaviour on testing at 350ºC, where the extruded state is the strongest. This behaviour is the same as that observed on testing similar composites processed using route A [15]. Fig. 6b shows that the material containing the milled reinforcement particles has a higher yield stress than the composites containing atomised particles at all test temperatures, but that the total strain that could be accumulated during ECAP processing without cracking was lower (5 passes (strain 3.5) only compared to 8 passes (strain 5.6)). It was mentioned in the experimental section that tensile test data and compression data gave identical yield stresses and data would be shown together. The similarity of tension and compression yield stresses may be confirmed by examination of the mechanical data seen in Fig. 6, remembering that data corresponding to as-extruded material – no ECAP passes – and that processed to 4-5 and 8 passes by ECAP are almost completely tensile test data, while material processed by ECAP to other strain levels were tested in compression.

Figure 7 shows the evolution of yield stress with testing temperature for the reference extruded materials containing 25 and 50% reinforcement particles, together with the materials processed by ECAP using route C. Stress values obtained using route A instead are similar and are not included in this Figure to avoid confusion. For extruded materials tested at room temperature to about 200ºC, an increase in the volume fraction of reinforcement from 25 to 50% produces only a slight (15%) increase in yield stress for atomised particles, slightly more (30%) for milled particles. Yield stresses of extruded material with 25% reinforcement are similar, independently of whether the particles are atomised or milled [14] and the latter data are not included in the present Figure. In contrast with these small strength gains, the increase in yield stress is much greater when the materials are processed by ECAP (75% increase in yield stress for composites with 25% volume fraction of atomised or milled particles). Although the reduction of particle size by milling strengthens the composite slightly, this is a small effect compared to the strengthening produced by matrix grain size refinement by the severe plastic deformation. It is also important to note that much of the strengthening achieved by ECAP is retained up to 200ºC. After deformation at that temperature, TEM observations, Figure 8, have confirmed that the microstructures were rather stable, with grain sizes changing only slightly (increasing from 800 nm to about 900 nm). These observations also showed that there were no oxide particles within the grains, thus confirming that the good retention of strength may be attributed to the stability of the fine grain structure.

4. Discussion

One of the most interesting aspects of the present study is the different grain size refinement achieved by severe plastic deformation depending on the ECAP route followed, with route A producing a greater reduction of grain size and faster strengthening. The deformation process is clearly different with slower dislocation accumulation in the aluminium matrix when using route C. This effect has been studied in Al-Mg alloys [16,17] where it was confirmed that the grain sizes were larger and the boundary misorientations lower when ECAP was carried out using route C. These authors explained this behaviour by the partial annihilation of dislocations that occurs with alternate passes, leading to a slower dislocation accumulation at the low angle boundaries which subdivide the original grains into smaller ones. As a consequence the subgrain structures produced have boundaries of slightly lower misorientation when route C is used. These results have been reconfirmed by our qualititative microstructural observations in the TEM (see Fig. 4) showing that, for the same number of ECAP passes (i.e. total accumulated strain), route A produces boundaries of slightly higher average misorientation than processing by route C where boundaries are more frequently imaged as very low-angle dislocation walls still.

At the same time, slower dislocation accumulation in the aluminium matrix of the present composites would lead to reduced stress concentrations at the matrix/reinforcement interface [15] which would produce less weakening at those interfaces. This could explain why the composites with atomised particle reinforcements can be processed up to eight ECAP passes before cracking using route C, whilst route A produced surface cracking after only four passes [15]. A similar effect occurs for composites containing the finer milled reinforcement particles, where route C could be continued to five passes but route A produced cracking after only two passes. In that case, the slower dislocation accumulation when using route C leads to only a small reduction of matrix grain size inside the smaller aluminium ligaments separating the closely-spaced reinforcement particles.

The evolution of aluminium matrix hardness with number of ECAP passes, Fig. 3b, confirms a faster increase when grain size reduction is more rapid (as for route A). The highest hardness values were achieved, however, for materials processed to many ECAP passes, for which the grain sizes were not always the finest, compare Figs. 3a and b. This indicates that there are other contributions to strengthening of the aluminium matrix, in addition to grain size refinement. A recent study carried out on Al6082 alloy separated the different strengthening contributions produced by ECAP, and showed that a high density of dislocations was retained inside the matrix which led to a similar extent of strengthening as that produced by the decrease in grain size [18]. In a similar way we can consider that the higher strengthening seen for some of the composites with moderate matrix grain size, Fig. 3, can be due to the higher free dislocation density within the grains produced during the larger number of ECAP passes when using route C.

A final aspect worth discussing is the large increase in yield stress of composites produced by severe plastic deformation, compared to that obtained by an increase in volume fraction or decrease in reinforcement size, Fig. 7. Since the reinforcement particles deform only elastically during tensile/compression testing, the large increase in yield stress measured for ECAP processed composites is due to the two hardening parameters described above, i.e. decrease in matrix grain size and increase in internal dislocation density. The faster decrease of yield stress with increasing temperatures for ECAP processed materials can be associated with the recovery of this deformation microstructure. The small strength loss (comparing ECAP processed materials and the equivalent as-extruded materials in Fig. 7) measured for temperatures up to 200ºC can be understood as caused by the recovery of the mobile dislocation density while the grain size remains rather stable to such temperatures. At temperatures above 250ºC the combination of both dislocation recovery and grain growth leads to a faster decrease in the strength of deformation-processed composites.

5. Conclusions

The evolution of microstructure of extruded Al-based composites reinforced with TiAl intermetallic particles has been examined when processing by equal channel angular pressing. Extruded materials exhibit a homogeneous distribution of reinforcement particles and good cohesion at the particle/matrix interfaces. Neither particle size and distribution nor interface cohesion are modified by ECAP processing. ECAP reduces the matrix grain size to about 500 nm when using route A and to about 750 nm when using route C, values much smaller than the grain sizes after extrusion.

The smaller extent of grain size refinement occurring when using route C, even after deforming to larger strain levels (strain of 5.6 for route C as compared with a strain of 2.8 for route A), has been attributed to the partial dislocation annihilation that occurs between alternate ECAP passes, slowing the rate of dislocation accumulation at low angle boundaries and, as a consequence, decreasing the rate of grain size refinement.

The hardness increase measured in the aluminium matrix during ECAP cannot be explained in terms of grain size reduction alone, and a significant contribution is attributed to the presence of mobile dislocations inside the grains produced by the severe deformation.

ECAP leads to a substantial increase (up to 75%) of the yield stress of the composites with increasing number of processing passes, being more rapid when route A is used. However, maximum strengthening is achieved after two to four passes in all cases. The strength increase due to ECAP is much larger than that obtained by increasing the volume fraction of reinforcement particles (from 25% to 50%) in extruded materials.

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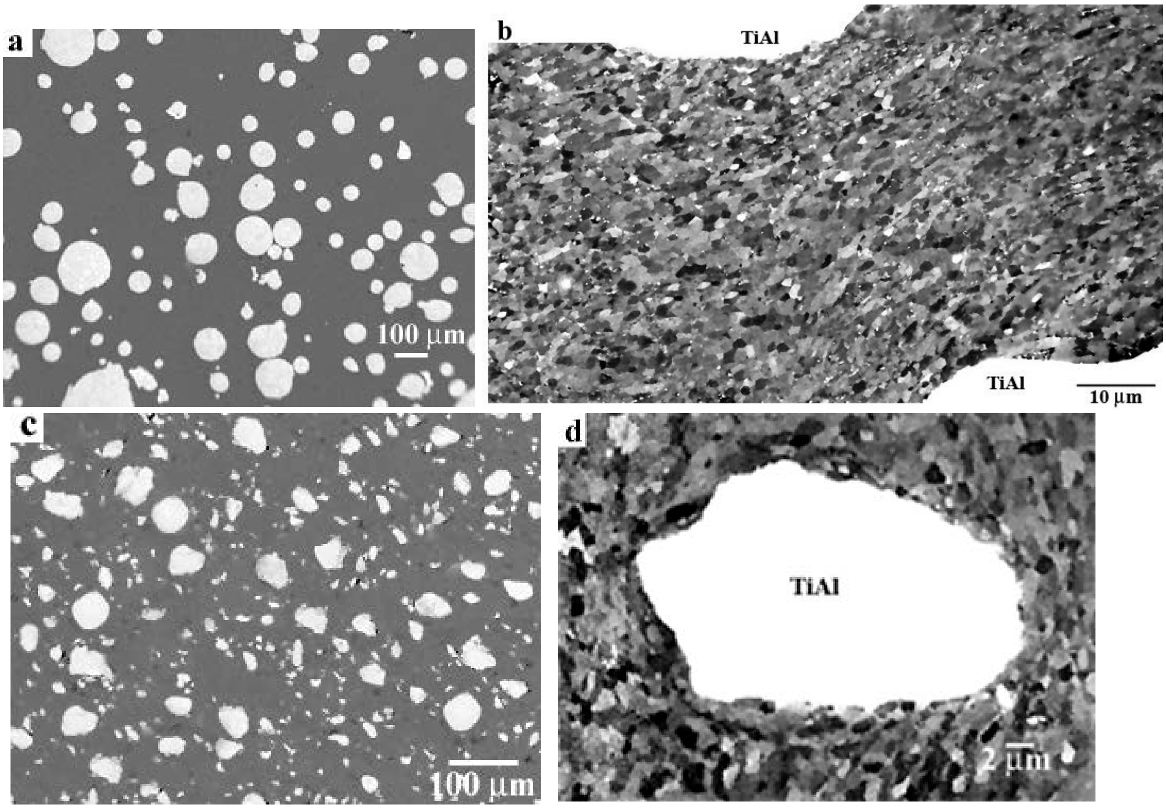

Figure 1. Particle distributions and ultrafine grain structures observed by SEM, using back-scattered electron contrast, of ECAP-processed composites containing 25% reinforcement: (a) and (b) material reinforced with atomised particles, after six passes; (c) and (d) material with milled reinforcement particles, after three passes.

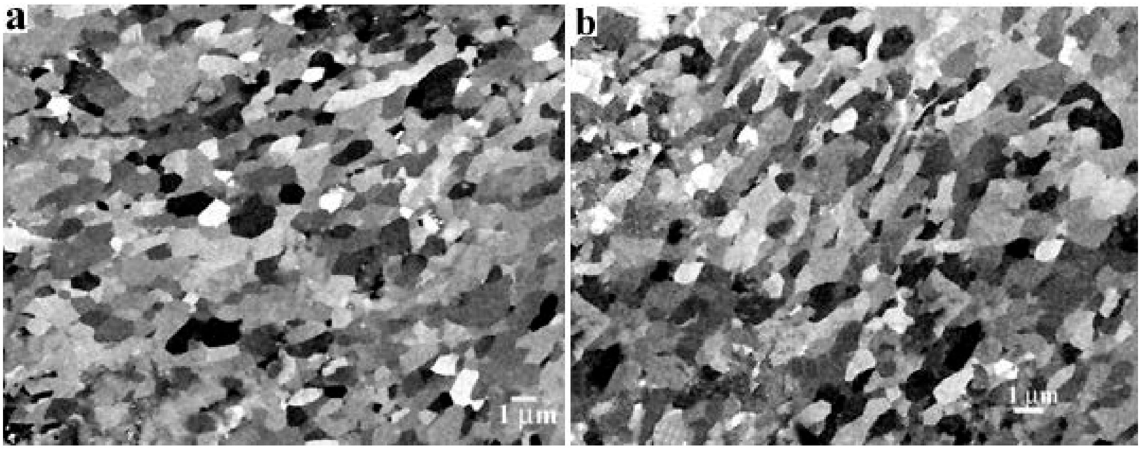

Figure 2. Aluminium matrix microstructure after severe deformation using route C: (a) material reinforced with atomised particles, after eight passes (b) material containing milled reinforcement particles, after five passes.

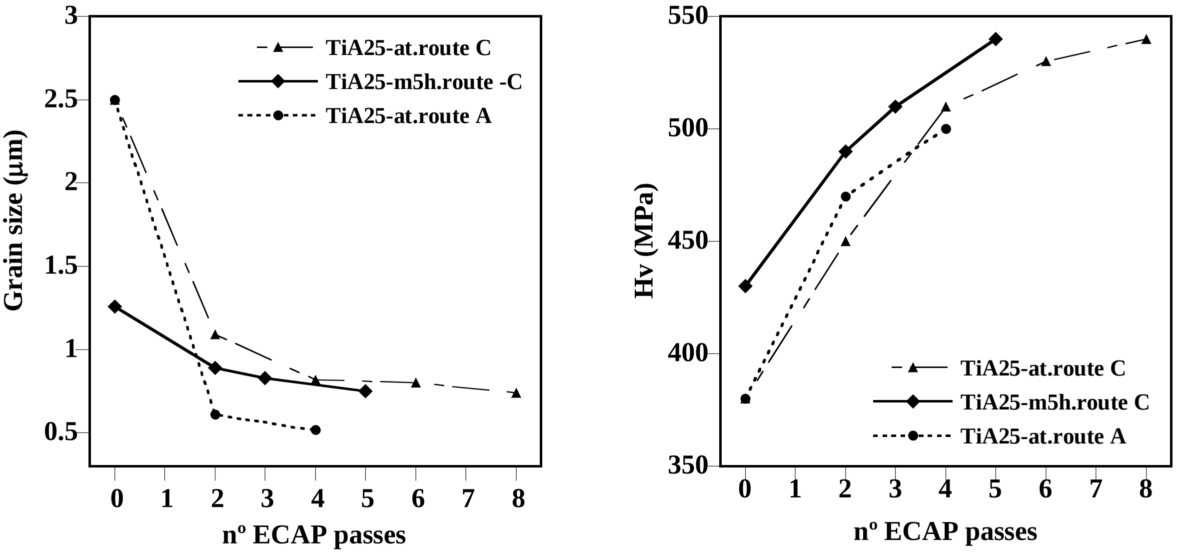

Figure 3. Evolution of grain size (a) and hardness (b) with number of ECAP passes depending of the route used (A and C) or the type of material. at.= atomised reinforcement; m5h= reinforcement particles obtained by milling for 5 h

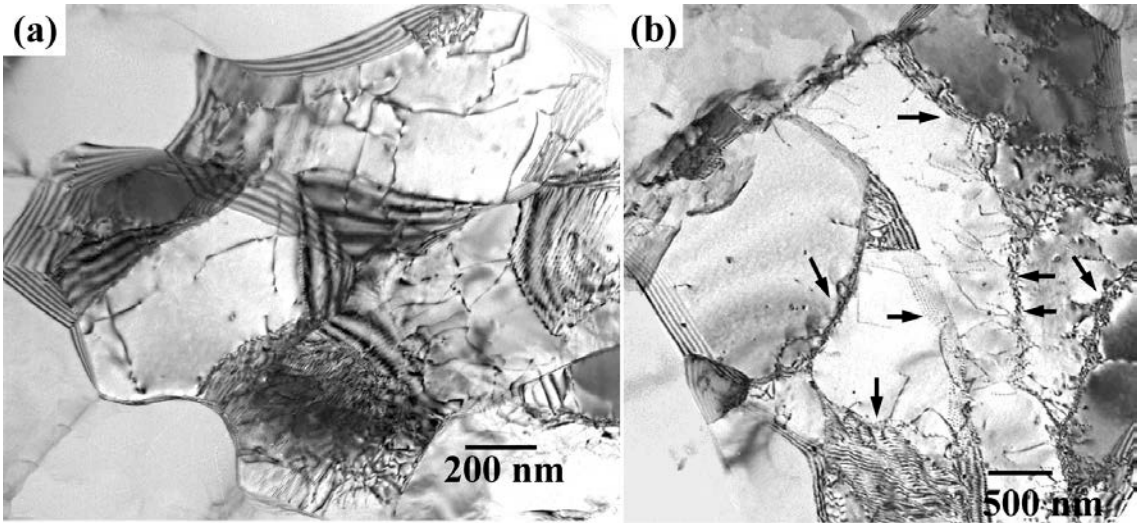

Figure 4. Microstructures observed by TEM of the aluminium matrix of composites reinforced with atomised particles, after four ECAP passes: (a) using route A produces higher angle boundaries characterised by fringe contrast; (b) using route C produces slightly lower angle boundaries, sometimes seen as dislocation walls (marked by arrows).

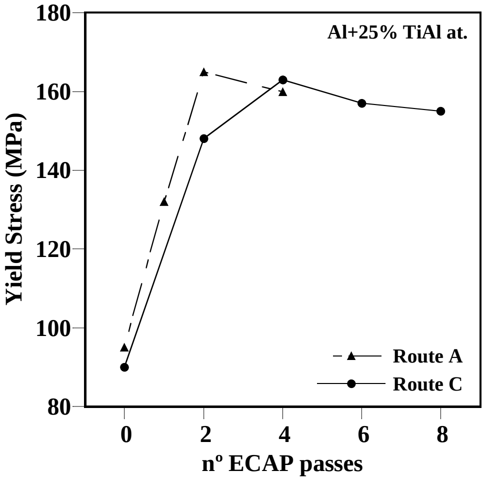

Figure 5. Evolution of room temperature yield stress with number of ECAP passes using two different routes in the material reinforced with 25% of atomised TiAl particles.

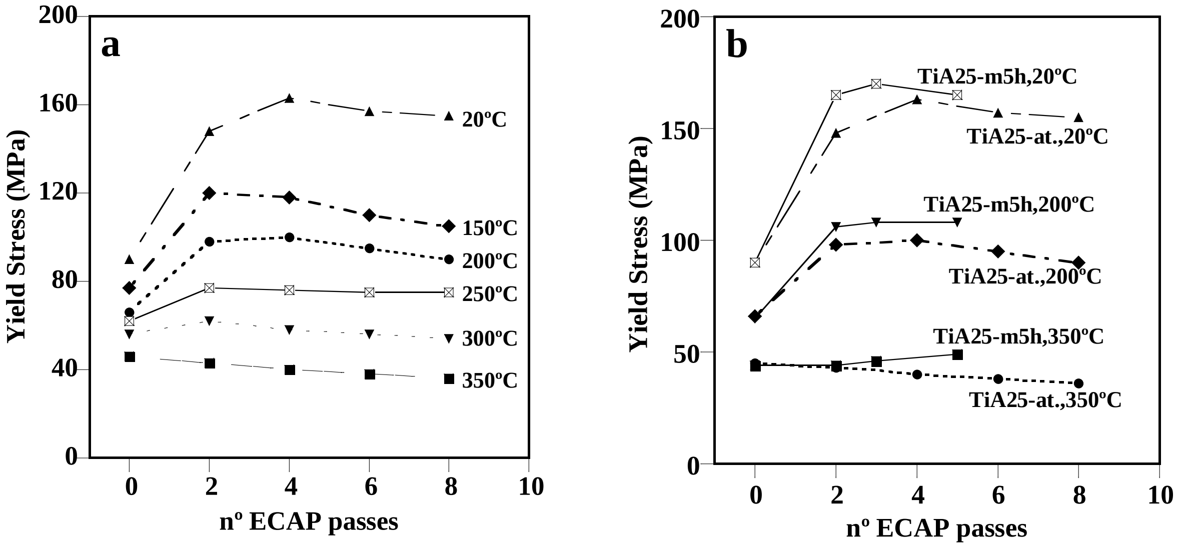

Figure 6. Evolution of yield stress, measured at both room temperature and several elevated temperatures, with number of ECAP passes using route C: (a) reinforced with atomised particles; (b) comparison of composites containing atomised (at) or milled (m5h) TiAl reinforcements. Test temperature is indicated. Materials in the as-extruded state (no ECAP passes), after 4-5 passes, and after 8 passes were tested in tension, while remaining materials were tested in compression.

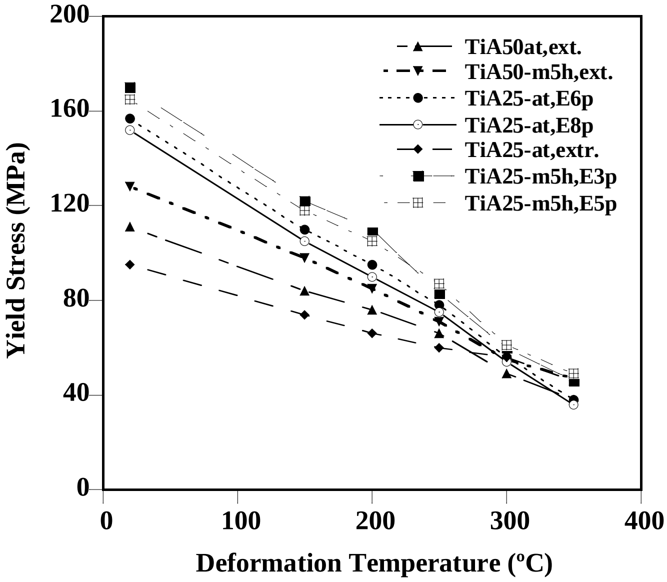

Figure 7. Variation of yield stress with test temperature for composites processed: as extruded reference state (ext); processed by ECAP (E) using route C to different passes (e.g. E6, E8); materials contained either 25% or 50% volume fraction of reinforcement TiAl particles, present either in the atomised state (at.) or after milling for 5h (m5h).

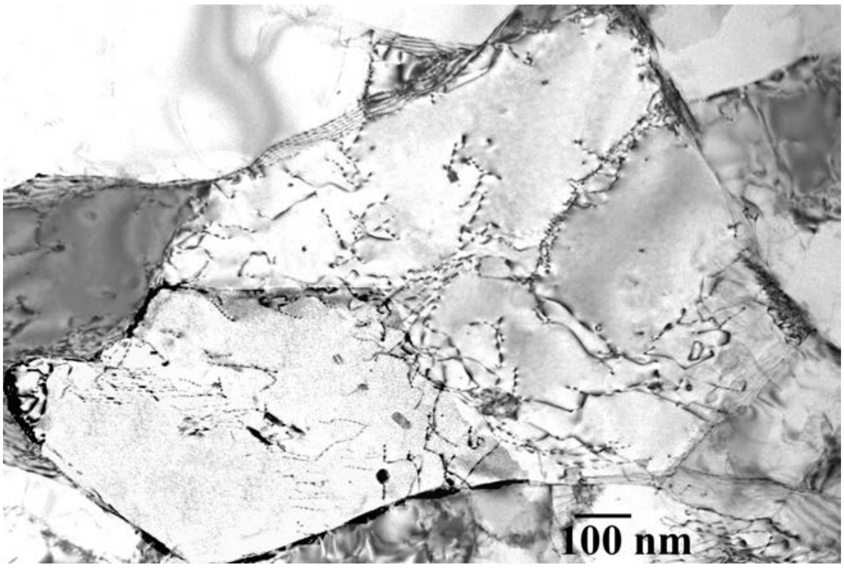

Figure 8. TEM microstructure observed after deformation at 200ºC of composite processed by six ECAP passes using route C. Note the absence of oxide particles inside the grain.