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Comparison of Al-Cu-Fe quasicrystalline particle reinforced Al composites fabricated by conventional casting and extrusion

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Aluminum matrix composites reinforced by $Al_{62}Cu_{26}Fe_{12}$ gas atomized powders were produced by conventional metallurgical processes, such as gravity casting with stirring and hot extrusion. This investigation was mainly focused on the dependency of the yield stress at room temperature as a function of the volume fraction of reinforcement, but other variables such as the matrix, coating layer around the particles, and processing were also investigated. For as-extruded composites, the addition of the Al-Cu-Fe particles improved the yield stress, although not dramatically owing to the large particle size. In contrast, it was found that the yield stress was considerably enhanced for the as-cast composites up to 10%(AlCuFe)p, while an asymptotic value was observed afterward. The dominant parameter appeared to be the strength of the matrix, which was found to be proportional to the volume fraction of the reinforcement. These results are discussed in relation with the possible strengthening mechanisms in order to estimate the role of the icosahedral and related crystalline phases on the increase of yield stress.

1. Introduction

Early investigations indicated that bulk quasicrystalline materials are extremely brittle at room temperature [1–3], which rules out their usage as structural materials, although their behavior becomes ductile enough to accommodate plastic deformation at high temperatures. However, numerous researches have been pursued to find potential applications, investigating their electronic [4], magnetic [5], and surface properties [6]. Dubois and colleagues [6, 7] exploited the surface properties of the icosahedral quasicrystalline materials such as high hardness, low friction coefficient, high wear resistance, and thermal stability to produce coatings for cookware. An alternative way to circumvent the brittleness of quasicrystals is to disperse these phases into a ductile matrix. The first utilization of the Al-Cu-Fe quasicrystalline particles as reinforcement in aluminum composites was reported by Tsai *et al*. [8], who noticed an improvement of the microhardness with an increase of the volume fraction of the icosahedral phase. Recently, a high strength steel with good ductility has been commercially produced by precipitation of the icosahedral quasicrystalline phase into maraging steels [9].

The strong demand for aluminum alloys with improved properties has led to the development of composites, such as Al/SiC and Al/Al_2O_3 composites, applied in automotive and aerospace industries. However, the integrity of the composites depends critically on the wettability between the reinforcement material and the matrix. Furthermore, degraded service performances at elevated temperatures and low fracture toughness remain weak points for these composites as well as the extreme difficulty in recycling.

Hence, the aim of this study was to evaluate the potential of the quasicrystalline phases as discontinuous reinforcement to produce aluminum composites. An Al-Cu-Fe alloy was chosen because of its low cost, relatively low density, high thermal stability, and compatibility with aluminum matrix. Since angular particles in discontinuous reinforced composites were found to be responsible for the premature crack initiation resulting from the high stress concentration at sharp

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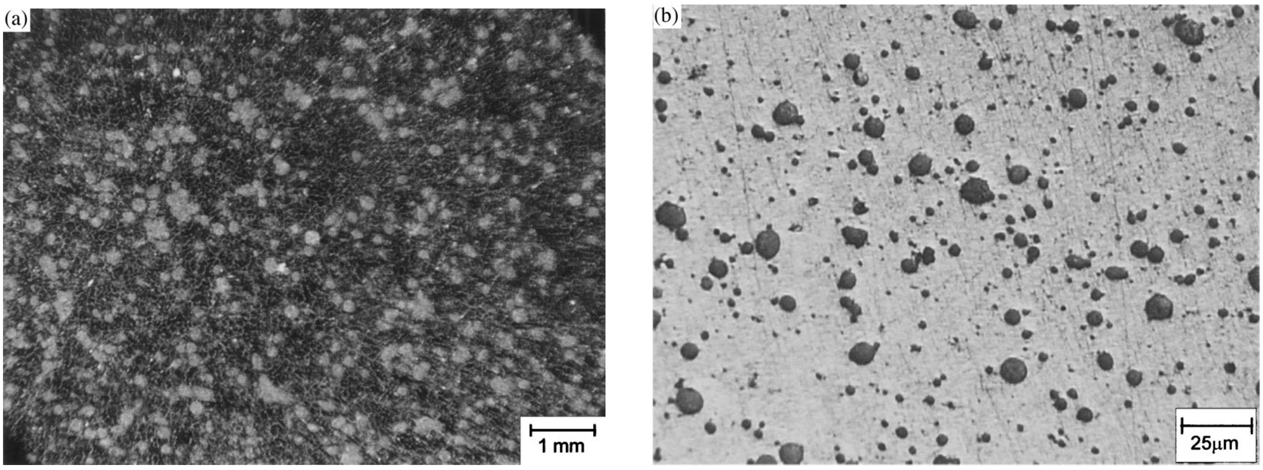

Figure 1 Representative microstructures of the Al/(AlCuFe)p composites; (a) as-cast and (b) as-extruded.

corner [10, 11], spherical particles of an $Al_{62}Cu_{26}Fe_{12}$ alloy were prepared by gas atomization technique. Aluminum matrix composites were produced by two different processes of conventional gravity casting with stirring and hot extrusion. Both processes were compared in terms of resultant particle distribution, microstructure and mechanical properties. The influence of the matrix and the role of a Ni coating layer onto the reinforcement particles were also examined during conventional casting. The strengthening mechanisms were assessed in order to estimate the role of the quasicrystalline phase and its related crystalline phases.

2. Experimental procedure

2.1. Fabrication of composites

$Al_{62}Cu_{26}Fe_{12}$ powders were prepared by gas atomization, and were subsequently sieved. A thorough structural characterization of the powders can be found elsewhere [12]. Larger powders of 74–149 μm in diameter were used for the casting process, while smaller powders of 38–73 μm in diameter were selected for the extrusion process.

For the fabrication of composites by casting method, the commercial purity aluminum (99.9 wt.%) was firstly melted in a high frequency induction furnace under a dynamic argon atmosphere at a temperature of 750°C. Then, $Al_{62}Cu_{26}Fe_{12}$ powders were mixed to the molten Al and the liquid mixture was poured into a pre-heated cylindrical steel mold (200°C). The volume fraction of the $Al_{62}Cu_{26}Fe_{12}$ quasicrystalline particles ((AlCuFe)p) was varied from 0 to 20%. The role of a Ni coating layer on the reinforcing particles was also evaluated by preparing composites of 15% volume fraction of (AlCuFe)p coated with a 5 μm thick Ni layer deposited by the non-electrolysis method. $Al_{96}Cu_4$ matrix - 5% (AlCuFe)p composites were fabricated, using identical conditions as described above, in order to analyze the role of the matrix on the resultant mechanical properties.

For the fabrication of the composites by powder metallurgy process, (AlCuFe)p were firstly mixed with Al powders by mechanical agitation, secondly placed into an aluminum tube of internal diameter of ∅ 60 mm, then degassed down to 10^{-3} Torr at 350°C, and finally extruded at 350°C to obtain a final rod-type specimen with a diameter of ∅ 15 mm.

2.2. Characterization

Microstructures were examined by means of X-ray diffractometry (XRD), optical microscopy (OM), scanning electron microscopy (SEM), and transmission electron microscopy (TEM). Reasonably homogeneous distribution of the particles was obtained in composites prepared by both conventional casting (Fig. 1a) and hot extrusion processes (Fig. 1b). However, a large size distribution of the particles was revealed in the extruded composites while small particles could not be observed in the as-cast composites.

Matrix microhardness was measured on the reinforced and unreinforced specimens, with indentations made in the particle-free regions of the reinforced specimens. At least 20 measurements were realized for each composite. Polished specimens of about $4 \times 4 \times 7$ mm^3 and $10 \times 10 \times 10$ mm^3 were prepared for compression and continuous indentation tests, respectively, and, for both tests, the mechanical loading was applied along the longitudinal axis. Continuous indentation is based on multiple indentation cycles on a polished surface by a spherical indenter of ∅ 2.4 mm. This technique has recently found many potential applications for assessing the *in-situ* mechanical properties of either large or small size components without affecting its integrity. However, the data analysis is not straightforward due to the complex stress field, and detailed descriptions regarding to the evaluation of the mechanical properties such as the yield stress and work hardening can be found in ref. [13]. Tensile tests were carried out on the hot extruded composites using cylindrical specimens of ∅ 6.4 mm and 25.4 mm gauge diameter and gauge length, respectively.

3. Results

3.1. Composites fabricated by conventional casting with stirring

Data of the 0.2% offset yield stress determined from compression and continuous indentation tests are

TABLE I Evolution of the yield stress determined by compression and indentation tests with the volume fraction of the (AlCuFe)p in the as-cast composites (expressed in MPa)

Volume fraction of (AlCuFe)p	5%	10%	15%	20%
Compression	144 ± 29	211 ± 15	178 ± 30	227 ± 20
Continuous indentation	135 ± 11	189 ± 10	188 ± 30	225 ± 24

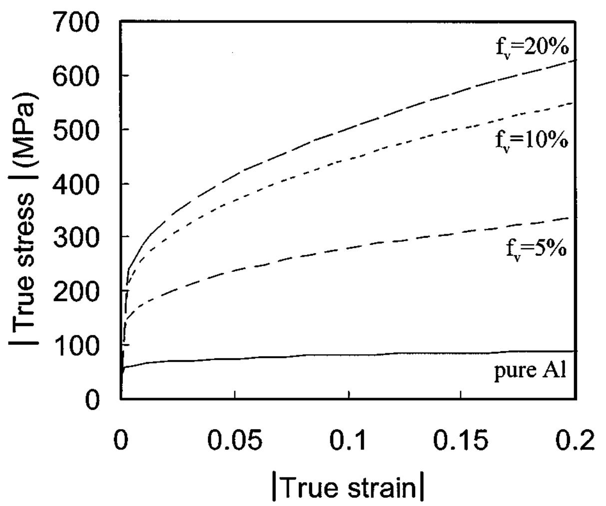

Figure 2 Stress-strain curves of the as-cast composites for different (AlCuFe)p volume fractions.

summarized in Table I, where the indicated values correspond to an average of at least three tests. Since this study was focused on the properties of composites without any consideration of heat-treatment effect, tensile tests were not carried out on as-cast specimens owing to the possible existence of casting defects in the ingots. These results revealed a marked scatter in the mechanical data, a common feature of metal matrix composites which can be partly attributed to the non-uniform distribution of the particles within the matrix.

Stress-strain curves determined from indentation tests are presented in Fig. 2. A significant increase of the yield stress (σ_y) and work hardening rate (n, defined as the exponent of the true plastic strain in $\sigma = K(\varepsilon_p)^n$) was achieved by the addition of the Al-Cu-Fe particles into the pure Al matrix. The evolution of the yield stress as a function of the volume fraction of the Al-Cu-Fe particles is shown in Fig. 3. Up to 10%, the dependency of this mechanical property with the volume fraction of (AlCuFe)p is consistent with previous works on aluminum matrix reinforced by ceramic particles [14], however, the yield stress seems to reach an asymptotic value for the volume fraction of (AlCuFe)p above 10%.

In order to investigate the influence of the matrix on the mechanical behavior of the composites, compression and indentation tests were performed on as-cast $Al_{96}Cu_4$/5%(AlCuFe)p composites. Although no significant effect of the matrix on the work hardening exponent could be observed, the yield stress of composites with an $Al_{96}Cu_4$ alloy was found to be approximately

TABLE II Values of the yield stress determined by tensile, compressive and indentation tests on the as-extruded Al/(AlCuFe)p composites

Al/10%(AlCuFe)p	Tension	Compression	Continuous indentation
Yield stress (MPa)	93.1 ± 1.4	95.9 ± 3.9	97.6 ± 10.4

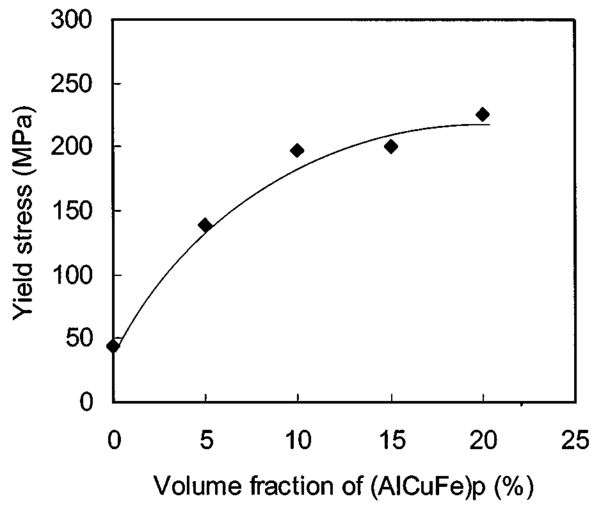

Figure 3 Evolution of the yield stress with the volume fraction of (AlCuFe)p.

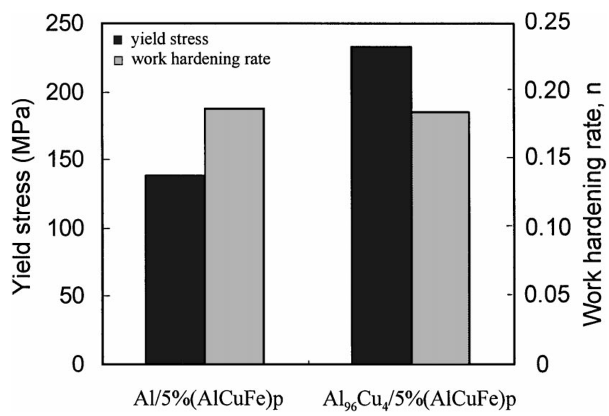

Figure 4 Comparison between the yield stress and the work hardening exponent determined in Al/5%(AlCuFe)p and $Al_{96}Cu_4$/5%(AlCuFe)p fabricated by conventional casting.

65% larger than the corresponding pure aluminum matrix composites (Fig. 4).

3.2. Composites fabricated by hot extrusion

Mechanical characteristics determined from tensile, compressive, and continuous indentation tests are summarized in Table II. Scatters of the tensile and compressive test data were relatively low, while the scatter in the indentation test data was equivalent to those of the as-cast Al/10%(AlCuFe)p composites. Nevertheless, these results indicate a slight difference between the yield stress measured under tensile and compressive loading. This tensile-compressive yield stress asymmetry observed in as-extruded composites can be explained by the state of residual stresses prior to loading. The mechanical residual stresses are induced during hot

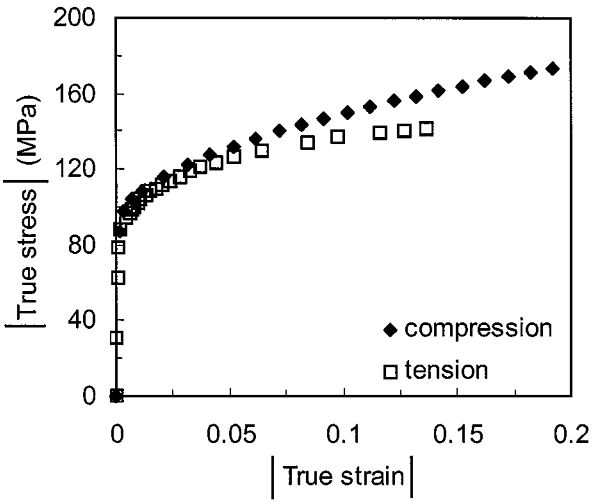

Figure 5 Comparison between compression and tension stress-strain curves of extruded composites.

extrusion process by the large extrusion ratio (25 : 1), and the thermal residual stresses are caused during the cooling down of the extruded rod to room temperature by the difference of thermal expansion coefficients between the matrix and particles [11].

The corresponding stress-strain curves obtained in compression and tension tests performed on extruded composites are compared in Fig. 5. Although the values of the yield stress determined by compression and tension tests are almost identical, the two stress-strain curves showed somewhat different behavior for large strain. This difference in mechanical behavior can be understood by a microstructural analysis. Fig. 6a and b show polished cross-sections of the specimens after tensile and compressive tests, respectively. Under large tensile strain, particles are broken in two with the development of cracks in the matrix (Fig. 6a). In tensile test, the primary mode of damage is therefore cracking of the medium and large size Al-Cu-Fe particles, as it was observed by Lloyd for Al/SiCp composites [15]. Fig. 6a, taken in the necking region, also reveals particle debonding (see arrow). For specimens tested in compression, a slight particle/matrix debonding could also be observed after large plastic strain (see arrows), but no cracked particles. A large work hardening cannot therefore be expected after the failure of the reinforcing particles under tensile loading, leading to the difference of stress-strain behavior shown in Fig. 5.

4. Discussion

By introducing particles into a pure metal or an alloy, the resulting yield stress is dependent on the direct strengthening provided by the particles, and on the strength of the matrix itself. The enhancement of properties due to the addition of particles can be better appreciated by subtracting the yield stress of the unreinforced matrix to the yield stress of the composite. On a commercial grade Al alloy reinforced with 10% of either SiC or Al_2O_3 particles, the increase of yield stress varies from approximately 50 to 110 MPa, depending on the matrix, the size of the particles and the heat-treatment conditions [11]. In this work it was found that, in Al/10%(AlCuFe)p composites, the average increase of the yield stress was about 44 MPa for the extruded composites, and about 150 MPa for the conventionally cast composites. Thus, the addition of the quasicrystalline forming particles into an aluminum matrix provided an efficient strengthening, despite the large particle size.

The strength of the matrix can be assessed through microhardness measurements of the matrix on reinforced and unreinforced composites, provided the indentation sizes are small enough to be far away from the reinforcement particles. The influence of the volume fraction of the particles on the matrix microhardness is clearly shown in Fig. 7. For composites prepared by the conventional casting method, a linear relationship was found with values of the microhardness similar to those determined by Tsai *et al.* [8] on Al(AlCuFe)p composites prepared by mechanical alloying and hot pressing technique. For an identical volume fraction of particles, the microhardness of the matrix was found to be larger in composites fabricated with an $Al_{96}Cu_4$ alloy than with pure aluminum. In contrast, the presence of (AlCuFe)p did not considerably increase the microhardness of the matrix in the hot extruded composites.

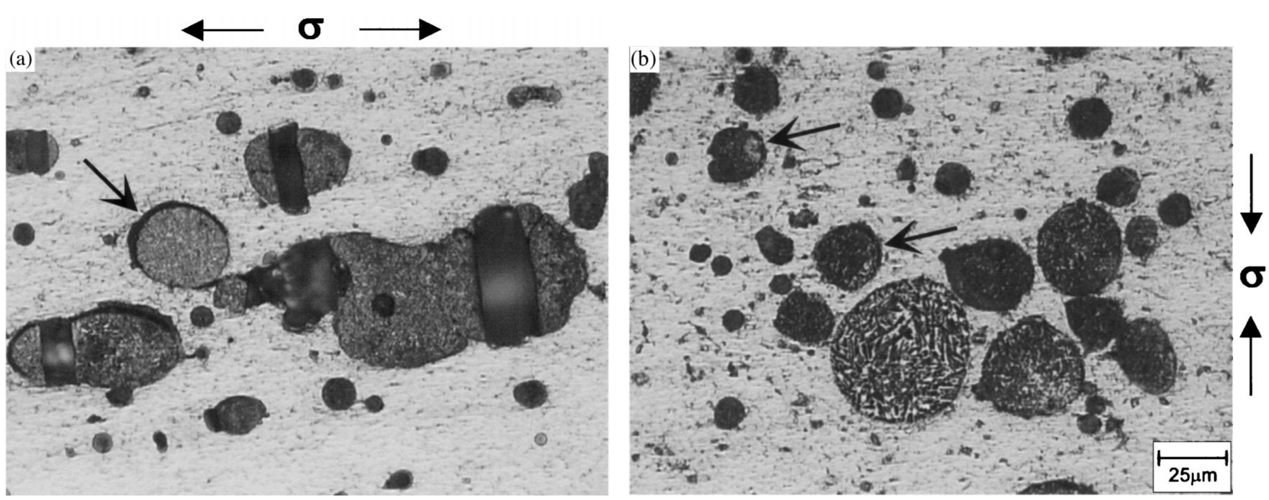

Figure 6 Cross-sectional view of composites prepared by hot extrusion; (a) after fracture in tension and (b) after compression test.

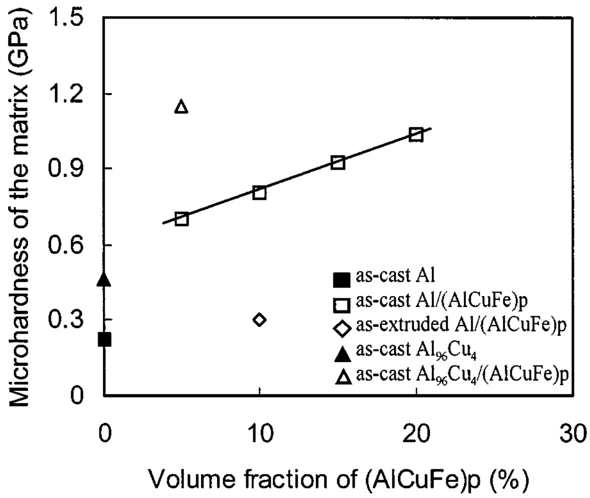

Figure 7 Evolution of the matrix microhardness with the volume fraction of (AlCuFe)p.

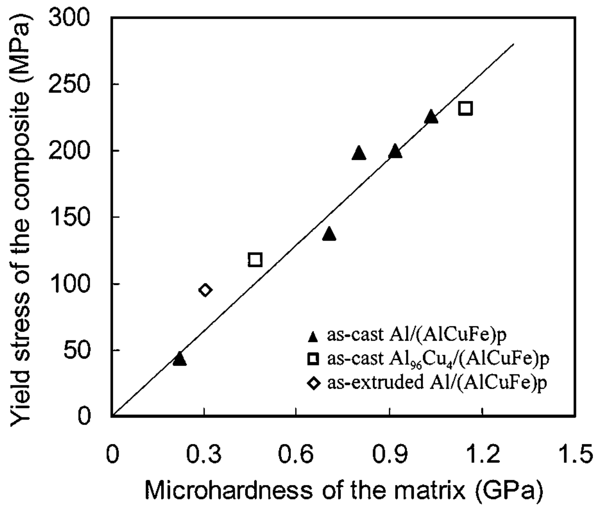

Figure 8 Evolution of the yield stress of the composites with the microhardness of the matrix.

For the aluminum composites reinforced by the Al-Cu-Fe particles with an average size of 5 μm, Tsai *et al.* [8] attributed the dependency of the microhardness to the dispersion of the icosahedral quasicrystalline phase (i-phase) for hot pressing performed at 400°C, and the ω-$Al_7Cu_2Fe_1$ crystalline phase for hot pressing at 600°C. However, the large discrepancy between Al/(AlCuFe)p composites prepared by conventional casting and hot extrusion indicated that the increase of the Al matrix microhardness was not solely due to the addition of (AlCuFe)p, but was also the consequence of structural modifications of the matrix occurring during the casting process, as it will be discussed later in more details.

In the absence of unified theories, the increase of yield stress brought by the particles can be estimated from models describing individual strengthening mechanisms [11] which are derived from the theories of continuum mechanics and of dislocation. In the continuum mechanics theories, the strengthening is governed by the particle shape and the volume fraction, based on the differences between the elastic properties of matrix and reinforcement, while dislocation hardening is dependent on the particle size as well as volume fraction. Due to the large particle size, and the almost similar elastic properties of the $Al_{62}Cu_{26}Fe_{12}$ alloy and the matrices, models such as the Orowan strengthening [16] and Eshelby's equivalent inhomogeneous inclusion [17] predict that the direct contribution of the Al-Cu-Fe particles is only about a few MPa. Other mechanisms, which take into account of the matrix strength, have to be considered.

The role of the matrix on the strength of the composite is shown in Fig. 8, where the data points give a linear correlation expressed as:

$$(\sigma_y)_c = 4.65\,(H_v)_m \quad (1)$$

where $(\sigma_y)_c$ is the yield stress of the composite, $(H_v)_m$ is the microhardness of the matrix (both variables being expressed in MPa). This result corroborates the conclusions made by McDanels [14] that, in composites with discontinuous reinforcement, although the yield stress of Al/SiC composites was dependent on the volume fraction of reinforcement, the primary factor was the strength of the matrix.

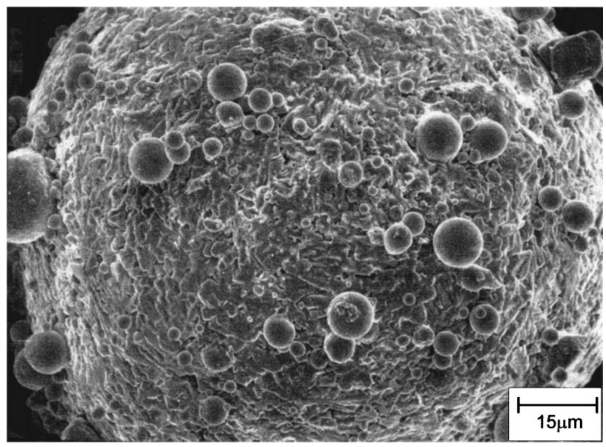

Figure 9 Optical microscopy showing the external aspect of an Al-Cu-Fe particle.

A careful observation of the powders revealed the presence of undesirable small size satellites found around large particles, satellites that could not be eliminated during sieving (Fig. 9). When mixed into the molten aluminum, the dissolution of these satellites led to a microstructural modification of the matrix (see Fig. 1a). Consequently, solid solution hardening is thought to play an important role in composites prepared by conventional casting, due to the dissolution of small Al-Cu-Fe particles into the matrix.

In order to investigate the role of the icosahedral phase on the strengthening, composites were fabricated by conventional casting with Ni coated Al-Cu-Fe particles. The larger content of the i-phase remaining in the particles can also be observed in Fig. 10b as compared to Fig. 10a, where the i-phase appears in gray while the dark phase corresponds to the β-phase, although no dramatic change could be revealed by XRD measurements (compare Fig. 10c and d). Furthermore, the observation of the matrix indicated that less dissolution of the particles has occurred with Ni coating onto the

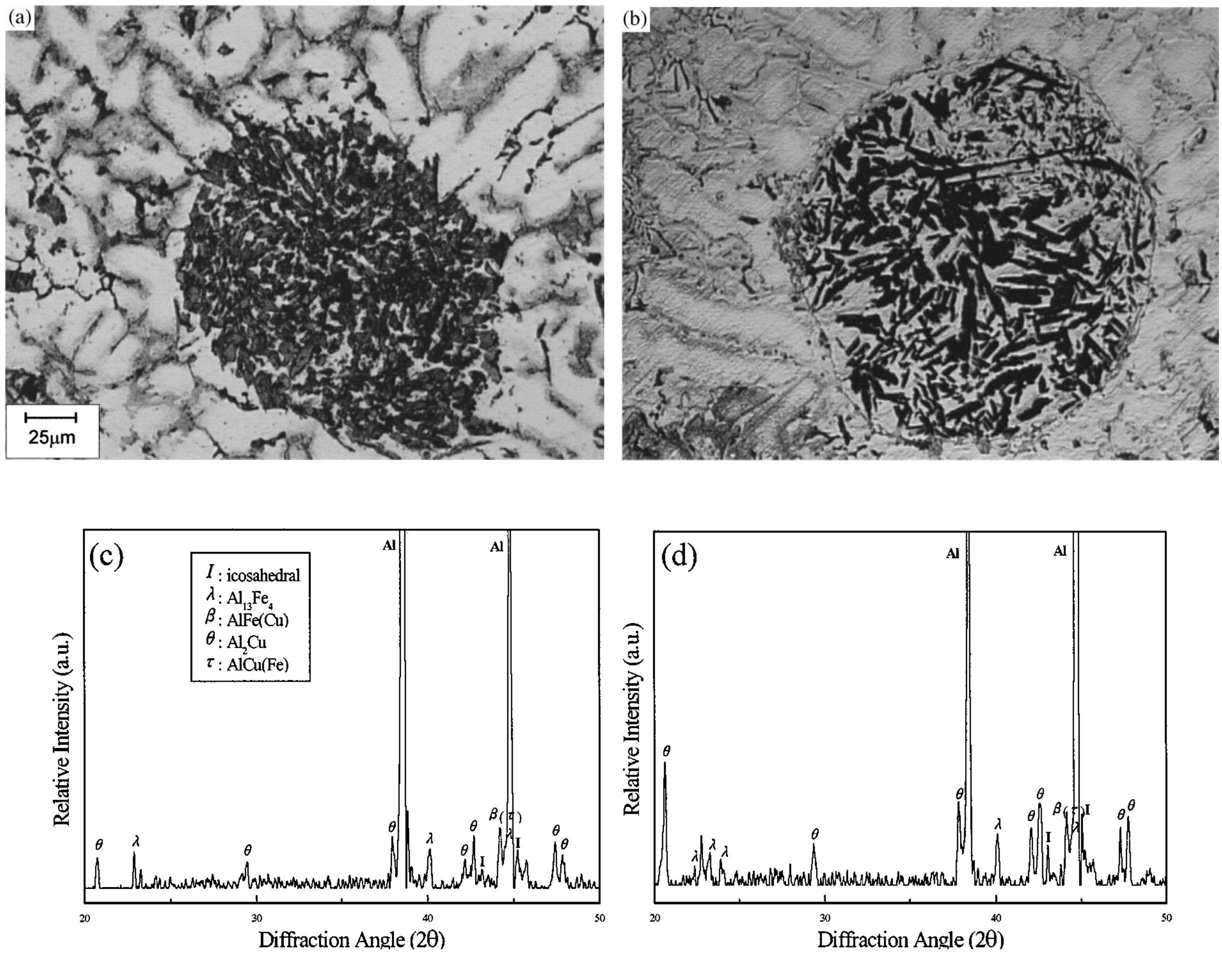

Figure 10 Al matrix composites prepared by conventional casting; (a) OM of Al/15%(AlCuFe)p, (b) OM of Al composites with Ni coated 15%(AlCuFe)p, (c) XRD of Al/15%(AlCuFe)p, and (d) XRD of Al composites with Ni coated 15%(AlCuFe)p.

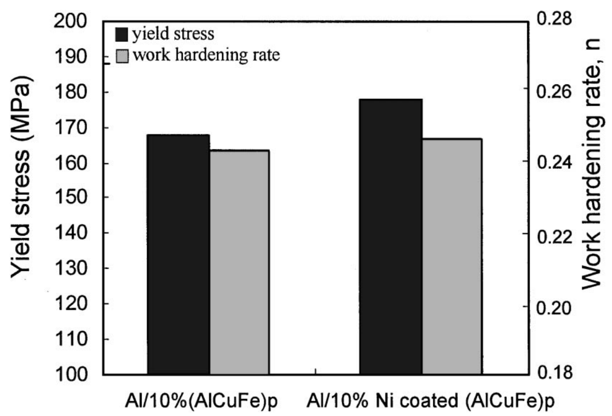

Figure 11 Influence of the Ni coating layer on the yield stress and work hardening of as-cast Al/15%(AlCuFe)p composites.

particles since the dendritic structure of the matrix is less marked. The effect of the Ni coating layer on the yield stress is shown in Fig. 11. An average increase of 6% of the yield stress was obtained while the work hardening did not vary significantly.

TEM observations confirmed the existence of the *i*-phase in the particles, but revealed a complex matrix microstructure, with the presence of λ (monoclinic $Al_{13}Fe_4$), β (b.c.c. CsCl type AlFe(Cu)), and ω ($Al_7Cu_2Fe_1$ tetragonal) phases which might contribute to the enhancement of the properties by precipitation hardening (see Fig. 12). Especially, the ω phase was newly observed in the as-cast composites, which had not been observed in the as gas-atomized powders. This ω phase could be formed by a reaction between the *i*-phase and aluminum matrix [18]. Therefore, the origin of the substantial increase of the yield strength cannot be solely attributed to the larger content of the dispersed icosahedral phases since it is accompanied by a change of the volume fraction of the other phases. Further TEM investigations are thus needed for a thorough determination of the strengthening mechanisms of the composites as a function of the volume fraction of the Al-Cu-Fe particles.

In comparison, no microstructural modification of the matrix was revealed by TEM in as-extruded composites. When mixed by mechanical agitation, the satellites observed in Fig. 9 were detached from the large Al-Cu-Fe particles and scattered into the matrix without affecting the microstructure of the composites but enlarging the particle size distribution (see Fig. 1b). Thus, the increase of the 0.2% offset yield stress in as-extruded composites can be accounted for as the superimposition of several mechanisms induced by load transfer, Orowan strengthening, increase of the dislocation density resulting from the mismatch of the thermal expansion coefficients between particles and matrix, reduction of the dislocation subgrain size and increase of the work hardening rate [19–21].

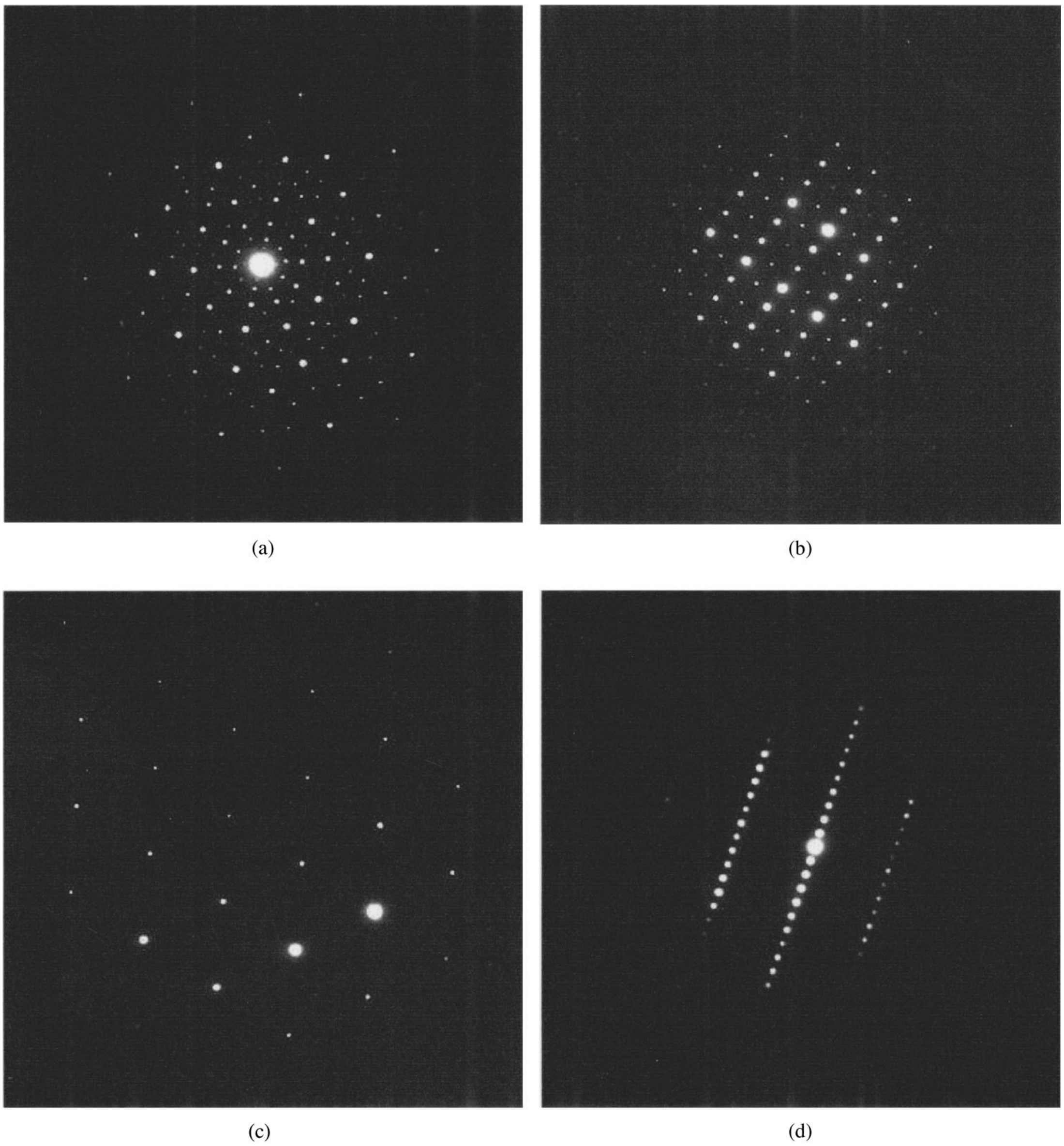

Figure 12 Selected area diffraction patterns of; (a) i-phase (5-fold), (b) λ-phase [001] zone axis, (c) β-phase [111] zone axis, and (d) ω-phase [120] zone axis.

5. Conclusions

Two conventional metallurgical processes were used for the fabrication of composites reinforced by Al-Cu-Fe quasicrystalline phase forming particles. Mechanical properties of the as-cast Al/(AlCuFe)p composites evaluated through compression and indentation tests indicated a significant increase of the yield stress especially in the range up to 10% volume fraction of the reinforcement. In contrast, a much lower increase of the yield stress could be observed in the hot extruded composites. For the as-cast composites, the major strengthening mechanisms were solid solution hardening resulting from the dissolution of small particles, precipitation hardening due to the formation of the ω phase, and dispersion hardening induced by the presence of the quasicrystalline phase. For the extruded composites, it was suggested that the strengthening occurred as a result of a decrease of the interparticle spacing, an increase of the dislocation density and an increase of the work hardening rate. Further microstructural investigations are required to quantity the role of the icosahedral and related crystalline phases on the increase of yield stress.

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