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An Al Alloy-Ti Microfilament Composite Manufactured by Co-Spray Forming and Accumulative Deformation Processing

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Spray forming with co-injection of a solid particulate phase to form a homogeneous distribution within the final spray formed billet has been studied as a new route to manufacturing metal-metal composites at large scale with negligible oxide. 12wt% Ti particles were co-injected into an atomised Al alloy droplet spray and co-deposited to form a 300kg billet at Peak Werkstoff GmbH, Germany. The microstructure comprised refined equiaxed α-Al grains (~5μm), spherical Si particles (~1μm) and uniformly distributed Ti particles (~80μm). Sections of the billet were extruded under a range of conditions into long strips 20mm wide and 6mm, 2.5mm and 1mm thickness. At high strains, the Ti particles were deformed into continuous fibres of a few microns in thickness. The large interfacial area between the fcc α-Al and hcp Ti inhibited dislocation motion and enhanced tensile properties. Accumulative roll bonding was then performed to higher total strains, while maintaining a constant cross-section, reducing the Ti fibres to sub-micron thickness. The fibres were studied by extraction after selective dissolution of the α-Al matrix. There was no interfacial reaction between α-Al and Ti or any measurable oxide formation.

Keywords: *Co-spray forming, metal-metal composite, extrusion, accumulated roll-bonding (ARB), electron backscattered diffraction (EBSD).*

1. Introduction

A metal-metal composite differs from a conventional metal matrix composite in that both the majority matrix and the minority "reinforcing" phase are ductile metals [1]. The constituent metals have limited mutual solubility and are generally either a face centred cubic (fcc) metal paired with a body centred cubic (bcc) metal such as Cu-Nb or Al-Nb, or a fcc metal paired with a hexagonal close-packed (hcp) metal such as Al-Ti [1]. These materials have been manufactured typically by mixing the constituent metals via powder metallurgy (PM) or by co-melting two metals that are miscible as liquid but immiscible as solid so that complete phase separation occurs during solidification. The PM or cast feedstock is then heavily deformed via a series of, or a repetitive process of extrusion, swaging, drawing and/or rolling into wire or sheet form producing an elongated filament morphology in the secondary phases. Metal-metal composites have been studied since the late 1970s [2] when it was discovered that following significant uniaxial deformation processing the strength increased beyond that which can be accounted for by the rule-of-mixtures. The precise mechanism of strengthening in these materials has not been identified unambiguously but the very large interfacial area between the two crystallographically dissimilar metals is likely to play a central role [2]. Ultimate tensile strengths of 2200, 1030 and 890MPa have been reported for Cu-18vol%Nb [2], Al-20vol%Nb [3] and Al-20vol%Ti [4] systems respectively. Among the severe plastic deformation processes, accumulative roll bonding (ARB) has the advantage of inducing high uniaxial strains without reducing the cross-sectional area.

This paper investigates co-spray forming as a route to producing Al based metal-metal composite feedstock ingots at a scale (~300kg per billet) significantly beyond previous PM and immiscible alloy system approaches. Furthermore spray forming offers advantages over PM in that significantly fewer

oxides are incorporated. Standard extrusion practices are used to deform the billets into strips of different thickness (deformation ratios), followed by accumulative roll-bonding to induce higher strains. Although the Al-Ti system studied here is similar to previous studies by powder metallurgy e.g. [4], we introduce another much harder, disperse third phase of pure Si particles that may play a role in the deformation of the minority Ti phase.

2. Experiments

2.1 Spray forming and extrusion

Nominal Al-12wt%Si alloy with co-injected 12wt%Ti powder (<150μm, D_{50} = 80μm) was spray formed at Peak Werkstoff GmbH, Germany. The composite billets of Ø300mm were machined to Ø297mm and then extruded using an eightfold die to Ø30mm bars at a deformation ratio of 12. The bars were cut into 50mm long sections and extruded into strips of different thickness at Oxford University, using the parameters shown in Table 1.

Table 1: Parameters used for the extrusion of Al-Si+Ti metal-metal composites.

Sample dimension	Extrusion parameters			
	Ratio	Temperature (˚C)	Force (kg)	Speed (mm/min)
Ø 30mm bar	12	350	22,000	600
2.5×20mm strip	162	380	62	5.20
1.0×20mm strip	408	415	62	1.27

2.2 Accumulative roll bonding (ARB)

ARB was conducted at the University of Buenos Aires using 1mm thick strip already with a true strain of ~6. Sample preparation involved strip degreasing in acetone and automatic sand brushing the faces to be bonded to remove any oxides. Both ends of the strips were fixed tightly together by stainless steel wire. ARB was carried out at room temperature and without lubrication, with a reduction in thickness of 50% per cycle (von Mises equivilant strain of 0.8). The roll diameter was 232mm and the speed was 2m/min. Any cracks generated at the strip edges were removed between each cycle. A total of 3 ARB cycles were carried out on the 1mm strip to give a total true strain of ~8.4.

2.3 Sample preparation

Metallographic as spray formed, extruded and rolled samples were prepared by: (1) hot mounting sections of interest in a PolyFast phenolic resin; (2) grinding using 320 grit SiC paper; (3) polishing using MD-Plan cloths with a 9μm diamond suspension; and (4) polishing using MD-Dur and MD-Chem cloths with colloidal silica + 10% hydrogen peroxide suspension. Thin foils from the extruded 1mm strip of <200nm were machined from bulk using a FEI 200 focused ion beam microscope and placed on a carbon coated Cu grid for transmission electron microscopy (TEM) study. In order to investigate the 3D morphologies of the Ti and Si phases after extrusion and ARB, the Al matrix was dissolved using butanol according to [5], followed by filtering. The Ti and Si phases remained intact on the filter paper and were dried and studied in the SEM.

2.4 Microstructural characterisation

SEM and EBSD analyses were carried out using a field emission gun JEOL 6500 SEM operating at 15keV for secondary or backscattered electron imaging, and at 20keV for EBSD data acquisition with samples tilted at 70° relative to the incident electron beam. EBSD scans were performed in a hexagonal scan pattern with a scan step size optimised for each magnification. An EDX TSL OIM acquisition system and analysis software was used for grain size and texture measurements. A grain was defined as having at least three data points in order to remove inaccuracies associated with the measurement of grains with very few data points [6]. The development of texture was also studied by

X-ray diffraction (XRD) on sections along and transverse to the extrusion direction in a Siemens D5000 diffractometer operating with Cu Kα X-ray of 1.5406Å wavelength.

2.5 Mechanical testing

Extruded strips were machined to an ASTM E8M (sub-size) tensile specimen and tests at ambient temperature were carried out at a cross-head speed of 0.5mm/min, corresponding to a nominal strain rate of 8.3×10^{-4} s^{-1}. The tensile direction was parallel to the extrusion direction.

3. Results and Discussion

3.1 As spray formed microstructure

During co-spray forming, the commercially pure Ti particles were injected into the atomised Al-Si droplet spray through a separate set of gas jets close to the point of atomisation. The Ti powder mixed with Al-Si alloy droplets primarily during atomisation of the Al-Si melt stream and co-deposited into the billet with Ti particles uniformly distributed in the Al-Si matrix, as shown in Figure 1a. The as-sprayed alloy composition was shown by chemical analysis to be close to the nominal composition, together with 0.13Fe.

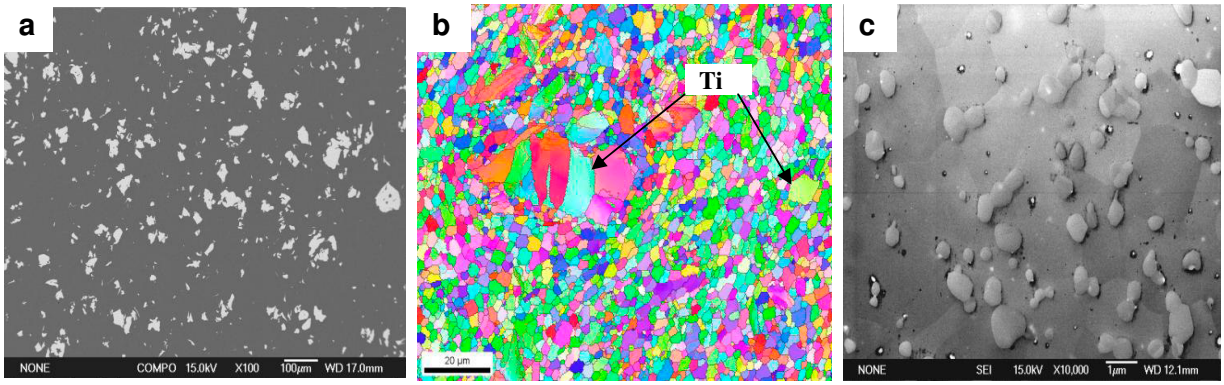

Fig 1: (a) Backscattered electron image of as-spray formed Al-12Si+12Ti showing the morphology of Ti particles and their uniform distribution in the α-Al; (b) an EBSD orientation map showing polycrystalline Ti particles and α-Al grains; and (c) secondary electron image showing the Al grains and divorced eutectic Si particles, primarily at α-Al grain boundaries.

The Ti particles had an irregular shape typical of manufacture by mechanical comminution. There was no obvious clustering or banding as reported in a co-spray formed Al-SiC_p composites [7]. Figures 1b and c show that the Al-Si matrix consisted of equiaxed α-Al grains 2-5µm in diameter and a large number of Si particles of <1µm diameter. The Si particles were often at the α-Al grain boundaries and were formed during a fully divorced eutectic reaction at 577°C, as previously reported in sprayed eutectic [8] and hyper-eutectic Al-Si binary alloys [9], and completely replacing the conventionally cast Al-12Si regular/irregular eutectic lamellar structure [10]. The removal of typically over 50% latent heat prior to deposition of the droplets [11, 12] created a huge number of droplets containing solid Al dendrites that subsequently underwent copious fragmentation and remelting [13] at deposition. The fragmented Al dendrites provided an intrinsic α-Al grain refining effect. Thus the as spray formed microstructure comprised an unusual hierarchy of Si particles of <1µm, a dilute α-Al matrix of grain size of 2-5µm, and Ti particles with D_{50} = 80µm.

3.2 Extruded microstructure

Figure 2a shows the microstructure of an extruded Ø30mm bar with the majority of the Ti particles slightly deformed and elongated along the extrusion direction (ED). Only a small fraction of the Ti particles underwent significant deformation and formed very thin fibres of a few microns

thickness, as highlighted by the arrow in Figure 2a. Figure 2b is an SEM image showing the deformation imposed on the Ti during extrusion through a 1mm die. Figure 2c shows an EBSD orientation map of the 2.5mm strip, with a single elongated polycrystalline Ti particle.

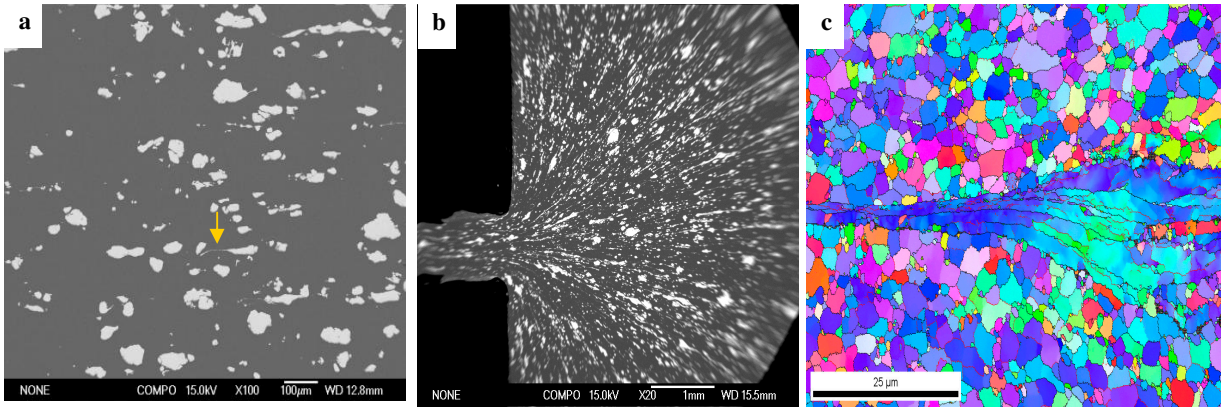

Fig 2: (a) Backscattered electron image of extruded Al-12Si+12Ti Ø30mm bar showing slightly elongated Ti particles; (b) backscattered image showing the extrusion of Al-12Si+12Ti through a rectangular 1mm slot die; and (c) an EBSD orientation map of a 2.5mm strip showing fine α-Al grains and a deformed Ti particle.

Figure 3 shows XRD traces comparing as spray formed Al-12Si+12Ti to the extruded 1mm strip. There was slight <111> texture in the α-Al in the as spray formed condition, due to a billet axial temperature gradient during final solidification [9], and no texture in the Si or Ti. In the 1mm thick strip and consistent with the EBSD data, there was again no strong texture in α-Al, Si or Ti. Although conventional Al alloys often develop a strong <111> or other α-Al texture during extrusion, texture development in spray formed and extruded hypereutectic Al-Si alloys studied in detail by X-ray pole figure analysis has previously been shown to be weak, even after an extrusion reduction of ~70% [14]. We propose particle stimulated nucleation (PSN) of dynamic recrystallization in Al-12Si due to the high number of ~1µm hard Si particles relieved accumulated strain energy and undermined the development of a pronounced sub-grain structure and macro-texture [15]. The α-Al grain size in this study was largely insensitive to the extent of deformation suggesting that PSN of recrystallization in the dilute α-Al by the Si was comparatively easy at all stages of processing.

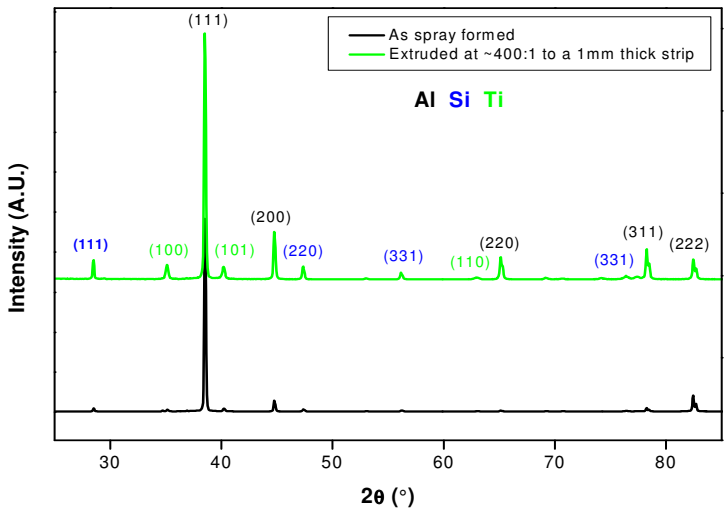

Fig 3: XRD patterns comparing the as-sprayed Al-12Si+12Ti material to the extruded 1mm strip showing that only weak <111> α-Al texture was induced after deformation to a strain of ~6.

Figure 4a shows that for a 1mm strip, over 50% of the Ti particles were deformed into fibres of thickness 0.5-2μm. Figure 4b shows Si particles a few microns apart producing indentations and kinks along a deforming Ti fibre. TEM observations in Figure 4c confirmed that unlike the α-Al that was easily recrystallized, the Ti fibres accumulated a comparatively high dislocation density. There was no discernible Al-Ti interfacial reaction nor interfacial oxide at any of the magnifications used in the TEM studies.

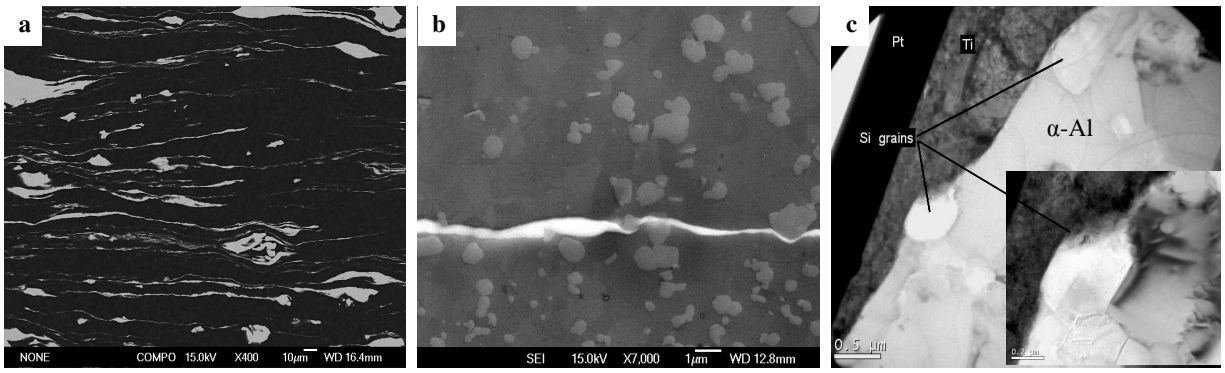

Fig 4: (a) Backscattered electron image of further extruded Al-12Si+12Ti 1mm strip showing heavily elongated Ti; (b) secondary electron image showing the interaction between Si particles and the elongated sub-micron Ti fibre; and (c) TEM image of the α-Al/Ti interface with embedded Si particles.

3.3 Mechanical properties

At a total strain of 6, the yield stress of Al-12Si+12Ti increased by over 30% relative to the extruded 30mm bar with a strain of 0.2, with no loss of elongation to failure of 17%. This was still a relatively low total strain and fraction of Ti in comparison with small batches of powder processed Al-20vol%Ti with strains >10 that showed dramatic strengthening [4]: because the matrix/fibre interface acts as a barrier to dislocation (slip) propagation, strengthening is proposed dependent on the fibre spacing in a Hall-Petch type relationship [16]. Nonetheless, these results provide encouragement for the feasibility of our spray forming based approach to scale-up.

Table 2: Mechanical properties of extruded 30mm diameter bar and 1mm thick strip.

	Strain	Yield Stress (MPa)	Elongation (%)
30mm bar	0.2	96	17
1mm strip	6	129	17

Despite the low Ti fraction, but in order to assess the response of the material to higher strains, strips of 1mm thickness were subjected to 3 ARB cycles resulting in the majority of Ti particles reduced to sub-micron thickness as shown in Figure 5a. There was no discernible fibre breakage, supporting previous ideas that the filamentary phase must be capable of a high recovery rate [1]. Figure 5b shows the Ti fibres after dissolution of the α-Al matrix. Many Ti fibres or ribbons were now over 500μm long with thickness as small as a few hundred nanometers, although some only slightly deformed larger Ti particles remained, deriving from the large spread of Ti sizes in the starting powder.

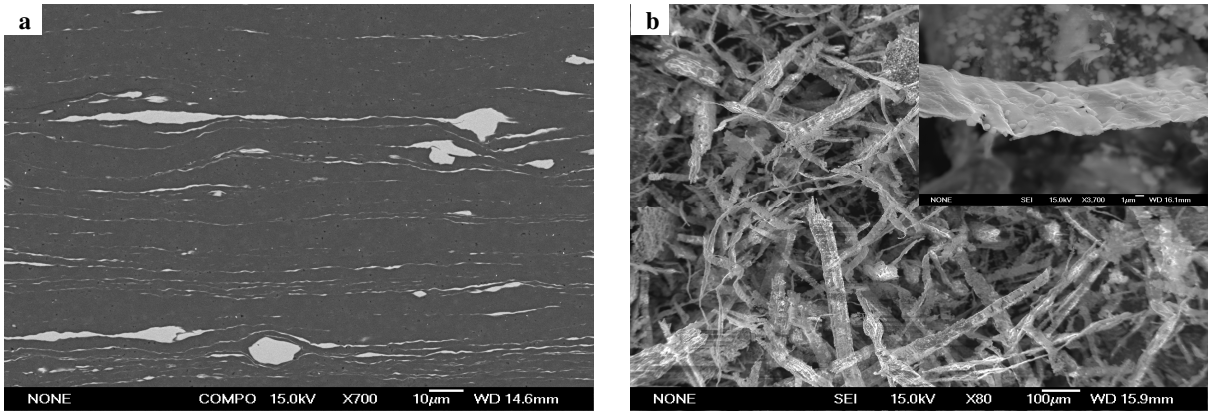

Fig 5: (a) Backscattered electron image of ARB Al-12Si+12Ti showing Ti fibres and ribbons of sub-micron thickness; and (b) SEM image of Ti fibres and ribbons after dissolution of the α-Al matrix.

Conclusions

Spray forming with co-injection has been shown to be an exciting route to the manufacture of metal-metal composites at large scale. The spray formed Al-12Si+12Ti comprised refined equiaxed α-Al grains (~5μm), spherical Si particles (~1μm) and uniformly distributed Ti particles (~80μm), and was free from measurable interfacial reactions or oxides. Deformation processing was carried out to a true strain of ~8.4 providing elongated Ti fibres of sub-micron thickness and α-Al grains that underwent particle stimulated dynamic recrystallization due to the Si particles to maintain a fine, near-constant grain size irrespective of total strain. It is suggested that during deformation, the Si particles also played a role in imparting stress to the Ti particles to produce fibres of sub-micron thickness. Preliminary tensile properties suggest that progressive Ti elongation provided modest strengthening. Larger Ti fractions and deformations are now being studied, with accumulative roll proving to be an encouraging deformation processing route.

Acknowledgements

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