2D SEM and 3D XCT investigation of in-situ Al-Cu-TiB_2 semi-solid forged composites

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Abstract

The present work compares the 2D and 3D distribution of TiB_2 particles in a semisolid processed Al-4.5%Cu-5wt.% TiB_2 in-situ composite prepared by flux assisted synthesis. The composite was synthesized by the reaction of K_2TiF_6 and KBF_4 salts in molten Al-4.5Cu alloy held at 800 ^oC for an hour. The extent of distribution of TiB_2 particles was investigated using Field Emission Scanning Electron Microscopy (FESEM) and X-ray computed tomography (XCT) to obtain 2Dand 3D images respectively. The studies indicated improved distribution of TiB_2 particles after semi-solid forging of composites (at 0.1 volume fraction of liquid and 50% reduction) as compared to as cast composites. The hardness of the semisolid forged composites showed a significant increase and is uniform in all directions. The increase in hardness could be attributed to particle fragmentation and its redistribution in the matrix. Further investigation will be needed to understand the mechanism of redistribution and investigate the mechanical properties of such composites in detail. Copyright © 2016 VBRI Press.

Keywords: X-ray computed tomography, semisolid forging, TiB_2 particles, metal matrix composite, clusters.

Introduction

Metal matrix composites (MMCs) have received attention owing to its application in automobile, military and aerospace sector [**1**]. MMCs combine the metallic properties with ceramic properties leading to significantly enhanced properties like high toughness and high strength modulus [**2**]. Aluminium-copper alloy is one of the commonly used age hardenable alloy due to its low density, low melting point, high specific strength and thermal conductivity [**3**]. The most commonly used reinforcing particles with aluminium matrix are SiC and TiB_2. TiB_2 is more inert as compared to aluminium matrix and thus do not form any brittle intermetallics at the interface. Further, TiB_2 has excellent properties such as high hardness (25-35 GPa), high melting point (3225°C), high elastic modulus (560 GPa), good thermal conductivity (60-120 W/m/K) and corrosion resistance which makes TiB_2 an ideal choice as a reinforcement in aluminium alloys [**4,5**]. Additionally, TiB_2 is a good heterogeneous nucleating agent for primary aluminium which helps in grain refining of the alloy [**6**]. Therefore, Al-Cu-TiB_2 composites are considered to be promising metal matrix composites as it results in improvement of properties by combining both the properties of aluminium-copper alloy and TiB_2 particles.

One of the major concerns in the preparation of MMCs is developing a processing method which can produce a minimal porosity with fine and uniform microstructure for improvement of properties. In the recent past quite a few studies have been reported on semisolid processing of MMCs [**7-14**] of which semisolid forging is found to be a promising method [**9-12**]. Semisolid forging involves forging a partially melted non-dendritic alloy slug in an open die to produce components near-to-their final shape. Some of the attributes are good surface finish, minimal porosity and fine and uniform microstructures which results in superior mechanical properties [**9-15**].This improvement in properties greatly depends on the behaviour of semisolid slurries in which the solid exists in the form of spheroidal particles and when sheared, flow like liquids[**15,16**].

A thorough literature review shows that although a lot of work has been done in the area of Al-Cu-TiB_2 in-situ composites, there is not much work with the semisolid forging process. In a study on Al–Cu–TiB_2 in situ composites synthesized by the reaction of molten alloy with halide salts, reported that addition of 5 wt.% TiB_2 resulted in high strength and ductility [**17**]. The semisolid state rolling is found to be helpful for refinement in the grain structure with improvement in average bulk hardness of in situ Al–4.5Cu-5wt.% TiB_2 composite. Semisolid state rolling also helps in uniform distribution of TiB_2 particles and $CuAl_2$ precipiatets in the Al–4.5Cu-$5TiB_2$ composite [**7,8,18**].The major limitation in the microstructural studies of all these works have been that the microstructural changes occurring on semisolid processing was done by 2D sectioning method. X-ray computed tomography overcomes this limitation by employing a high intensity X-ray beam to penetrate through a small sample which is recorded by a high speed digital camera and by reconstructing the 3D model of the microstructure [**15**]. In an X-ray computed tomography (XCT) study of Al–4.5Cu-$5TiB_2$ composite with 0.3 volume fraction of liquid (V_{fl}), the porosity was found to be reduced on thixoforging [**19**].

In the present work, Al-4.5Cu-$5TiB_2$ composite is prepared in-situ by Flux Assisted Synthesis (FAS) technique and forged in a semisolid temperature range. The Al-4.5Cu-$5TiB_2$ composite was semisolid forged at 0.1 volume fraction of liquid (V_{fl}). Further, the TiB_2 particle size distribution was investigated using both 2D Scanning Electron Microscopy (SEM) and 3D X-ray computed tomography(XCT) techniques. Also, the hardness of as cast composites and semisolid forged composites were compared with the microstructures.

Experimental

Preparation of Al-4.5 wt% Cu-5 wt% TiB_2 in-situ composite

Molten Al-4.5Cu alloy was prepared by mixing commercially pure aluminium with 99.7% purity (Hindalco) and copper with 99.9% purity (Hindalco) at 800 °C. The Al-4.5Cu-$5TiB_2$ composite was synthesized by Flux Assisted Synthesis (FAS) technique which consists of addition of halide salts, K_2TiF_6 and KBF_4(Madras aluminium co.), to molten Al-4.5Cu alloy at 800 °C and a reaction time of one hour. The exothermic reaction within the melt results in formation of titanium-diboride (TiB_2) particles. The melt was stirred intermittently (every 10 minutes) to ensure complete reaction and homogenous distribution of TiB_2 particles. After a reaction time of one hour, the lighter dross was decanted and the melt was degassed using C_2Cl_6. The composite melt was finally poured into rectangular mild steel mould. The casting was later machined to produce samples with dimension of 10 x 10 x 60 mm^3.

Semisolid processing of Al-4.5 wt% Cu-5 wt% TiB_2 in-situ composite

The sample for semisolid forging was soaked at 623°C corresponding to its 0.1 liquid fraction (hereafter referred as $0.1V_{fl}$) for 10 minutes prior to forging. The semisolid forging was carried out using 80 ton hydraulic press, at 150 Kg/cm^2 load with a ram speed of 20 mm/sec. The specimen was subjected to forging up to 50% deformation in the semisolid state at 623^oC corresponding to $0.1V_{fl}$. The K-type thermocouples were used to monitor the temperature with an accuracy of ± 2 °C.

Mechanical testing- Hardness measurement

The hardness of the as cast *in-situ* Al-4.5Cu-5wt%TiB_2 composite and semisolid forged *in-situ* Al-4.5Cu-5wt%TiB_2composite were measured using a Vickers hardness tester (Zwick Roell ZHV) at a load of 5 Kgf. The hardness was measured at ten different locations in all three x, y, and z directions after fine polishing and is indicated in the micrographs along three directions.

Microstructural characterization

Scanning electron microscopy

Microstructures of the samples in all three directions were investigated individually after polishing using standard metallographic procedures. The samples were etched with Keller's Reagent (5 ml HNO_3+ 3 ml HCl + 2 ml HF in 190 ml distilled water). Electron backscattered images were used for the investigations. Micrographs were captured using a field emission scanning electron microscope (FESEM) using Zeiss, Carl Zeiss SMT AG instrument coupled with energy dispersive X-ray spectrometer (EDS). The individual micrographs were then reconstructed into a 3D model for analysis.

X ray computed tomography (XCT)

X-ray computed tomography was carried out using Zeiss versa machine at WMG, University of Warwick, UK. Cylindrical samples of 1 mm diameter were prepared for XCT to allow sufficient sample penetration at the resolution required. The sample was loaded into the scanner with parameters shown in **Table 1**. To achieve the sub-micron resolution a 20x lens optic at the detector was used. Since the as cast sample contains significantly larger features with agglomeration of TiB_2 particles, a lower resolution was employed by binning pixels. The advantage of binning pixels is the increased amount of flux received by the detector that leads to smaller scan times. The detector consists of 2000 x 2000 pixels, which given binning, results in a 380 µm and 820 µm field of view for semisolid forged composite and as cast samples respectively. The images were reconstructed using the Zeiss reconstruction software that employs a filtered back projection algorithm **[20]**. The resultant volumes were then segmented and analysed in Avizo 9.0 (FEI, USA; http://www.fei.com/software/avizo3d). Initially the 'normalise greyscale' function was employed across the volumes such that the greyscale value of the matrix and particle phases were similar. The volume was segmented into different phases using a watershed based algorithm as described in seminal work by Digabel and Lantuejoul [**21**]. In this algorithm the volume is progressively filled from initial markers according to rate of change of voxel greyscale values.

Table 1. X-ray tomography scanning parameters.

Parameter	As cast	0.1 V_{fl}, 50% reduction
Voltage (kV)	110	110
Current (µA)	79	79
Exposure (s)	15	32
Filter (Quartz, mm)	1	1
Number of Projections	3201	3201
Pixel binning	X2	X1
Voxel size (nm)	820	190

In this manner it can be thought of the filling of a topographical map where the height corresponds to a greyscale value; if the greyscale gradient becomes high then the filling propagates slower, and where it is too sharp it will be selected by a different marker. Initial greyscale values for the matrix and particle phase were 20000-25000 and >30000 respectively. With the segmentation, complete analysis of the selections were performed to determine volume fractions, particle diameter and particle volume. As is known, a direct evaluation of error in segmentation of phases is difficult to establish and is dependent on the scan quality, specifically the sharpness of material boundaries. To attempt to quantify such error bounds, the watershed was repeated with ±1000 grey values which resulted in variations less than 0.02% of the total volume.

Results and discussion

The FESEM micrographs in back scattered mode was taken to distinguish TiB_2 particles from aluminium matrix. **Fig. 1(a)** shows the as-cast microstructure of Al-4.5Cu-5TiB_2 composite. Clustered network of TiB_2 particles (white phase) of varying size can be seen randomly distributed in the matrix in all three directions. It is evident from the images that microstructure is uniform and isotropic in all three x, y, and z directions. Forging reduction of Al-4.5Cu-5TiB_2 composite in semisolid state resulted in significant fragmentation of TiB_2 particle clusters and redistribution of TiB_2 particles as shown in **Fig. 1(b)**. Microstructure of composite in the direction of forging is different from the microstructure in the direction normal to forging. The α-aluminium grains are elongated and TiB_2 particles were found to be aligned along the direction normal to forging. Further, large clusters of TiB_2 particles were also present, while a major fraction of clusters were well dispersed in the microstructure. The fragmented TiB_2 particles were redistributed by the viscous drag of 0.1 volume fraction of intergranular liquid when subjected to compression during forging.

The hardness values as marked in **Fig. 1(a, b)** shows that the hardness of in-situ Al-4.5Cu-5TiB2 composite is increased by 23% after semisolid forging. The increase can be attributed to a better distribution of TiB2 particles, reduction in cluster size of TiB2 and closure of pores that were present in the as cast composite leads to increased density after forging **[22-25]**.

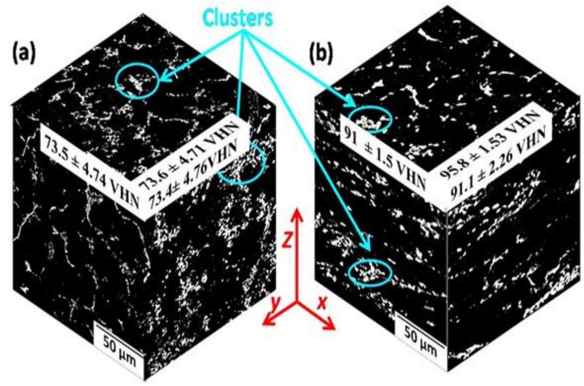

Fig. 1. FESEM micrographs of in-situ Al-4.5Cu-5TiB_2 composite in x, y, and z directions (a) as cast condition (b) 0.1 volume fraction of liquid, V_{fl} and 50% reduction (z is the direction of forging). The white labels on the micrographs represent Vickers hardness values.

The hardness of semisolid forged in-situ Al-4.5Cu-5TiB_2 composite **Fig. 1(b)** is found to be ~5% higher in forging direction z than the normal x and y directions. As observed in **Fig. 1(b)** the grain flow is normal to the forging direction. The dislocations undergo numerous deflections as it moves across the sample due to the resistance provided by the particles aligned along these grain boundaries. Each of these deflections requires more energy and makes the material more resistant to deformation. Hence, hardness is found to be higher in the forging direction.

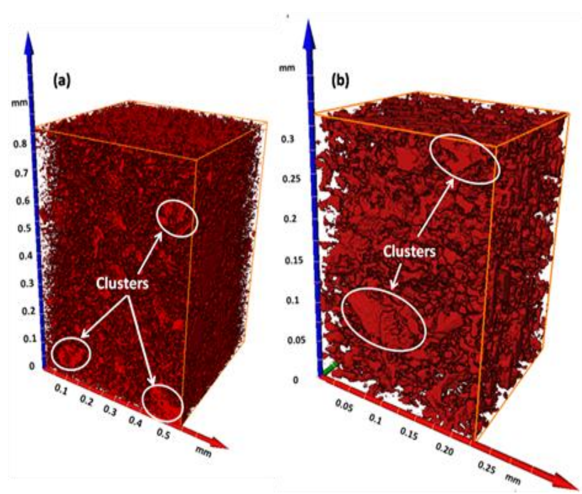

Fig. 2. 3-D XCT showing TiB_2 particle clusters in Al-4.5Cu-5TiB_2 composite (a) as cast condition (b) after deformation in semisolid state.

Fig. 2 shows the 3D reconstructed XCT images of TiB_2 particles in Al-Cu-TiB_2 composites. The TiB_2 particles in as cast condition **(Fig. 2(a))** is found to be more clustered or agglomerated than after semisolid forging **(Fig 2(b))**. A cluster is a collection of particles separated by a distance of less than the order of the particle equivalent diameter (i.e. < 2 µm) [**26**]. The magnification used for semisolid forged condition is twice than that of as cast condition since fragmented TiB_2 particles in forged condition is much smaller than the clustered form in as cast condition.

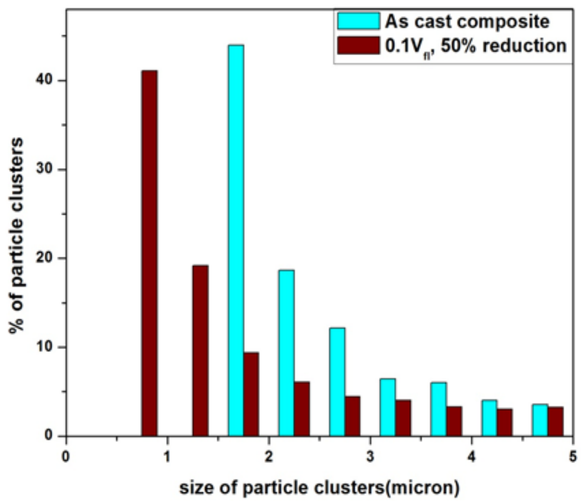

Fig. 3. Size distribution of TiB_2 particle clusters obtained from XCT of Al-4.5Cu-5TiB_2 composites.

The size distribution of TiB_2 particle clusters obtained from XCT data **(Fig. 3)** shows that 60% of TiB_2 particle clusters are in the size range of 0.5-1.5 micron after semisolid forging, whereas the TiB_2 particle clusters in as cast condition is greater than 1.5 microns. The size of 70% particle clusters in semisolid forged condition and 45% particle clusters in as cast condition is less than the equivalent diameter of TiB_2 particles (< 2µm). Therefore, these particle clusters can be considered as individual TiB_2 particle rather than clusters. The 25% more TiB_2 particle clusters (> 2 µm) in as cast condition compared to semisolid forged condition implying that semisolid forging has resulted in significant fragmentation of the TiB_2 particle clusters.

Quantified results of TiB_2 particle size distribution shows the fragmentation of TiB_2 particles on forging and its redistribution by the viscous drag of the intergranular liquid. This could be the possible reason for the enhancement of the hardness on semisolid processed sample compared to as cast in-situ Al-4.5Cu-5TiB_2 composite.

This study demonstrates the effectiveness of semisolid forging process for producing fine-grained castings with uniform distribution of reinforcement particles and improvement in mechanical properties. It is evident that the semisolid forging has the dramatic effect of enhancing the particle size distribution compared to the in-situ castings; however, the particle size distribution can be further improved by increasing the volume fraction of liquid during forging process or by changing the amount of deformation.

Conclusion

The preliminary 3-D XCT investigation of semisolid forged Al-4.5Cu-5TiB_2 composite in the semisolid state (containing a liquid fraction of 0.1) shows that the forging in semisolid state improves the distribution of TiB_2 particles in the matrix. A significant fragmentation of TiB_2 clusters followed by its distribution led to an increase in hardness by ~23% on semisolid forging. While SEM studies show that the distribution of TiB_2 clusters is uniform in the matrix, 3-D XCT studies shows the extent of particle clustering in the composite. Hence, it is can be concluded that 3-D XCT can be used as an effective tool to optimize the process parameters to obtain a microstructure devoid of TiB_2 clustering.

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Author's contributions

Conceived the plan: mc, am, ps; Performed the expeirments:jw, jm; Data analysis: mw, jm; Wrote the paper: jm, am, ps. Authors have no competing financial interests.

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