A REVIEW ON THE FABRICATION TECHNIQUES OF ALUMINIUM MATRIX NANOCOMPOSITES

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Abstract – *In recent year, metal matrix composites have been considered as materials that offer better mechanical properties compared to conventional alloy. Recently, the developments of metal matrix nanocomposites (MMNCs) have become more attractive in various applications. However, the synthesis of MMNCs by conventional casting method has shown a limitation due to low wettability of the reinforcement phase by the molten metal. This paper is aimed at reviewing the best result techniques to fabricate the aluminium matrix nanocomposite (AlMNCs). However, each of these techniques has their own advantages and disadvantages. This review conclude that the powder metallurgy (PM) as the best technique certainly for mass production and cost effectiveness.*

Keywords: Aluminium matrix nanocomposites; Metal matrix nanocomposites; Fabrication techniques; Powder metallurgy

1.0 INTRODUCTION

Aluminium (Al) matrix composites exhibit higher mechanical properties than unreinforced Al alloys and had been widely studied since 1920s. Due to their lightweight, high Young's modulus, high specific strength and high wear resistance, it had been used for wide range of application such as sporting goods, electronic packaging, armours, nuclear, biotechnology, aerospace, marine, automotive and transport industries [1-4]. Furthermore, the advantages of Al and its alloys over others composite's matrix are high specific strength and stiffness, good damping capacities, dimensional stability and good machinability. To strengthen the metal matrix, it is of interest to use nano-sized ceramic particles, while maintaining good ductility, high temperature creep resistance and better fatigue [5]. Recently, metal matrix nanocomposites (MMNCs) have become more attractive in various applications because of their better mechanical properties over conventional microparticle reinforced. Nanocomposites are composite material that has at least one of its dimensions in the size of less than 100 nm [6]. Low wettability of ceramic nanoparticles with the molten metal matrix is the main issue to fabricate the MMNCs which do not allow the production of MMNCs by usual casting processes. In fact, small powder aggregates are prone to produce cluster which they can lose the ability to be dispersed homogeneously in the matrix for an optimal strengthening potential [7]. In order to overcome this issue, several alternative techniques have been proposed.

2.0 ALUMINIUM (Al) ALLOY AND AVAILABLE REINFORCEMENT

Various species of nano-sized oxides (Al_2O_3, ZrO_2, MgO, $FeTiO_3$), and carbides (SiC, B_4C) have been employed as reinforcement agents which due to low density and melting point, very good specific strength and thermal conductivity of Al [7-8]. Carbon nano-materials, such as fullerenes, carbon nanotubes, and graphenes have been well attended as reinforcements for the Al matrix composites, due to their lightweight nature and superior strength/stiffness stemming from strong sp2 C—C bonds [9]. Recent work in the field of carbon was developed by the discovery of carbon nanotubes (CNTs) by Iijima in 1991 although CNTs might have been synthesised in 1960 by Bacon. A multi-walled carbon nanotube (MWCNT) is fabricated of many single-walled carbon nanotubes (SWCNT) structured in a concentric manner [10]. Based on size and shape as well as their excellent in mechanical properties such high elastic modulus, tensile strength and aspect ratio, SWCNTs and MWCNT are found as a new kind of reinforcement material for the production of novel MMC in the scientific community [11][12]. Silicon carbide nanowires (SiCNWs) have been attracting considerable attention as a novel type of reinforcement filler due to their outstanding properties such as high thermal stability, high thermal conductivity, superior mechanical properties and chemical inertness. However, the improvement in materials' properties by using SiC NWs as fillers in metal matrix composites has not been thoroughly studied yet [13].

3.0 PROCESSING TECHNIQUES OF ALUMINIUM MATRIX NANOCOMPOSITES

The processing technique to fabricate aluminium matrix nanocomposites (AlMNCs) can be classified into three main groups:

i. solid state processes include high energy ball mill and powder metallurgy (PM) techniques with modifications in the processing step such as, hot isostatic pressing, cold pressing, extrusion followed by a sintering treatment [7];
ii. liquid-state (casting) processes include stir casting, ultrasonic-assisted casting; and
iii. semi-solid processing include combination of rheocasting and squeeze casting, semi-solid route stir casting

The restrictions of the second and third groups occur from difficulties in combining the two phases thoroughly, hard determination of critical temperature for infiltration, issues due to fluidity and/or wettability at matrix-reinforcement interface, in addition to harmful reactions at the interface [8].

3.1 Solid state process

3.1.1 High energy ball milling

Several solid production methods were developed to fabricate MMNCs. In particular, different PM techniques were successfully performed in this field. Some papers discussed on high-energy ball milling known as mechanical alloying (MA) which a powder metallurgy technique is comprising in repeated cold welding, fracturing and re-welding of particles in a high energy ball mill. This practice is of primary importance since it allows achieving a better distribution of nano-powder into the composite by breaking up the ceramic clusters [7]. It is

important to note that obtained mechanical properties of mechanically alloyed products strongly depend on the consolidation parameters and milling time. Although there are different methods to compact powders, hot pressing produces net-shape and cost-effective procedure with controlled microstructure and mechanical properties [8].

Al powder (99.8% pure) and Al_2O_3 powders (99.99% pure, 50 and 150nm sizes) were combined under argon atmosphere inside a glove box to reduce any contamination. High-energy milling of powders was conducted to produce the composite powders which uphold a ball to- powder weight ratio of 10:1. About 0.5–1.0wt.% of stearic acid which used as stearic acid was applied as a process control agent (PCA). Testing of the mechanically milled powders established uniform distribution of the reinforcement phase. This has been performed for volume contents up to 50% and particle sizes up to 50 nm [14].

3.1.2. Consolidation followed by Sintering Route

The synthesis of two nanocomposite systems of (Al-20wt.%Al_2O_3) and (Al-10wt.% Al_2O_3 - 10wt.% ZrO_2) was conducted by grinding under argon gas atmosphere at 300 rpm milling rotation speed and ball to powder ratio of 10:1. Stearic acid (PCA) was used to prevent the aggregation of powders during milling. In order to achieve high dense samples, consolidation of the milled particulates was performed in two stages including pressing and sintering of the compacts. Pressing the powders was done with the applied pressure was 600MPa and 5min of the load hold time. For sintering, the compressed powders were kept at 600°C for 45min and nitrogen gas was applied to avoid oxidation of aluminium. As result, the density of the green compacted powders is lower than the density of sintered compacted powders which may be attributed to reduction of pore space during sintering process and/or due to grain refining. Sintering process play an important role in obtaining higher density materials. Furthermore, the microhardness increased after the addition of reinforcement particles and milling which can be referred to some reasons. Increasing the high energy ball milling duration will achieve higher deformation and increases work hardening of powders. Furthermore, the addition of hard reinforcing particles might improve the work hardening rate of the matrix resulting in increase in hardness [15].

SiC NWs and pure Al metal powder were mixed in different wire/Al ratio varying from 5% to 15% by volume. In order to get a homogenous mixture, the SiC NWs were ultrasonically diffused in ethanol for 10min before vibration-mixing with the Al powders for 30min. The mixture was dried at 120°C and sieved into a fine powder. The prepared composite powders were hot pressed at 600°C under pressure of 40 MPa for 1hr in argon gas. It is indicated that the density of the composite specimens is higher than that of the monolithic SiC NWs and monolithic Al means SiC NWs were well distributed within the Al matrix. Due to the addition of the SiC NWs, the wear performance of metal based material is significantly enhanced as composites with 15% vol of SiC NWs show 76.95% decrease in wear rate compared to pure Al [13].

Many of the studies on Al-CNT have been performed using the PM method. A study has accounted a 350% enhance in the compressive yield strength with 1.6vol% CNT addition, which, owing to a homogeneous distribution of CNTs achieved by the nanoscale dispersion method. The homogeneous distribution and good interfacial bonding of CNTs increasing them directly on Al powder through the chemical vapour deposition (CVD) method before compacting and sintering. With addition of 6.5vol.-% CNT, they also have achieved increase in hardness tensile strength. Aluminium powder (99.9% pure) and MWCNTs (formed by the

spray pyrolysis method) were mixedin an ultrasonic bath for 5min and milled in a high-energy shaker mill and the milling media to powder weight ratio was 5:1. There is no addition of process control agent and argon gas was applied while all milling runs were performed. Consolidated products were done with the applied pressure was 950MPa and 2min of holding time. Compacted specimens were pressure-less sintered during 3hrs at 823K inside vacuum (2 Torr). As a result, the relative densities of the nanocomposites were 1–5% lower than the theoretical values. Furthermore, the σ_{max} increased as the milling time and MWCNT concentration increased. The nanocomposite prepared with 2hrs of milling time and 0.75wt.% MWCNT reaching the maximum value which around 77Hv compared with the shorter milling time (1hr) which the most significant strengthening effect acting on the nanocomposites is the effect of the nanocrystalline state [9].

Al (particle size smaller than 63μm) and Al_2O_3 (α-alumina powder with 99.5% pure, average size of about 27-43nm) nanocomposite have been undergo high energy planetary ball mill and then were fabricated under 420MPa pressure and holding time was 5min. For sintering, the compressed powders were kept at 624-626ºC for 45min under argon gas atmosphere and then the samples were furnace cooled. This studied revealed that the agglomerations of particles would be removed by increasing milling time. At 4hrs milling time, alumina agglomerations were observed yet, but they are smaller and less than no milled powders. However, a uniform distribution of the reinforcement particles can be caused by increasing the milling time, dissolution of particles agglomerations and reduction of distances between them. It can be noted that, this milling time particles have been under deformation and cold welding therefore, flattened particles with high aspect ratio were formed. This is because Al particles are soft and their sizes are increased by cold welding. High sintering temperature (625ºC) and difference in thermal expansion coefficient of Al and Al_2O_3 produce thermal stress. The stress will be disappeared by dislocations production and cause increasing in dislocations density means, important for strength enhancement. Agglomeration of the alumina particles cause stress concentration and unexpected fracture thus, strength is increased by uniform distribution of the alumina particles [16].

Recent study, pure Al powder (minimum of 99.7% pure, average size between 10 and 100μm and reforcing agent of SiC (average size of 200nm) and Al_2O_3 (average size of 60nm) ceramic nanoparticulates were used. Several Al-based nanocomposites containing up to 5vol.% of SiC and Al_2O_3 nanoparticulates were prepared using conventional powder metallurgy (P/M) route. Both Al powder and nanoparticulates in addition to 0.5-1.5wt.% paraffin lubricant wax were placed mechanically mixed until a homogeneous mixture is achieved. The powders were cold compaction under 500MPa pressure. The compacted powders were sintered under argon inert gas atmosphereat at 600°C for 100min. After sintering, the nanocomposites were subjected to hot extrusion at 500°C using the extrusion die. As result, when the volume fraction of the nanoparticulates dispersed into the Al matrix is increased, the agglomeration percent of the nanoparticulates also tends to increase which found to be concentrated on the grain boundaries of Al grains. Furthermore, it has been found that the Al/Al_2O_3 nanocomposites exhibited better nanoparticulates distribution than Al/SiC nanocomposites. The Al/SiC nanocomposites exhibited more agglomeration percent and the agglomerations size was found to vary between 0.5 and 10μm in size. Although small agglomerates in Al/SiC and Al/Al_2O_3 nanocomposites still existed in the matrix, the agglomerates have been greatly improved when compared with the severe agglomerates in nanocomposites fabricated using traditional mechanical stirring method [17].

Other study conducted on CNT which was dispersed in a solvent and sonicated for 20min and it is been evaporated by heating the solvent. Al and the reinforcement of CNT, GR were blended in ball milling furnace for 10min at 200rpm which essential for uniformity of the product. Samples were compacted under load of 135KN for 2min. For sintering, the green compacts were heated up from room temperature to 660.32°C 45min under nitrogen atmosphere to avoid the surface contamination. Shrinkage of AL+CNT+GR and AL+CNT+GR+ND was accurse during sintering as a result of pore size reduction. The density values of AL+CNT+GR+ND composition are very near to the density value of base metal while the density values of AL+CNT+GR composition are less compared to the density values of AL+CNT+GR+ND composition. For AL+CNT+GR+ND composition, the hardness values are high compared to AL+CNT+GR composition. The wear resistance increases with the amount of reinforcements as AL+CNT+GR and AL+CNT+GR+ND composition wear resistance high for 2% [12].

3.1.3. Cold Isostatic Pressing (CIP)

Al powder (99.98% pure, 25μm), Si powder (98.5% pure, 25μm) and two types of nanoparticles of γ-Al_2O_3 (99.98% pure, 50nm) and rutile-TiO_2 (99.8% pure, 30nm) were prepared. Dry mixing method was used to mix the powder of alloy (Al-12%Si) with both Al_2O_3 and TiO_2 or with single Al_2O_3 or TiO_2. The mixing time was for 4hrs at milling speed of 650rpm to get good particles distribution. Cold compaction was carried out at 10Mpa pressure followed by sintering process at 520ºC for 90min with argon flow rate 2L/min. As result, the sample with 4wt% Al_2O_3 shown the highest hardness because of nano Al_2O_3 has higher hardness compared to TiO_2. It is proved that mechanical milling produces uniform dispersion of the reinforcement nanoparticles in the Al-12wt%Si matrix. The nanocomposites reinforced with single addition of 4wt% Al_2O_3 nanoparticles shown the highest hardness and higher wear resistance compared with base alloy and other nanocomposites [6].

3.1.4. Hot Isostatic Pressing (HIP)

Al–graphene composite powders were fabricated by consolidation process via hot isostatic pressing (HIP). HIP was done at 375°C for 20 min. Samples of pure Al, 0.1wt% graphene and 1.0wt% MWNT was made. Due the greater interfacial contact area, the dispersion of graphene is complicated compared to CNTs thus low weight fraction of graphene was selected. After HIP, the billets were preheated to 550°C for 4hrs and then extruded on a 50tonne extrusion press. The presences of MWCNTs increase the tensile strength of Al by up to 12%. However, during processing, graphene was tending to form aluminium carbide leads to decrease the hardness and tensile strength of Al. The formation of the aluminium carbide is because of the nature of grapheme that produced by thermal exfoliation/reduction of graphite [18].

3.1.5. Extrusion prosses

Al alloy 6061/SiC nanocomposites were produced by powder metallurgy using ball milling process followed by secondary forming processes namely extrusion. All the billets were extruded at 37°C and the extrusion ratio (ER) was 6:1 which produced 1.8 of a true strain. Other studied have analysed on the microstructure of the effect of the extrusion temperature of Al alloy matrix composites conclude that the increasing values of the extrusion

temperature up to a certain point, the particles are much more uniformly distributed. If the temperature is not high enough, a homogeneous distribution of the particles will not occurs as it failed to assist the flow of the matrix alloy under the applied stress resulting in. Furthermore, a partial melting of the matrix alloy at the grain boundaries happens if extrusions are carried out at a relatively high temperature that causing the particles to get suspended in the melt matrix near the grain boundaries. It has been reported that, the number of pores is decreased effectively and interfacial bonding strength between matrix and reinforcement particles is improved by the extrusion method [4].

3.2. Liquid-state prosesses

3.2.1. Stir Casting Method

Al alloy A356 reinforced with MgO nanoparticles have been fabricated via stir casting method. Density measurements of fabricated samples revealed that the bulk density of samples increases with increasing the vol.% of MgO up to 2.5% at 800, 850 and 950°C. However, the density for cast sample at 800 and 950°C were decreased due to the pores formation and agglomeration at high content of reinforcement. In fact, the incremental trend was dominant at 850°C for the bulk density. The values obtained for bulk densities of the nanocomposites are remarkably close to the corresponding theoretical density values. The uniform presence of MgO in Al matrix with no signs of formation of other intermetallic phases has been approved by XRD phase analysis. Furthermore, the reinforcement particles that uniformly distributed in the matrix alloy, in spite of regional agglomerations [19].

The stir casting technique is performed to fabricate Al (99.7% pure) and nano sized Ilmenite ($FeTiO_3$) particulate composites. As result, the Ilmenite particles are even distribution through some of the clusters were formed nevertheless good distribution of particles was achieved. As the reinforcement is increased, the tensile and hardness values were also increased. It can be observed that 5wt% nano Ilmenite reinforcement exhibit the maximum tensile strength and hardness values and it can be conclude that the mechanical properties, both hardness and tensile strength has been increased compared to the cast Al as maintained ductility nature of matrix material [20].

A studied has been conducted on Al alloy reinforced with nano-ZrO_2 particulates were dispersed by Disintergrated Melt Deposition (DMD) technique followed by hot extrusion. Al alloy was heating up to 720°C and the preheated (200°C) reinforcement was added. To get the uniform distribution of the reinforcement, Al alloy and preheated reinforcement was well stirred at 450rpm by crate the vortex. In secondary processing, extrusion of the deposited monolithic and nanosized ZrO_2 containing Al matrix was conducted in a hydraulic press at 250°C. Microstructural analysis showed fairly uniform distribution of reinforcement in a fine grain refinement with least porosity formation. As reinforcement content is increased up to 12% revealed improve in mechanical properties as well as fracture toughness since the increasing trend in matrix hardness. A higher constraint to the localized deformation during indentation due to presence of ZrO_2 and reduced grain size [21].

3.2.2. Ultrasonic cavation method

Boron carbide, B_4C (size 50nm) particulates are used as its density is close to the Al, and thus the particulate will not tend to agglomerate during the process. AA2024 Al alloy was heating

up to 638°C as well as sonication process. B_4C particles are added at various weight percentages to the melt during the cavitation process in argon atmosphere during melting. The metal is poured into a die and plates are cast. The as cast plates are heat treated (solutionised at 560°C for 1hr) and ageing is done at 160°C for 12hrs. The heat treated plates were undergone for rolling process to remove the casting defects as well for grain elongations. The hardness, ultimate tensile strength and yield strength of the nanocomposites were improved significantly by the presence of B_4Cp particles which increased by 14%, 6%, 11%, respectively [22].

3.3. Semi-solid processes

For this process, only few works are available include combination of rheocasting followed by squeeze casting and the other is semi-solid route stir casting, even if this method has been applied widely in fabrication of the conventional method.

3.3.1. Combination of rheocasting and squeeze casting method

The A356/Al_2O_3 nanoparticles were prepared using a rheocasting technique followed by squeeze casting techniques. A356 alloy was melted at 680°C was allowed to cool to the semi-solid temperature of 602°C as the liquid/solid fraction was about 0.7 and the stirring was started at approximately 1000rpm. Before stirring, Al_2O_3 nanoparticles was preheated up to 400°C for 2hrs and were added inside stirring which had formed the vortex. After that, during the agitation, preheated Al_2O_3 were mixed into the matrix. The agitation was stopped after completing the addition of Al_2O_3 nanoparticles. Then, the molten mixture was poured into preheated tool steel mould and immediately squeezed during solidification using a hydraulic press of 50tonne capacity for 5min. The nanocomposites then were heat treated at 540°C for 3hrs and then quenched into cold water. After cooling, nanocomposite samples were artificially aged at 160°C for 12hrs. As result, the A356/Al_2O_3 containing up to 3% of 60nm Al_2O_3 nanoparticles showed better thermal conductivities than A356/Al_2O_3 contain of 200nm nanoparticles. Compared with the A356 base alloy, the A356/Al_2O_3 showed lower electrical conductivities as increasing the volume fraction and the size of the Al_2O_3 nanoparticles from 60 to 200nm [23].

3.3.2 Semi-solid route stir casting method

Sample of purified MWCNTs is mixed with pure Al powder (avg. size 100μm) with a weight ratio of 1:6 respectively and stearic acid is then added as PCA. The MWCNTs are dispersed with the Al powder by ball milling at 200rpm for one hour and the mixture is then compacted at 90MPa for 10min. In order to improve the wettability of the matrix alloy, a 0.5% weight fraction of pure magnesium which serves as a surfactant is added to the melt. Stirring process is performed at 500rpm for 1min in semi-solid state (590-600°C) after the preinfiltration of the preform by the molten metal for 30sec. Argon gas is purged to prevent hydrogen entrapment during melt processing. The tensile strength increases from 155.4MPa for the monolithic alloy to an average value of 201.2MPa for 1.5wt% MWCNTs reinforced composite which the maximum value is 208MPa. The increased of the tensile strength due to increasing of the dislocation density in the matrix around the MWCNTs resulting of thermal expansion mismatch between the MWCNTs and the matrix [24].

A356 Al alloy was melted at 660°C then was allowed to cool to the semisolid temperature of 601°C and a 0.75% weight fraction pure Mg of is added to the melt in order to improve the wettability. Al/MWCNTs blocks or billets introduced into the melt and stirring was continued for 1 min in semi-solid state after the preinfiltration of the preform by the molten metal for 1min to produce homogenous mixture at temperature of the slurry was 620°C. The agitation was stopped after completing the addition of Al/MWCNTs billets or blocks, and the molten mixture was poured into preheated low carbon steel mould (250°C) and immediately squeezed during solidification. The pouring temperature for process was 601°C, and the speed of impeller was 750rpm. Well uniformly dispersed of MWCNTs into the melt alloy, resulted in good distribution and less agglomeration which improved mechanical properties of the castings. During cooling to RT and the difference of the coefficients of thermal expansion (CTE) between the matrix and CNTs, MWCNTs help in strengthening and hardening the matrix by increasing the matrix alloy dislocation density. This behaviour may be also partly attributed to the grain refinement that accompanied by addition of MWCNTs and stirring action. Due to grain refinement and effects of the strengthening of nanoparticles, the weight fraction of MWCNTs increases as the hardness increased [25]. The A356 matrix alloy reinforced with 1.5wt% MWCNTs revealed the ultimate tensile and yield strength properties and elongation percentage due to the uniformly distributed of reinforcement and grain refinement of Al matrix and the elongation percentage. The compressive strengths of nanocomposites have been increased with increasing MWCNTs weight fraction compared to A356 Al alloy at optimal value of 1.0wt%. Large amount of CNTs have poorly affected the material strength due to cluster formation of the particles or poor wettability with the matrix [26].

4.0 CONCLUSSION

This paper presents a review on various techniques applied for the fabrication of AlMNCs. The technique include high energy ball milling, PM route followed by sintering and secondary treatment such as CIP, HIP and extrusion, stir casting, ultrasonic cavation, combination of rheocasting and squeeze casting and semi-solid route stir casting method. The challenge on fabrication of AlMNCs such as low wettability of molten metal, it can be conclude that powder metallurgy as the most suitable method for manufacturing of AlMNCs. In order to achieve improvement in the mechanical properties, it is believed that on sintering process by powder metallurgy route should be further explored. .

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