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In-situ formed, ultrafine Al-Si composite materials: ductility

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ABSTRACT

Purpose: The work objective includes optimization of the casting production and heat treatment processes that will simultaneously maximize the combination of strength, hardness, and ductility for hypereutectic Al-Si compositions with Si volume fractions of as much as 25 vol.%. In addition, such an in-situ formed composite alloy will attain a unique combination of low production cost, high potential recycled content, and functional characteristics suitable for mission critical aerospace and vehicular applications.

Design/methodology/approach: The unique High Pressure Die Casting Universal Metallurgical Simulator and Analyser (HPDC UMSA) was used for melting, cyclic melt treatment, and solidification of the hypereutectic Al-Si-X (A390). The produced as-cast structures contained colonies of nano-diameter Si whiskers and other morphologies, and absence of primary silicon particles. Heat treated structures rendered nano and ultrafine metal matrix composites.

Findings: New developed as-cast Al-Si materials containing nano-diameter Si whiskers, without primary silicon particles required ultra short time heat treatment to result in nano and ultrafine metal matrix composite, rendering their hardness, strength and wear resistance, and the same time retaining toughness and ductility.

Research limitations/implications: The cast samples were produced in laboratory conditions and potential tensile strength was estimated from empirical correlation with micro-hardness measurements. In the future, the comprehensive mechanical properties need to be tested.

Practical implications: These ultrafine Si, Al-MMCs can be net-shape formed by modified HPDC technology or consolidated from spray-atomized alloy powder.

Originality/value: Optimization of the entire production process for the hypereutectic Al-Si alloy compositions achieved a uniform distribution of ~ 25 vol.% of ultrafine Si particles in ductile FCC-Al matrix further reinforced by age hardening with nano-scale spinodal GP-zones. The associated mechanical property and ductility improvements will open a wide range of critical lightweighting components in transportation: aerospace, terrestrial vehicle and marine to the optimized hypereutectic Al-Si alloys. Presently, these components do not use the commercial HPDC A390 alloys due to their limited ductility and strength. Proposed new technology will allow conversion of various cast airspace alloys with ultrahigh mechanical properties to the automotive applications.

Keywords: Ultrafine Al-Si composite, Al metal matrix, Hypereutectic Al alloys, Mechanical properties, High ductility

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PROPERTIES

1. Hypereutectic Al-Si-Cu-Mg alloys (A390 type)

Al-Si alloys are the "bread-and-butter" of the Al foundry industry with 2 million tons/year in North America used in transportation applications. Hypereutectic Al-Si (A390 type) alloys are used for high hardness, low wear, cast components. A390 alloy automotive applications include pistons, cylinder liners and liner-less engine blocks. High Si content reduces thermal expansion coefficient, improves thermal conductivity by 3X compared with cast iron.

As illustrated in Figure 1, the microstructure of heat treated commercial A390 consists predominantly of spheroidized eutectic Si(2) ≈ 5 µm particles dispersed in an Al matrix also containing Al-Si-Fe-Cu intermetallics. However, it also contains 8-10 vol.% of thermally stable primary Si(1) particles that are 15-50 µm diameter and agglomerates of these particles extending to ≈120 µm. Non-uniform distribution of primary Si(1) crystals leads to areas of high Si(1) particle concentration. These areas combine into critical flaws of >1 mm that limit fracture strength, elongation of the commercial HPDC hypereutectic Al-Si-X components.

As-cast properties are predominantly controlled by primary and secondary Si morphology. In current casting alloys a coarse microstructure induces internal flaws and stress concentrations that reduce the ductility and strength of cast parts. These deficiencies limit engineering component applications.

Suppression of primary Si formation and reduction of secondary Si size is necessary for better mechanical properties and require that process conditions for alloying, liquid metal treatment, solidification and heat treatment be optimized.

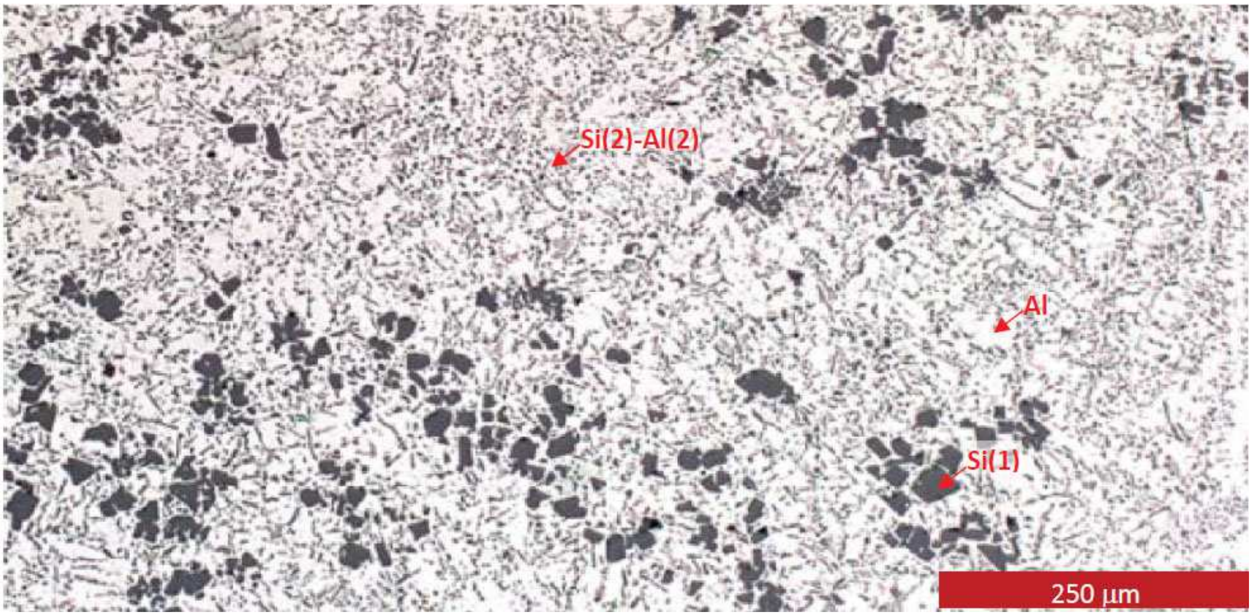

Fig. 1. Typical microstructure of high-pressure die cast and heat-treated A390 component (Al 20Si 3Cu alloy) provided by the Yamaha Motors Co. (Light optical micrograph (LOM), horizontal field-of-view (HOV) = 1,000 µm) [1]

2. Ultrafine Si dispersion-hardened, artificial aging-strengthened Al metal matrix composites (MMC)

A new family of hypereutectic Al-(18-28 wt.%) Si-X alloys feature as-cast 20-30 vol.% of in-situ formed nano-diameter Si whiskers [1,2]. On solution treatment these whiskers being fragmented and spheroidize into individual sub-micron equiaxed Si particles in a ductile FCC-Al solid solution matrix containing X. The material can be formed or used in this ductile form. However, it can be further strengthened by an aging process step which allows formation of coherent Guinier-Preston X containing intermetallic zones through a process of spinodal decomposition. This results in ≈5 vol.% of highly strained, coherent nano-scale X-intermetallic precipitates that are particularly effective in reinforcing the matrix by pinning down the dislocation motion through the Al matrix. Effective spinodal reinforcing phases contain Cu, or Zn, or Mg+Si (see Fig. 2).

Primary Si(1) phase formation is suppressed by a combination of Si modifier addition and a high cooling rate during solidification. The as-solidified structure is dominated by colonies of nano-diameter Si fibres/whiskers, and absence of bulk primary silicon particles. Other alloying components tend to solidify late and form thin, but nearly continuous layers of brittle Al intermetallics at the boundaries between the Al-Si whisker colonies. The ternary alloy as-solidified structure is hard but brittle with the macro-crack following the intermetallic layer. Heat treatment is necessary to thermally stabilize the ultrafine structure of the Si filler, to dissolve the intermetallics in the Al matrix and to strengthen it through uniform precipitation of nano-scale spinodal precipitates. Solution treatment spheroidizes the Si whiskers forming uniform dispersion of ultrafine equiaxed Si particles that are mainly smaller than 1 μm (see Fig. 3). It also dissolves intermetallics in the Al matrix, and homogenizes the alloying element concentration in this ductile FCC Al matrix. The material is most ductile in this solution treated and quenched temper and at this stage is suitable for both consolidation and forming. Solution heat-treated micro-structures of >20 vol.% uniform dispersion of ultrafine mainly <1 μm Si(2) spheroids in the Al matrix lead to exceptional hardness and wear resistance. The strength can be further enhanced by spinodal decomposition and precipitation hardening of the Al matrix through aging heat treatment.

During solution heat treatment Cu intermetallics dissolve in Al leaving ductile and formable solid-solution Al matrix. In subsequent aging Cu forms coherent nano-scale precipitates in the Al matrix increasing both yield and ultimate strength. Ultrafine Si particles do not crack during deformation promoting work hardening of the surrounding Al matrix, and allowing high ultimate fracture strength and elongation [4]. Nanoscale intermetallic precipitates strengthen Al matrix increasing composite yield strength and ultimate strength while retaining substantial ductility.

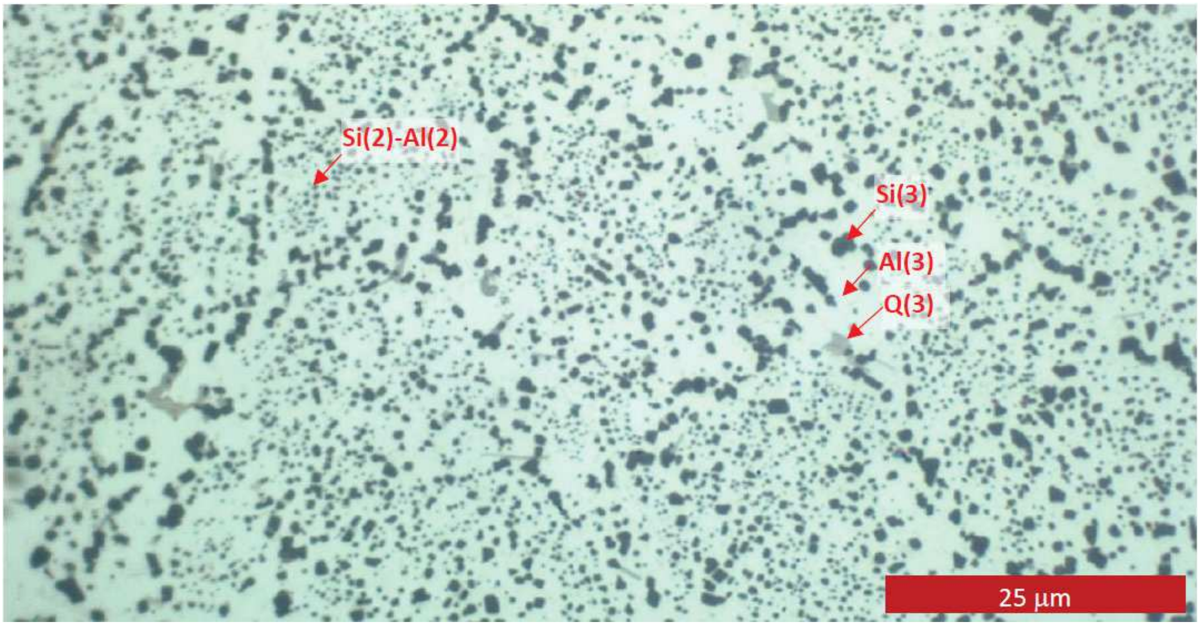

Fig. 2. Typical microstructure of squeeze-cast and heat-treated (Al 20Si 3Cu 0.5Mg 0.5Fe) in-situ formed ultrafine metal matrix composite using unique Universal Metallurgical Simulator and Analyser Platform. (LOM, HOV = 100 μm), [1,7]

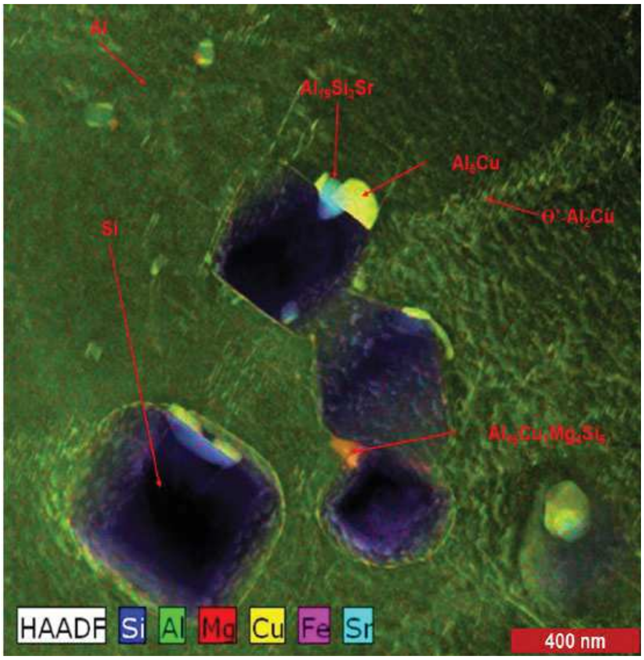

Fig. 3. Squeeze-cast and heat-treated (Al 20Si 3Cu 0.5Mg 0.5Fe) in-situ formed ultrafine metal matrix composite. (HAADF-STEM, HOV = 2 μm), [1]

3. Composite production process

A technique that in-situ forms Si dispersoids and spinodal precipitates in-situ allows the use of conventional high cooling rate casting technologies that include continuous strip casting, high pressure die casting, or a combination of powder atomization and powder consolidation through forging or extrusion. The production cost would be comparable to the currently produced commercial alloys by these casting and forming technologies.

Figure 4 shows the schematic of the production process for the in-situ Al-Si-X composite. Each of the process steps are similar to conventional industrial metal casting technology or metal powder processing. Integrated melting, alloying and melt treatment allow for saving of modifier additions. Melt treatment focuses on dissolution and removal of all the heterogeneous nucleation sites for the primary hypereutectic intermetallic crystals. These are further suppressed by addition of alkali and/or alkaline earth element as modifying agent. This agent also modifies the secondary Si structure repressing plates and promoting formation either nanoscale branched Si dendrites or individual needles. Success in obtaining the desired as-cast structure depends on high cooling rate solidification. Solution heat treatment fragments and spheroidizes the secondary Si needles and dendrite branches forming uniform distribution of sub-micron Si grains. It further places the alloying elements in solution in the FCC-Al matrix making the material sufficiently ductile for shape forming or powder consolidation through hot extrusion. After quenching an aging treatment promotes formation of uniform distribution of spinodal coherent nanoscale precipitates throughout the FCC-AL matrix, that can double the yield and ultimate strengths while retaining most of the matrix ductility. Optimized control of each process step is required to obtain the desired ultrafine composite microstructure and to get the hoped-for improvements in physical, mechanical properties.

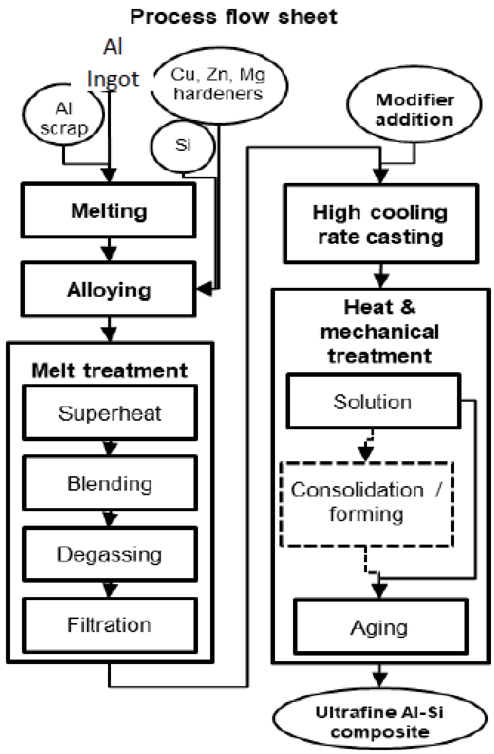

Fig. 4. Composite production process schematic

3.1. Low cost process

Ultrafine composite production process steps (including usage of ordinary ingots) are conventional, allowing a target of significant improvement in specific mechanical properties WITHOUT significant increase in component cost. Starting material costs are low due to common, low-cost alloying elements: Al and Si, with minor additions of Cu, Zn, Mg or Sr. All these elements are common in current generation of foundry alloys.

3.2. Ultra fast process

Ultra short process step times make the composite production process scalable to the high-volume production of automotive components. Solidification takes <10 sec, solution treatment <10 min and artificial aging <10 min.

3.3. Easily recyclable

In contrast to most dispersion-strengthened metal matrix composites, this ultrafine Si dispersion in Al matrix is easily recyclable. Both new and old Al-Si composite scrap can be simply melted and included in the next batch of the same material. The ultrafine Si dispersion easily and quickly dissolves in the Al melt. This allows loss-free utilization of the scrap with no negative impact on either processing or product properties.

4. Product properties

The new material is a low-density composite with higher mechanical properties as follows:

Density: The composite retains low density of Al matrix (Tab. 1).

Low composite density helps in attaining high specific property values.

Table 1.
Typical in-situ composite density

Component	Content, vol.%	Density, g/cm^3
Al	60%-70%	2.7
Si	30%-20%	2.3
θ-Al_2Cu	balance	4-5
Q-$Al_5Mg_8Cu_2Si_6$		
π-$Al_8FeMg_3Si_6$		
Composite	100%	2.6-2.8

Wear: of heat treated ultrafine MMC material was 30% lower than the commercial cast and heat treated HPDC Al 20Si 3Cu alloy. Wear in heat treated ultrafine Si, Al-MMC materials was 51% lower as compared to the as-cast structure.

Microhardness-Tensile Strength: Tensile test samples were NOT yet produced. Potential tensile strength was estimated from empirical correlation with macro-hardness measurements (Tab. 2).

In HPDC cast commercial engine blocks the strength potential is not attained due to combination of internal flaws and brittleness caused by large and agglomerated primary Si(1) grains, and undissolved intermetallics. Composite in which Si is present as ultrafine Si(2) particles uniformly distributed in ductile matrix is likely to approach the potential UTS much more closely.

Table 2.
Macrohardness and potential tensile strength of commercial A390 castings and of in-situ ultrafine composite of similar composition

	Hardness		Potential Ultimate Tensile Strength, UTS
	Rockwell	Brinell	MPa
15 mm A390 engine block wall	72	130	435
7 mm A390 engine block wall	78	134	475
A390 in-situ composite	86	169	570

Ductility: In the base hypereutectic A390 alloy the ductility is limited by the large size and non-uniform distribution of the primary Si crystals. These Si crystals crack during deformation and these microcracks link to cause brittle failure at ≈1% elongation. Current process prevents the primary Si crystal nucleation and growth allowing substantial undercooling. It appears that at sub-liquidus temperature melt crosses the Si spinodal surface allowing spontaneous uphill diffusion which leads to simultaneous formation of multitude of individual nano-diameter Si whiskers in the Al melt. Al follows solidifying as a ductile face-centre-cubic matrix between the Si whiskers.

The A390 alloy contains 4-5 wt.% of Cu which combines with Al and Mg to form brittle θ-Al_2Cu and Q-$Al_5Mg_8Cu_2Si_6$ intermetallics in the residual melt between the Al-Si whisker colonies. The final solidification rate is substantially slower and these intermetallics form crystals whose extent is limited by the inter-colony space. This introduces weak-brittle planes into the as-cast composite structure which is neither strong nor ductile in as-cast condition.

However, sub-solidus solution heat treatment quickly dissolves these intermetallics in the FCC-Al matrix. The resulting FCC-Al matrix phase is solid-solution strengthened by Cu and Mg solute atoms, but the material recovers most of the ductility of the FCC matrix allowing a possibility of consolidation and forming the material in this solution-treated state. Si nano-whiskers also change during the solution treatment. While they do not dissolve, the whiskers spheroidize leaving a uniform distribution of 20-30 vol.% of hard sub-micron equiaxed diamond-structured Si particles. These contribute to material hardness, low density, thermal conductivity and wear resistance but remain small enough and well-distributed enough to permit plastic deformation of the surrounding metal matrix without cracking and not to limit the elongation of the composite material. Results of a calculation shown in Figure 5 based on Orowan dislocation bowing mechanism shows that for 30 vol.% of uniformly distributed, non-deformable particles their contribution to material shear strength is only 20 and 10 MPa for Si particles of 0.5 and 1 μm diameter. This is insignificant compared to matrix yield strength >300 MPa, and thus it is reasonable to expect little impact of well distributed Si particles in that size range on matrix plastic deformation and ultimate elongation.

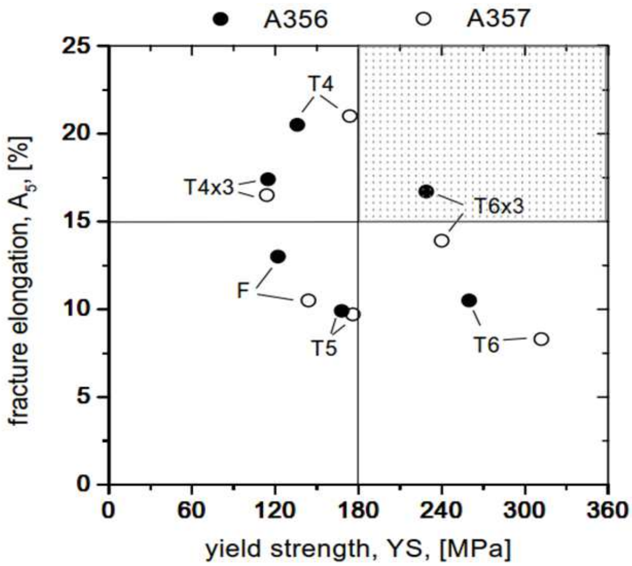

Fig. 5. Dependence of fracture elongation and YS on heat treatment for thixiocast A356 and A357 alloys. Note little change in elongation between T4x3 and T6x3 that share the short 3 min solution and spheroidization treatment. Ogris [4]

The matrix of the solution-treated Al-Si-Cu MMC has an Al-Cu composition which is well known for its age strengthening/hardening behavior. The aging heat treatment below the θ-Al_2Cu phase spinodal surface allows formation of Cu-rich Guinier-Preston zones on small nano-scale uniformly throughout the FCC-Al matrix volume. These zones transform to θ” and θ’-Al_2Cu precipitates coherent with the FCC-Al matrix that have a strengthening effect due to their small size, uniform distribution and high lattice mismatch strain. Spinodal precipitates uniformly distributed in the Al matrix allow the material to strengthen and work harden, without sacrificing ductility.

The Al matrix for an in-situ composite with A390 composition specification has matrix composition after solution heat treatment like AA 2024 wrought alloy specification. That material solution heat treated, quenched and naturally aged (T4 temper) has 20% ductility with 325 MPa yield strength (YS) and 472 MPa ultimate strength (UTS). Precipitation strengthening of AA 2024 by artificial aging to T6 temper increases YS to 395 MPa and UTS to 495 MPa while retaining 13% elongation [3].

5. Discussion

In most literature examples either poor distribution or excessive size of the reinforcing particles leads to loss of ductility in the metal matrix composite. However, there are some examples where excellent distribution of nanoscale reinforcement led to enhanced ductility. Precipitate strengthening through spinodal decomposition described above is one such example where formation of a uniform distribution of coherent nanoscale precipitates retains much of the ductility of the metal matrix.

Ogris [4] in his thesis work optimized the ductility of hypoeutectic A356/A357 (Al 7Si 0.3/0.6Mg) alloys. In these alloys modification of the Si(2) structure by Sr under fast thixiocasting solidification resulted in nano-diameter fibrous Si(2) dendrites in Al(2) matrix nearly indistinguishable from Si(2) structure observed in our work. In our case Si (2) volume fraction was ≈25 vol.% where as in A356/7 this volume fraction is limited to <15 vol.% by the eutectic composition. Further, in thixiocast A356/7 the Si(2)-Al(2) fibrous structure occupies only ≈50 vol.% of the total volume that is between the primary Al(1) globules, where as in the A390 based material Si(2)-Al(2) the occupies >90 vol.% of the total material. Ogris started with a conventional 12 h solution treatment at 540°C, followed by cold water quench for T4 or additional artificial aging of 4 h at 160°C for T6 temper. During the 12 h solution treatment Ogris observed very quick disintegration and spheroidization of the fibrous Si(2) dendrites into equiaxed submicron Si grains followed by Oswald ripening growth of these grains to >5 μm. Ogris observed that for T4 and T6 tempers the macro-crack initiated in and grew through the “eutectic” regions cracking the Si grains and bypassing the primary Al globules. He reduced the size of the spheroidized Si grains to <1 μm by reducing the solution heat treatment to 3 min followed by quenching for T4x3 and by 4 h artificial aging for T6x3. These modified T4x3 and T6x3 tempers resulted in optimum microstructure for thixiocast A356/7 alloys where the submicron Si particles did not crack, the fracture path followed Al matrix and did not discriminate between the primary Al globules and the Si particles containing "eutectic" regions of the microstructure. In both these regions at T6x3 temper Al matrix was strengthened by artificial aging which generated coherent spinodal Mg_2Si precipitates uniformly distributed throughout the Al matrix in both primary and "eutectic" regions. These coherent Mg_2Si precipitates account in A356 for doubling of the YS from 115 to 229 MPa, increase of UTS from 232 to 317 MPa and almost no change in of ductility 17.4+/-1% and 16.7+/-0.5% between T4x3 to T6x3 tempers. This clearly demonstrates that under optimum conditions submicron Si particles and nano-sized coherent precipitates have little effect on the material ductility in spite their significant contributions to modulus, strength, hardness, wear and impact resistance. The current hypereutectic A390 type in-situ composite expands this ideal distribution of submicron Si grains throughout the Al matrix volume, while increasing Si particles volume fraction to ≈25 vol.%. It also increases the volume fraction of coherent precipitates to ≈5 vol.% of θ'-Al_2Cu. While neither one of these changes will increase ductility, they do improve strength and hardness.

Khorshidi et. al. [5] reports on tensile properties of hypereutectic Al matrix composite containing 15 vol.% of Mg_2Si intermetallic. The intermetallic structure is modified by addition of 0.15 wt.% of Li. The modifier addition reduces the primary $Mg_2Si(1)$ particles from ≈30 μm to ≈3 μm and refines the secondary $Mg_2Si(2)$ structure from eutectic plates to branched coral like structure in FCC-Al matrix with nanometer scale branch diameters. This refinement of the $Mg_2Si(1,2)$ structure is reflected in increase in both UTS and elongation respectively from 230 to 315 MPa and from 2 to 7%. Khorshidi did not optimize the heat treatment of this composite for maximum ductility and strength by spheroidization of $Mg_2Si(2)$ "corals" and spinodal growth of β'-Mg_2Si precipitates in Al matrix.

Zhang et. al. [6] report on pure copper reinforced with nanoscale graphene flakes where they were able to obtain excellent flake distribution. Introduction of 4 vol.% of graphene into pure Cu led to Cu grain size reduction to 2 μm, doubling of the YS to 220 MPa, 40% increase in UTS to 287 MPa, 90% increase in toughness to 126 MJ/m^3 all combined with 36% increase in elongation to failure into a superplastic value of 54%.

The A390 based in-situ composite metal matrix materials provide a unique combination of absence of primary Si(1) grains with excellent distribution of high-volume fraction of secondary, sub-micron Si(2) hard particles in a matrix that can be further reinforced with nanoscale coherent precipitates. While we are unlikely to achieve Zhang's result of improving on the ductility of a pure FCC-Cu metal matrix, we expect a vast improvement over the poor ductility of a conventional A390 alloy casting and are aiming for ductility in the range ≈16% already demonstrated by Ogris for optimally heat treated thixiocast A356 or A357. We believe that, like Ogris, our ultra short solution treatment will allow us to improve on the ductility of conventional AA 2024 (13% @ T6 and 20% @ T3) metal matrix. This would also be a significant improvement over results of Khorshidi et. al. in both strength and ductility despite significantly increased volume fraction of the hard phase.

6. Conclusions

Patent-pending process was developed for production of a new family of in situ-formed ultrafine Si, Al-MMCs, hardened by high volume fraction of uniformly distributed ultrafine Si dispersion, and strengthened by nanoscale spinodal coherent precipitates. These ultrafine Si, Al-MMCs can be net-shape formed by slightly modified HPDC or consolidated from spray-atomized alloy powder. Low-cost alloying elements and low-cost industrially practiced process steps make ultrafine Si-Al MMCs affordable with the production costs like conventional HPDC Al alloys. Developed technology is applicable to other alloy systems. The ultrafine Si-Al MMCs exhibit low density, high hardness, low wear. Its strength and ductility is expected to be vastly improved over conventional casting of the same hypereutectic composition. Due to the uniform distribution of the submicron hard Si crystals we expect ductility of the composite to approach that of the FCC-Al alloy matrix alloy. The ultrafine Si, Al-MMCs are fully recyclable and capable of high recycled content. These ultrafine Si, Al-MMCs will be developed and optimized for lightweighting components in transportation: aerospace, terrestrial vehicle and marine.

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