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A Review on Heat Treatment Factor and Precipitations to Improve the Third Generation of Aluminum Lithium Alloys Used in Aeronautic Applications

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ABSTRACT

Aluminum lithium (Al-Li) alloys have attracted attention for military applications and aeronautic industry because of their properties especially low density compared to conventional aluminum alloys, the excellent characteristics that are attributed to the lithium addition, and to the different types of precipitations that have a radical effect on mechanical properties of Al-Li alloys. The 3rd generation of Al–Li alloys has been improved to reduce the defects of previous generations by combining the density reduction with high strength. This paper provides a review on the third generation of Al-Li alloys and its applications, the different types of precipitations, heat treatments and their influence on mechanical properties and grain refinement. Most early studies that have been realized by researchers over the last few years on aluminum lithium alloys for additive manufacturing are also reviewed in this paper.

1. Introduction

As early as the mid-1920s, many studies on Lithium addition to Aluminum was reported. More attention was garnered particularly to the aeronautics and aerospace field [1]. The first generation of aluminum lithium alloys was used in military aircraft in 1957 in the form of 2020 Al-Li plate used by Alcoa in 1958 in the wings of the Navy's Vigilante aircraft [2-3], and it comes down to the advantages that lithium offers on the reduction of the density of aluminum alloys [4], compared to the conventionally commercial 7xxx and 2xxx series aluminum alloys [5-6]. Lithium is the lightest metal element with Lattice constant of a = 0.35023nm density of 0.534 g.cm^3, atomic number of 3, body-centered cubic structure and high plasticity [7]. It shows that with the addition of 1wt% of lithium to aluminum, elastic modulus will increase by 6% and the alloy density will decrease by 3% [8-7]. Researchers often find problems with understanding the laws of compression of lithium aluminum alloys that always changes with the production process, work parameters, heat treatment so on. These parameters influence the mechanical properties such as anisotropy, hardness, and elastic limit. The objective of this paper is to make a review on the various approaches that have been

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used to improve the strength, precipitations, and toughness of lightweight Aluminum-lithium alloys and anisotropic control. In particular, the relevant mechanical problems due to the formation of different types of precipitation and the influence of aging are examined with the aim of clarifying the main mechanisms of the alloys mentioned. Recent research results are reviewed, although most early studies as well as current work are also mentioned to provide continuity, Al-Li alloys for additive manufacturing are also reviewed.

2. Development of Al-Li alloys

2.1 The first and second generation of Aluminum Lithium alloys

After the first utilization of the 1st generation of Al-Li alloys (2020 in 1958s developed by Alcoa) with so many advantages like: high creep resistance between 150°-200°C [9], and high strength, however, this alloy had a poor ductility [10-11], which aims to develop the 1420 Al-Li alloys with low density, a good weldability and stiffness. The 1421 alloy was also made with higher values of yield stress and ultimate strength, but the main defect of this alloys is the poor toughness caused by shearing of Al_3Li which considered as the most strengthening phase [12]. The 2nd generation Al-Li alloys is created for aerospace and aircraft applications with the objective of reducing density (for 8% to 10%) compared to traditional aluminum alloys [13-15], and stiffness improvement [16,17]. Therefore, the main objective is to develop materials with enhancements in both mechanical properties and lifetime cycle that can be used in the construction of wings and fuselage. Accordingly, in the 1970s and 1980s. The majority of prior research has focused on the optimization of Silicon and Iron contents that have the primary effect on ductility and toughness, the same for Manganese that was replaced zirconium to produce Al_3Zr precipitates for ductility, toughness, and grain refinement [18,19]. The increase in the percentage of lithium can generate an increase in size and volume fraction of δ', which is responsible on strengthening, [20] and was the main reason for increasing lithium percentage in the second generation of Al-Li-X alloys as shown in Table 1, but this higher % addition of lithium caused several advantages that limited their usage like fracture toughness, corrosion, anisotropic and fatigue [21,22].

Table 1
Densities, alloying elements and the developers on the three generations of Aluminum lithium alloys [9].

Alloy	Li wt%	Cu wt%	Mg wt%	Ag wt%	Zr wt%	Sc wt%	Mn wt%	Zn wt%	Al wt%	Density g/cm^3	Place. Data
First generation											
2020	1.2	4.5					0.5			2.71	Alcoa 1958
1420	2.1		5.2		0.11					2.47	Soviet, 1965
1421	2.1		5.2		0.11	0.17				2.47	Soviet, 1965
Second generation (Li > 2wt%)											
2090	2.1	2.7			0.11					2.59	Alcoa 1984
2091	2.0	2.0	1.3		0.11					2.58	Pechiney 1985
8090	2.4	1.2	0.8		0.11					2.54	EAA 1984
1430	1.7	1.6	2.7		0.11	0.17				2.57	Soviet 1980s
1440	2.4	1.5	0.8		0.11					2.55	Soviet 1980s
1441	1.95	1.65	0.9		0.11					2.59	Soviet 1980s
1450	2.1	2.9			0.11					2.60	Soviet 1980s
1460	2.25	2.9			0.11					2.60	Soviet 1980s
Third generation (Li< 2wt%)											
2195	1.0	4.0	0.4	0.4	0.11					2.71	LM/Reynolds, 1992
2196	1.75	2.9	0.5	0.4	0.11		0.35 max	0.35 max		2.63	LM/Reynolds, 2000
2297	1.4	2.8	0.25 max		0.11		0.3	0.5 max		2.65	LM/Reynolds, 1997
2397	1.4	2.8	0.25 max		0.11		0.3	0.10		2.65	Alcoa, 2002
2098	1.05	3.5	0.53	0.43	0.11		0.35 max	0.35		2.70	McCook-Metals, 2000
2198	1.0	3.2	0.5	0.4	0.11		0.5 max	0.35 max		2.69	Reynolds/ McCook-Metals/Alcan, 2005
2099	1.8	2.7	0.3		0.09		0.3	0.7		2.63	Alcoa, 2003
2199	1.6	2.6	0.2		0.09		0.3	0.6		2.64	Alcoa, 2005
2050	1.0	3.6	0.4	0.4	0.11		0.35	0.25 max		2.70	Pechiney/ Alcan 2004
2296	1.6	2.45	0.6	0.43	0.11		0.28	0.25 max		2.63	Alcan 2010
2060	0.7	3.95	0.85	0.25	0.11		0.3	0.4		2.72	Alcoa 2011
2055	1.15	3.7	0.4	0.4	0.11		0.3	0.5		2.70	Alcoa 2012
2065	1.2	4.2	0.5	0.30	0.11		0.4	0.2		2.70	Constellium 2012
2076	1.5	2.35	0.5	0.28	0.11		0.33	0.30 max		2.64	Constellium 2012

2.2 Third generation of Aluminum Lithium alloys

The 3rd generation of Al–Li alloys has been improved to reduce the defects of previous generations [23]. Reducing inspection and maintenance, weight savings, and performance are the main reasons for the development of this generation were tailored to cover the requirements of aeronautic and military industry [2]. Figure 1 shows an example of the Al-Li alloys applications in the manufacturing of aircrafts. As it is seen in Table 1, the 3rd contains lower amounts of Lithium (<2%) and a high Cu/Li ratio compared to the 2nd generation of Al-Li alloys [24]. It was noted that decreasing lithium amounts can positively influence the thermal stability and toughness of aluminum lithium alloys [14,25,26].

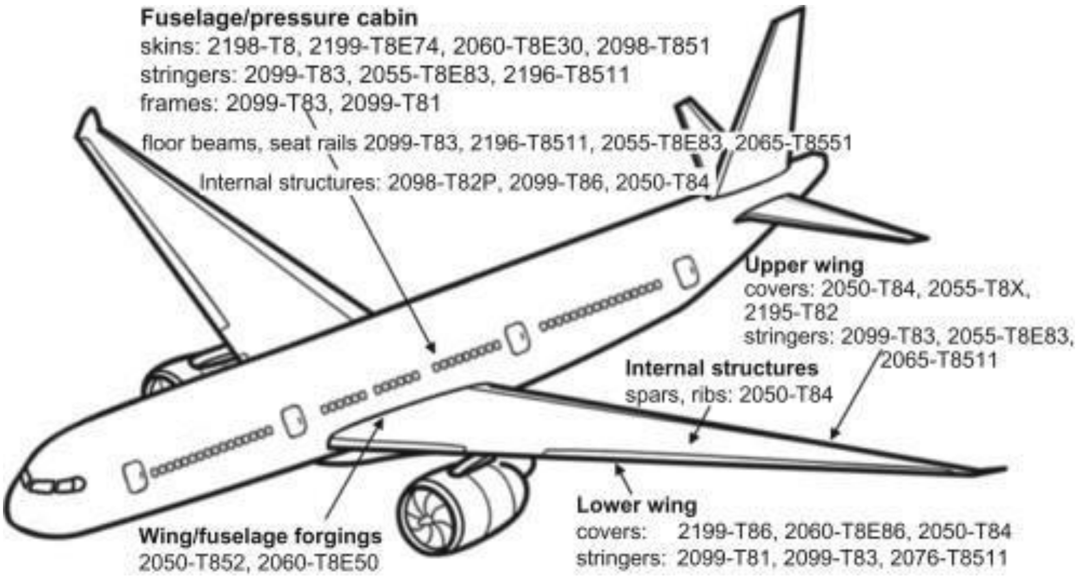
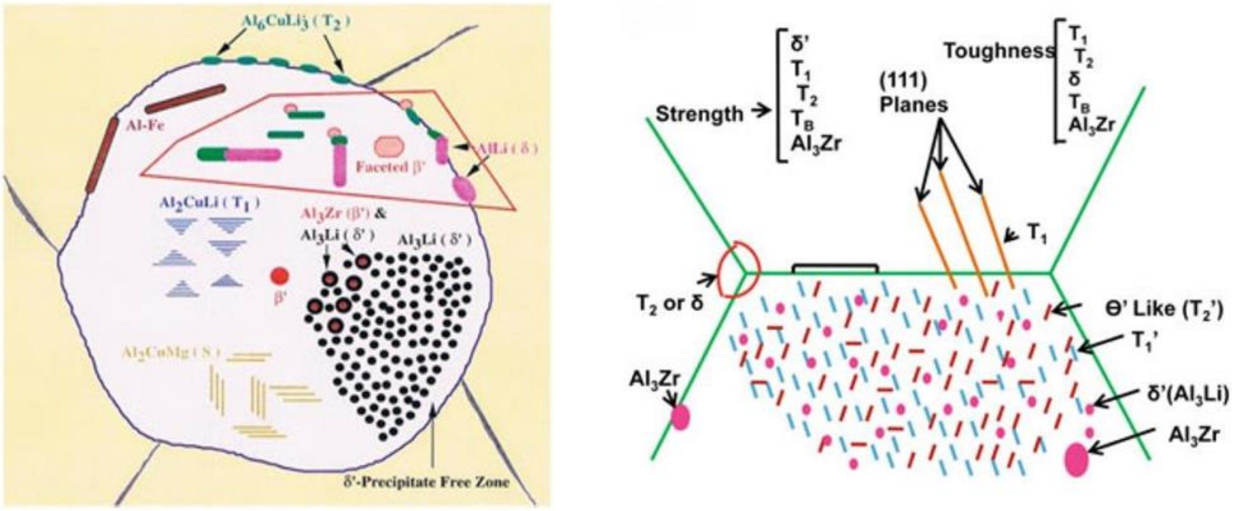


Fig. 1. Various uses of the third generation of Al-Li alloys in the aircraft structure [27]

3. Effect of Heat Treatment and Precipitations on Mechanical Properties of Al-Li Alloys

Several studies focused on the mechanical characteristics of Al-Li alloys. The initial microstructure is the essential parameter in the determination of ductility, cracking resistance, strength, and fracture toughness [28]. The Ce+Zr addition can enhance mechanical characteristics of Al-Cu-Li alloys, the alloys with Ce and Zr can increase yield strength and ultimate tensile strength [29,30]. Si and Fe can negatively influence the properties like toughness fracture, affect strength and work hardening by coursing intermetallic compounds that can be seen at grain boundaries [17,31,32]. The addition of Mg and Cu alloying elements can give rise to phase of the S' (A12CuMg) that mainly depends on the percentage of Li element and on the presence of reinforcing SiC particles phase [33], which can improve strength and tensile ductility [9]. It was noted that the hardness increasing with a good combination of toughness and strength can be caused by the addition of different amounts of rare earths for the Al–Li–Cu alloys [34]. A peak in the strength was observed for 1.3 wt% Li of Al-Li-Cu-Zr alloys with trace additions of Mg and Ag compared to various amounts of lithium from 0wt% Li to 1.3wt% Li [35]. The tensile properties of 2099 Al-Li alloy is enhanced by homogenization treatment compared to as-cast alloy by the dissolution of interdendritic phases, uniform deformation caused by the decrease of dendritic structures that can enhance ductility, reduction of grain boundaries phases which positively influences the propagation of micro-cracks under stress at grain boundaries that increase strength [36]. For the Al-Li 2198-T8 alloys using High Pressure Torsion HPT, the strength and hardness increase because of dislocation strengthening and grain refinement [37], and its plasticity is inversely proportional with the tensile strength in case of processing by Friction Spot Welding (FSpW) [38]. The Schematics of typical microstructural for Al–Li alloys are presented in Figure 2.



(a) (b)

Fig. 2. Schematics of typical microstructural features in (a) second and (b) third generation Al–Li alloys [39]

3.1 Effect of heat treatment on macroscopic Anisotropy and texture in Al-Li Alloys

Several parameters can influence the anisotropy of Al-Li alloys such as crystallographic texture, shearing of the Al_3Li phases and the resultant temperature field direction related to the current stress states, recrystallization degree, type and history of the deformation process before artificial ageing, the alloying elements which are the main parameters for the distribution and the precipitations size [9]. Al-Li alloys presents high anisotropy than Al alloys, and its due to the coherent ordered δ' phase (up to 20%) [40]. For the 1445 Al-Li alloy sheet; the non-recrystallization is caused by $Al_3(Sc,Zr)$ Nano-sized which can appear with a large volume when being solutionized of 575 °C, and pin the grain boundaries, dislocations and subgrain boundaries while the main recrystallization model is subgrain coalescence and increase [41]. Controlling sheet metal's anisotropy can improve its formability and plastic anisotropy [42]. For 2195 Al-Li alloy cold-rolling sheet, the investigation for the anisotropy during aging treatment shows that the anisotropy decreases during aging time as long as over-aging is not reached [43,44]. During the sheet metal forming of Al-Li alloys anisotropy affects the final formed shape. Bouchaâla *et al.,* [45] studied the impact of anisotropic and isotropic yield functions of AA2090 Al-Li alloy on the thickness distribution during sheet metal forming process. Many phenomenological yield functions have been suggested for the prediction of plastic anisotropy for deep drawing process (e.g Hill [46], Barlat *et al.,* [47-48], Bron and Besson [49]).

3.2 Age Hardening Behavior

Hekmat *et al.,* [50] mentioned that the as cast AA2195 alloys exhibit a very low density and strength, and it is not recommended to be used directly in the mold application. The different states of aging and rate of deformation influence the adiabatic shear behaviors of 2195 Al-Li alloy, The peak-aged (heat treatment at 500 °C for 30 min, quenching in water and subsequently aged at 180 °C for 4 h) 16 h (peak-aged), 40 h (over-aged), the two samples had different properties, it is observed that the over-aged samples have a high sensitivity to adiabatic shear, while the peak-aged samples exhibit more resistance to adiabatic shear [51]. During deformation, the ability of the various precipitates to resist dislocation motion is the essential factor for the strengthening response of age- hardenable Al-Li-Cu-X alloys [52]. Walker *et al.,* [53] have worked on the impact of incomplete solution treatment on 2195 Al-Li alloy. They have demonstrated that the strength value is higher in case of full heat treatment and the incomplete heat treatment can reduce size and volume of T_1 precipitates that can be nucleated on the dislocation structures during cold work. T_1 precipitation can be formed after ageing treatment and can decrease copper content in solid solution in Al-Cu-Li alloys [54]. For the Al-4Cu-1Li-0.25Mn-alloy; the age hardening can influence the thickness of Al_2Cu precipitate, which is the major phase, it increases continuously with time (aging at 17h – 180 °C) and the Al_2CuLi phase growth slowly with time [55]. While for the Al-1.3Li-5.8Cu-0.4Mg-0.4Ag-0.14Zr-0.3Ce an achievement of excellent mechanical properties and microstructure evolution during hot treatment (520 °C at 30 min) was caried out due to a large amount of Al_2CuLi precipitate and a few needle phases like Al_2Cu) that can be observed [56].

3.3 Effect of heat treatment on precipitate structure in Al-Li alloys

The mechanical characteristics of third generation Al–Li alloys are dramatically affected by the precipitates in their microstructures [57]. Figure 3 presents the effect of different phases on mechanical properties for Al-Li alloys. In this section, pertinent aspects of phase equilibria and precipitation reactions in these alloys are reviewed, the improvement of precipitation of Ө' at the expense of Ө'', and structure of the homogeneously nucleated zones can be changed because of the presence of small amounts of lithium [58]. The plasticity and toughness of Al-Cu-Li alloys can be improved by double aging from high to low (165 °C, 10 h) + (140 °C, 35 h) due to the growing of the T_1 phase (the major phase with a non-uniform size distribution) after the first ageing step and promoted the formation of new precipitates [59]. The Al_2MgLi and δ_0 phases may form in Al-Mg-Li alloys but in complex Al-Li alloys containing Cu, Al_2CuMg (S_0) forms and can eliminate the formation of Al_2Cu [60]. For the 2A97 aluminum lithium alloy, Guinier-Preston zone (GP zone) can be formed after aging without pre-deformation for 12h at 165°C, with cubic precipitates can only be observed after aging at 40h, The T_1 precipitates in 2A97 alloy aged at 150°C for 4 h for alloys aged with 6% pre-deformation at 150°C which is higher than the alloy aged at 165°c without pre-deformation [61].

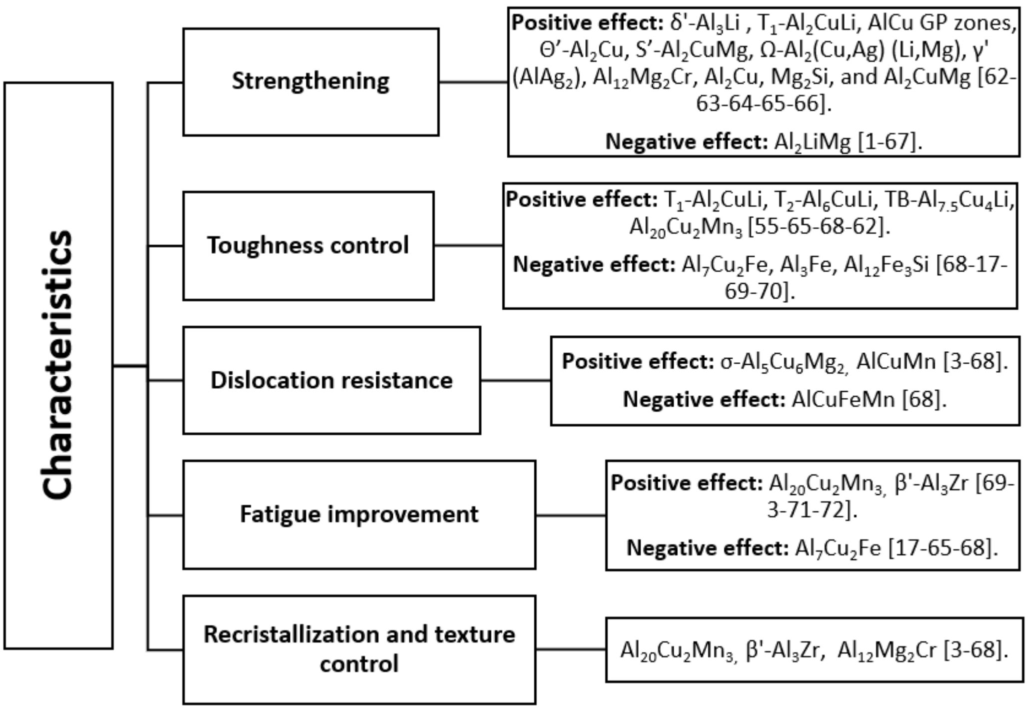


Fig. 3. Root cause - Effect of different phases on mechanical properties for Al-Li alloys

Depending on the ratio Cu/Li, the major strengthening phase Al_2CuLi (T_1) and Al_2Cu (θ') can be formed by the additions of Cu, in addition; Li additions form the coherent Al_3Li (δ0) and the Zr additions form the coherent Al_3Zr phase this precipitation which can eliminate recrystallization and thus generate a strong deformation at the texture [52-73]. The Al–Cu–Li alloys can present high amount of δ' precipitate as compared to Al–Mg–Li alloys [40]. Strengthening of Al_3Li is depending on many mechanisms such as modulus hardening and order hardening, coherency and surface hardening [74]. The Table 2 presents the impact of alloying elements for the Al-Cu-Li alloys.

Table 2
The impact of alloying elements on Al–Cu–Li Alloys [3]

Alloying Element	Impact
Li and Mg	Increase strength, decreasing density, and solid solution
Sc, Mn, Zr, and Cr	Texture and grain size control due to dispersoid formation.
Cu	Increase strength, and solid solution.
Zn	Increasing strength, corrosion, and solid solution.
Ag	Nucleation agent, coats the T_1 precipitate.
Ti and B	Grain refinement
Na, Si, Fe, and K	Impurities, adversely affect mechanical properties

3.4 Grain Refinement

Final grain size control is the main influencing factor to enhance mechanical properties for aluminum alloys in general. Grain refinement of the 3rd generation Al-Li alloys has been investigated by so many researchers [19-75-76-77]. Xinxiang *et al.,* have worked on the effect of cerium and zirconium microalloying addition in Al-Cu-Li alloys, they proved that the intermetallic dispersoids can be refined by Ce addition after homogenization [29]. There are four methods for grain refinement: Severe Plastic Deformation (SPD), addition of Grain Refiner (GR), Rapid Solidification (RS), Vibration and Stirring (VS) during solidification [78]. The presence of Zr on Al-Li alloys can control grain structure during high temperature by promoting Al_3Zr_Al_3Li dispersoids, which minimize the planar slip and improve ductility [79]. For the 2099 alloys, the dendritic structures can disappear after two-steps of homogenization treatment with a degreasing of segregation at the grain boundaries with residual AlCuFeMn/AlCuMn particles around it [36]. Liu *et al.,* [76] concluded that for the 2195 Al-Li alloy, the grain size increases with annealing at a lower temperature (300–350 °C), and it increase when the annealing temperature rose (350–400 °C) as well as the deformation texture [76-75-74-75]. Suresh *et al.,* in the investigation on effect of Sc addition on the evolution on the texture of AA2195 alloys during thermo-mechanical processing, they concluded the Sc addition reduce the grain size and enhanced precipitation kinetics with hardness and strength improvement as well as the presence of fine $Al_3(Sc, Zr)$ dispersoids [75].

4. Aluminum lithium alloys for additive manufacturing

Additive manufacturing processes have regularly been at the heart of the news in recent years, it is opposed to subtractive manufacturing processes (machining for example) or by deformation (forging for example) by making it possible to manufacture layer parts by layer from a 3D file [80-82]. Regarding manufacturers in the aeronautics and space sector, additive manufacturing plays a very important role, with a view to significantly reducing part manufacturing time, while maintaining great flexibility in design with well-defined mechanical and metallurgical properties [83,84]. This technology has the advantage of not using any tools during the manufacturing process and makes it possible to produce in a very short time (a few days) small series of functional parts with complex morphologies [81-85]. According to NF ISO / ASTM 52900, additive manufacturing is the process of assembling materials to manufacture parts from 3D model data, generally layer by layer [86-80]. There are seven main additive manufacturing processes which have the same principle, which is the manufacture of parts by adding material layer by layer namely: Binder Jetting, Directed Energy Deposition, Powder Bed Fusion, Sheet Lamination, Material Extrusion, Material Jetting, and Vat Photo Polymerization [86]. Very few studies are the subject of the 3rd generation of aluminum lithium alloy for additive manufacturing. Zhong *et al.,* [87] investigated microstructure and mechanical properties of wire arc additive manufacturing (WAAM) for Al-Li 2050 alloy, they concluded that the micro-hardness can be improved by post-deposited solution treatment and artificial aging (T6), after post deposited heat treatment; a dispersedly distribution of the θ (Al_2Cu) and δ' (Al_3Li) secondary phases at the grain boundary are observed. Most often, the cooling rate is characterized by the dendritic fineness which can be estimated by DAS (Dendrite Arm Spacing), also an increase in grain size is explained by the increase in heat flow per unit length which justifies the fines of the grain size of additive manufacturing processes compared to conventional processes (Figure 4) [88-90]. Liu *et al.,* have studied the Al-14 at % Li (atomic %) high lithium alloy as cast and laser powder bed fusion (L PBF), the cooling rate is higher for the laser power bed fusion process which leads to a uniform Li distribution in the primary α phase through solute trapping rather than to the formation of the brittle δ–Al-Li phase, which is prevented, which can improve the hardness [91]. Urekli *et al.,* demonstrated that for the additive manufacturing (L PBF) process using binary Aluminum lithium alloys, the elastic modulus radically increases with increasing of lithium content, and the high cooling rate is the most important parameters to reduce the negative effect of δ–Al-Li phase on yielding an inhomogeneous microstructure and poor mechanical characteristics [92]. Raffeis *et al.,* investigated the microstructures of an AA2099 Al-Cu-Li alloy for the Laser Powder Bed Fusion process (LPBF), the T_1 phase cannot be nucleate on dislocations without appropriate heat treatment, the preheating gave birth to two main precipitation, T_1 (Al_2CuLi) and T_B ($Al_{7.5}Cu_4Li$) [93]. Xin *et al.,* studied the effect of heat treatment process on mechanical properties microstructures of laser additive manufactured 5.02w%Cu-1.04%wLi aluminum lithium alloys, they concluded that α(Al) matrix and T_B(Al_7Cu_4Li) are the mainly phases on as deposited microstructure with small amount of Cu rich phase in the grain boundary [94]. The T_B phase and copper-rich disappear after annealing, with Al-Cu-Fe impurity phase presented in the grain boundary. The solid solution quenching and heat treatment enhance tensile strength and microhardness of Al-Li alloys compared to as-deposited alloys [94]. Jiao *et al.,* investigated the heat treatment microstructures, T_B phase and its influence on the micro-hardness of laser additive manufactured Al-Cu-Li alloys [95], they have confirmed results in the [94] ref, they observed also that during aging at 400 °C; the micro-hardness decreases before it reaches the maximum value [95].

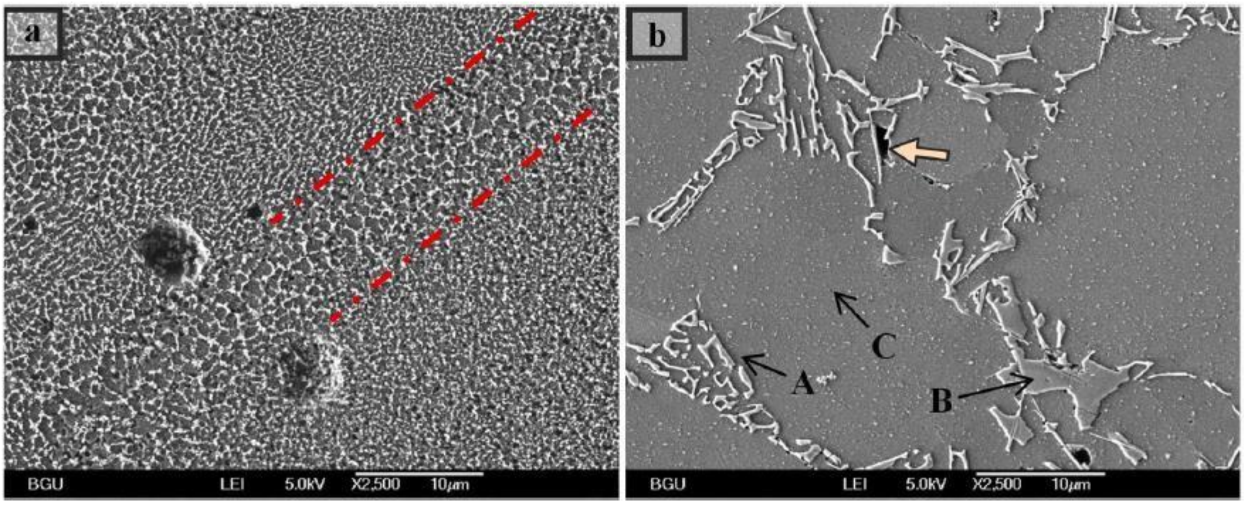


Fig. 4. Microstructure of AlSi10Mg manufactured by (a) SLM, (b) foundry; With (A) the Al-Si eutectic, (B) Si dispersed in the Al matrix and (C) the intermetallic phases containing Fe [90]

5. Conclusion

This paper aimed to review and summarize studies on the third generation of Al-Li alloys, especially on the different type of precipitations, the influence of the addition elements and their impact on the microstructure and mechanical properties. Research and development priorities have been discussed in the literature, which aimed to conclude that

i. The mechanical properties of 3rd generation Al–Li alloys are dramatically affected by the precipitates in their microstructures.
ii. Al-Li alloys exhibit different types of precipitations which can be varied depending on different parameters: addition elements, Cu / Li ratio, manufacturing process, and heat treatment. The control of those parameters contributes an excellent characteristic of the Al-Li alloys.
iii. Final grain size control is the main influencing factor to enhance mechanical properties for aluminum alloys in general.
iv. Additive manufacturing processes present many limitations and challenges compared to conventional manufacturing processes. Manufacturing parameters, process defects (anisotropy, porosity, etc.), and manufacturing cost are the greatest challenges for industrial applications of additive manufacturing, particularly in the aeronautic industry.

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