

## Literature review

### Suitable new aluminium alloys for wire Manufacturing

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## CHANGE CONTROL

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## 1. SCOPE

In the framework of the CleanSky project IAWAS, this report has been issued in order to support the deliverable "D1.1 – Suitable new aluminium alloys for wire manufacturing" aiming to define the specification for the new wire to be manufactured for Laser Beam Welding (LBW) and Wire Arc Additive Manufacturing (WAAM) applications.

Other 3 reports will be delivered by SONACA, Selectarc and Cranfield University, addressing respectively Laser Beam Welding (LBW), drawing wire, and WAAM (Wire + Arc Additive Manufacturing). All together with the feedbacks from the first LBW and WAAM test results will allow the completion of D1.1.

The parallel goal of this written review is to provide an overall view of the current state of the art alloys that could be suitable for LBW and WAAM technologies.

## 2. METALLURGICAL BACKGROUNDS

The development of materials has accompanied the expansion of technologies since the 19<sup>th</sup> century. Aluminium, discovered by Saint Claire Deville in 1854 was first considered as a noble metal. The electrolyze technology discovered by Heroult and Hall in 1886 opened its way to mass market.

Aerospace took advantage at a high level of the synergies between an industry and a material, in that case aluminium. As a mere detail, the Wright brothers' flight in 1903 used aluminium in the crankcase of the engine. However, it is in the 1930 years that this metal has found its main use for aerospace when aircraft's conception turned into fully metallic aerostructures. Aircraft manufacturers turned naturally towards aluminium alloys as they have a unique compromise of properties between density and good overall mechanical, damage and corrosion resistance properties. This induced weight reduction, which is a key point for aircraft performances.

From the 1930 to 1980 years, a variety of alloys / tempers came out from intensive developments both from Aluminium producers and aircraft's manufacturers : see figure 1 that gives an example of typical materials used in an aircraft at the end of the 70's. Mainly 2xxx (Al-Cu) and 7xxx (Al-Zn-Mg-Cu) series alloys are used. Many alloys / tempers combinations have been developed in order to get the best properties for given parts : compression strength for upper wing skin, tension and damage tolerance for lower wing skin, corrosion resistance for lower part of fuselage, etc...

A major turn on aerostructure conception raised in the early 80's as long carbon fiber resin reinforced composites became mature enough so that structural parts can be produced with. Table 1 and figure 2 give proportions of material over the last 40 years. The percentage of Aluminium alloys has decreased from almost 70% to 20% roughly.

Aircraft	Launch date	Al	Ti	Composites	Steel	others
A310	1978	67%	5%	10%	13%	5%
A320	1987	58%	6%	20%	13%	3%
A350	2013	19%	14%	53%	6%	8%

Table 1 : evolution of materials proportion for Airbus family

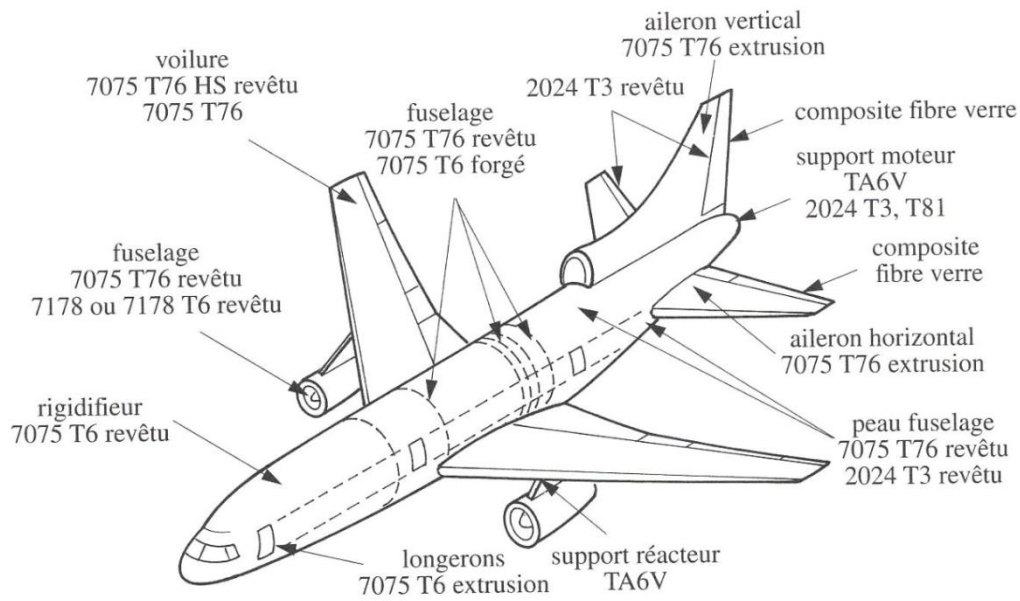
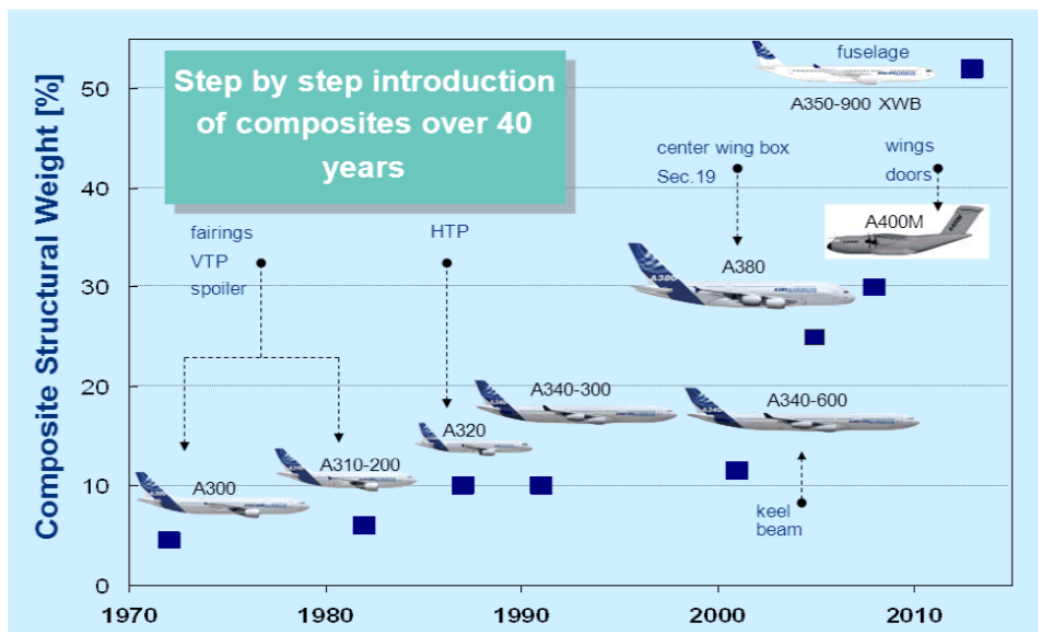


Figure 1 : typical use of materials at the end of the 70's



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Figure 2 : composites introduction on Airbus family over the years.

As a reaction to this main trend, a new alloy family came up in order to compete with composites by offering a lower density, i.e. a lower weight and similar properties to 2xxx and 7xxx series alloys : the Aluminium – Lithium alloys.

R. J. RIOJA and J. Liu [1] give a comprehensive review of the evolution of this alloy family taken into account the so-called first, second and third generations of these alloys. The chemical compositions are given in table 2, and plotted as Cu and Cu:Li proportions in figure 3.

Early first and second generations Al-Li alloys (ex. – 2020 and 2090) displayed high amounts of anisotropy with respect to mechanical properties; however, third generation Al-Li type alloys have sought to mitigate the degree of anisotropy by increasing the Cu:Li ratio as well as adding other carefully calculated alloy additions [2]. Generally, these alloying additions alongside careful thermomechanical processing techniques, help mitigate the problems of early Al-Li alloys by controlling the optimizing the size and distribution of T1,  $\delta'$ , and  $\theta'$  [3].

This current 3<sup>rd</sup> generation of Al-Li have the following alloying additions :

- Li is added in order to increase the strength and reduce the density of the alloy. As stated above, Li combines with Cu to form several Al-Cu-Li type precipitates, but is also generally present in the microstructure as  $\delta'$  ( $\text{Al}_3\text{Li}$ ), which helps strengthen the alloy.
- Mg is added chiefly for solid solution strengthening purposes, although it has been shown that Mg can substitute for Li to form  $\text{Al}_2(\text{Cu, Li-Mg})$  T1 precipitates.
- Cu additions help form the main strengthening precipitates T1 ( $\text{Al}_2\text{CuLi}$ ) and the  $\theta'$ -type ( $\text{Al}_2\text{Cu}$ ) precipitates. Cu also combines with Li to form the T2 precipitate ( $\text{Al}_6\text{CuLi}_3$ ), which helps increase the toughness of the alloy [4].
- Ag is added for solid-solution and precipitation strengthening. Ag atoms substitute Li atomic positions in the T<sub>1</sub> structure leading to  $\text{Al}_2(\text{Cu-Ag})(\text{Li-Mg})$ . However this alloying element has a strong impact on alloy's cost.
- Zn has been shown to improve the corrosion resistance of Al-Cu-Li alloys ; however, Zn is not believed to be involved in any wide-spread precipitation process and is thought to remain in solid solution.
- Zr and Mn control recrystallization and texture. All 3<sup>rd</sup> generation alloys exhibit the maximal 0.11% Zr. Above this percentage, Zr is retains in solid solution without any beneficial effect.
- Fe and Si as impurities affect fracture toughness, fatigue, and corrosion. The lower are this elements, the higher fatigue properties will be.
- Ti is used as a grain refiner during solidification of ingots. Rods of AT5B, containing  $\text{TiB}_2$  are introduced during castin the refine the grain structure.
- Na and K are impurities which affect fracture toughness.

	Li	Cu	Mg	Ag	Zr	Sc	Mn	Zn	Approximate Date
<b>1st generation</b>									
2020	1.2	4.5					0.5		Alcoa 1958
01420	2.1		5.2		0.11				Soviet 1965
01421	2.1		5.2		0.11	0.17			Soviet 1965
<b>2nd generation (Li ≥ 2 pct)</b>									
2090	2.1	2.7			0.11				Alcoa 1984
2091	2.0	2.0	1.3		0.11				Pechiney 1985
8090	2.4	1.2	0.8		0.11	0.17			EAA 1984
01430	1.7	1.6	2.7		0.11				Soviet 1980s
01440	2.4	1.5	0.8		0.11				Soviet 1980s
01450	2.1	2.9			0.11				Soviet 1980s
01460	2.25	2.9			0.11	0.09			Soviet 1980s
<b>3rd generation (Li &lt; 2 pct)</b>									
2195	1.0	4.0	0.4	0.4	0.11				LM/Reynolds 1992
2196	1.75	2.9	0.5	0.4	0.11		0.35 max	0.35 max	LM/Reynolds 2000
2297	1.4	2.8	0.25 max		0.11		0.3	0.5 max	LM/Reynolds 1997
2397	1.4	2.8	0.25 max		0.11		0.3	0.10	Alcoa 1993
2198	1.0	3.2	0.5	0.4	0.11		0.5 max	0.35 max	Reynolds/McCook 2005
2099	1.8	2.7	0.3		0.09		0.3	0.7	Alcoa 2003
2199	1.6	2.6	0.2		0.09		0.3	0.6	Alcoa 2005
2050	1.0	3.6	0.4	0.4	0.11		0.35	0.25 max	Pechiney 2004
2060	0.75	3.95	0.85	0.25	0.11		0.3	0.4	Alcoa 2011
2055	1.15	3.7	0.4	0.4	0.11		0.3	0.5	Alcoa 2012

Table 2 : chemical compositions of Al-Li [1]

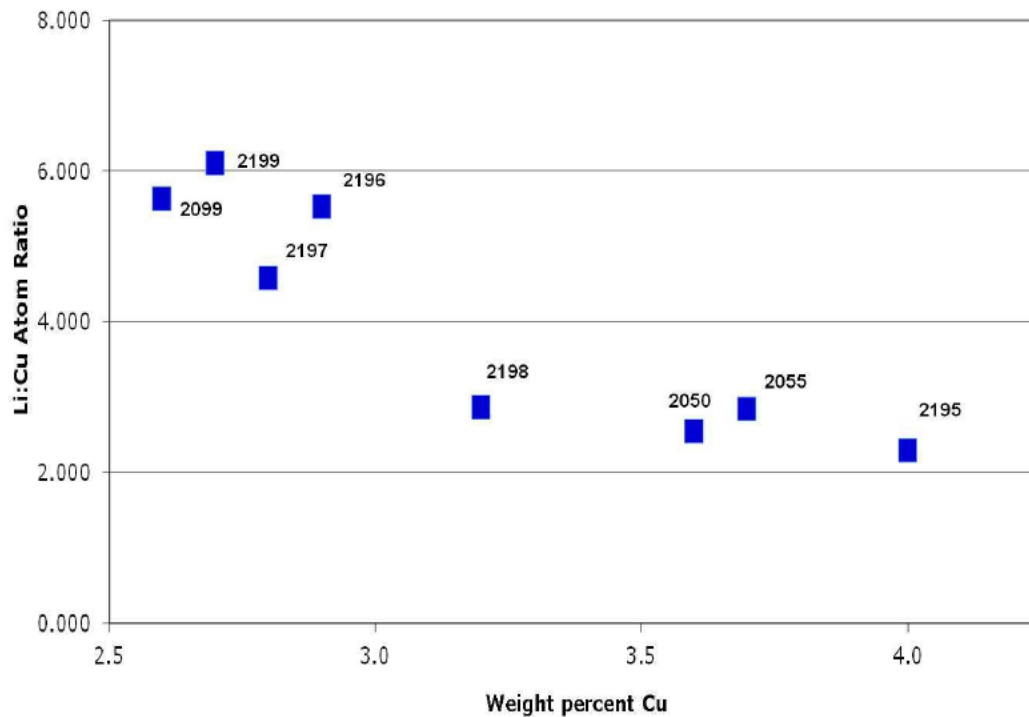


Figure 3 : Commercial Al-Cu-Li aluminum alloys plotted with respect to Li:Cu contents [5]

## 3. KEY CHARACTERISTICS NEEDED

Aluminium alloys have the capability of never-ending reinventing applications. Integral structures have been engineered 20 years ago : thick plates being machined in order to end up with a single part including thin cover and stiffeners. Its goal is to replace an aerostructure produced with a thin plate, an extrusion and rivets bounding them together [6]. Again, weight reduction is targeted through this application.

However such thick plates have a great inconvenient as the mechanical properties in the core of the plate decrease when the thickness increase [7].

Therefore innovative solutions have to be found : the idea driven by the latest innovations is not to take away material by machining thick plates, but in opposite to add materials from a thin plate :

- laser beam welding stick the stiffener to the plate itself by welding the 2 together, without the need of rivets and horizontal part of stiffeners : see figure 4.
- local additive manufacturing of aluminium to create walls and stiffeners at the right positions : see figure 5.

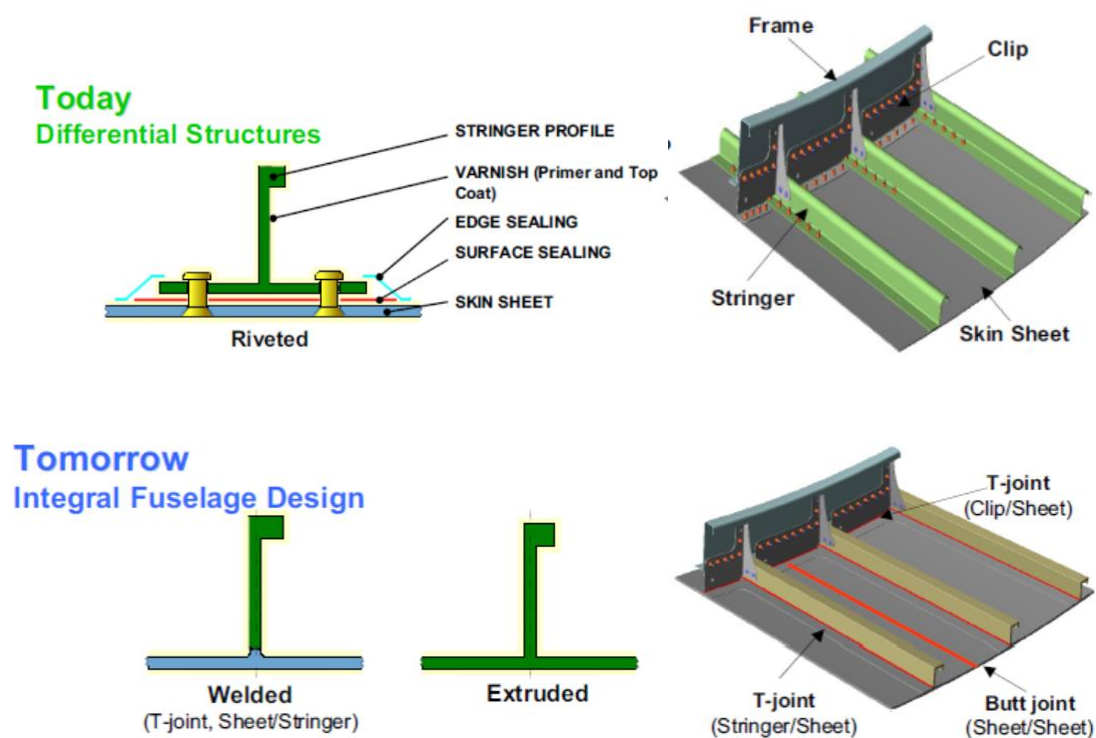


Figure 4 : LBW technology for integral structures [8]



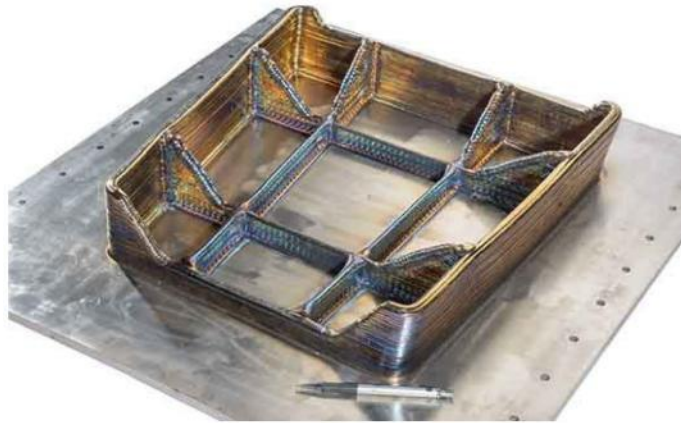


Figure 5 : Example of titanium near-net-shape WAAM part [9]

Both technologies use wires in order either to weld (filler metal) or to add material. The major points that we are facing in this project are :

- The design of chemical compositions that would minimize defects created by such technologies. This will go in addition with the process improvements that the project will generate.
- The ability to manufacture such alloys as wires.

## 3.1 Defects generated by the 2 technologies

The comprehensive literature reviews performed by the University of Cranfield and Sonaca give the major defects generated by the LBW and WAAM : see Table 3. The idea is here to see how the chemical compositions of alloys can be adjusted in order to help decreasing the amount of these defects, in conjunction with the improvements of these technologies expected in this IAWAS project.

One point is however clear : one shall remain within the Al-Li metallurgy. Only these alloys can compete with composites with their lower density.

Defect	LBW	WAAM	comments	Ways of improvements <sup>1</sup>
Porosity	✓	✓	Same phenomenon and root cause for both	T
Hot cracking / solidification cracking	✓	✓		T/C
Liquation cracking	✓		in the partially melted zone	T/C
Loss of mechanical properties	✓		To some extent, both technologies can end up with low mechanical properties : either by losing them under the effects of heat treatments with LBW or because heat treatments are not possible for WAAM	C
corrosion	✓			T/C
Loss of alloying elements		✓		T

Table 3 : summary of undesired effects for LBW and WAAM

## 3.1.1 Porosity

A widespread issue in fusion welding of aluminium is porosity : this issue is also a major problem in AM. Hydrogen solubility in aluminium is much higher at liquid (0.69 cm<sup>3</sup>/100g) than solid state (0.036 cm<sup>3</sup>/100g). At liquid state, hydrogen from the atmosphere and wire and substrate surface contamination is dissolved into the melt pool. During solidification, the solubility of hydrogen drops and gas pores are formed: as aluminium solidify very quickly, most of these pores are trapped in the material.

The high presence of porosity in the weld induces a loss of mechanical properties.

## 3.1.2 Hot cracking

Due to their high thermal expansion coefficient Al-Li alloys are very sensitive to hot cracking, also known as solidification cracking. It is one of the major defects that can occur during metal solidification. It is the result of an inadequate melt feeding and/or a large solidification interval of the alloy that initiate micro-pores and severe deformation during the metal solidification leading to the opening and propagation of cracks.

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<sup>1</sup> T : technology induced improvement, C : chemical compositions induced improvement

## a) Weldable 5xxx series alloys

5xxx series alloys (Al-Mg) are known as weldable alloys : It means that they can be fusion welded without being too susceptible to hot cracking. Filler wires of approximately the same composition as the base material are used for welding of 5xxx-alloys. In such alloys, mechanical properties are obtained by work-hardening (cold deformation) and microstructure.

The Influence of alloying elements Zr, Sc and Mn has been studied by Johansen [10]. These 3 elements form small precipitates (dispersoids) that retard recrystallisation and control grain size when this one happens. Smaller grain sizes are favorable to properties. The addition of Scandium seems more interesting than Zr in controlling recrystallisation as it seems that segregations of Zr are larger than those of Sc.  $Al_3Zr$  particles were distributed rather heterogeneously as compared to the  $Al_3(ScZr)$ -particles.

## b) Influence of dispersoids on hot cracking

Hot cracking can be reduced when changing the grain structure during solidification : promote small grain structure with nucleants [11], i.e. change from columnar dendritic to equiaxed cellular grains.  $TiB_2$  are used as nucleants (rods addition during casting). They may be introduced during WAAM if double wires are used.

$Al_3Zr$  are also used as grain refiner. However all Al-Li contains Zr to a sufficient level.

The addition of Scandium in conventional alloys have proven its ability to reduce hot cracking during solidification [12] : a content above 0.25% prevented hot cracking in 7108. 2<sup>nd</sup> generation Al-Li had Scandium inside, but no 3<sup>rd</sup> generation. This could be an option to test such effect.

## c) Other interesting metallurgy

### Non heat-treated Al-Li

Concerning Al-Li, an alloy 1424 was welded using the TIG and  $CO_2$  laser techniques with a filler metal resembling the material itself but with scandium [13]. Results were promising and no hot cracking have been observed.

### Al-Mg-Li

Following the idea of Al-Mg weldable alloys, a type of alloys seems interesting : Al-Mg-Li. Some literature exists on such compositions. Eberl and Bes [14] has shown that interesting mechanical properties can be obtained with Mg [4.0% – 5.0%], typically 4.5%, and Li [1.0% – 1.6%], typically 1.4% other elements being Zr [0.05% – 0.15%], typically 0.12%. Such alloys are air quenched, so interesting for WAAM and LBW.

### 3.1.3 Reduction of loss of mechanical properties

Most Al-Li have to be water quenched and artificially aged in order to exhibit right properties. This means that any local or global heating above typically 150°C (LBW) or any process where no heat treatment will be performed at all (WAAM) would end up with low properties and undesirable effects.

There is however a family of alloy that can be "air quenched" : Al-Mg-Li (-Zn – Zr – Sc). The latest on this alloy family is the 1424 [15].

This alloy is told as weldable and corrosion-resistant. It has been created based on two Russian alloys 1420 and 1421 by reducing the lithium content and additional alloying with zinc. Alloy 1424 is a promising material for welded fuselages instead of the unweldable alloy 2024; the former has an obvious advantage over the latter where the density and the elasticity modulus are concerned.

In comparison with 2024 T3, this alloys has 10% lower density, +12% in Young's modulus, +15% increased yield strength and reduced fatigue crack growth rate : -70% at  $\Delta K = 25\text{MPa}\sqrt{\text{m}}$  [8].

Concerning the chemical composition, table 4 gives the ranges found in literature. This alloy has no copper inside. This means that it doesn't follow Al : Li stoichiometry rules.

[15] states that this alloy can be age hardened. The use of three-stage aging at 85 – 120°C provides a finely dispersed  $\delta'$ -phase in the low-temperature stage I ( $t_1$ ), its sufficiently large volume in stage II ( $t_2 > t_1$ ), and additional segregation of  $\delta'$ -particles in stage III ( $t_3 < t_2$ ) aimed at diminishing the supersaturation of the solid solution. In this state the principal hardening  $\delta'$ -phase ( $\text{Al}_3\text{Li}$ ) and the excess phase S1 (S1') ( $\text{Al}_2\text{MgLi}$ ) are in equilibrium with the solid solution.

In the Tempus' study [8], it seems that the 1424 is used as plate and not filler. However, the addition of scandium seems to indicate that the application was welded structure.

The composition given by [13] has been used in a study where an alloy 1424 has been welded using the TIG and CO<sub>2</sub> laser techniques with a filler metal resembling the material itself (with scandium). After aging treatment, the welded / unwelded TYS ratio was 0.74.

	Li (%)	Mg (%)	Zn (%)	Cu (%)	Si (%)	Mn (%)	Zr (%)	Sc (%)
[15]	1.5 – 1.9	4.1 – 6	0.1 – 1.5	na				
[8]	1.5 – 1.75	5.0 – 5.6	0.4 – 0.7	-	0.08 max	-	0.05 – 0.3	0.01 – 0.06
[13]	1.63	5.35	0.65	-	0.05 max	-	0.09	0.08

Table 4 : chemical compositions found in literature for 1424 alloy

Structure produced by WAAM could therefore being only aged : [15] proposes for instance a triple step aging the rage 85 – 120°C. Experiments needs to be performed in order to find the right treatment.

## 3.2 Ability to manufacture such wires

The typical process is given in figure 6.

- Casting of Al-Li alloys represents technological challenges. The surface of the molten metal exhibits high oxidation. Therefore, furnaces, channels and molten surface of billets shall be protected by inert gas.
- The homogenisation treatment aims at reducing the chemical segregation that has occurred during the casting itself. Only local segregation at the scale of the grain are so lowered. Segregation at the scale of the billet itself will remain.
- Billets are then cut to length and scalped using mechanical saw and turning machines.
- These cut billets are then heated, usually using induction furnaces at approx. 400°C : metal is still solid but have high ductility in order to be extruded.
- For the extrusion, a die of 6mm has been used. The end product after extrusion is then a 6mm bar, 12 to 15m long, straight.
- For the use of the project, such bars have been coiled on a mandrel. No link between bars can be done into UAC facility.

Main issues may arise at SelectArc (not described in the figure 6) depending on their process :

- Welding each coiled bar together may be difficult as electrical welding is used. Indeed, Al-Li alloy doesn't respond well? to electrical conductivity. The addition of silver (Ag) might help in increasing this electrical response.
- Al-Li can start precipitation hardening at room temperature. This is especially true for high straightening potential alloy such as 2196. Annealing shall be performed immediately before wire drawing. 1424 alloy will react differently as main alloying element Mg remains in solid solution at room temperature.
- Wire shaving may also be difficult as Al-Li drawn wire before annealing will be texturized and will have very low ductility in ST-direction (perpendicular to the wire length).

## 4. CONCLUSION / FURTHER WORK

This literature review gives some indications of alloys that could be tested in order to produce wires for LBW and WAAM. A strong cooperation between project's partners is however necessary as alloy reaction during LBW and WAAM is related to the process itself :

- 1424 alloy might be an interesting candidate. Addition of Sc and Ag may be discussed. Moreover, for WAAM, a second wire of  $TiB_2$  rod may be added if the 2 wires WAAM technology is used.
- Al-Mg alloy with the addition of Li seems also interesting as Li will lower the density whereas this alloy may remain weldable as being based on the Al-Mg chemistry.
- More classical Al-Mg alloy may also be studied. The addition of Sc and / or Zr to promote fine and weld distributed dispersoids will help in controlling the grain structures and therefore properties.

Whatever the alloy selected, a fine tuning of chemical composition and heat treatments will be necessary. Some numerical simulation could be done then in order to reduce experimental trials.

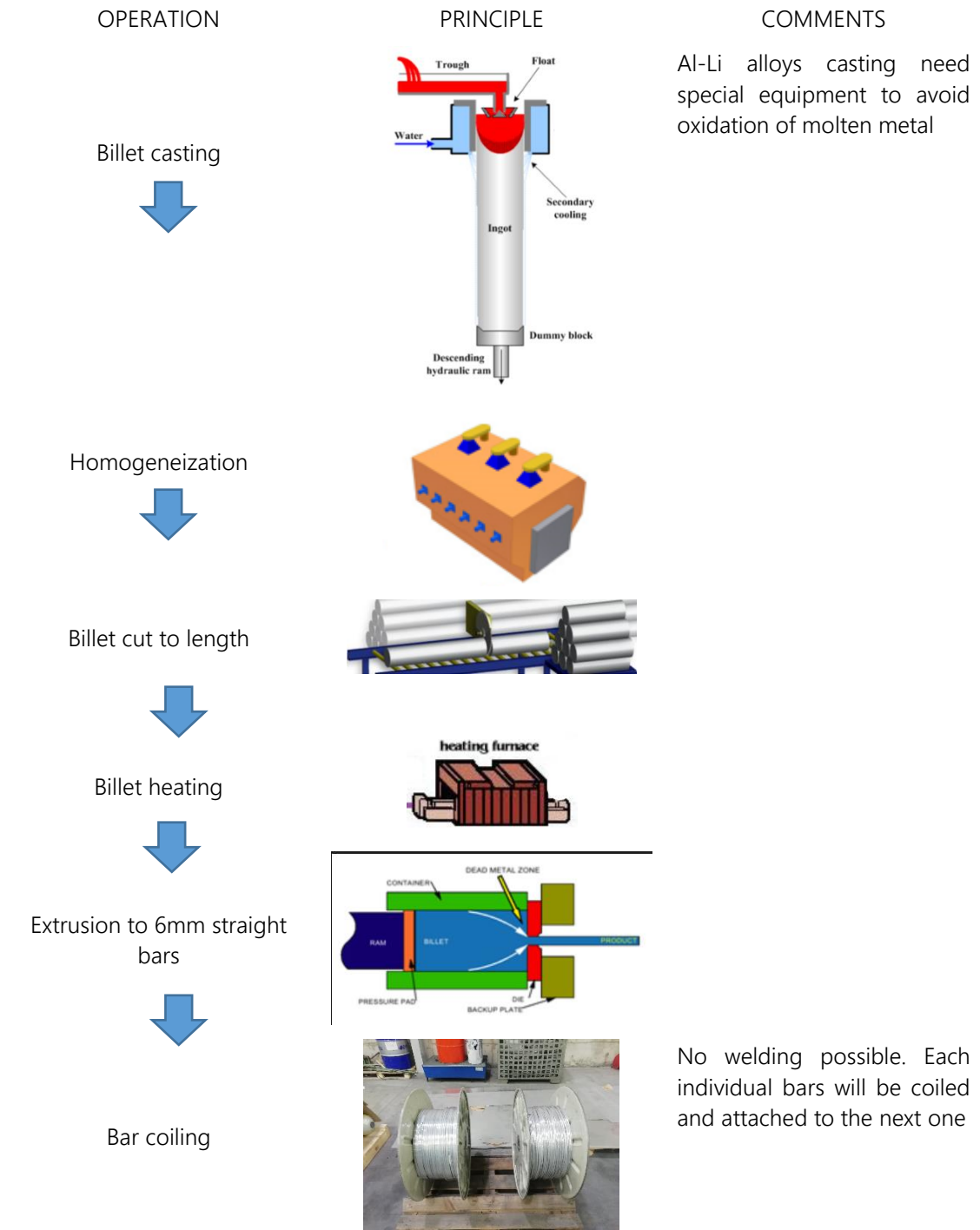


Figure 6 : principle of coil manufacturing at UAC

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