



(51) International Patent Classification:

C22C 21/06 (2006.01) C22C 1/04 (2023.01)  
B33Y 70/00 (2020.01) C22C 21/00 (2006.01)  
B33Y 80/00 (2015.01)

(21) International Application Number:

PCT/EP2024/079827

(22) International Filing Date:

22 October 2024 (22.10.2024)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

2316254.8 24 October 2023 (24.10.2023) GB

(71) Applicant: **ALLOYED LIMITED** [GB/GB]; Unit 15, Oxford Industrial Park, Yarnton, Kidlington, Oxfordshire OX5 1QU (GB).

(72) Inventors: **ROSE, Rory Hans Jacob**; c/o Alloyed Limited, Unit 15 Oxford Industrial Park, Yarnton, Kidlington,

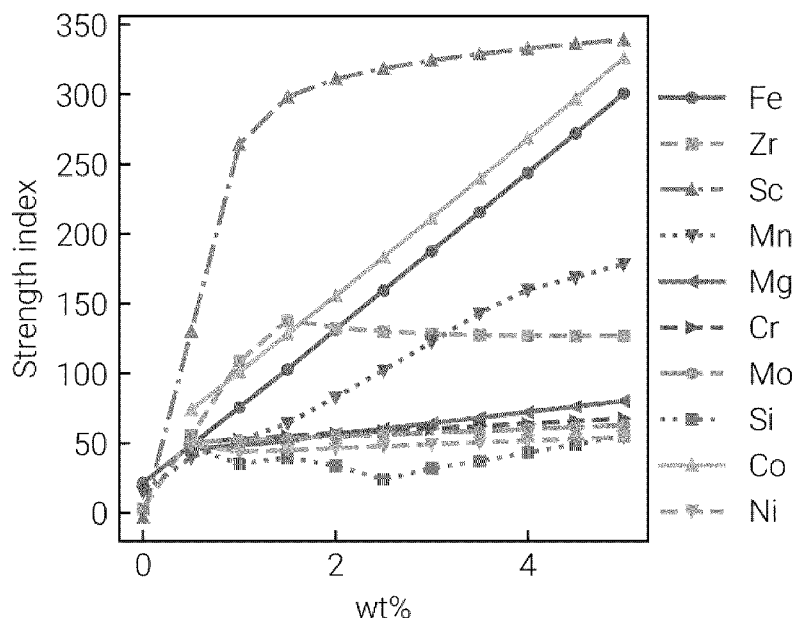
Oxfordshire OX5 1QU (GB). **FORGHANI, Farsad**; c/o Alloyed Limited, Unit 15 Oxford Industrial Park, Yarnton, Kidlington, Oxfordshire OX5 1QU (GB). **KHORSAND, Shohreh**; c/o Alloyed Limited, Unit 15 Oxford Industrial Park, Yarnton, Kidlington, Oxfordshire OX5 1QU (GB). **PEARSON, Charles Nicholas**; c/o Alloyed Limited, Unit 15 Oxford Industrial Park, Yarnton, Kidlington, Oxfordshire OX5 1QU (GB). **ADOLE, Onuh John**; c/o Alloyed Limited, Unit 15 Oxford Industrial Park, Yarnton, Kidlington, Oxfordshire OX5 1QU (GB). **HALL, Dylan Nesta**; c/o Alloyed Limited, Unit 15 Oxford Industrial Park, Yarnton, Kidlington, Oxfordshire OX5 1QU (GB).

(74) Agent: **J A KEMP LLP**; 80 Turnmill Street, London EC1M 5QU (GB).

(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG,

(54) Title: ALUMINIUM ALLOY

Fig. 1



(57) Abstract: An aluminium alloy comprising: in weight percent: 0.45 to 1.1 wt.% iron; at least two of erbium, hafnium, niobium, titanium, scandium, zirconium in an amount of up to 0.6wt% each; and erbium, hafnium, titanium, scandium, zirconium in total sum of 0.4 to 1.2wt; 0.5 to 2.5 wt.% magnesium; 0.9 to 3.0 sum of one or more of the elements selected from the list: manganese, chromium, molybdenum, vanadium, cobalt; 0.0 to 0.45 sum of one or more of the elements selected from the list: silicon, copper, calcium, nickel; the balance being aluminium and unavoidable impurities with a total of less than 0.5 wt.%.

WO 2025/087901 A1

KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY,  
MA, MD, MG, MK, MN, MU, MW, MX, MY, MZ, NA,  
NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO,  
RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH,  
TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS,  
ZA, ZM, ZW.

**(84) Designated States** (*unless otherwise indicated, for every kind of regional protection available*): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SC, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

**Published:**

- *with international search report (Art. 21(3))*
- *in black and white; the international application as filed contained color or greyscale and is available for download from PATENTSCOPE*

## ALUMINIUM ALLOY

**Introduction**

5 It has been difficult to develop high-temperature aluminium alloys suitable for additive manufacturing. This has been due to a combination of difficulty in controlling cracking during manufacturing and controlling spatter for dispersion strengthened alloys. Cracking during manufacturing can in part be controlled by careful process optimisation and product geometry alterations to avoid large cracks. However, these techniques have limits and an alloy which has  
10 less susceptibility to cold cracking during additive manufacturing is desired. Spattering occurs when metal droplets are discharged on the metal surface during additive manufacturing. The droplets solidify and become stuck on the metal surface after cooling down. This can lead to the introduction of defects in the product.

15 AlSi01Mg is an aluminium alloy comprising about 10 wt.% silicon and about 0.5 wt.% magnesium. This alloy is well suited to additive manufacturing and exhibits little spatter. However, the high-temperature (300°C) yield strength and ultimate tensile strength are disappointing at about 70 MPa.

20 Scalmalloy has very high room temperature yield strength and ultimate tensile strength after T5 aging (e.g. at 325°C for Scalmalloy ) conducted for a few hours (e.g. 4 hours). The microcracking performance of this alloy during additive manufacturing is reasonable, however the alloy suffers from poor spatter performance. Additionally, the high-temperature yield strength and ultimate tensile strength are disappointing. Scalmalloy has less than 0.2 wt.% iron,  
25 about 0.3 wt.% zirconium, about 0.7 wt.% scandium, and about 5 wt.% magnesium with a very small addition of silicon.

The present invention relates to an aluminium alloy which has a unique balance of high-temperature strength combined with resistance to microcracking during additive manufacturing  
30 and spatter performance.

The present invention provides an aluminium alloy comprising (or consisting of): 0.45 to 1.1 wt.% iron; at least two of erbium, hafnium, niobium, titanium, scandium, zirconium in an amount of up to 0.6wt% each; and erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 0.4 to 1.2wt; 0.5 to 2.5 wt.% magnesium; 0.9 to 3.0 sum of one or more of the elements selected from the list: manganese, chromium, molybdenum, vanadium, cobalt; 0.0 to 0.45 sum of one or more of the elements selected from the list: silicon, copper, calcium, nickel; the balance being aluminium and unavoidable impurities with a total of less than 0.5 wt.%.  
5

In an embodiment the alloy comprises 2.0 wt.% or less magnesium, preferably 1.9 wt.% or less magnesium, more preferably 1.8 wt.% or less magnesium, even more preferably 1.6 wt.% or less magnesium and most preferably 1.5 wt.% or less magnesium. Such an alloy is more resistant to spatter.  
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In an embodiment the alloy comprises 0.55 wt.% or more magnesium, preferably 1.0 wt.% or more magnesium, more preferably 1.35 wt.% or more magnesium, most preferably 1.4 wt.% or more magnesium. Such an alloy has higher strength.  
15

In an embodiment the alloy comprises 0.55 wt.% or less scandium, preferably 0.5wt.% or less scandium and more preferably 0.475 wt.% of scandium. Such an alloy has higher high temperature strength after heat treatment.  
20

In an embodiment the alloy comprises 0.55 wt.% of zirconium, preferably 0.5wt.% or less zirconium and more preferably 0.475 wt.% of zirconium. Such an alloy has higher high temperature strength after heat treatment.  
25

In an embodiment the alloy comprises 0.95 wt.% or less iron, preferably 0.9 wt.% or less iron, preferably 0.8 wt.% or less iron, more preferably 0.75 wt.% or less iron, most preferably 0.7 wt.% or less iron. Such an alloy is more resistant to cold cracking.

In an embodiment the alloy comprises 1.0 wt.% or more of sum of one or more of the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, preferably 1.1 wt.% or more sum of one or more of the elements in the list: manganese, chromium, molybdenum, vanadium,  
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cobalt, more preferably 1.5 wt.% or more sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt. Such an alloy has increased strength.

5 In an embodiment the alloy comprises 2.5 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, preferably 2.25 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, more preferably 2.0 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, and most preferably 1.9 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt. Such an alloy increased ductility and so suffers even less from cold cracking.

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In an embodiment the alloy comprises 0.2wt.% or more scandium, preferably 0.25 wt.% or more scandium, more preferably 0.3 wt.% or more scandium, more preferably 0.35 wt.% or more scandium, most preferably 0.4 wt.% for more scandium. Such an alloy has increase strength without suffering an increase in cold cracking.

15

In an embodiment the alloy comprises 0.2wt% or more scandium, preferably 0.25 wt.% or more zirconium, more preferably 0.3 wt.% or more zirconium, more preferably 0.35 wt.% or more zirconium, most preferably 0.4 wt.% for more zirconium. Such an alloy has increase strength without suffering an increase in cold cracking.

20

In an embodiment the alloy comprises 0.5 wt.% or more of iron, preferably 0.55 wt.% or more of iron, and most preferably 0.6 wt.% or more of iron. Such an alloy has increased strength.

25 In an embodiment the alloy comprises a sum of zirconium and scandium in wt.% of 0.4wt.% or more, preferably 0.6 wt.% or more, more preferably 0.8 wt.% or more and most preferably 0.85 wt.% or more. Such an alloy has increased strength.

In an embodiment the alloy comprises scandium and zirconium. These are the most preferred elements to offer strengthening after heat treatment but which advantageously tend to stay mostly in solid solution during additive manufacture.

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In an embodiment the alloy comprises at least two of erbium, hafnium, niobium, titanium, scandium, zirconium in an amount of 0.55wt.% or more each, preferably of 0.5wt.% or more each and most preferably of 0.475wt.% or more each. Such an alloy has increased strength.

- 5 In an embodiment the alloy comprises erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 0.5wt% or more, preferably containing erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 0.6wt% or more, more preferably containing erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 0.7wt% or more, more preferably containing erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of  
10 0.8wt% or more, most preferably containing erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 0.85wt% or more. Such an alloy has increased strength.

- In an embodiment the alloy comprises erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 1.1wt% or less, preferably containing erbium, hafnium, niobium, titanium,  
15 scandium, zirconium in total sum of 1.0wt% or less, more preferably containing erbium, hafnium, niobium, titanium, scandium, zirconium in total sum of 0.95wt% or less. Such an alloy is less likely to suffer from significant Al<sub>3</sub>X precipitation during additive manufacture.

- In an embodiment where of those elements of erbium, hafnium, niobium, titanium, scandium and  
20 zirconium present, at least two, preferably three, are present in an amount of 0.25wt.% or more each, preferably wherein of those elements of erbium, hafnium, niobium, titanium, scandium and zirconium present, at least two, preferably three, are present in an amount of 0.3wt% or more each and most preferably wherein of those elements of erbium, hafnium, niobium, titanium, scandium and zirconium present, at least two, preferably three, are present in an amount of in an  
25 amount of 0.35wt% or more each. Such an alloy has increased strength.

- In an embodiment the microstructure of the alloy has a uniform dispersion of transition metal dispersoids, with the majority of any scandium, zirconium, erbium, hafnium, niobium and titanium in solid solution with optionally some primary precipitation of Al<sub>3</sub>X where X is one or  
30 more of scandium, zirconium, erbium, hafnium, niobium and titanium. Such an alloy is less likely to suffer from cold cracking but yet have high strength.

In an embodiment the microstructure of the alloy comprises transition metal dispersoids and a dispersion of Al<sub>3</sub>X precipitates homogeneously through the microstructure where X is one or more of scandium, zirconium, erbium, hafnium, niobium and titanium with optionally some primary precipitation of Al<sub>3</sub>X where X is one or more of scandium, zirconium, erbium, hafnium, niobium and titanium. Such an alloy has high temperature strength yet can be manufactured by additive manufacturing followed by heat treatment, without cold cracking.

The term “consisting of” is used herein to indicate that 100% of the composition is being referred to and the presence of additional components is excluded so that percentages add up to 100% by weight. Unless states otherwise, all quantities of alloying elements in the aluminium alloy are given in weight percent.

The invention will be more fully described, by way of example only, with reference to the accompanying drawings in which:

15

Figure 1 is a graph showing a calculated strength index of aluminium alloys with addition of various elements in varying quantities

Figure 2 shows results showing the increase in yield strength (converted from hardness) with increasing iron content in an aluminium-iron alloy;

Figure 3 is a micrograph showing a crack in an additively manufactured article from an aluminium alloy comprising 1.2 wt% iron, balance aluminium;

Figure 4 shows results showing the yield strength converted from hardness in a different alloys comprising aluminium, magnesium, scandium and zirconium. The x axis represents the sum of scandium and zirconium in the alloy and the y axis the yield strength;

Figure 5 shows formation of primary Al<sub>3</sub>X precipitates in an aluminium-0.7 wt.% Sc – 0.6 wt.% Zr (ABD-S420) in the as additive manufactured state;

30

Figure 6 shows results showing the increase in yield strength (converted from hardness) with increasing magnesium content in an aluminium-magnesium alloy;

Figure 7 shows the variation in solidus temperature for a series of aluminium alloys alloyed with different elements and in different amounts where 0 wt.% represents the composition in the range of example alloy 1 (M420) without any of the element. For example M420 without magnesium has solidus around 650 °C and boiling point around 2500 °C;

Figure 8 shows the variation in boiling temperature of aluminium alloys when alloyed with different elements and in different amounts where 0 wt.% represents the composition in the range of example alloy 1 (M420) without any of the element as in figure 7;

Figure 9 shows on the y axis the influence on elongation in an aluminium alloy of several different elements in varying amounts on the x axis;

Figure 10 shows results showing the increase in yield strength (converted from hardness) with increasing manganese content in an aluminium-manganese alloy;

Figure 11 shows results showing the increase in yield strength (converted from hardness) with increasing cobalt content in an aluminium-cobalt alloy;

Figure 12 shows experimental results of variation in yield strength with temperature for different alloys of aluminium in different heat-treated states.

## **25 The Present Invention**

High-temperature aluminium alloys (for example aluminium alloys with a yield strength of above 175 MPa at 250°C) are generally dispersion strengthened alloys in which micro-scale dispersoids are uniformly distributed in the microstructure. In the as formed state, this alloy has a uniform dispersion of sub-micron transition metal dispersoids through the melt pool. These phases are characterised using scanning electron microscopy with mean (area) equivalent circle diameter of less than 1µm, preferably less than 500nm and more preferably less than 200nm. The

majority of Sc and Zr are in solid solution with potentially some primary precipitation of Al<sub>3</sub>X. In the T5 heat treated condition, the dispersoids are slightly coarsened and there is a fine dispersion of nanoscale Al<sub>3</sub>X precipitates homogeneously through the microstructure (i.e. these phases are characterised using scanning electron microscopy with mean (area) equivalent circle diameter of less than 10nm and more preferably less than 5nm). Mg is always in solid solution in both conditions.

Forming products and articles from high-temperature aluminium alloys by additive manufacturing has proved difficult. This is because the dispersoid intermetallics reduce elongation which can lead to cracking. Hot cracks can develop during solidification of deposited alloy and typically propagate between grains of the material. Hot cracks are primarily controlled by the solidification behaviour of the alloy including the solidification range and size of grains. Alloys with wide solidification ranges and large grains tend to suffer more from hot cracking. Hot cracking can be prevented in the present alloy using conventional approaches. Cold cracks (sometimes called delayed cracks) on the other hand are less understood and develop after solidification, form as a result of residual stresses caused by thermal contraction and propagate between both grains and through grains. Cold cracks are particularly important for producing complex and large geometries as residual stresses build up as more material is printed and are exacerbated by sharp cross section changes which concentrate stress.

20

An additional difficulty with aluminium alloys for additive manufacture is spatter performance.

The present inventors have developed several insights in aluminium alloy design to enable a high-temperature aluminium alloy to be formed by additive manufacturing with improved resistance to cracking and with lower chance of spatter.

25

The insights made by the inventors include:

- (i) Alloying elements are added to strengthen aluminium through the formation of dispersoids to increase high temperature strength
- (ii) Alloying elements are limited to avoid a large drop in ductility (which would otherwise result in cold cracking)

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- (iii) Limiting certain elements so as to limit the formation of Al<sub>3</sub>X type dispersoids during additive manufacturing, allowing the Al<sub>3</sub>X precipitates to form on subsequent T5 heat treatment
- (iv) reducing solidus temperature to reduce residual stress and thereby cold-cracking
- 5 (v) increasing work hardening rate also to reduce cold-cracking
- (vi) limiting certain elements to maintain a high boiling point to reduce spatter
- (vii) Controlling certain elements so as to reduce the coefficient of thermal to mitigate cold cracking.

10 The inventors have devised a composition which utilises the above insights (which in some cases compete with each other) to develop the aluminium alloy of the present invention.

Generally adding alloying elements to aluminium typically introduces dispersoids and this results in an increase in high-temperature strength. Figure 1 is a graph showing predicted  
15 strength index which was generated based on experimental results and literature data which considers microstructural characteristics of the alloys, indicating the relative strengthening effect of several common alloying elements, within the composition space considered, used to increase the strength of aluminium through the formation of dispersoids. This graph and the results in figures 2,4,6,10 and 11 are for specimens prepared by casting and scanning a laser on the cast  
20 surface to imitate the process microstructure and cooling rate of additive manufacture

Reasons for introducing or not each of the elements listed, and in what quantity, is discussed below.

25 Starting first with iron, iron is a common alloying element in aluminium alloys. Figure 2 shows how increasing amounts of iron at least up to 4% significantly increases the yield strength of an aluminium alloy. A minimum amount of 0.45 wt.% iron is added to the alloy of the present invention to take advantage of iron's strong influence on tensile strength. Preferably iron is present in an amount of 0.50 wt% or more, and more preferably 0.55 wt% or more, or even 0.60  
30 wt% or more, further to increase the tensile strength of the alloy.

On the face of it therefore, increasing the iron content of the aluminium alloy is advantageous. Table 2 confirms that for alloy Al-1.2Fe, a reasonable high-temperature yield strength and ultimate tensile strength is achieved for an alloy with 1.2wt.% iron. However, to print large and complex components, the present inventors have found that increasing the amount of iron in the alloy can result in cracking due to the formation of dispersoids during the additive manufacturing process which leads to the reduction in ductility. Iron was also found to increase in the thermal expansion coefficient; increasing residual stresses. Figure 3 is micrograph of a cold crack which developed in an aluminium alloy comprising 1.2 wt.% iron (example Al-1.2Fe of tables 1 and 2) additively manufactured. Therefore, in the present invention the amount of iron is limited to 1.1 wt% or less at which level microcrack due to cold cracking are not expected to form. Example alloy ABD-M420, which has an amount of iron of 0.72 wt.%, is shown in Table 2 to achieve an acceptable level of microcrack tolerance (0.8). In an embodiment, the amount of iron is preferably reduced further to a maximum of 0.95 wt.% or less or even 0.8wt.% or less or 0.75 wt.% or less.

The present inventors have sought ways further to increase the high-temperature yield strength and ultimate tensile strength and in order to do this, the inventors investigated adding further alloying elements to add to the aluminium-iron alloy.

As can be seen from Figure 1, scandium and zirconium strongly increase the strength of an aluminium alloy, particularly at low concentrations. Figure 4 shows the measured yield strength of various aluminium-magnesium-zirconium-scandium alloys plotted against the sum of scandium and zirconium content. This shows a more or less linear increase in yield strength with sum of scandium and zirconium, due to dispersoid formation.

However, additive manufacturing experiments have shown that if either scandium and zirconium are present in an amount of more than 0.6 wt.% this can lead to microcracking due to cold cracking and can lead to the formation of large primary Al<sub>3</sub>X dispersoids which limits the beneficial strengthening effect of Sc and Zr while increasing elemental cost significantly. This is apparent from the results in table 2, particularly comparing ABD-S420 and ABD-S450, both with low iron content, but having much poorer microcracking tolerance than ABD-M420. The inventors have determined that this due to the precipitation of Al<sub>3</sub>X type precipitates during the

additive manufacturing process leading to cold-cracking because of the resulting reduction in ductility. Figure 5 is a micrograph showing the presence of Al<sub>3</sub>X type precipitates in example ABD-S420 in the as formed additive manufactured state.

5 However, surprisingly, the inventors have found that if scandium and zirconium are each limited to 0.6 wt%, Al<sub>3</sub>X type precipitates do not appear to be stable during the additive manufacturing process (the metastable state is that scandium and zirconium remain in solid solution). This means that the aluminium alloy can be formed into a product using additive manufacturing without precipitation of scandium and zirconium dispersoids, thereby meaning that scandium  
10 and zirconium do not contribute to a reduction in ductility during additive manufacturing.

The scandium and zirconium can then be formed into nanoscale precipitates during T5 heat treatment (e.g. 400 C for 4h for ABD-M420). That is, the metastable state at concentrations of up to 0.6 wt.% of scandium and up to 0.6wt.% of zirconium appears to be solid solution. On the  
15 basis of this understanding it has been possible for the inventors to take advantage of the good strengthening properties of zirconium and scandium (figures 1 and 4) without deleteriously impacting cold-cracking performance during additive manufacturing. Preferably, scandium is present in an amount of 0.55 wt.% or less or 0.5wt.% or less and/or zirconium is preferably present in an amount of 0.55 wt.% or less or 0.5wt.% or less. This yet further reduces the  
20 probability of precipitation of dispersoids resulting from the presence of those elements during additive manufacturing and therefore reduces the likelihood of cold-cracking. For the same reason, in a further preferred embodiment, scandium is limited to 0.475 wt.% or less and/or zirconium is limited to 0.475 wt.% or less.

25 In order to increase the high-temperature YS and UTS (and also increase the room-temperature YS and UTS) it is desirable to include zirconium in an amount of 0.2 wt.% or more and scandium in an amount of 0.2 wt.% or more. Preferably, each element is present in an amount of 0.25 wt.% or more, and more preferably 0.30 wt.% or more. In further embodiments, scandium and/or zirconium is present in an amount of 0.35 wt.% or more. Figure 4 shows that such an  
30 alloy results in increased strength. Preferably one or the other (most preferably both) amounts of scandium and zirconium are high. Thus preferably the aluminium alloy comprises a sum of

zirconium and scandium in wt.% of 0.6 wt.% or more, preferably 0.8 wt.% or more and most preferably 0.85 wt.% or more.

Erbium, hafnium, niobium and titanium are expected to behave in a similar way to Sc and Zr, namely to stay in solid solution at low concentrations during additive manufacturing and to precipitate during T5 heat treatment to form nanoscale precipitates and thereby significantly increase high temperature strength. Therefore, in an embodiment, the alloy contains at least two of erbium, hafnium, titanium, scandium, niobium and zirconium in an amount of up to 0.6wt% each, and a sum of erbium, hafnium, titanium, scandium, niobium and zirconium in of 0.4 to 1.2wt%. In another embodiment, the alloy contains at least two of erbium, hafnium, titanium, scandium and zirconium in an amount of up to 0.6wt% each, and a sum of erbium, hafnium, titanium, scandium and zirconium in of 0.4 to 1.2wt%. Scandium and zirconium are the most preferred of those alloying elements. The best properties are achieved by Sc and Zr due to a core shell type precipitate where advantage is taken of the combination of very high nucleation rate from Sc and a low growth rate due to the Zr shell. In an embodiment of those elements of erbium, hafnium, titanium, scandium, niobium and zirconium present, at least two, preferably three, are present in an amount of 0.25wt.% or more each, preferably 0.3wt.% or more each and most preferably in an amount of 0.35wt.% or more each. In an embodiment of those elements of erbium, hafnium, titanium, scandium and zirconium present, at least two, preferably three, are present in an amount of 0.25wt.% or more each, preferably 0.3wt.% or more each and most preferably in an amount of 0.35wt.% or more each. In an embodiment the aluminium alloy comprises a sum of erbium, hafnium, titanium, scandium, niobium and zirconium in wt.% of 0.5wt.% or more, preferably 0.6 wt.% or more, more preferably 0.7wt.% or more, more preferably 0.8 wt.% or more and most preferably 0.85 wt.% or more. In an embodiment the aluminium alloy comprises a sum of erbium, hafnium, titanium, scandium and zirconium in wt.% of 0.5wt.% or more, preferably 0.6 wt.% or more, more preferably 0.7wt.% or more, more preferably 0.8 wt.% or more and most preferably 0.85 wt.% or more. As the sum increases, the high temperature strengthening effect after heat treatment increases.

Because the presence of these elements increases high temperature strength after heat treatment, preferably the aluminium alloy contains aluminium alloy contains at least two of erbium, hafnium, titanium, scandium, niobium zirconium in an amount of 0.55wt.% or more each,

preferably of 0.5wt.% or more each and most preferably of 0.475wt.% or more each. Preferably the aluminium alloy contains aluminium alloy contains at least two of erbium, hafnium, titanium, scandium, zirconium in an amount of 0.55wt.% or more each, preferably of 0.5wt.% or more each and most preferably of 0.475wt.% or more each.

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In an embodiment the aluminium alloy contains erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 1.1wt% or less, preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 1.0wt% or less, more preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.95wt% or less. In an  
10 embodiment the aluminium alloy contains erbium, hafnium, titanium, scandium, zirconium in total sum of 1.1wt% or less, preferably containing erbium, hafnium, titanium, scandium, zirconium in total sum of 1.0wt% or less, more preferably containing erbium, hafnium, titanium, scandium, zirconium in total sum of 0.95wt% or less. Such alloys have reduced propensity to form Al<sub>3</sub>X type primary precipitates. The formation of such precipitates is preferably avoided as they  
15 tend to use up these elements and so reduce the precipitation of fine Al<sub>3</sub>X precipitates after heat treatment.

Particularly where the majority of the strengthening effect after heat treatment is achieved by the presence of scandium and/or zirconium, erbium, hafnium, niobium, titanium can be present in a  
20 sum of 0.2wt% or less, preferably in a sum of erbium, hafnium, niobium, titanium of 0.1wt% or less, more preferably in a sum of erbium, hafnium, niobium, titanium of 0.05wt% or less, and in an embodiment even being substantially free of erbium, hafnium, niobium and titanium. This is advantageous as hafnium is expensive and as mentioned elsewhere, the combination of zirconium and scandium achieves very fine precipitates. In an embodiment the alloy is  
25 substantially free of niobium except the extent to which it is present as an unavoidable impurity. In such an alloy, where the majority of the strengthening effect after heat treatment is achieved by the presence of scandium and/or zirconium, erbium, hafnium, titanium can be present in a sum of 0.2wt% or less, preferably in a sum of erbium, hafnium, titanium of 0.1wt% or less, more preferably in a sum of erbium, hafnium, titanium of 0.05wt% or less, and in an embodiment even  
30 being substantially free of erbium, hafnium and titanium.

Figure 1 shows the beneficial effect of magnesium on the ultimate tensile strength and it provides strength without sacrificing ductility. Many commercial aluminium alloys contain magnesium because of this effect. Figure 6 shows experimentally the increase in yield strength (based on a conversion from measured hardness) achieved by adding increasing amounts of magnesium into aluminium.

Therefore, the present invention contains at least 0.5 wt.% magnesium in order to take advantage of its good strengthening properties. Magnesium also has the effect of reducing the solidus and increasing the work hardening rate of the alloy, which is beneficial in avoiding cold-cracking. However, excessive amounts of magnesium have been found by the present inventors to result in poor spatter performance. This can be seen in Table 2 in the case of AlMgTy90 which contains 13.8 wt.% magnesium and in scalmalloy which contains 4.7 wt.% magnesium. The inventors have determined that the mechanism for spatter appears to be the formation of vapour plumes and these result in spatter defects. The vapour plumes occur due to the lowering of the boiling temperature of the alloy resulting from the addition of magnesium. The effect of reduction in boiling point by magnesium compared to other alloying elements for ABD-M420 (see table 1) is illustrated in Figure 8 which is generated using CALPHAD predictions of the temperature for 1% of Gas phase assuming equilibrium conditions.

The present inventors have determined that an amount of 2.5 wt.% or less magnesium avoids the difficulty with spatter. Preferably, magnesium is limited to 2.0 wt.% or less. It can be seen from Figure 8 that reducing the magnesium content even further will further increase the boiling point of the alloy, thereby reducing the chance of spatter. Therefore, reducing the magnesium content to 1.9 wt.% or less or 1.8 wt.% or less or even 1.6 wt.% or less will have advantages in reducing the chance of spatter. In an embodiment, magnesium is limited to 1.5 wt.% or less. As can be seen in Figure 8, at such a concentration the deleterious effect of magnesium in lowering the boiling temperature of the alloy is the same as that of the other main alloying elements used in the present invention.

On the other hand, Figure 7 (generated in the same way as figure 8 and for ABD-M420 compositional variations) shows the reducing effect magnesium has on the solidus temperature. The inventors have found that magnesium has an even greater effect on reducing the non-

equilibrium solidus temperature than the equilibrium solidus illustrated in figure 7. This appears to be the result of the change and shape of the Sheil curve. This lowers the residual stress in the additively manufactured product, thereby resulting in less cold-cracking. For this reason and to take advantage of the additional strengthening effect of magnesium, magnesium is present in an amount of 0.5 wt.% or more. Adding more magnesium further increases those effects so that preferably magnesium is added in an amount of 0.55 wt.% or more and more preferably 1.0 wt.% or more. In an embodiment magnesium is added in an amount of 1.35 wt.% or more to make the most of the advantages in reducing cold-cracking offered by magnesium additions.

Furthermore, Figure 9 shows the negative impact of elongation (a ductility drop) of the concerned elements. This index is generated by combining experimental and literature data to account for the effect of microstructural characteristics on elongation utilising CALPHAD predicted non-equilibrium microstructures. A drop in ductility is to be avoided as it can lead to cold cracking.

It is desired to add more of the alloying elements of figure 1 in order further to increase the strength of the alloy, so long as other properties are not deleteriously affected.

Figure 9 shows that silicon has the least impact on ductility although it is clear from Figure 1 that silicon also has the least impact on strength. Therefore, the present invention allows an amount of silicon of up to 0.45 wt%, although this is not preferred. Preferably, silicon is present in 0.1 wt.% or less and most preferably is excluded from the alloy because it can react with scandium and zirconium and decreasing the strengthening effect of these elements by stopping the formation of the beneficial Al<sub>3</sub>X precipitates during ageing.

Nickel is also shown as having a low impact on reduction in ductility in Figure 9. However, like silicon, nickel has a small impact on an increase in strength. Therefore, nickel is allowed up to a maximum of 0.45 wt%, preferably of 0.05 wt% or less, and most desirably the alloy is nickel free.

In order further to increase the strength of the aluminium alloy, additions of one or more of manganese, chromium, molybdenum, vanadium and cobalt in an amount of up to 3.0wt.% in

sum are added, preferably up to an amount of 2.5 wt.% in sum are added. Figure 1 shows that cobalt and manganese have the greatest effect of increasing strength of those alloys, although molybdenum and chromium also add a little in strength. Vanadium is expected to act in a similar way to chromium and cobalt. However, of those five elements, manganese and cobalt are preferred. In fact, manganese is more preferred than cobalt because cobalt, similar to molybdenum, has been found to increase the melting point of the aluminium alloy greatly. Increasing the melting point of the alloy makes it much harder to atomise an electrode of the alloy in the process for making the powder used in additive manufacturing.

Molybdenum is shown in figure 9 to have a low impact on elongation, along with manganese and cobalt. However, molybdenum is not as strong a strengthening element as either manganese or cobalt (figure 1) and is therefore not preferred. Chromium has a relatively large impact on reducing ductility (Figure 9) whereas its effect on increasing strength is relatively limited. Therefore, chromium is the least preferred of the group of elements, manganese, chromium, molybdenum, vanadium and cobalt which are present in some in an amount of 3.0wt.% in sum or 2.5 wt% in sum or less. To reduce the impact of reduction in ductility, in an embodiment the sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt is 2.25 wt.% or less, even 2.0 wt.% or less and more preferably 1.9 wt.% or less.

The sum of manganese, chromium, vanadium, cobalt and molybdenum is preferably 0.9 wt% or more in order to achieve a minimum strengthening effect on the alloy. In an embodiment, the sum of those elements is 1.0 wt% or more, preferably 1.1 wt% or more and in one embodiment even 1.5 wt% or more. In an embodiment, manganese is chosen from the group of those five elements and is present in an amount of 0.7 wt% or more and/or 2.0 wt% or less.

Figures 10 and 11 show the effect on yield strength of manganese and cobalt additions to aluminium. Generally, it shows increasing strength with increasing additions. However, the additions of those elements are limited due to their effect on reducing ductility as explained with reference to Figure 9.

Small additions of other elements commonly alloyed with aluminium are permissible. The sum of these additions is 0.45 wt% or less, preferably 0.1 wt.% or less, though in an embodiment the

- alloy is substantially free of additional elements. These additions include the already mentioned silicon and nickel. Other additions could be copper; however, this is an element which forms intermetallics in the as built condition and requires expensive heat treatment to remove them before the formation of beneficial theta phase so that preferably copper is limited to 0.24 wt.% or less and preferably the alloy is copper free and calcium however calcium is likely to reduce the boiling point of the alloy leading to difficulties with spatter and calcium is very reactive in the molten state so it becomes difficult to work with the melt during atomisation. Preferably calcium is present at most at 0.4wt% but preferably the alloy is calcium free.
- 10 In addition, the alloy may contain small amounts of other incidental impurities of any element not listed in the above section. For example, these impurities may be unavoidable impurities which are difficult to remove from aluminium. Such impurities may be present with a total amount of less than 0.5 wt.%.
- 15 The following two tables show some commercially available alloys and some experimental alloys. Table 1 is a list of the alloys with their composition and Table 2 shows the yield strength, tensile strength and elongation at room-temperature and where measured at high-temperature. The occurrence or not of microcracks and spatter defects is also shown. Alloy ABD-M420 is an alloy of the present invention and exhibits high room temperature YS and UTS, very good high temperature YS and UTS and acceptable elongation at both room temperature and high temperature. The occurrence of microcracks during additive manufacture is limited and the spatter performance is good. None of the other alloys listed in the table perform as well in all those areas.

25 **Table 1.** Chemical composition of assessed commercial and experimental alloys (wt.%)

Alloys	Al	Fe	Zr	Sc	Mg	Si	Cu	Ti	Mn
AlSi10Mg	89.12	-	-	-	0.48	10.4	-	-	-
Scalmalloy	93.65	0.14	0.27	0.69	4.7	0.07	-	-	-
F357	92.5	-	-	-	0.5	7	-	-	-
AlMgty 90	86.2	-	-	-	13.8	-	-	-	-
Al-1.2Fe	98.8	1.2	-	-	-	-	-	-	-
ABD_CAM	98.36	-	0.83	0.49	-	-	-	0.32	-
ABD_H50	96.7	1.78	0.57	0.45	-	-	-	-	-
ABD-H251	96.7	1.9	1.4	-	-	-	-	-	-
ABD-H252	95.31	1.9	0.74	0.55	1.5	-	-	-	-
ABD-H253	90.3	3	-	0.9	1	4	-	0.8	-
ABD-S400	95.82	1.47	0.47	0.6	1.64	-	-	-	-

ABD-S420	97.03	0.66	0.6	0.64	0.89			0.18	-
ABD-S450	97.39	0.66	0.33	0.8	0	-	0.48	0.34	-
ABD-M420	95.1	0.72	0.44	0.44	1.42	-	-	-	1.88

**Table 2.** Properties of assessed commercial and experimental alloys

Alloys	YS	YS <sup>1</sup>	UTS	UTS <sup>1</sup>	EI%	EI% <sup>1</sup>	Macro-crack tolerance <sup>2</sup>	Spatter
	23 °C (MPa)	300 °C (MPa)	23 °C (MPa)	300 °C (MPa)	23 °C	300 °C		
AlSi10Mg	204	70	358	73	7.2	41.4	1	Excellent
Scalmalloy <sup>3</sup>	> 480	> 54	> 510**	> 77.5	> 13	-	0.8	Poor
F357 <sup>3</sup>	> 265	-	> 330	-	> 11	-	0.20	Good
AlMgty 90 <sup>3</sup>	> 200	-	> 410	-	> 35	-	0.41	Very poor
Al-1.2Fe	203	142	230	147	16.9	16.6	0.25	Excellent
ABD_CAM	192	43	209	55	16.7	21.9	0.41	Excellent
ABD_H50	250	206.5	296	223	15.95	11.7	0.23	Excellent
ABD-H251	277.5	179.5	323	202	14.6	17.6	0.14	Excellent
ABD-H252	358.5	195	409	201	12.3	8.6	0.14	Good
ABD-H253	413	189	467	222	3.4	21.5	0.10	Good
ABD-S400	391	-	427	-	8.7	-	0.20	Good
ABD-S420	447	-	455	-	7.1	-	0.41	Good
ABD-S450	447	-	392	-	7.1	-	0.35	Excellent
ABD-M420	428	172	477	179	12.5	5.2	0.8	Good

<sup>1</sup> The high-temperature data are reported in as-built conditions, while room temperature data are reported in T5 (direct aged) conditions, unless specified otherwise by asterisks.

<sup>2</sup> The macro-crack tolerance has been normalized to a scale ranging from 0 to 1, where 1 represents the lowest susceptibility to macro-crack formation during the printing process.

<sup>3</sup> The data are from the supplier datasheet.

5

10 Table 3 below lists the limits of the present inventive alloy and preferred limits in wt.% for the main alloying elements, with Ti, V, Si, Cu, Ca and Ni being present in sum up to 0.45wt.%, but the alloys are preferably sum less than 0.1wt.% and most preferably free of Si, Cu, Ca and Ni and in one embodiment free of all six elements. Table 4 lists the even more preferred limits.

Table 3:

	Fe	Sum of two or more of Zr, Sc, Er, Hf, Ti with each limited to 0.6wt% (preferably at least Sc and Zr)	Mg	Sum of Mn, Cr, Co, Mo, V (preferably Mn, Co, most preferably Mn)
Min	0.45	0.4	0.5	0.9
Max	1.1	1.2	2.5	2.5
Preferred min	0.5	0.5	1.0	1.1
Preferred max	0.95	1.0	1.8	2.0
More preferred min	0.6	0.6	1.35	1.5
More preferred max	0.8	0.85	1.5	1.9

Table 4:

	Fe	Zr	Sc	Mg	Mn and/or Co
Min	0.45	0.2	0.2	0.5	0.9
Max	1.1	0.6	0.6	2.5	3.0
Preferred min	0.5	0.25	0.25	1.0	1.1
Preferred max	0.95	0.5	0.5	1.8	2.0
More preferred min	0.6	0.35	0.35	1.35	1.5
More preferred max	0.8	0.475	0.475	1.5	1.9

5 Table 5 lists the limits of another specific preferred alloy.

Table 5:

	Fe	Zr	Sc	Mg	Mn (optionally Co or a Co/Mn mixture instead of Mn)
Min	0.45	0.2	0.2	0.5	0.9
Max	1.1	0.6	0.6	2.5	3.0
Preferred min	0.5	0.3	0.3	1.0	1.4
Preferred max	0.9	0.55	0.55	1.8	2.2
More preferred min	0.6	0.4	0.4	1.4	1.75
More preferred max	0.7	0.5	0.5	1.6	1.95

## CLAIMS

1. An aluminium alloy comprising, in weight percent:  
0.45 to 1.1 wt.% iron;  
5 at least two of erbium, hafnium, titanium, scandium, niobium, zirconium in an amount of up to 0.6 wt.% each; and  
erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.4 to 1.2 wt.%;  
0.5 to 2.5 wt.% magnesium;  
10 0.9 to 3.0 wt.% sum of one or more of the elements selected from the list: manganese, chromium, molybdenum, vanadium, cobalt;  
0.0 to 0.45 wt. sum of one or more of the elements selected from the list: silicon, copper, calcium, nickel;  
the balance being aluminium and unavoidable impurities with a total of less than  
15 0.5 wt.%.
2. The aluminium alloy of claim 1, comprising 2.0 wt.% or less magnesium, preferably 1.9 wt.% or less magnesium, more preferably 1.8 wt.% or less magnesium, even more preferably 1.6 wt.% or less magnesium and most preferably 1.5 wt.% or less magnesium.  
20
3. The aluminium alloy of claim 1 or 2, comprising 0.55 wt.% or more magnesium, preferably 1.0 wt.% or more magnesium, more preferably 1.35 wt.% or more magnesium, most preferably 1.4 wt.% or more magnesium.
- 25 4. The aluminium alloy of any preceding claim, comprising 0.55 wt.% or less scandium, preferably 0.5wt.% or less scandium and more preferably 0.475 wt.% of scandium.
5. The aluminium alloy of any preceding claim, comprising 0.55 wt.% of zirconium, preferably 0.5wt.% or less zirconium and more preferably 0.475 wt.% of zirconium.  
30

6. The aluminium alloy of any preceding claim, comprising 0.95 wt.% or less iron, preferably 0.9 wt.% or less iron, preferably 0.8 wt.% or less iron, more preferably 0.75 wt.% or less iron, most preferably 0.7 wt.% or less iron.
- 5 7. The aluminium alloy of any preceding claim, comprising 1.0 wt.% or more of sum of one or more of the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, preferably 1.1 wt.% or more sum of one or more of the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, more preferably 1.5 wt.% or more sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt.
- 10 8. The aluminium alloy of any preceding claim, comprising 2.5 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, preferably 2.25 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, more preferably 2.0 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt, and most preferably 1.9 wt.% or less sum of one or more the elements in the list: manganese, chromium, molybdenum, vanadium, cobalt.
- 15 9. The aluminium alloy of any preceding claim, comprising 0.2wt.% or more scandium, preferably 0.25 wt.% or more scandium, more preferably 0.3 wt.% or more scandium, more preferably 0.35 wt.% or more scandium, most preferably 0.4 wt.% for more scandium.
- 20 10. The aluminium alloy of any preceding claim, comprising 0.2wt% or more scandium, preferably 0.25 wt.% or more zirconium, more preferably 0.3 wt.% or more zirconium, more preferably 0.35 wt.% or more zirconium, most preferably 0.4 wt.% for more zirconium.
- 25 11. The aluminium alloy of any preceding claim, comprising less than 0.1 wt.% of the sum of one or more of the elements selected from the list: silicon, copper, calcium, nickel.
- 30 12. The aluminium alloy of any preceding claim, wherein the aluminium alloy is substantially free of silicon, copper, calcium, nickel, except as unavoidable impurities.

13. The aluminium alloy of any preceding claim, comprising 0.05 wt.% or less nickel, preferably free of nickel.
14. The aluminium alloy of any preceding claim, comprising 0.25 wt.% or less copper,  
5 preferably free of copper.
15. The aluminium alloy of any preceding claim, comprising 0.5 wt.% or more of iron, preferably 0.55 wt.% or more of iron, and most preferably 0.6 wt.% or more of iron.
- 10 16. The aluminium alloy of any preceding claim, comprising 0.10 wt.% or less of silicon, preferably silicon free.
17. The aluminium alloy of any preceding claim, comprising 0.4 wt.% or less of calcium, preferably calcium free.  
15
18. The aluminium alloy of any preceding claim, comprising a sum of zirconium and scandium in wt.% of 0.4wt.% or more, preferably 0.6 wt.% or more, more preferably 0.8 wt.% or more and most preferably 0.85 wt.% or more.
- 20 19. The aluminium alloy of any preceding claim, comprising scandium and zirconium.
20. The aluminium alloy of any preceding claim, comprising a sum of erbium, hafnium, niobium, titanium of 0.2wt% or less, preferably comprising a sum of erbium, hafnium, niobium, titanium of 0.1wt% or less, more preferably comprising a sum of erbium, hafnium, niobium,  
25 titanium of 0.05wt% or less, preferably being substantially free of erbium, hafnium and titanium.
21. The aluminium alloy of any preceding claim, containing at least two of erbium, hafnium, titanium, scandium, niobium, zirconium in an amount of 0.55wt.% or more each, preferably of 0.5wt.% or more each and most preferably of 0.475wt.% or more each.  
30
22. The aluminium alloy of any preceding claim, containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.5wt% or more, preferably containing erbium,

hafnium, titanium, scandium, niobium, zirconium in total sum of 0.6wt% or more, more preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.7wt% or more, more preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.8wt% or more, most preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.85wt% or more.

23. The aluminium alloy of any preceding claim, containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 1.1wt% or less, preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 1.0wt% or less, more preferably containing erbium, hafnium, titanium, scandium, niobium, zirconium in total sum of 0.95wt% or less.

24. The aluminium alloy of any preceding claim, wherein of those elements of erbium, hafnium, titanium, scandium, niobium and zirconium present, at least two, preferably three, are present in an amount of 0.25wt.% or more each, preferably wherein of those elements of erbium, hafnium, titanium, scandium, niobium and zirconium present, at least two, preferably three, are present in an amount of 0.3wt% or more each and most preferably wherein of those elements of erbium, hafnium, titanium, scandium, niobium and zirconium present, at least two, preferably three, are present in an amount of in an amount of 0.35wt% or more each.

25. The aluminium alloy of any preceding claims, wherein the microstructure of the alloy has a uniform dispersion of transition metal dispersoids, with the majority of any scandium, zirconium, erbium, hafnium, niobium and titanium in solid solution with optionally some primary precipitation of Al<sub>3</sub>X where X is one or more of scandium, zirconium, erbium, hafnium, niobium and titanium.

26. The aluminium alloy of any of claims 1-24, wherein the microstructure of the alloy comprises transition metal dispersoids and a dispersion of Al<sub>3</sub>X precipitates homogeneously through the microstructure where X is one or more of scandium, zirconium, erbium, hafnium, niobium and titanium with optionally some primary precipitation of Al<sub>3</sub>X where X is one or more of scandium, zirconium, erbium, hafnium, niobium and titanium.

27. The aluminium alloy of any preceding claim, wherein any magnesium is in solid solution.

28. The aluminium alloy of any preceding claim, wherein the alloy is substantially free of niobium.

5

29. An article comprising the aluminium alloy of any of the preceding claims.

30. The article of claim 29, wherein the article is an additively manufactured article.

Fig. 1

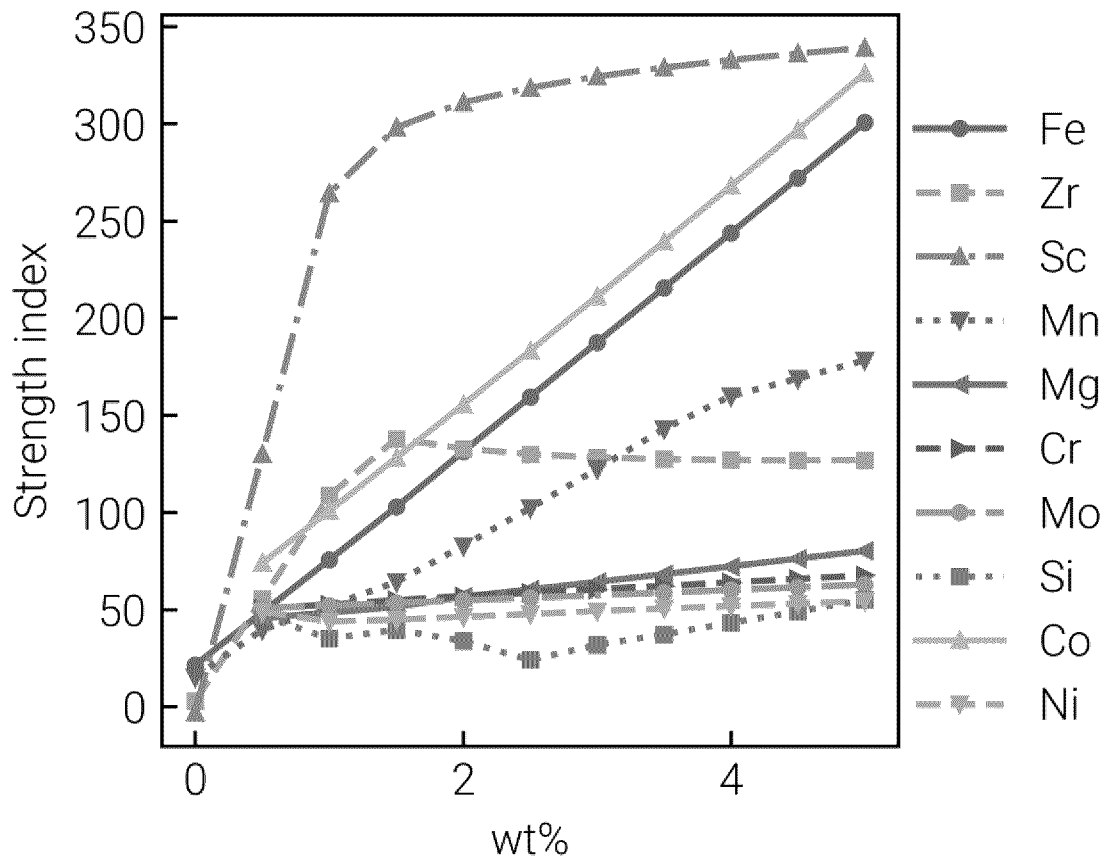


Fig. 2

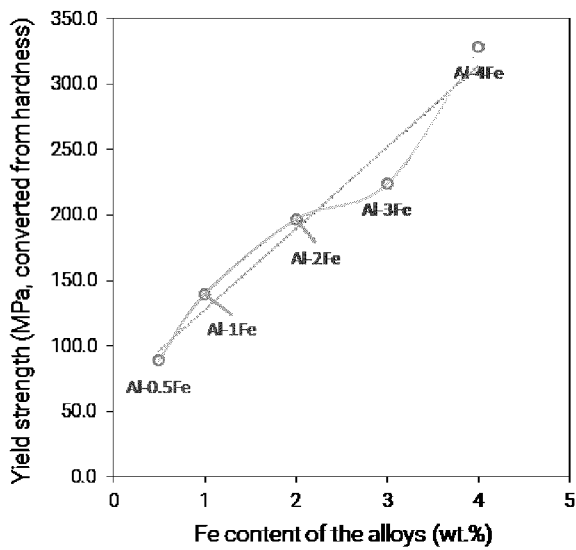


Fig. 3

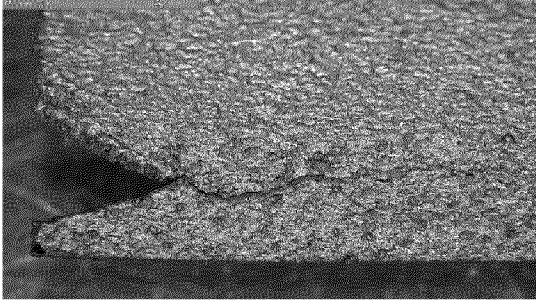


Fig. 4

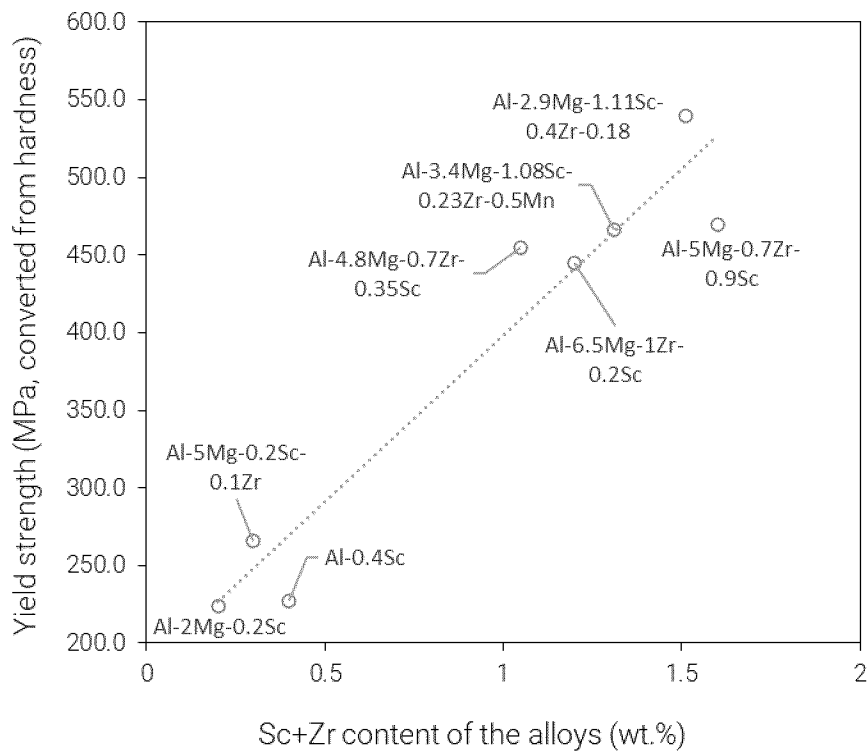


Fig. 5

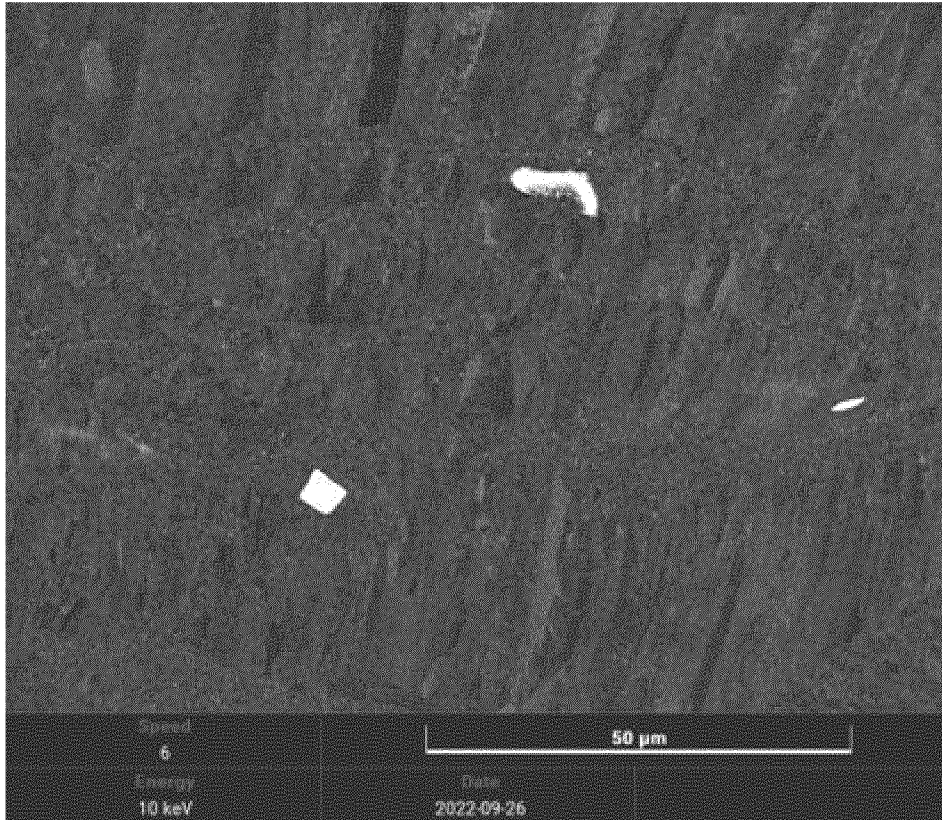


Fig. 6

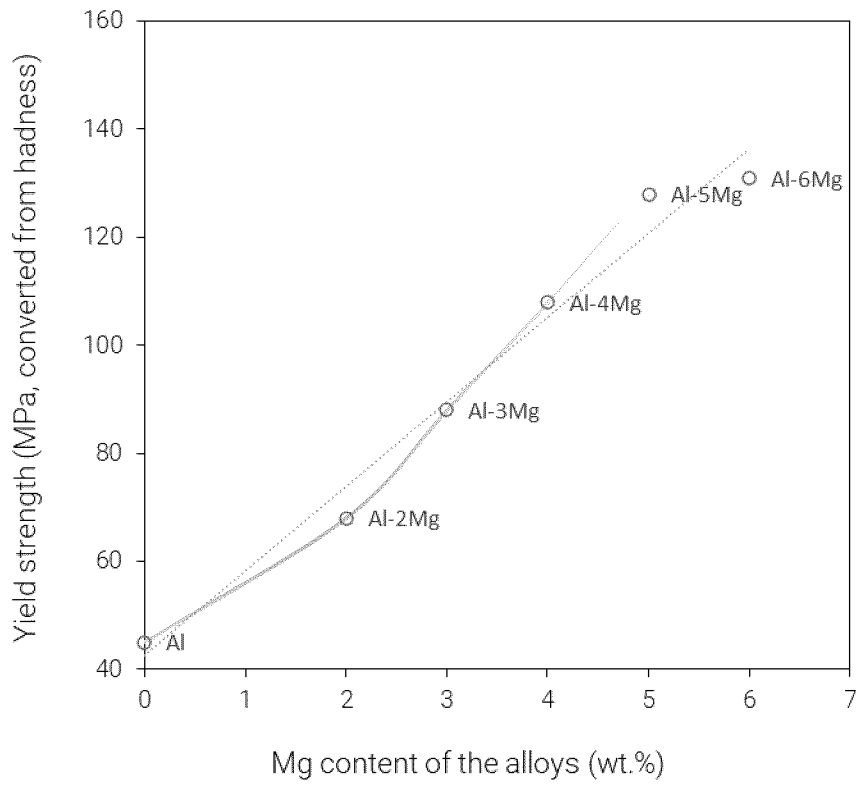


Fig. 7

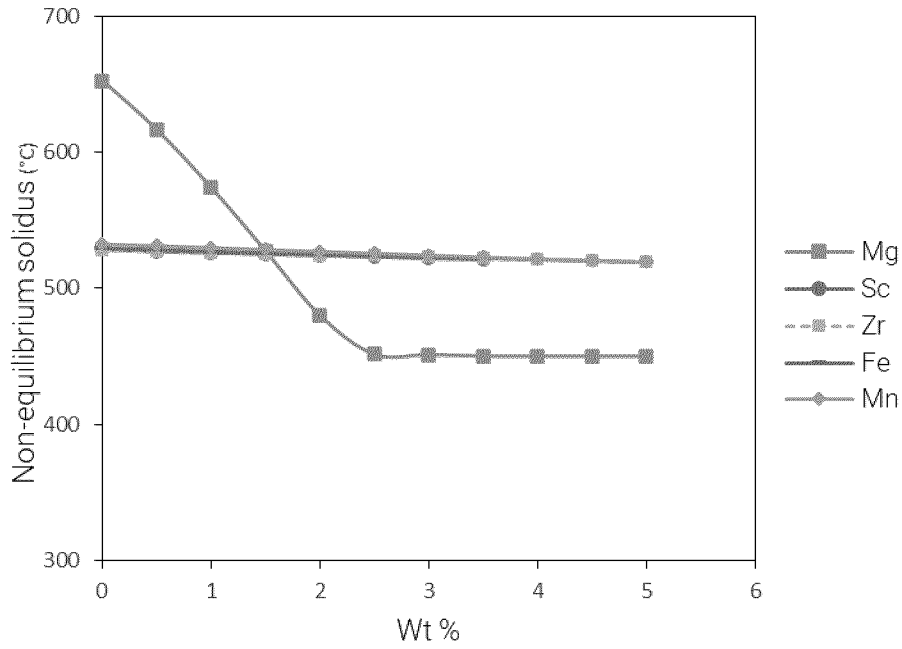


Fig. 8

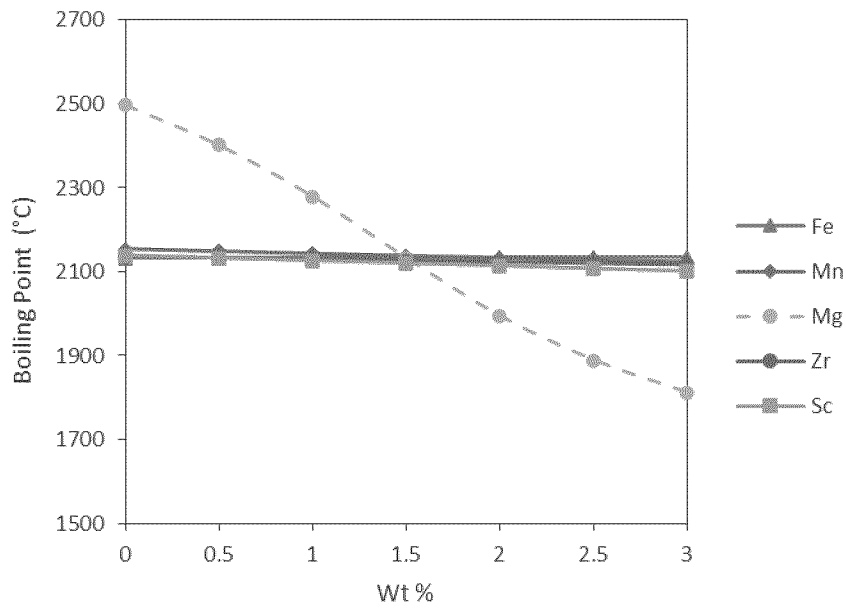


Fig. 9

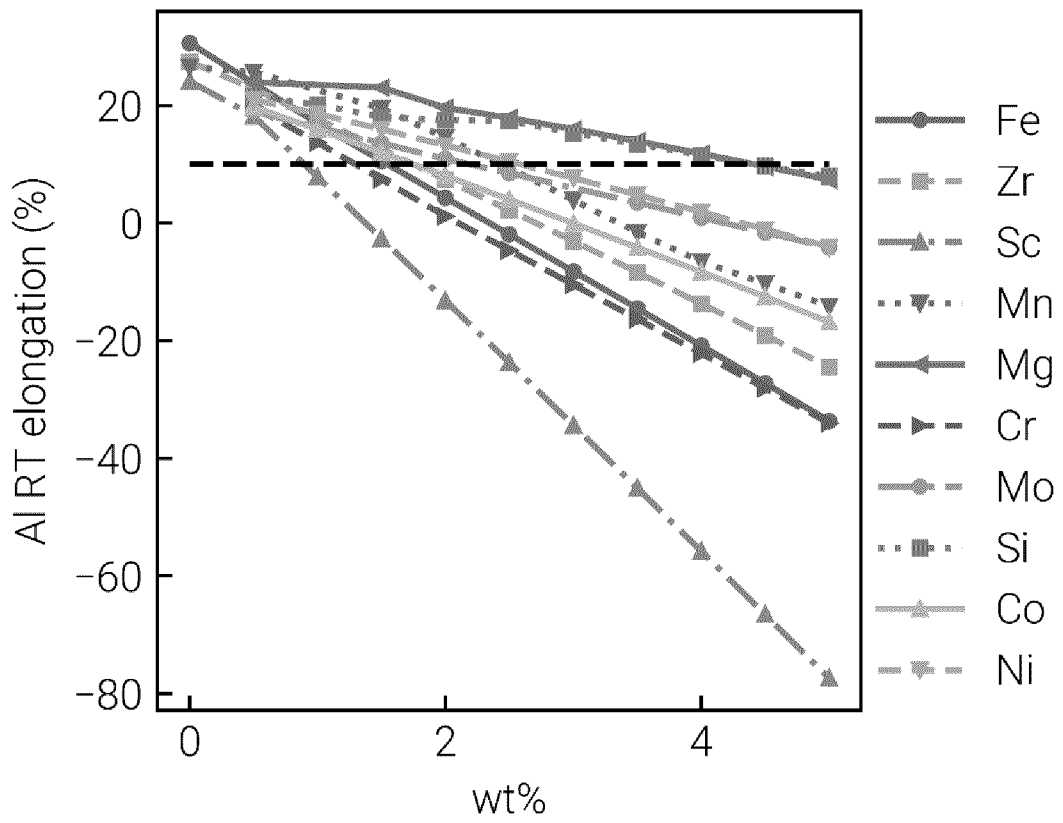


Fig. 10

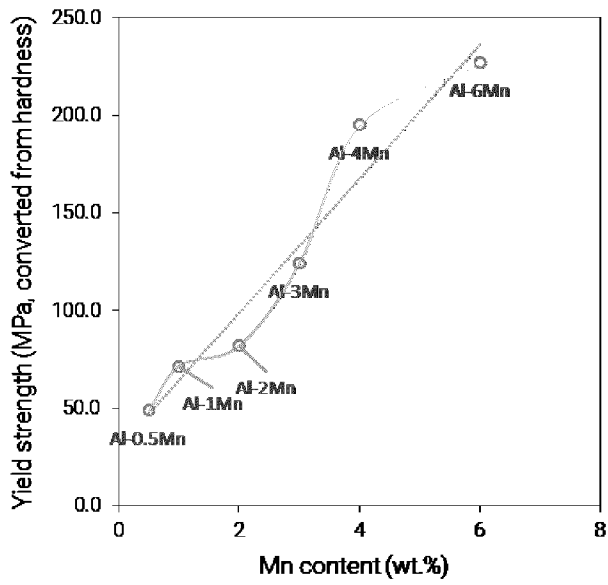


Fig. 11

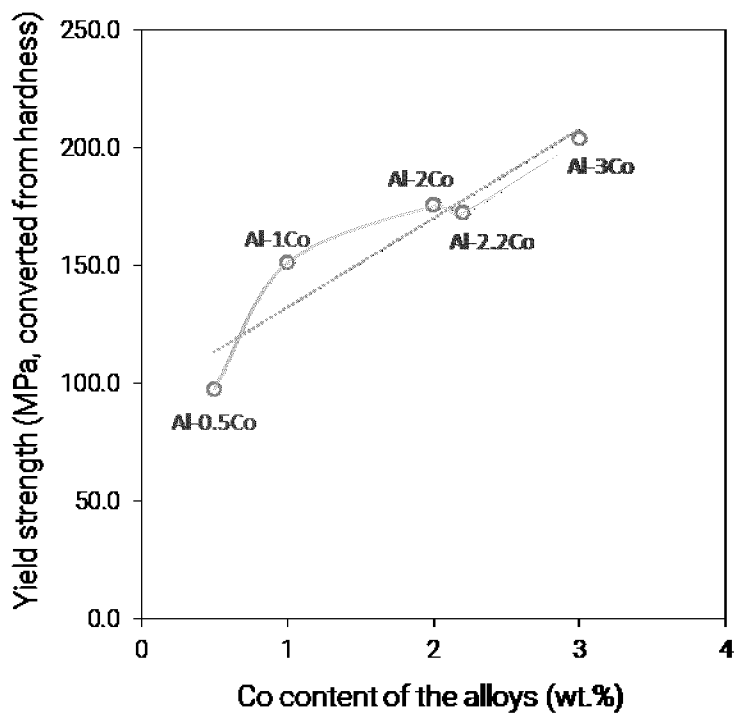
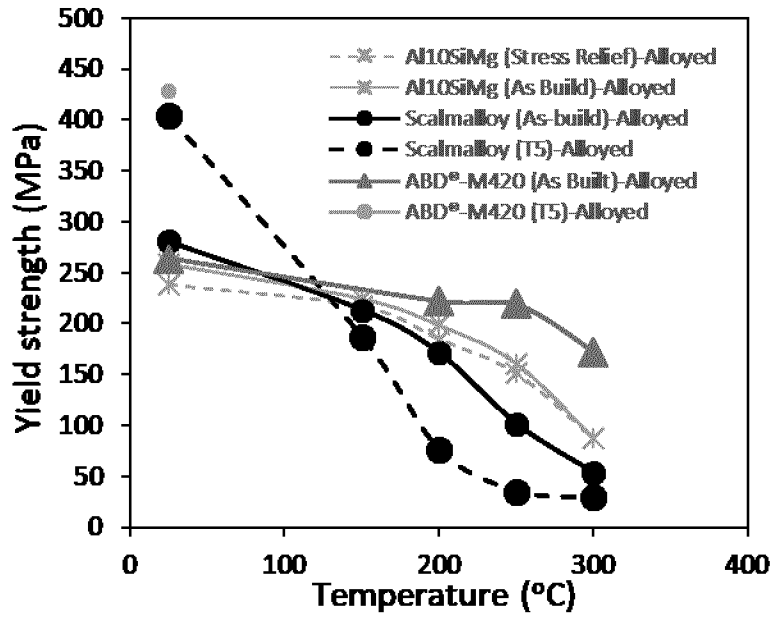


Fig. 12



# INTERNATIONAL SEARCH REPORT

International application No  
PCT/EP2024/079827

<b>A. CLASSIFICATION OF SUBJECT MATTER</b>		
INV. C22C21/06	B33Y70/00	B33Y80/00
ADD.	C22C1/04	C22C21/00
According to International Patent Classification (IPC) or to both national classification and IPC		
<b>B. FIELDS SEARCHED</b>		
Minimum documentation searched (classification system followed by classification symbols) B33Y C22C		
Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched		
Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)  EPO-Internal, COMPENDEX, CHEM ABS Data		
<b>C. DOCUMENTS CONSIDERED TO BE RELEVANT</b>		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>LI ZHEN ET AL: "Improvement in the mechanical properties and creep resistance of Al-Mn-Mg 3004 alloy with Sc and Zr addition", MATERIALS SCIENCE, vol. 729, 18 May 2018 (2018-05-18), pages 196-207, XP093243243, AMSTERDAM, NL ISSN: 0921-5093, DOI: 10.1016/j.msea.2018.05.055 Section 2. Experimental Procedures example Code SZ30; table 1 Section 3.1 figures 6-7 Section 3.4.1; table 2</p> <p style="text-align: center;">----- -/-</p>	1-30
<input checked="" type="checkbox"/> Further documents are listed in the continuation of Box C. <span style="margin-left: 200px;"><input checked="" type="checkbox"/> See patent family annex.</span>		
<p>* Special categories of cited documents :</p> <p>"A" document defining the general state of the art which is not considered to be of particular relevance</p> <p>"E" earlier application or patent but published on or after the international filing date</p> <p>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</p> <p>"O" document referring to an oral disclosure, use, exhibition or other means</p> <p>"P" document published prior to the international filing date but later than the priority date claimed</p> <p>"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</p> <p>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone</p> <p>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</p> <p>"&amp;" document member of the same patent family</p>		
Date of the actual completion of the international search		Date of mailing of the international search report
26 January 2025		06/02/2025
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer  Neibecker, Pascal

## INTERNATIONAL SEARCH REPORT

International application No

PCT/EP2024/079827

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 2004/057047 A1 (NIPPON LIGHT METAL CO [JP]; TAKAHASHI TERUE [JP] ET AL.) 8 July 2004 (2004-07-08)	1-8, 10-17, 19,20, 22,23, 25-30
A	example Alloy B; table 1 page 2 - page 4 claims 1-2 -----	9,18,21, 24

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No

PCT/EP2024/079827

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
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