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INFLUENCE OF Cu ON THE MICROSTRUCTURE DEVELOPMENT OF AlSi7MgCu ALLOY

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Abstract

Designing and characterisation of AlSi7MgCu alloy with extra addition of Cu (up to 1.435 wt.%) represents a challenge in order to achieve advanced mechanical properties already in ascast state. Microstructural investigation of AlSi7MgCu alloy reveals a wide range of complex reactions and possible intermetallic phases due to the interaction of alloying and trace elements. An extra addition of Cu (up to 1,435 wt.%) as a secondary alloying element initiates an additional interaction with transition elements Fe, Mn and secondary alloying element Mg. Evolution of microstructure and determination of solidification sequence enables detail overview of solidification path in both states, as-cast and heat-treated. Enrichment of solidification process with complex intermetallic phases reveals following constituents: dendrite network; iron-based needle-like Al₅SiFe and / or complex Chinese script formation $Al_{15}(Fe,Mn,Cu)_3Si_2;$ main eutectic $(\alpha_{A1}+\beta_{Si});$ complex eutectic clusters of Al₈Mg₃(Fe,Mn,Cu)Si₆ and Al₅(Fe,Mn,Cu)₂Mg₈Si₆ phase and secondary eutectic phase precipitations α_{Al} +Mg₂Si and α_{Al} +Al₂(Fe,Mn,Cu). Infiltration of common transition elements such as Fe and Mn in Cu bearing phases' resulted in high total content of transition elements (Fe+Mn+Cu). Microstructural investigation also indicates continuous interaction of Fe, Mn and Cu in formation of wide range of intermetallic phases' through the whole solidification process.

The nature (morphology and alloying elements interaction) of formed intermetallic phases comprehends to the tensile mechanical properties development due to strong connections and interactions during solidification process as a whole.

Keywords: AlSi7MgCu alloy, copper, microstructure, transition elements

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1. INTRODUCTION

The increasing demand toward lightweight aluminium alloys have expanded its application in automotive industry due to downsizing effect and therefore reducing the CO_2 emissions [1,2,3]. Safety-critical aluminium alloys castings have been exposed to high demands of the market related to long term stability in aggressive engine environment. The mechanical properties, hot cracking susceptibility and other quality feature of aluminium components are strongly dependent from chemical composition [4,5,6]. Therefore, the innovative approach to the designing of modified chemical composition of aluminium alloys with higher tensile strength, thermal stability, corrosion resistance becomes significant for structural components in automotive industry.

Excellent castability, recyclability, low cost manufacturing, high specific strength and its favourable relationship to weight, especially in the heat-treated state, indicate conventional AlSi7Mg alloy as a frequent choice for complex geometry castings with high mechanical properties [7,8,9,10]. The solidification sequence of hypoeutectic AlSi7Mg alloys begins with development of primary aluminum dendrites α_{Al} and formation of dendritic network, followed by eutectic reaction ($\alpha_{Al} + \beta_{Si}$) on the primary grains α_{Al} or independently on present nucleants rich on iron and/or other impurities with different crystallographic orientation [11]. The way of eutectic occurs determines the amount and morphology of eutectic phase, and also the porosity ratio in the microstructure. The primary and most important alloying element in conventional AlSi7Mg alloy is Si which is characterized by high fluidity and reduction in shrinkage, followed by Mg responsible for strength increase [12,13,14].

Magnesium addition up to a 0.7 wt. % has a strengthening effect through the precipitation of $\alpha_{Al}+Mg_2Si$ eutectic phase and/or Mg-rich intermetallics with other alloying elements mostly due to transformation of the deleterious Al₃FeSi platelets into a Chinese script phase with a composition Al₈Mg₃FeSi₆. [13,15,16]. The yield strength, tensile strength and elongation of the as-cast Al-Si-Mg alloys can vary by the content of Mg [17]. The yield strength increasing of Mg content, but showed no significant difference with increasing of Mg₂Si phase ratio. Conversely, the elongation was decreased with the increase of Mg and Mg₂Si ratios in this alloy [17].

Also, Cu is commonly used as an alloying element to increase the strength of cast alloys, especially when heat treatment is applied. In Al-Si alloys, Cu is usually added in levels between 1.5 - 3.5 % and forms the intermetallic phase Al₂Cu [18,19]. On the other hand when Cu forms a continuous network at the grain boundaries the consequence will be a serious reduction in ductility [19,20]. Additionally, copper decreases significantly the melting point and eutectic temperature of the alloy. Therefore, the copper increases the solidification range of the alloy, and facilitates the condition of porosity formation [21,22,23]. Literature survey reveals a number of investigations related to the influence of alloying elements on the AlSi7Mg alloy properties [24,25,26].

The conventional AlSi7Mg alloy, corresponded to the numerous standards (EN 1706, IDM 4234) [27,28], has been investigated previously [29].

Designing of new chemical composition of AlSi7MgCu alloy with extra addition of Cu (up to 1,435 wt.%) represents a challenge in order to achieve advanced properties. A wide range of complex reactions and intermetallic phases occurs due to numerous alloying (Si, Mg, Cu) and trace elements (Fe, Mn) interaction. Obtained advanced mechanical properties are strongly

depended from the complex microstructure development based on particular elements interaction. Although calculation of phase stability indicates solidification sequence determination, the interaction with other elements brought out numerous combinations. Those combinations cover up infiltration of additional elements in regular, commonly present phases. The aim of this investigation is to determine the influence of copper on microstructure development.

2. EXPERIMENTAL

Characterization of newly designed AlSi7MgCu alloy has been performed in as-cast and heat-treated state [9,10,10].

An AlSi7MgCu alloy melt was prepared in an induction furnace ABB IMTK 2000 with the ingot and return ratio in charge material 1: 1. After melting at a temperature of $770 \pm 5 \degree$ C, degassing of the melt was performed with the nitrogen (N₂) using a MTS 1500 - Foseco equipment. Melt treatment was performed through inoculation with AlTi5B master alloys and modification with AlSr10 master alloy. Chemical composition analysis was performed on an optical emission spectrometer ARL-3460.

Heat treatment was performed following the regime: heating from room temperature to the annealing temperature of 480° C for 2 hours, and the retention of the final annealing temperature during 8h, followed by air cooling.

Previous investigation comprehends development of equilibrium phase diagram, simultaneous thermal analysis and mechanical properties analysis [8,9,10]. Detail metallographic investigations were performed using light microscopy (Olympus GX 51) and microstructural investigations (SEM/EDS) using scanning electron microscopes Tescan Vega TS 5136 MM equipped with energy dispersive spectrometer Bruker.

Samples for metallographic investigation were prepared by standard metallographic preparation procedure by grinding and polishing, followed by etching in 0.5% HF.

3. RESULTS AND DISCUSSION

Chemical composition of AlSi7MgCu alloy with extra addition of copper has been designed and compared with previously investigated conventional AlSi7Mg alloy [29], as shown in Table 1.

Element, wt.%	Si	Fe	Cu	Mn	Mg	Ti	Sr
AlSi7Mg	7.008	0.101	0.130	0.010	0.320	0.139	0.0121
AlSi7MgCu	7.527	0.235	1.435	0.076	0.348	0.147	0.0223

Table 1. The chemical compositions of AlSi7Mg / AlSi7Mg(Cu) alloys

An AlSi7MgCu alloy is in line with the EN 42000 AC standard for AlSi7Mg alloy in relation to the content of the base alloying elements Si and Mg, and trace elements such as Fe and Mn [27]. Deviation has been implemented with significant increase in Cu content.

Modelling of newly designed AlSi7MgCu alloy by ThermoCalc (TCW 5.0) program resulted with previously calculated equilibrium phase diagram [10]. Interaction of alloying and trace elements reveals wide range of intermetallic phases, followed with calculation of solidification sequence of AlSi7MgCu. Calculated equilibrium solidification sequence of AlSi7MgCu alloy is shown in Table 2 [10].

Reaction description	Reaction			
Liquidus temperature, T ₁	$L \rightarrow L_1 + \alpha_{Al}$			
Eutectic temperature, T _e	$L_1+\alpha_{Al} \rightarrow L_2+ \alpha_{Al}+(\alpha_{Al}+\beta_{Si})$			
Precipitation of secondary intermetallic phases temperature, T ₁	$L_2+(\alpha_{AI}+\beta_{Si}) \rightarrow L_3+(\alpha_{AI}+\beta_{Si})+Al_{15}(FeMn)_3Si_2$			
Precipitation of secondary intermetallic phases temperature, T ₂	$L_3 \rightarrow L_4 + Al_5 Cu_2 Mg_8 Si_6$			
Precipitation of secondary intermetallic phases temperature, T ₃	$L_4 + Al_5 Cu_2 Mg_8 Si_6 \rightarrow L_5 + Al_8 FeMg_3 Si_6$			
Precipitation of secondary intermetallic phases temperature, T ₄	$L_5 \rightarrow L_6 + Al_7 Cu_2 M$			
Precipitation of secondary intermetallic phases temperature, Solidus temperature, T _s	L ₆ →Al ₂ Cu			

Table 2. Calculated equilibrium solidification sequence of AlSi7MgCu alloy [10]

Microstructure of AlSi7MgCu alloy in as-cast (F) and heat-treated (T) state was investigated using light microscopy, as shown in Figure 1 [9].



Figure 2. Micrographs of AlSi7Mg(Cu) alloy obtained by light microscopy [9]

A smaller magnification (100x) reveals uniform distribution of primary dendritic network with evenly distributed interdendritic areas in both states, as-cast (F) and heat-treated (F), respectively. Higher magnification indicates rougher and broken dendritic branches dotted with iron-bearing intermetallic phases with needle-like morphology, (Al₅SiFe), and coarse secondary intermetallic phases at grain boundaries in as-cast state (F). The highest magnification (1000x) indicates the presence of under-modified eutectic (mixed fiber and lamella morphology). Heat-treated state (T) indicated uniformly distributed fiber morphology of main eutectic ($\alpha_{Al}+\beta_{Si}$). Last solidifying phases have been noticed at grain boundaries. Theirs morphology and colour reveals secondary eutectic phases $\alpha_{Al}+Al_2Cu$ (platelets particles and clusters) and $\alpha_{Al}+Mg_2Si$ (thin ramified black particles) in as-cast (F) state. Heat-treated state (F) reveals fine fragmented secondary intermetallic phases' at the grain boundaries.

Comparison of microstructural constituents' distribution and size revealed with scanning electron images at higher magnification is given in Figure 3.



Figure 3. Scanning electron images of AlSi7MgCu alloy in a) as-cast (F) and b) heat-treated state (T)

Beside dendrite network, first to evaluate is iron-based needle-like Al₅SiFe and / or complex Chinese script formation Al₁₅(Fe,Mn,Cu)₃Si₂. Appropriate ratio of Cu and Mg precipitate in a metal matrix, while the bulk cohesively forms complex eutectic clusters of Al₈Mg₃(Fe,Mn,Cu)Si₆ and Al₅(Mn,Fe,Cu)₂Mg₈Si₆ phase. Solidification ends with secondary eutectic phase precipitations α_{AI} +Mg₂Si and α_{AI} +Al₂(Fe,Mn,Cu). Heat treatment has a positive influence on uniform distribution and refining of intermetallic phases'. Morphological detrimental intermetallic phases' enriched in iron known as β phase (Al₅SiFe) are no longer present in that particular morphology. Also, classical Al₂Cu clusters have been resolved in heat-treated state.

Analysis of microstructure development was performed due to Cu content phases in both samples type. Energy dispersive stoichiometry analysis reveals significant copper enrichment of characteristic intermetallic phases. An overview of Mg, Cu and total transition elements (Fe+Mn+Cu) content in as-cast (F) and heat-treated (T) state is presented in Figure 4.



Figure 4. Analysis of scanning electron images of AlSi7MgCu alloy by energy dispersive spectrometry in a) as-cast (F) and b) heat-treated state (T)

SEM/EDS investigation enables determination modification of solidification path induced by increased copper content. Magnesium, copper and other significant transition elements (Fe, Mn) content range in both states has been determined as shown in Table 3.

Reaction description	Intermetallic phase	Mg, wt.%		Cu, wt.%		Fe+Mn+Cu, wt.%	
		F	Т	F	Т	F	Т
$L \rightarrow L_1 + \alpha_{A1} + Al_5 SiFe + Al_{15} (Fe, Mn, Cu)_3 Si_2$	Al ₁₅ (Fe,Mn,Cu) ₃ Si ₂	0.88- 2.80	1.12	2.65- 6.34	22.77	26.04	32.54
$L_2 \rightarrow L_3^+$ Al ₅ (Fe,Mn,Cu) ₂ Mg ₈ Si ₆ + Al ₈ Mg ₃ (Fe,Mn,Cu)Si ₆	Al ₅ (Fe,Mn,Cu) ₂ Mg ₈ Si 6	1.53- 33.0 3	2.58- 13.0 9	1.61- 40.22	7.3- 13.41	2.65- 41.42	7.96- 14.16
$L_2 \rightarrow L_3^+$ Al ₅ (Fe,Mn,Cu) ₂ Mg ₈ Si ₆ + Al ₈ Mg ₃ (Fe,Mn,Cu)Si ₆	Al ₈ Mg ₃ (Fe,Mn,Cu)Si ₆	0,82- 6,07	0.85- 5.29	4.87- 35.3	3.26- 28.35	15.83 - 36.68	15.37 - 32.93
$L_4 \rightarrow \alpha_{Al} + Al_2(Fe,Mn,Cu)$	α _{Al} +Al ₂ (Fe,Mn,Cu)	0.71- 5.02	0.89- 2.33	30.86 - 45.96	30.23 - 43.98	32.07 - 51.89	31.88 - 45.55

Table 3. Microstructural analysis of copper bearing phases

Both states indicated wide range of common transition (Fe, Mn) and secondary elements (Mg, Cu) content. Heat-treated state revealed decreasing of magnesium, copper and (Fe+Mn+Cu) content in general when compared to as-cast state, due to homogenisation process as well as resolving of Mg and Cu in matrix [9]. Wide range of Mg and Cu content in particular phases is dependent to the phases' position in solidification process hierarchy and final size. Infiltration of common transition elements such as Fe and Mn in Cu bearing phases' resulted in high total content >30 wt.%, except for Al₅(Fe,Mn,Cu)₂Mg₈Si₆ with high Mg content. Although, Fe and Mn bearing phase are considered to be a high-temperature phases', the investigation results indicate continuous interaction of Fe, Mn and Cu in formation of wide range of intermetallic phases' through the whole solidification process.

Previous investigation indicates determination of solidification sequence and characterization of AlSi7MgCu alloy [8,9,10], indicating the significant increase in yield strength and tensile strength of innovative AlSi7MgCu alloy in as-cast state and significant increase of elongation in heat-treated state. Detail microstructure investigation relating to the Cu bearing phases' reveals stronger interaction of transition elements in solidification sequence through participation in forming the intermetallic phases.

4. CONCLUSION

Microstructural investigation of AlSi7MgCu alloy reveals a wide range of complex reactions and possible intermetallic phases due to the interaction of alloying and trace elements. An extra addition of Cu (up to 1,435 wt.%) as a secondary alloying element initiates additional interaction with transition elements Fe, Mn and secondary alloying element Mg. Evolution of microstructure and determination of solidification sequence enables characterization of solidification path in both states, as-cast and heat-treated.

Microstructural investigation reveals following constituents: dendrite network; iron-based needle-like Al₅SiFe and / or complex Chinese script formation Al₁₅(Fe,Mn,Cu)₃Si₂; main eutectic ($\alpha_{Al}+\beta_{Si}$); complex eutectic clusters of Al₈Mg₃(Fe,Mn,Cu)Si₆ and

 $Al_5(Fe,Mn,Cu)_2Mg_8Si_6$ phase. Solidification ends with secondary eutectic phase precipitations $\alpha_{Al}+Mg_2Si$ and $\alpha_{Al}+Al_2(Fe,Mn,Cu)$.

Appropriate ratio of Cu and Mg also precipitate in a metal matrix, especially in heat-treated state. Ratio of Cu and Mg developed in particular intermetallic phases reveals wide range of theirs content, which is connected to the phases' position in solidification process hierarchy and final size. Infiltration of common transition elements such as Fe and Mn in Cu bearing phases' resulted in high total content of transition elements (Fe+Mn+Cu)>30 wt.%, except for Al₅(Fe,Mn,Cu)₂Mg₈Si₆ with high Mg content. Although, Fe and Mn bearing phase are considered to be a high-temperature phases', the investigation results indicate continuous interaction of Fe, Mn and Cu in formation of wide range of intermetallic phases through the whole solidification process.

The nature (morphology and alloying elements interaction) of formed intermetallic phases comprehends to the tensile mechanical properties development due to strong connections and interactions during solidification process as a whole.

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