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THE EFFECTS OF MAGNESIUM AND TITANIUM ADDITIONS ON THE MICROSTRUCTURE AND PROPERTIES OF AS-CAST AL-5 % Cu ALLOYS

VPLIV VSEBNOSTI MAGNEZIJA IN TITANA NA MIKROSTRUKTURO IN LASTNOSTI LITIN IZ Al-5 % Cu ZLITIN

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The effect of the mass fraction of magnesium content in the range 1–5 %, on the microstructure and properties of Al–Cu–Mg alloys (with a copper content of 5 %) was examined. The as-cast microstructure was modified by the addition of AlTi5B1 to obtain 0.25 % of titanium in the alloys. Using X-ray powder diffraction we established that the tetragonal intermetallic compound Al_2Cu and the orthorhombic intermetallic compound Al_2CuMg are formed across the whole range of magnesium additions.

The effect of magnesium and titanium contents on the microstructure was monitored quantitatively using automatic image analysis to measure the linear intercept grain size, the secondary dendrite arm spacing (DAS), the size of the eutectic cells (Le), as well as the size distribution and the volume fractions of the α-solid solution and the eutectic. In alloys with a high magnesium content the average values of the DAS and the grain size were found to decrease. In contrast, with a lot of magnesium the average eutectic cell length and the eutectic volume fraction were found to increase.

The changes in the chemical composition of the alloy cause changes in the microstructure that are reflected in the Brinell hardness and the compression strength. The compression strength and hardness increase with the content of magnesium and titanium.

Key words: Al-Cu-Mg alloys, as-cast structure, intermetallic phases Al_2Cu and Al_2CuMg, lattice parameters, geometrical parameters, hardness, compression strength

Raziskan je vpliv masnega deleža magnezija v razponu od 1 % do 5 % na mikrostrukturo in lastnosti zlitin AlCuMg s 5 % bakra. Strjevalna mikrostruktura je bila modificirana z dodatkom AlTi5Bi1 s ciljem doseči v zlitinah vsebnost 0,25 % titana. Z uporabo rentgenske difrakcije je bilo ugotovljeno, da nastajata v vsem območju dodatkov magnezija tetragonalna intermetalna faza Al_2Cu in ortorombična faza Al_2CuMg.

Vpliv magnezija iz titana na mikrostrukturo je bil kvantitativno ocenjen z avtomatsko analizo slike in določene so bile: linearna intercepcijska dolžina, širina sekundarnih dendritnih vej (DAS), velikost evtektičnih celic (Le) in velikostna porazdelitev ter volumenski delež trdne raztopine α. V zlitinah z visokim magnezijem sta bila manjša DAS in velikost zrn, medtem ko sta bila večja povprečna velikost evtektičnih celic in volumenski delež evtektika.

Spremembe v kemični sestavi in mikrostrukturi se izražajo v trdoti po Brinellu in v tlačni trdnosti, ki rasteta s povečanjem vsebnosti magnezija in titana.

Ključne besede: zlitine Al-Cu-Mg, strjevalna struktura, intermetalne faze, Al_2Cu in Al_2CuMg, parameter kristalne mreže, geometrični parametri, trdota, tlačna trdnost

1 INTRODUCTION

An excellent strength vs. density ratio combined with good formability and corrosion resistance make Al–Cu–Mg alloys potential candidates for a number of industrial applications [1].

Developed many years ago for applications in the field of aeronautics, these alloys have been considered for a wide range of other applications. Because of their high specific strength they are mainly considered as a substitute for iron-based materials in structural parts used in the transportation industry. Several technical compositions have already been standardized and new alloys based on the Al–Cu–Mg system are being considered and developed [2].

In the binary aluminium-copper system the aluminium-rich terminal solid solution is in equilibrium with the intermetallic phase θ, with the approximate composition Al_2Cu, since some solid solubility exists. The addition of magnesium allows the formation of more intermetallic compounds, such as Al_2CuMg, Al_6CuMg_4, AlCuMg and $Al_5Cu_6Mg_2$ [3]. Magnesium increases the strength [3] and hardness of the alloys, and especially in castings this is accompanied by a decrease in the ductility and the impact resistance. Titanium is added as a grain refiner and it is very effective in reducing the grain size [4]. It also produces a better dispersion of insoluble constituents, porosity and non-metallic inclusions, resulting in a significant improvement in the mechanical properties of the material. Since grain size controls the distribution [3] of the porosity and the constituents, the mechanical properties of these alloys are very sensitive to grain size. Standard industrial aluminium-copper alloys solidify with the formation of a dendritic structure; however, the tendency to form a globular structure at higher copper contents was also reported [1,2] and confirmed in our previously published work [5,6].

In this paper we have examined the as-cast microstructure in the Al–Cu–Mg system over a wide range of magnesium and titanium contents in order to determine the effect of the content of both elements on the microstructure and properties of Al–Cu–Mg alloys. Depending on the alloy composition (i.e., the Cu content and the Cu/Mg ratio), a different phase distribution and, consequently, different material characteristics can be obtained. The characterization of six different Al–Cu–Mg alloys with the mass fraction of copper 5 % and Cu/Mg ratios of 5:1; 1.66 : 1 and 1:1 was performed with X-ray powder diffraction, quantitative microstructure analysis, optical and electron microscopy. The hardness and the compression strength were also measured.

2 EXPERIMENTAL

The investigated materials were aluminium-copper-magnesium alloys with the chemical compositions shown in **Table 1**. In these alloys aluminium is the primary constituent, and in the cast alloys the basic structure consists of cored dendrites of an aluminium solid solution with a variety of constituents at the grain boundaries or interdendritic spaces forming a brittle, more or less continuous, network of eutectics. Copper has been the most common alloying element almost since the beginning of the aluminium industry, and a variety of alloys in which copper is the major addition were developed. Magnesium is usually combined with copper. The constituents formed in the alloys containing only one or more of copper, magnesium, etc. are soluble ones. In an AlCu5Mg1 alloy in which the copper : magnesium ratio is in the range 8:1 to 4:1 the main hardening agents are Al_2Cu and Al_2CuMg, and both of these phases are active. In the AlCu5Mg3 and AlCu5Mg5 alloys with the Cu/Mg ratio in the range between 4:1 and 1:1, Al_2CuMg controls the properties.

The experimental work can be divided into two phases. The first phase consisted of melting and casting the samples with different compositions from the aluminium-copper-magnesium system with the addition of 0.00–0.25 % Ti (AlTi5B1) as a modifier. The second phase included the characterization of the samples obtained by previous melting and casting with X-ray powder diffraction, quantitative microstructure analysis, and examination in an optical and an electron microscope (JCXA-733). The properties of these materials, including the hardness and compression strength, were also determined.

The X-ray diffraction analysis was performed on the alloys AlCu5Mg, AlCu5Mg3 and AlCu5Mg5 using a wide range of angles (2θ from 5° to 100°), a step size of 0.02° and a holding time of 0.50 seconds at each step. A diffractometer with a graphite monochromator and a constant-divergence slit (D) of 1mm was used. The current and the voltage of the X-ray tube during the analysis were 30 mA and 40 kV, respectively. The width of the receiving slit (R) was 0.1 mm, corresponding to fine focussed X-ray tubes. The radiation was Cu $K\alpha_1/\alpha_2$ ($\lambda\alpha_1$ = 0.154060 nm and $\lambda\alpha_2$ = 0.154438 nm).

Using a QUANTIMET 500MC automatic image analyser and a linear-interception measuring method we were able to measure the grain size (minimum, maximum and average values), the dendrite arm spacing (DAS) [7], and the eutectic cell length (ECL), to determine the relative standard measuring errors (RSEs) for all parameters, as well as the distribution by grain size and volume fractions of the α-solid solution and the eutectic.

3 RESULTS AND DISCUSSION

The mass fraction of copper in the standard Al–Cu–Mg alloys was up to about 5 %, slightly below the value of 5.65 % that represents the maximum solid solubility of copper in aluminium at the eutectic temperature of 548 °C. The investigated Al–Cu–Mg alloys have cellular structures and contain the soluble phases Al_2Cu and Al_2CuMg in various amounts and at various locations in the microstructure, depending on the thermal history of the specimen.

The **Figures 1–6** were obtained using an optical microscope. The microstructure was also investigated in a

Tabela 1: Kemična sestava v masnih deležih raziskanih zlitin Al-Cu-Mg
Table 1: Chemical composition in mass fraction of the investigated AlCuMg alloys (%)

Type of sample	Al %	Fe %	Si %	Cu %	Zn %	Mg %	V %	Cr %
AlCu5Mg1 (0 % Ti)	bal.	0.14	0.08	5.353	0.067	1.064	0.001	0.001
AlCu5Mg1 (0.25 % Ti)	bal.	0.14	0.06	4.915	0.060	0.917	0.006	0.001
AlCu5Mg3 (0 % Ti)	bal.	0.17	0.07	5.113	0.067	3.266	0.001	0.002
AlCu5Mg3 (0.25 % Ti)	bal.	0.17	0.07	5.089	0.063	2.983	0.005	0.002
AlCu5Mg5 (0 % Ti)	bal.	0.19	0.07	5.435	0.072	5.814	0.000	0.003
AlCu5Mg5 (0.25 % Ti)	bal.	0.18	0.08	4.901	0.063	5.077	0.005	0.003

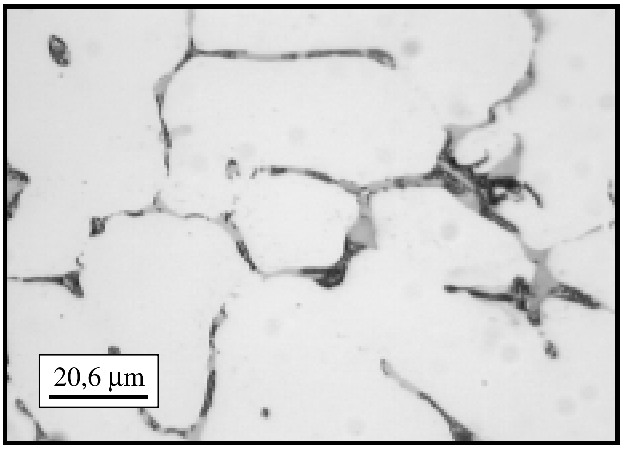

Figure 1: Microstructure of the AlCu5Mg1 alloy without any addition of Ti (Etched with Keller's reagent)
Slika 1: Mikrostruktura zlitine AlCu5Mg1 brez dodatka titana, jedkano po Kellerju

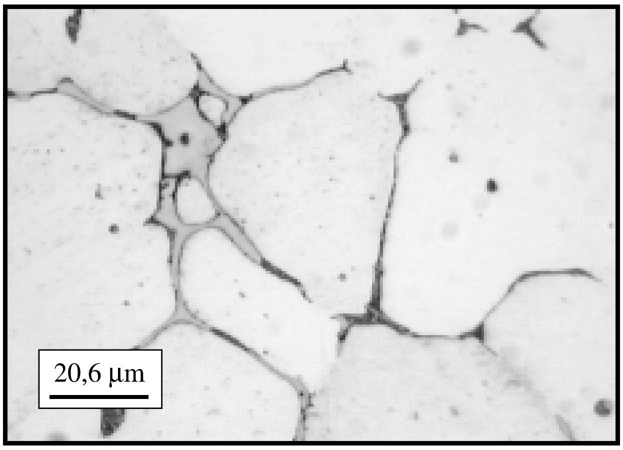

Figure 2: Microstructure of the AlCu5Mg1 (0.25 % Ti) alloy (Etched with Keller's reagent)
Slika 2: Mikrostruktura zlitine AlCu5Mg1 (0,25 % Ti), jedkano po Kellerju

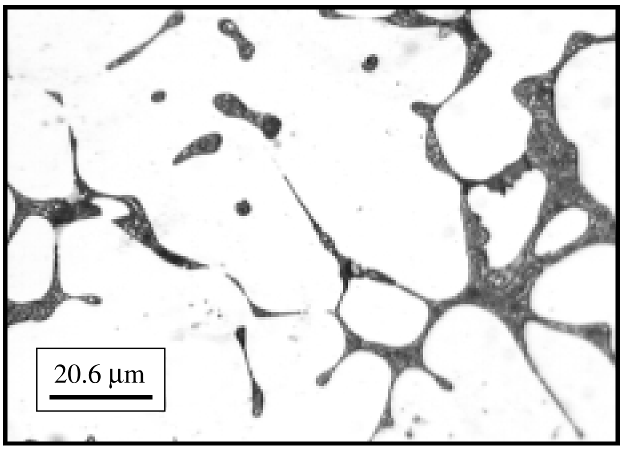

Figure 3: Microstructure of the AlCu5Mg3 alloy without Ti addition (Etched with Keller's reagent)
Slika 3: Mikrostruktura zlitine AlCu5Mg3 brez dodatka titana, jedkano po Kellerju

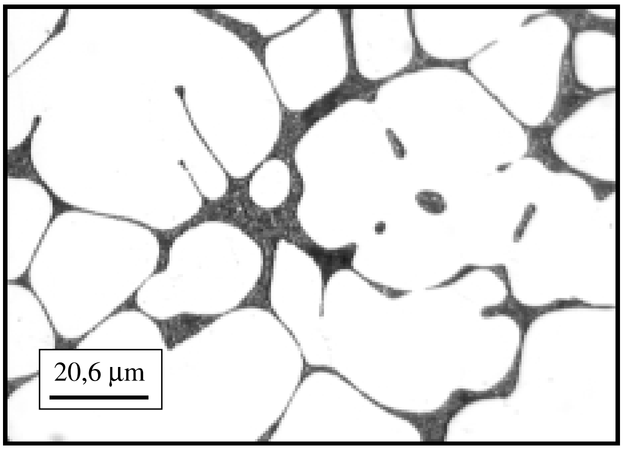

Figure 4: Microstructure of the AlCu5Mg3 (0.25 % Ti) alloy (Etched with Keller's reagent)
Slika 4: Mikrostruktura zlitine AlCu5Mg3 (0,25 % Ti), jedkano po Kellerju

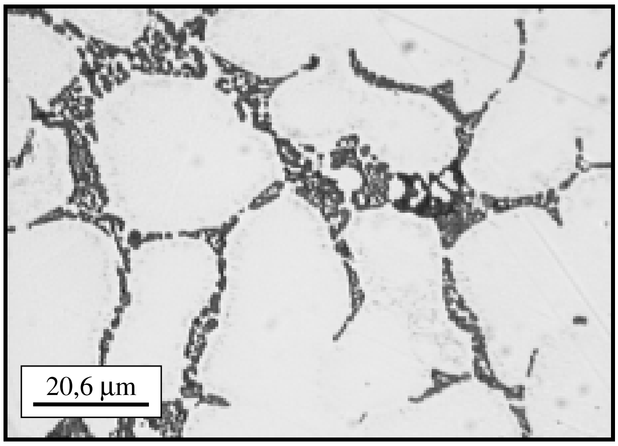

Figure 5: Microstructure of the AlCu5Mg5 alloy without Ti addition (Etched with Keller's reagent)
Slika 5: Mikrostruktura zlitine AlCu5Mg5 brez dodatka titana, jedkano po Kellerju

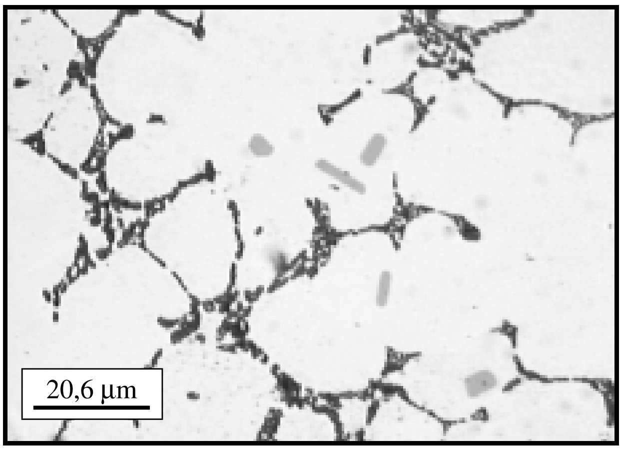

Figure 6: Microstructure of the AlCu5Mg5 (0.25 % Ti) alloy (Etched with Keller's reagent)
Slika 6: Mikrostruktura zlitine AlCu5Mg5 (0,25 % Ti), jedkano po Kellerju

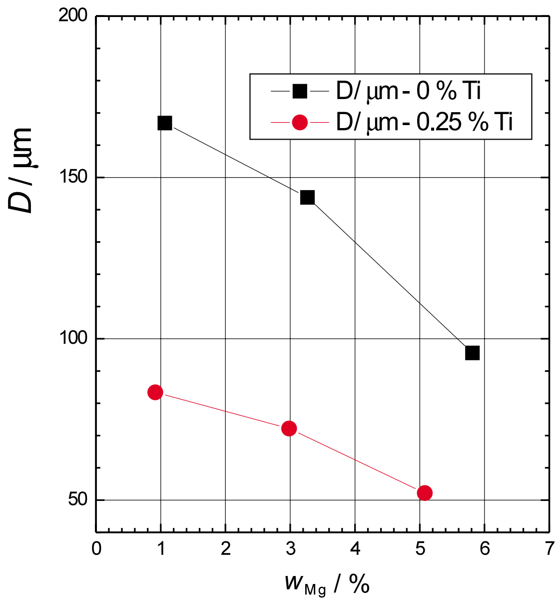

Figure 7: Grain size *D* for different magnesium and titanium contents in AlCuMg alloys
Slika 7: Velikost zrn *D* za različno vsebnost magnezija in titana v zlitinah AlCuMg

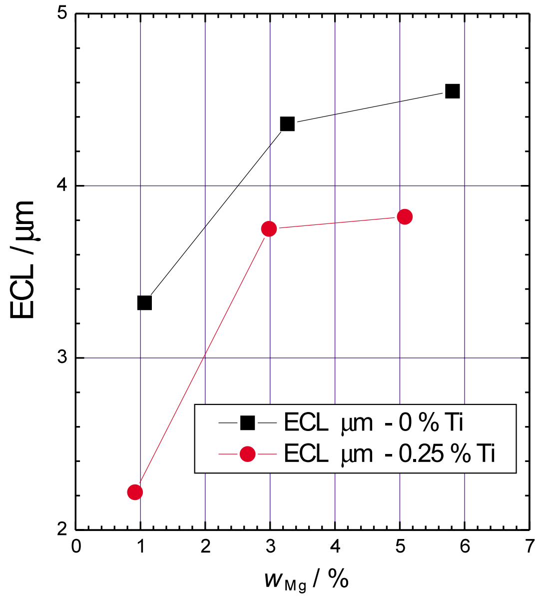

Figure 9: Eutectic cell length and volume fraction of eutectic for different magnesium and titanium contents in AlCuMg alloys
Slika 9: Velikost evtektične celice in volumenski delež evtektika za različno vsebnost magnezija in titana v zlitinah AlCuMg

JCXA-733 electron microprobe analyser. The current and the voltage during the analysis for the copper, magnesium and titanium were $1\cdot10^{-8}$ A and 20 kV, respectively. Kα radiation was used. The content of copper and magnesium in the white phase is low. X-ray analysis showed the presence of magnesium in the eutectic grey phase, while copper is found in the bright phase. Titanium is present in platelets in some eutectic areas in the white phase. Isolated particles containing titanium are also found dispersed in the interior of the matrix grains.

3.1 Quantitative analysis of the microstructure

Using automatic image analysis the grain size (**Figure 7**), the dendrite arm spacing (DAS) (**Figure 8**), the eutectic cell length (ECL) (**Figure 9**), and the volume fractions of the α-solid solution and the eutectic were assessed.

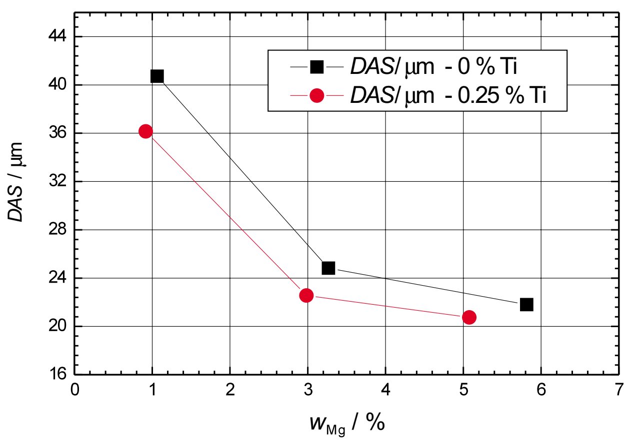

Figure 8: Dendrite arm spacing (*DAS*) for different magnesium and titanium contents in AlCuMg alloys
Slika 8: Velikost dendritnih vej (*DAS*) za različno vsebnost magnezija in titana zlitinah AlCuMg

The measurement of the dendrite arm spacing was performed in the same manner as the grain size measurement: using the intercept method. The dendrite arm spacing is an important microstructure characteristic of the cast aluminium-copper-magnesium alloys.

The grain size, which is expressed as the mean grain diameter, as well as the distribution of the dendrites and the eutectic depend on the casting parameters [4], the melt temperature and the solidification rate, all of which affect the properties of the alloys. Furthermore, the addition of the grain refiner (AlTi5B1) produced the nearly equiaxed structure shown in the micrographs. The addition of titanium and boron in form of the alloy AlTi5B1 produces particles of TiB_2 in the melt [4]. These particles are then nuclei for the $TiAl_3$ phase that affects the solidification. Titanium and aluminium produce a peritectic reaction with the $TiAl_3$ and the solid peritectic acts as a solidification nucleus for pure aluminium and its solid solutions. The reduction in grain size and dendrite arm spacing and the improvement in microstructure uniformity as result of the addition of the grain refiner are shown in **Figures 1–6**. The size and shape of the grains are also affected by the added magnesium. For the same content of titanium in the alloy and with an increased amount of magnesium, the average values of the dendrite arm spacing and the grain size are decreased and a fine, uniform grain size, as shown in the micrographs, is obtained. Furthermore, in the alloys containing a lot of magnesium the average values of the eutectic cell length and the volume fraction

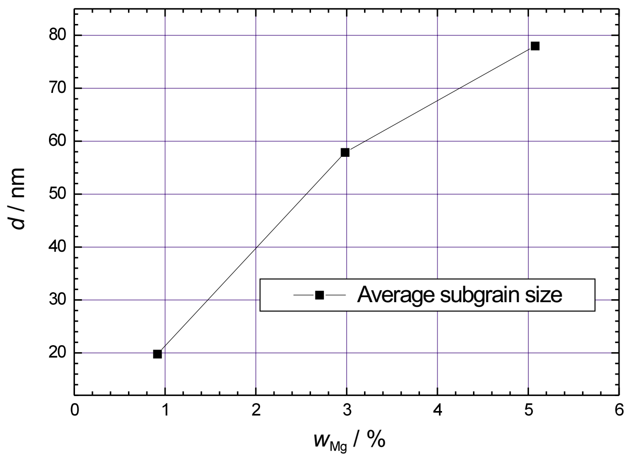

Figure 10: Average sub-grain size (*d*/nm) in the crystallographic direction [112] for different magnesium contents in the alloys (0.25 % Ti)

Slika 10: Povprečna velikost podzrn (*d*/nm) v kristalni smeri [112] za različno vsebnost magnezija v zlitinah

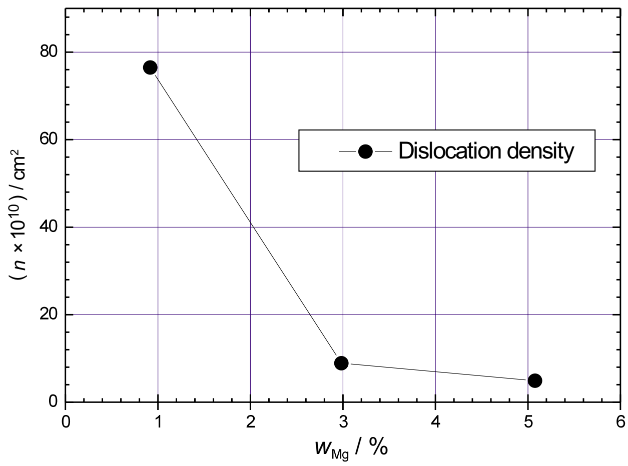

Figure 12: Dislocation density *n* in the direction [112] for different magnesium contents in the alloys with 0.25 % Ti

Slika 12: Gostota dislokacij v smeri [112] za različno vsebnost magnezija pri zlitinah z 0,25 % Ti

of the eutectic were found to increase. The chemical composition affects the microstructure through its influence on phase relations.

3.2 XRD analysis

Using X-ray diffraction we established that the tetragonal intermetallic compound Al_2Cu and the orthorhombic intermetallic compound Al_2CuMg are formed across the whole range of magnesium additions. The lattice parameters determined for the tetragonal intermetallic compound Al_2Cu are as follows: for the AlCu5Mg1 alloy, a = 0.6034 nm, c = 0.4869 nm and V = 0.1773 nm^3; for the AlCu5Mg3 alloy, a = 0.6030 nm, c = 0.4877 nm and V = 0.1774 nm^3; for the AlCu5Mg5 alloy, a = 0.6058 nm, c = 0.4880 nm and V = 0.1791 nm^3. The lattice parameters determined for the orthorhombic intermetallic compound Al_2CuMg are as follows: for the AlCu5Mg1 alloy, a = 0.3990 nm, b = 0.9209 nm, c = 0.7128 nm and V = 0.2621 nm^3; for the AlCu5Mg3 alloy, a = 0.4051 nm, b = 0.9333 nm, c = 0.7064 nm and V = 0.2671 nm^3; for the AlCu5Mg5 alloy, a = 0.4011 nm, b = 0.9283 nm, c = 0.7109 nm and V = 0.26474 nm^3. The lattice parameters determined for the tetragonal intermetallic compound Al_2Cu and for the orthorhombic intermetallic compound Al_2CuMg are in agreement with the data from the literature. On the JCPDS card 25 0012 the parameters for Al_2Cu are a = 0.6065 nm, c = 0.4873 nm and V = 0.17928 nm^3 and on the JCPDS card 28 0014 the parameters for Al_2CuMg are a = 0.4000 nm, b = 0.9250 nm, c = 0.7150 nm and V = 0.26455 nm^3.

From the X-ray diffractograms the following microstructural parameters were calculated: the average

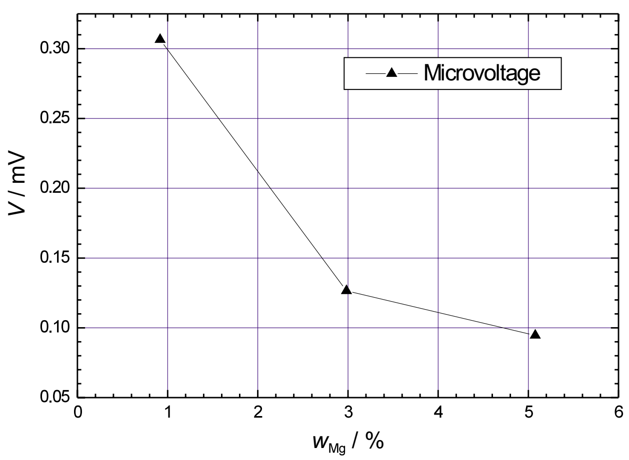

Figure 11: The microvoltage *V* in the crystallographic direction [112] for different magnesium contents in the alloys with 0.25 % Ti

Slika 11: Mikrovoltaža v kristalni smeri [112] za različno vsebnost magnezija v zlitinah z 0.25 % Ti

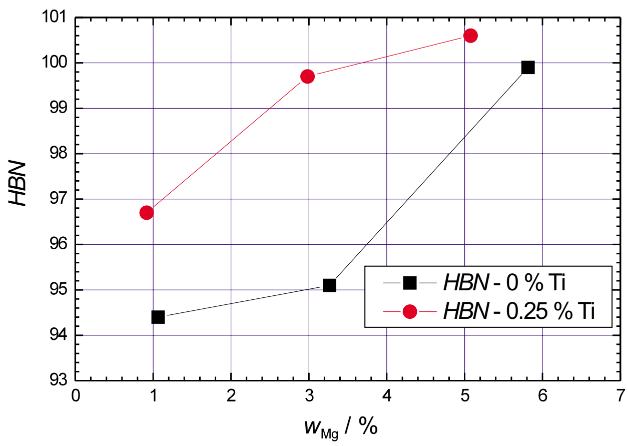

Figure 13: Hardness *HBN* of the AlCuMg alloys with different contents of magnesium and titanium

Slika 13: Trdota zlitin AlCuMg za različno vsebnostjo magnezija in titana

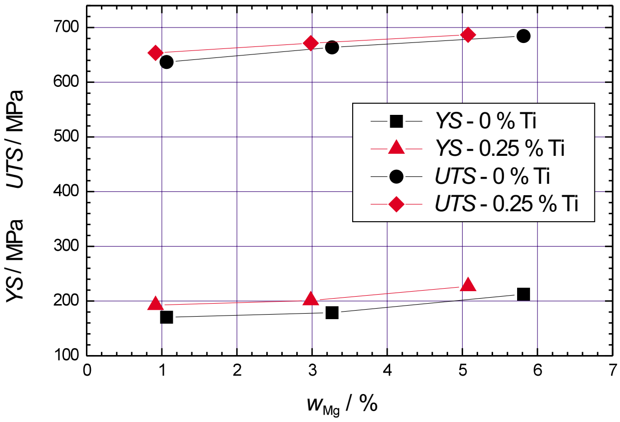

Figure 14: Compression strength of AlCuMg alloys with different contents of magnesium and titanium
Slika 14: Tlačna trdnost zlitin AlCuMg z različno vsebnostjo magnezija in titana

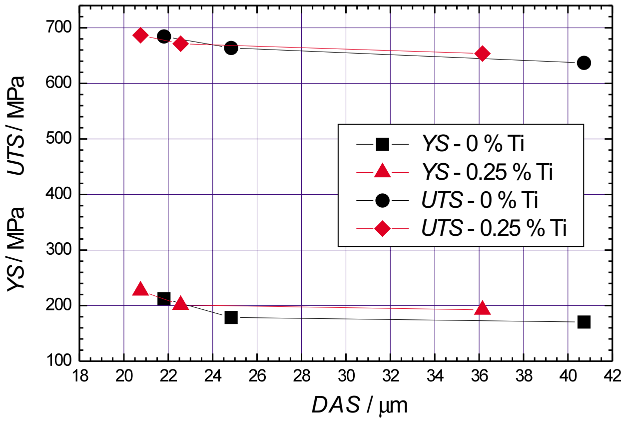

Figure 15: Compression strength of AlCuMg alloys in dependence of the secondary dendrite arm spacing (*DAS*)
Slika 15: Tlačna trdnost zlitin AlCuMg v odvisnosti od velikosti sekundarnih dendritnih vej

sub-grain size (**Figure 10**), the microvoltage (**Figure 11**) and the dislocation density (**Figure 12**).

The sub-grain size is the area of the crystal-grain lattice from which the X-rays are coherently diffracted. The sub-grains are separated with dislocation walls and have a space orientation that is different by several angle minutes. Using X-ray diffraction of polycrystals, the sub-grain is defined as a range of quantitative values, starting from the average length in a definite crystallographic direction, through the average volume, to their dimensional distributions. In alloys containing a lot of magnesium the average sub-grain size in the crystallographic direction [112] was found to increase.

Microvoltages are the most-often used parameter of crystal-lattice deficiency and represent the deviations in the distance, d, between two crystal planes that have identical {hkl} indices in a determined crystallographic direction. This kind of crystal-lattice deficiency is the result of the distribution of dislocations or differences in the chemical composition of the alloy.

The dislocation density is also a parameter of lattice defectiveness. It is most often defined as the minimum density of dislocation-free areas compared to the number of dislocations on the sub-grain boundaries.

The X-ray examination of the different aluminium-copper-magnesium alloys showed very high microvoltage values, which were to be expected because of the way the alloys were manufactured and the method used for their investigation.

3.3 Mechanical properties

Figures 13, 14 and 15 show the Brinell hardness and the compression strength as a function of magnesium content for the Al–Cu–Mg alloys with and without any added titanium. The changes in the chemical composition of the alloy cause changes in the microstructure that are reflected in the Brinell hardness and the compression strength. The hardness of the modified alloy is slightly higher than the hardness of the alloy without a modification treatment (**Figure 13**). By increasing the content of magnesium the hardness and compression strength also increase. The increase in the compression strength is only the result of increasing the amount of magnesium, and the influence of titanium is very small (**Figure 14**). It is well known that the strength of the face-centred cubic lattice in metals (and aluminium alloys) is almost insensitive to a change in grain size, sub-grain size or the secondary dendrite arm spacing [3], but a significant effect on the strength of these alloys from results from the dissolve strengthening of magnesium in aluminium. With increased amounts of magnesium in the alloy, the average values of the dendrite arm spacing are decreased and the compression strength increases. This means that the increase in the compression strength is mostly the result of an increase in the content of magnesium (**Figures 13–15**), while the strength of these alloys is almost independent of any change in the grain size, the sub-grain size or the secondary dendrite arm spacing.

4 CONCLUSIONS

Based on our findings the following conclusions can be drawn about the effect of the amount of magnesium and titanium on aluminium-copper-magnesium alloys:

– Using X-ray diffraction we established that the tetragonal intermetallic compound Al_2Cu and the orthorhombic intermetallic compound Al_2CuMg are formed across the whole range of magnesium additions.
– For the same content of titanium in the alloy and with increased amounts of magnesium, the average values of the dendrite arm spacing and the grain size are decreased. Also, in alloys containing a lot of magnesium the average values of the eutectic cell length and of the volume fraction of the eutectic were found to increase.

- For the same magnesium content the average grain size and dendrite arm spacing are decreased with an increasing titanium content. With the addition of AlTi5B1 a modification of the solidification structure and a smaller solidification grain size are obtained. We have confirmed that titanium is a very effective grain refiner.
- With increasing the magnesium content the sub-grain size is increased and the dislocation density is decreased.
- The compression strength and the hardness increase with the content of magnesium.
- The hardness of the modified alloy is slightly higher than the hardness of the alloy without any modification treatment. Any increase of the compression strength is only the result of increasing the content of magnesium, and the influence of titanium is very small.

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