Journal of Mining and Metallurgy, 39 (3‡4) B (2003) 509-526

CHARACTERIZATION OF MICROSTRUCTURE AND PROPERTIES OF ALCUMG ALLOYS

B. Zlaticanin[1], S. Duric[2], B. Jordovic[3] and B. Radonjic[1]

[1]University of Montenegro, The Faculty of Metallurgy and Technology, Cetinjski put bb, 81000 Podgorica, Serbia and Montenegro
[2]University of Belgrade, The Faculty of Mining and Geology, 11000 Belgrade
[3]The Faculty of Technical, 32000 Cacak, Serbia and Montenegro

(Received ; accepted)

Abstract

The effect of magnesium content, in the interval range from 1 mass.% to 5 mass.%, on the microstructure and properties of aluminium – copper – magnesium alloys was examined. The as-cast structure was modified by the addition of the AlTi5B1 to give alloys containing 0 to 0.25 mass. % titanium. Using X-ray powder diffraction we established that the tetragonal intermetallic compound Al_2Cu and orthorhombic intermetallic compound Al_2CuMg are formed across the whole range of magnesium additions.

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

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

Keywords: aluminium· copper· magnesium alloy, as-cast structure, intermetallic phases Al_2Cu and Al_2CuMg, lattice parameters, geometrical parameters, hardness and compression strength

1. Introduction

Excellent strength vs. density ratio, formability and corrosion resistance, make AlCuMg alloys a potential candidate for a number of industrial applications [1].

Developed in the early times in the aeronautical field, they have been then considered for a wide range of different applications, even though, due to their high specific strength, they are mainly considered as a substitute of iron-based materials for structural parts in the transportation industry. Several technical compositions are presently standardized and new alloys based on that metallic system are now 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, which has approximately the formula Al_2Cu**,** although some solid solubility exists. The addition of magnesium allows the formation of more intermetallic compounds, such as Al_2CuMg, Al_6CuMg_4, AlCuMg and $Al_5Cu_6Mg_2$. Magnesium increasesŠlĆthe strength and hardness of the alloys, but, especially in castings, this is accompanied by a decrease in ductility and impact resistance. Titanium is added as grain refiner and it is very effective in reducing the grain size. This reduction in grain size results in a better dispersion of insoluble constituents, porosity and nonmetallic inclusions, giving a decided improvement in mechanical properties. Since grain size controls the distribution[2] of porosity and 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, a tendency to form with a globular structure at higher copper contents was reported[1,2] and confirmed in our earlier unpublished work.

In this paper we have examined the as-cast structure in the AlCuMg system over a wide range of magnesium and titanium contents in order to investigate the effects of magnesium and titanium contents on the microstructure and properties of AlCuMg alloys. Depending on the alloy composition (say Cu content and Cu/Mg ratio), different phase distributions and consequently different material characteristics can be obtained. Characterization of eight different AlCuMg alloys having a copper content 5 mass.% and a Cu/Mg ratio 5; 2,5; 1,66; 1,25 and 1 respectively, is done by X-ray powder diffraction, quantitative microstructure analysis, by optical and electron microscope and by determined hardness and compression strength.

2. Experimental

The investigated materials were aluminium-copper-magnesium alloys with the chemical composition as 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 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 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, the both are active, in the AlCu5Mg2, AlCu5Mg3, AlCu5Mg4 and AlCu5Mg5 alloys in which the Cu/Mg ratio is in the range between 4:1 and 1:1 Al2CuMg controls the properties.

Experimental work can be divided in two phases. The first phase consists of melting and casting of samples with different compositions in the aluminium-copper-magnesium system, with addition of 0 mass.% to 0,25 mass.% Ti (AlTi5B1) as a modifier. The second phase includes characterization of samples obtained by previous melting and casting with X-ray powder diffraction, quantitative microstructure analysis, optical and electron microscope JCXA-733. The properties of these materials have been also examined including: the determination of hardness and compression strength measuring.

Table 1. Chemical composition of the investigated aluminium-copper-magnesium alloys (in mass. %)

Type of sample	%Al	%Fe	%Si	%Cu	%Zn	%Mg	%V	%Cr
AlCu5Mg1(0%Ti)	93.28	0.14	0.08	5.353	0.067	1.064	0.001	0.001
AlCu5Mg1(0,25%Ti)	93.54	0.14	0.06	4.915	0.060	0.917	0.006	0.001
AlCu5Mg2(0,25%Ti)	92.55	0.16	0.06	4.903	0.061	1.924	0.005	0.002
AlCu5Mg3(0%Ti)	91.29	0.17	0.07	5.113	0.067	3.266	0.001	0.002
AlCu5Mg3(0,25%Ti)	91.28	0.17	0.07	5.089	0.063	2.983	0.005	0.002
AlCu5Mg4(0,25%Ti)	90.12	0.15	0.15	5.030	0.063	4.235	0.004	0.002
AlCu5Mg5(0%Ti)	88.25	0.19	0.07	5.435	0.072	5.814	0.000	0.003
AlCu5Mg5(0,25%Ti)	89.38	0.18	0.08	4.901	0.063	5.077	0.005	0.003

The X-ray diffraction analysis was performed on the aluminium-copper-magnesium alloys: AlCu5Mg1, AlCu5Mg2, AlCu5Mg3, AlCu5Mg4 and AlCu5Mg5, using a wide range of angles (2θ) from 5 to 100^o with a step size of $0{,}02^o$ 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 30mA and 40kV, respectively. The width of the receiving slit (R) was 0,1mm, corresponding to fine focussed X-ray tubes. The radiation was the Cu $K\alpha_1/\alpha_2$, doublet ($\lambda\alpha_1$ = 1,54060 − and $\lambda\alpha_2$ = 1,54438 −).

Special attention was given to an assessment of the different structural parameters by quantitative microstructure analysis, using an automatic device for image analysis, QUANTIMET 500MC, and linear measuring method. Using automatic image analysis we were able to measure the grain size (minimum, maximum and average values - see Table 2), the dendrite arm spacing[3]-DAS (see Table 3 and Figs. 1 and 2), the eutectic cell length-Le (see Table 4 and Figs. 3 and 4), the relative standard measuring errors (RSE) for all mention parameters, as well as the distribution by grain size and volume fractions of the α-solid solution and the eutectic.

Table 2. Grain size for different magnesium contents in aluminium-copper-magnesium alloys

Type of sample	Average, μm	min, μm	max, μm	RSE, %
AlCu5Mg1 (0%Ti)	166.85	30	375	3.278
AlCu5Mg1 (0,25%Ti)	83.41	30	195	3.465
AlCu5Mg2 (0,25%Ti)	79.37	15	155	2.932
AlCu5Mg3 (0%Ti)	143.81	30	300	2.657
AlCu5Mg3 (0,25%Ti)	72.18	30	135	2.982
AlCu5Mg4 (0,25%Ti)	63.24	15	140	3.987
AlCu5Mg5 (0%Ti)	95.62	30	210	2.342
AlCu5Mg5 (0,25%Ti)	52.18	30	105	2.768

Table 3: Dendrite arm spacing (DAS) for different magnesium contents in aluminium-copper-magnesium alloys

Type of sample	average, μm	min, μm	max, μm	RSE, %	Vv,αh.s. vol%
AlCu5Mg1 (0%Ti)	40.72	0.83	252	2.89622	91.350
AlCu5Mg1 (0,25%Ti)	36.15	0.83	243	2.66070	91.294
AlCu5Mg2 (0,25%Ti)	27.10	0.83	188	2.23466	86.967
AlCu5Mg3 (0%Ti)	24.83	0.83	147	2.12997	83.252
AlCu5Mg3 (0,25%Ti)	22.56	0.83	166	2.18237	86.783
AlCu5Mg4 (0,25%Ti)	21.66	0.83	165	1.97073	85.407
AlCu5Mg5 (0%Ti)	21.81	0.83	132	2.11418	85.614
AlCu5Mg5 (0,25%Ti)	20.75	0.83	124	2.11840	86.092

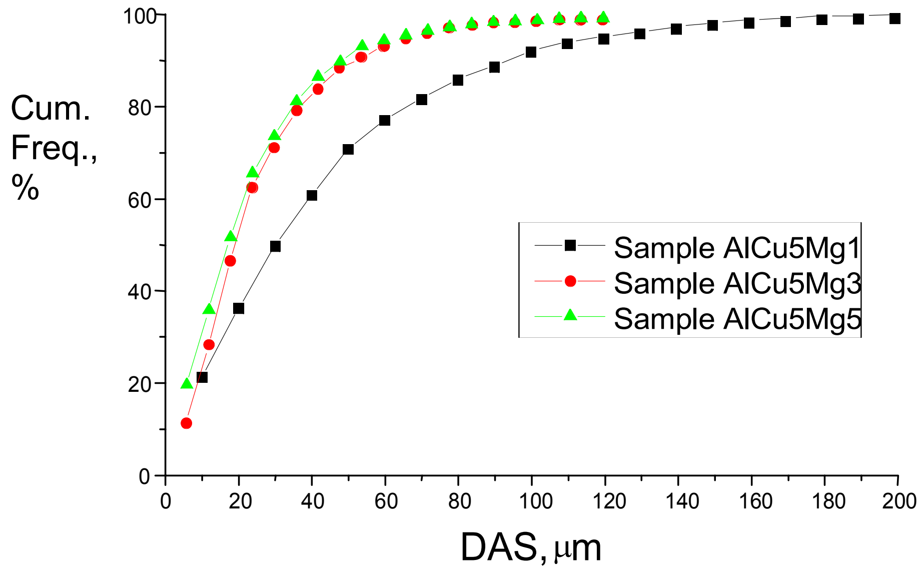


Fig. 1 Comparative cumulative frequency for the non-modified alloys

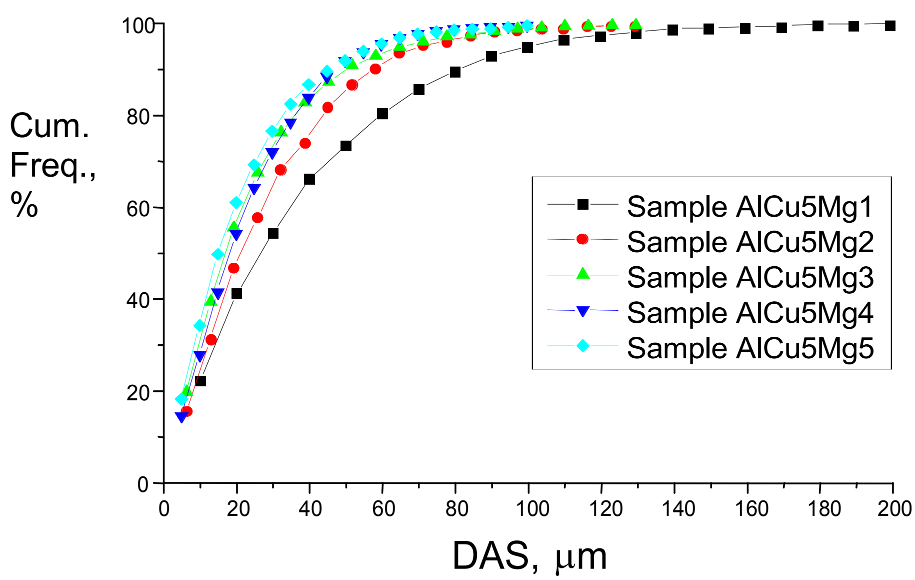


Fig. 2 Comparative cumulative frequency for the modified alloys

Table 4. The eutectic cell length-Le and volume fraction of eutectic for different magnesium contents in aluminium-copper-magnesium alloys

Type of sample	average, μm	min, μm	max,μm	RSE, %	Vv,e. vol%
AlCu5Mg1 (0%Ti)	3.32	0.32	27.74	3.06795	8.648
AlCu5Mg1 (0,25%Ti)	2.22	0.32	31.29	3.00930	8.052
AlCu5Mg2 (0,25%Ti)	3.67	0.32	33.87	3.05049	13.017
AlCu5Mg3 (0%Ti)	4.36	0.32	79.03	2.82088	16.745
AlCu5Mg3 (0,25%Ti)	3.75	0.32	39.03	3.04168	13.214
AlCu5Mg4 (0,25%Ti)	3.76	0.32	33.87	2.79790	14.588
AlCu5Mg5 (0%Ti)	4.55	0.32	38.23	2.40883	14.382
AlCu5Mg5 (0,25%Ti)	3.82	0.32	32.06	2.34632	13.905

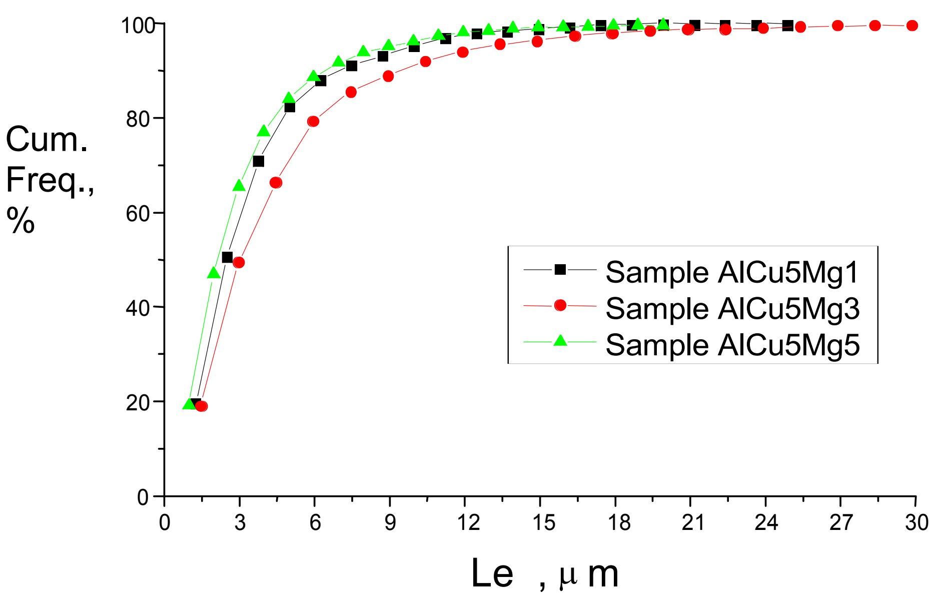


Fig. 3 Comparative cumulative frequency for the non-modified alloys

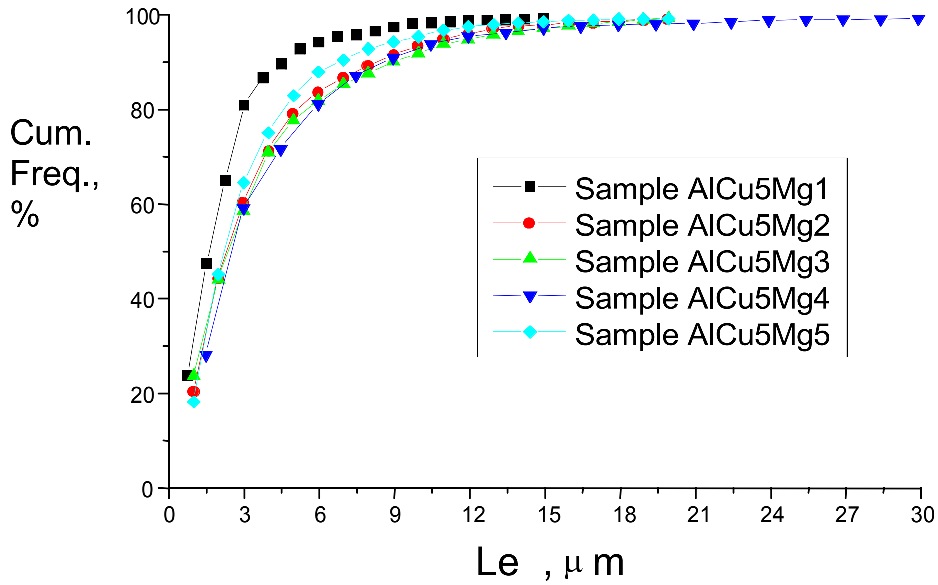


Fig. 4 Comparative cumulative frequency for the modified alloys

3. Results and Discussion

The copper content in the standard AlCuMg alloys (by EN AC 21000 or by 3.2502.00 JUS C.C2.300) was up to about 5 mass. %, slightly below the value of 5,65 mass. % that represents the maximum solid solubility of copper in aluminium at the eutectic temperature of 548°C. These investigated AlCuMg alloys have dendritic-cellular structures. Micrographs show dendrites of aluminium solid solution as the primary phase, with a eutectic mixture filling the interdendritic spaces. The eutectic is of the divorced type · particles of a second phase in a solid solution. The second phase can be an intermetallic compounds that contain aluminium and one or more alloying elements (Al_2Cu and Al_2CuMg); intermetallic compounds that do not necessarily contain aluminium (Mg_2Cu or $MgCu_2$); or an alloying element, such as copper or magnesium, depending on the composition of the alloy. Cast AlCuMg alloys contain soluble phases: Al_2Cu or Al_2CuMg which appear in various amounts and at various locations in the microstructure, depending on the thermal history of the specimen.

The Figs. 5-12 obtained by applying optical microscope. Besides, microstructure was observed by applying electron microscope JCXA-733. The current and the voltage during the analysis for the copper, magnesium and titanium were 1x10^{-8} A and 20kV, respectively, and we applied Kα radiation. The content of copper and magnesium in the white phase is low. X-ray analysis showed the presence of magnesium in the eutectic gray phase, while copper is found in the bright phase. Titanium is present in platelets in some eutectic area in white phase, isolated particles of containing titanium are found also dispersed in the interior of the matrix grains.

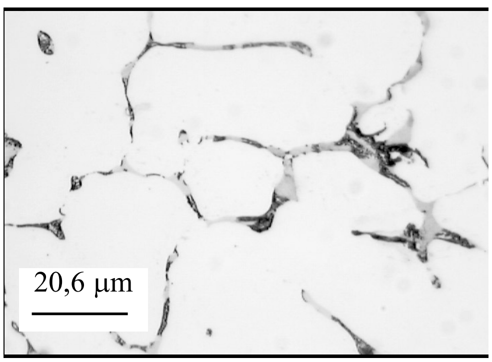


Figure 5. Microstructure of $AlCu_5Mg_1$ (0%Ti) alloy, Keller's reagent

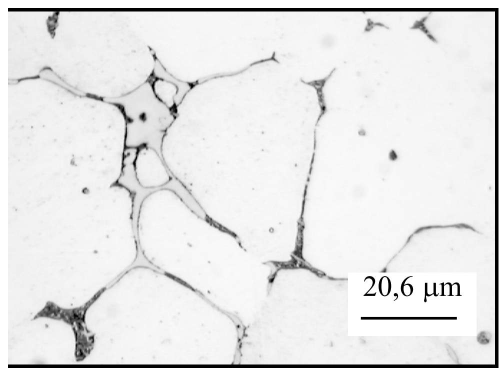
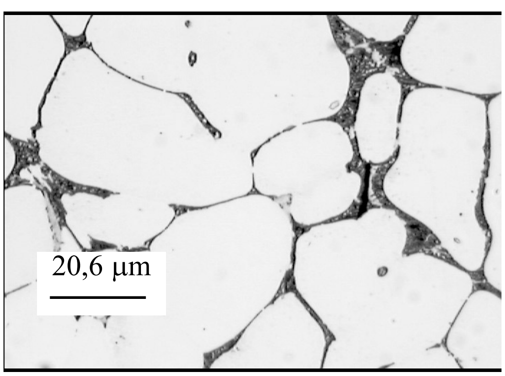


Figure 6. Microstructure of $AlCu_5Mg_1$ (0,25%Ti) alloy, Keller's reagent



Figyre 7. Microstructure of $AlCu_5Mg_2$ (0,25%Ti) alloy, Keller's reagent

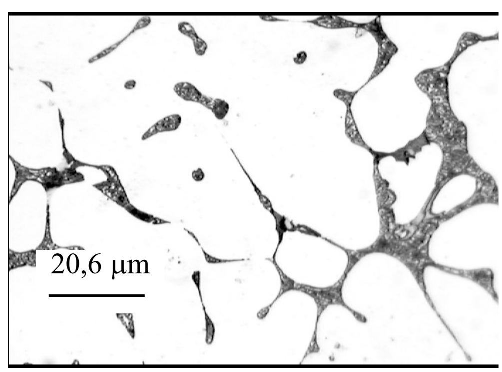


Figure 8. Microstructure of $AlCu_5Mg_3$(0%Ti) alloy, Keller's reagent

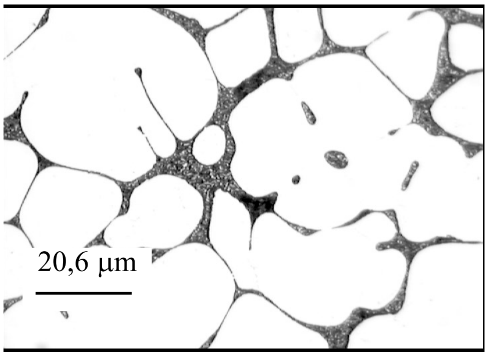


Figure 9. Microstructure of $AlCu_5Mg_3$ (0,25%Ti) alloy, Keller's reagent

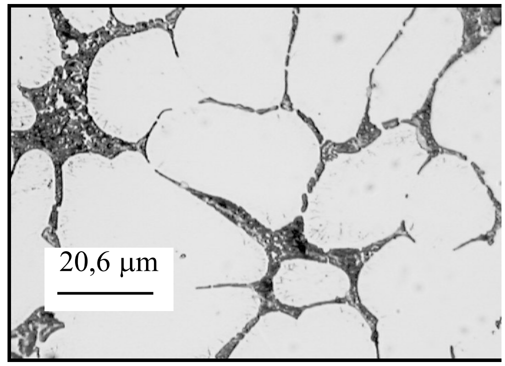


Figure 10. Microstructure of $AlCu_5Mg_4$ (0,25%Ti) alloy, Keller's reagent

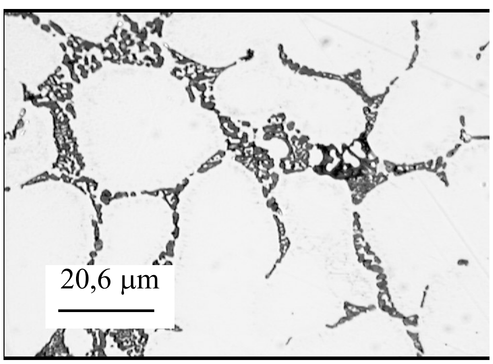


Figure 11. Microstructure of $AlCu_5Mg_5$ (0%Ti) alloy, Keller's reagent

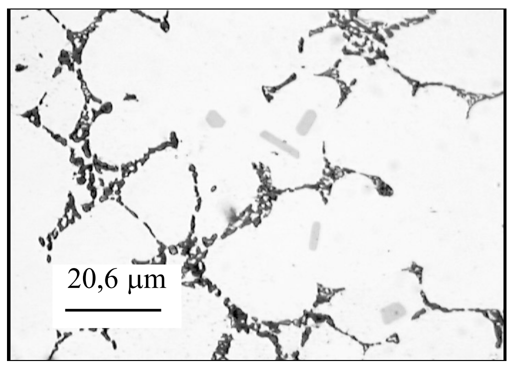


Figure 12. Microstructure of $AlCu_5Mg_5$ (0,25%Ti) alloy, Keller's reagent

3.1 The X-ray analysis

Using X-ray diffraction we established that the tetragonal intermetallic compound Al_2Cu and orthorhombic intermetallic compound Al_2CuMg are formed across the whole range of magnesium additions. Obtained results for the lattice parameters of tetragonal intermetallic compound Al_2Cu are: for AlCu5Mg1 alloy a = 6,034 Å, c = 4,869 Å and V = 177,3 Å^3; for AlCu5Mg2 alloy a = 6,036 Å, c = 4,864 Å and V = 177,2 Å^3; for AlCu5Mg3 alloy a = 6,030 Å, c = 4,877 Å and V = 177,4 Å^3; for AlCu5Mg4 alloy a = 6,054 Å, c = 4,889 Å and V = 179,2 Å^3; for AlCu5Mg5 alloy a = 6,058 Å, c = 4,880 Å and V = 179,1 Å^3. Obtained results for the lattice parameters of orthorhombic intermetallic compound Al_2CuMg are: for AlCu5Mg1 alloy a=3,990 Å, b =9,209 Å, c =7,128 Å and V = 262,1 Å^3; for AlCu5Mg2 alloy a=3,993 Å, b =9,210 Å, c =7,129 Å and V = 262,2 Å^3; for AlCu5Mg3 alloy a=4,051 Å, b =9,333 Å, c =7,064 Å and V = 267,1 Å^3; for AlCu5Mg4 alloy a=4,011 Å, b =9,291 Å, c =7,116 Å and V = 265,2 Å^3; for AlCu5Mg5 alloy a=4,011 Å, b =9,283 Å, c =7,109 Å and V = 264,74 Å^3. Obtained results for the lattice parameters of tetragonal intermetallic compound Al_2Cu and for the lattice parameters of orthorhombic intermetallic compound Al_2CuMg are agree with published data from literature: JCPDS card 25 0012 for Al_2Cu are a = 6,065 Å, c = 4,873 Å and V = 179,28 Å^3; and JCPDS card 28 0014 for Al_2CuMg are a =4,000 Å, b =9,250 Å, c =7,150 Å and V = 264,55 Å^3.

From the X-ray diffractograms (Figs. 13-17) the microstructural parameters have been calculated: the average sub-grain size (Table 5), the microvoltage (Table 6) and the dislocation density (Table 7).

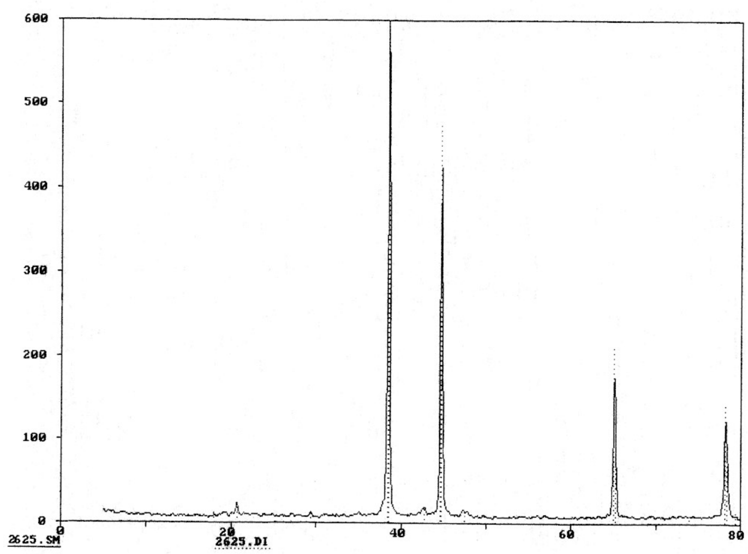


Figure 13. Diffractogram of AlCu5Mg1 alloy

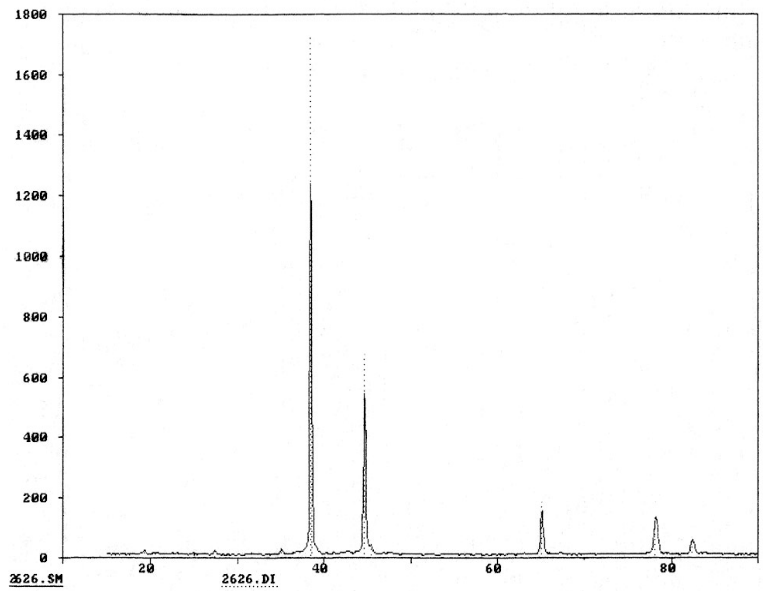


Figure 14. Diffractogram of $AlCu_5Mg_2$ alloy

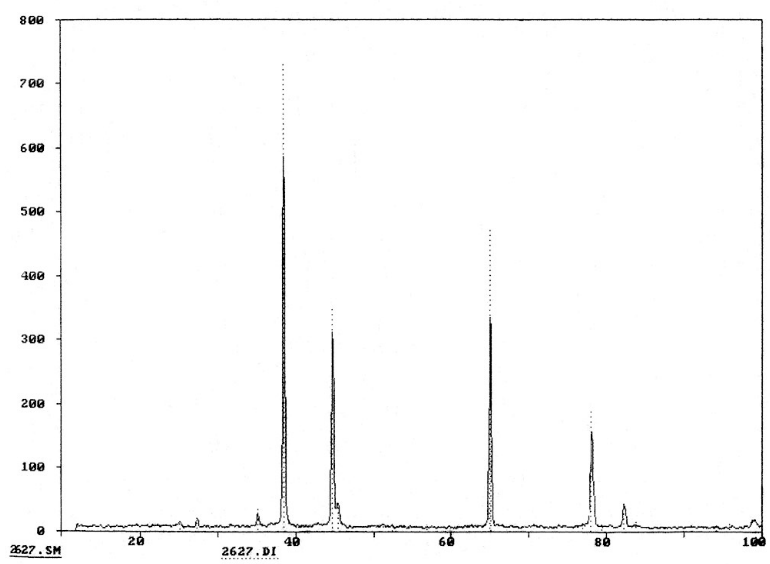


Figure 15. Diffractogram of $AlCu_5Mg_3$ alloy

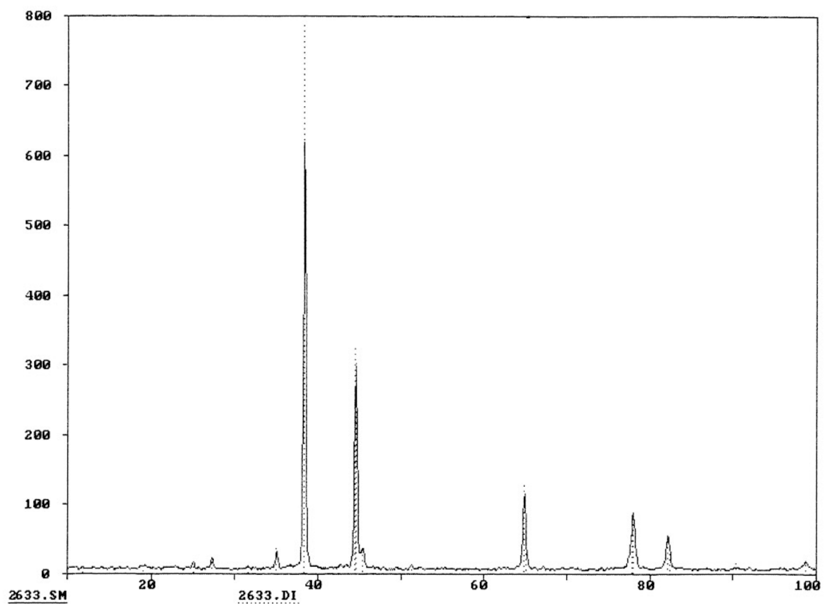


Figure 16. Diffractogram of $AlCu_5Mg_4$ alloy

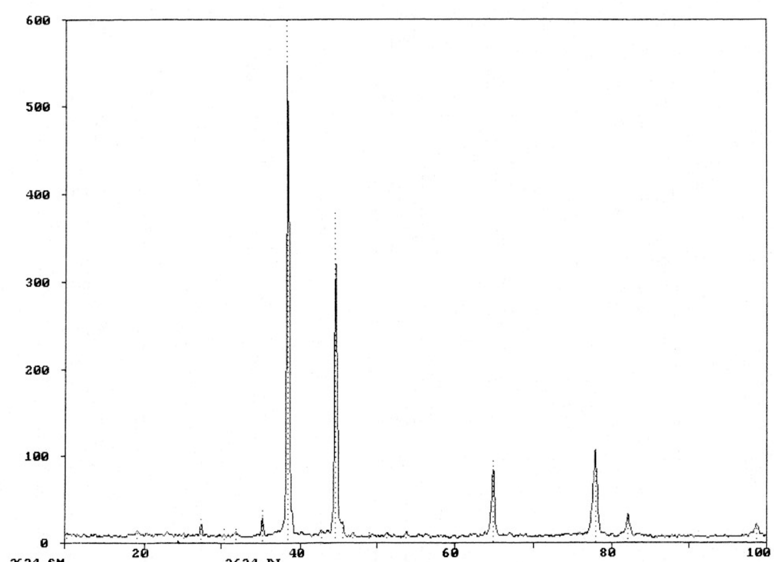


Figure 17. Diffractogram of $AlCu_5Mg_5$ alloy

Table 5. Average sub-grain size in the crystallographic direction [112] for different magnesium contents in aluminium-copper-magnesium alloys

Type of sample	Average sub-grain size, Å
AlCu5Mg1	198
AlCu5Mg2	463
AlCu5Mg3	579
AlCu5Mg4	772
AlCu5Mg5	780

Table 6. The microvoltage in the crystallographic direction [112] for different magnesium contents in aluminium-copper-magnesium alloys

Type of sample	Microvoltage (mV)
AlCu5Mg1	0.3065
AlCu5Mg2	0.1583
AlCu5Mg3	0.1265
AlCu5Mg4	0.0950
AlCu5Mg5	0.0946

The sub-grain is the range of the lattice of the crystal grain from which the X-rays are coherently diffracted. The sub-grains are separated by dislocation walls and have a space orientation which 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 high magnesium the average sub-grain size in the crystallographic direction [112] were found to increase.

Table 7. Dislocation density in the direction [112] for different magnesium contents in aluminium-copper-magnesium alloys

Type of sample	Dislocation density, cm^{-2}
AlCu5Mg1	76.5×10^{10}
AlCu5Mg2	13.9×10^{10}
AlCu5Mg3	8.9×10^{10}
AlCu5Mg4	5.0×10^{10}
AlCu5Mg5	4.9×10^{10}

Microvoltages are the most-often used parameter of crystal-lattice deficiency and represent the deviations in the distance d between two crystal planes having identical šhklć indices in a determined crystallographic direction. This kind of a 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 the 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 (Table 6), which were expected because of the way the alloys were manufactured and the method used to investigate them.

3.2. Quantitative microstructure analysis

Using automatic image analysis we were able to measure the grain size, dendrite arm spacing-DAS, the eutectic cell length-Le and volume fractions of the α-solid solution and the eutectic.

Measurement of dendrite arm spacing is accomplished in the same manner as grain size measurement, that is, by the intercept method. Dendrite arm spacing is an important consideration in cast aluminium-copper-magnesium alloy microstructures. From the results of these measurements, information can be obtained regarding the rate of solidification of the material and therefore some indication of the strength of the material. For example, the finer the dendrite arm spacing is, the higher the strength.

Measured grain sizes are expressed in the mean diameter per grain.The grain size and the distribution of dendrites and eutectic depend on the casting parameters [4], the melt temperature and the solidification rate, which also affect the properties of the alloys. Also, addition of a grain refiner (AlTi5B1) resulted in the nearly equiaxed structure shown in micrographs. The addition of titanium and boron in form of the alloy AlTi5B1 are used to produce particles of TiB_2 in the melt. These particles are then the 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 also improvement in structure uniformity as a result of adding a grain refiner is shown in figs. 5-12 and Table 2. Also, size and shape are affected by the addition of magnesium. With increased amounts of magnesium for the same content of titanium in the alloy, the average values of the dendrite arm spacing and grain size are decreased, and we obtain a fine, uniform grain structure, as shown in micrographs and Table 2-3. Also, in alloys containing high magnesium the average values of the eutectic cell length and volume fractions of the eutectic were found to increase (Table 4). Chemical composition affects structure through its influence on phase relations.

3.3 Mechanical properties

The Brinell hardness and the compression strength are shown in Table 8. The changes in chemical composition of the alloy cause changes in the structure that are reflected in the Brinell hardness and the compression strength. The hardness of the modified alloy is higher than the hardness of the alloy without any modification treatment. By increasing the content of magnesium and titanium the hardness and compression strength also increase.

Table 8. Hardness and compression strength of aluminium-copper-magnesium alloys with different amounts of magnesium

Type of sample	HBaverage	$R_{p0,2}$ (MPa)	R_m (MPa)
AlCu5Mg1 (0%Ti)	94.4	170.5	636.9
AlCu5Mg1 (0,25%Ti)	96.7	192.4	653.7
AlCu5Mg2 (0,25%Ti)	97.8	196.8	671.1
AlCu5Mg3 (0%Ti)	95.1	178.7	663.7
AlCu5Mg3 (0,25%Ti)	99.7	201.0	671.4
AlCu5Mg4 (0,25%Ti)	99.8	208.6	681.7
AlCu5Mg5 (0%Ti)	99.9	212.6	684.5
AlCu5Mg5 (0,25%Ti)	100.6	226.9	686.6

4. Conclusions

Based on our findings we can draw the following conclusions about the effect of the content of magnesium on aluminium-copper-magnesium alloys:

-Using X-ray diffraction we established that the tetragonal intermetallic compound Al_2Cu and orthorhombic intermetallic compound Al_2CuMg formed across the whole range of magnesium additions.

-With increased amounts of magnesium for the same content of titanium in the alloy, the average values of the dendrite arm spacing and grain size are decreased. Also, in alloys containing high magnesium the average values of the eutectic cell length and volume fractions of the eutectic were found to increase. (see Table 2-4).

-With the same chemical composition but increased titanium content, the average values of the grain size and dendrite arm spacing are decreased (see Table 2-4). With the addition of AlTi5B1 a modification to the solid ification structure and smaller solidification grains are obtained. We confirmed that titanium is a very effective grain refiner.

-Compression strength and hardness increase with the content of magnesium and titanium (Table 8).

References

1. L.F. Mondolfo, *Aluminium Alloys: Structure and Properties*, Butterworth and Co (Publishers) Ltd, London (1976) 253.
2. X. Yang, J.D. Hunt and D.V. Edmonds, *A quantitative study of grain structures in twin-roll cast aluminium alloys*, part II: AA 3004, Aluminium, 69(2)(1993)158-162.
3. AM. Samuel, FH. Samuel, *Effect of Alloying Elements and Dendrite Arm Spacing on the Microstructure and Hardness of an Al-Si-Cu-Mg-Fe-Mn (380)*, Journal of Materials Science, 7(4)(1995)1698-1708.
4. B. Radonjic, *Directionality of Cast Aluminium Structure*, Aluminium, 58(11)(1982)646-649.