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MECHANICAL PROPERTIES OF HIGH COPPER CONTAINING Al-Cu-Si CAST ALLOYS AT ELEVATED TEMPERATURE

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ABSTRACT

The automotive applications of heat treatable AlCu cast alloys are designed for performance at high temperature, which can be improved using specially alloying element as well as different heat treatment. In this research work an effect of alloying element such as Cu, Ni and heat treatment on microstructures and mechanical properties of AlCu alloy were investigated using optical microscope (LIMI), scanning electronic microscope (SEM) equipped with energy dispersive X-ray detection (EDS), Differential scanning calorimetry (DSC) and mechanical testing. It was found that during solidification most of the elements Cu, Mg and Ni segregated at the grain boundaries in the form of Al_2CuMg or Al_7Cu4Ni and a very small part of Cu and Ni element dissolved in the FCC-Al matrix. The latter is not completely reclaimable for strengthening mechanisms in FCC-Al matrix in its as-cast state. The results of mechanical test show that yield strength and ultimate tensile strength increases after heat treatment significantly compared with the as-cast state. Furthermore the mechanical properties at elevates temperature (240°C) is higher with higher Cu+Ni+Si containing alloys compared with the less Cu+Ni+Si containing alloy.

KEYWORDS

Precipitation, Ni, Intermetallic phases, Thermodynamic simulation, SEM

INTRODUCTION

Due to the high strength-to-weight ratio, high strength and ductility at room- and elevated temperature, AlCuSi cast alloys are widely applied in the automotive industries specially for producing cylinder head and engine block, where modified alloys with enhanced metallurgical properties are always needed to satisfy the ever increasing demands for the applications. The AlCu based cast alloys can be strengthened by age hardening. The precipitation processes from which this hardening is derived can be attributed to the presence of selected trace element additions or micro alloying, which can change the process or kinetics of precipitation in age hardenable AlCu cast alloys (Polmear, 1998; Morris, 1991). Furthermore their strength is mainly controlled by the aging process through the precipitation and growth of very fine precipitates of the θ′-phase (a semi-coherent, metastable form of the equilibrium Al_2Cu phase) from the Al and Cu Guinier-Preston (GP) zones. Several studies have shown that solute clustering occurs prior to the precipitation of GP zones, and this modifies both the nature and kinetics of the precipitation process. The typical size of GP zones is in the order of tens of nanometers, and the chemical composition of these precipitates is not easily measured, even with a typically analytical electron microscope (Polmear, 1989).

The typically microstructure of as-cast AlCuMgSi alloys consists of FCC Al-Matrix, eutectic Si, Mg_2Si, θ-Al_2Cu, S-Al_2CuMg, Q- $Al_5Cu_2Mg_8Si_6$ and different Fe- Containing intermetallic phases, which is preferably determined by the Cu content.

Numerous studies have determined that the adding of alloying elements such as Ni, Zr etc. can improve the microstructures, mechanical properties, thermal stabilities and hot tearing resistance of AlCu based alloys. This is attributed to grain refinement and the presence of the coherent dispersion-strengthening phase such as Al_3Ni, with an L_{12} crystal structure. The L_{12} crystal structure (FCC) has the largest lattice parameter misfit with Al (a=4.215 Å and d = +4.08% at room temperature), thereby improving creep resistance by enhancing elastic interactions with dislocations (Karnesky, 2006; Marquis, 2002; Krug, 2011; Christopher, 2012).

The solid solubility of Ni in Al does not exceed 0.04%. Over this amount, it is present as an insoluble intermetallic, usually in combination with Fe. Ni (up to 2%) increases the strength of high-purity aluminum but reduces ductility due to the formation many complex intermetallic phases such as Al_3Ni, Al_3CuNi, Al_7Cu_4Ni, Al_9FeNi. These phases have much bigger contributions to the elevated-temperature properties of AlCu piston alloys, owing to their better thermal stability, mechanical properties, morphologies and distributions (Lia, 2010; Asghar, 2011). It is generally considered that the complex eutectic which formed in last section of solidification at the grain boundaries are the weakest areas in alloys at elevated temperature. At elevated temperature, the thermally stable intermetallic like Cu- or Ni-containing phases could impose drag on boundaries and help to increase the elevated-temperature strength (Lia, 2010; Asghar, 2011).

The aim of this research work is investigation of the mechanical properties of AlCuSi cast alloys at elevated temperature without adding expensive element such as Ce, Co, Ag and evaluates their suitability as a potential alternative to the AlSiCu cast alloys traditionally used for the production of cylinder heads and engine blocks etc.

SIMULATION AND EXPERIMENTAL

The investigation of AlCu alloys for high temperature application require investing huge efforts in the selection of the alloying elements such as Cu, Mg, Si, Ni and the determination of potential compositions as well as their heat treatments. This effort and cost can be reduced by simulation of material based on thermodynamic considerations.

ThermoCalc calculations are based on thermodynamic data which is supplied in a database. There exists a wide selection of high quality databases for various purposes that include many different materials.

In this study an intermetallic phases depended on alloying system was calculated by the software ThermoCalc, the database used was TTAl8. For the forecast of precipitated phases in the as-cast microstructure at room temperature equilibrium conditions were chosen. Three AlCuSi cast alloys were selected for this work: one without Ni and two containing 1 wt.% Ni (Table 1).

Table 1. Nominal range of alloy composition

Content [wt.%]	Cu	Mg	Si	Fe	Mn	Ni	Ti
Alloy 1, AlCu4Mg1	4.0	1.0	0.5	0.3	0.7	0	0.2
Alloy 2, AlCu9Mg1	9.0	1.0	0.5	0.3	0.7	0.5	0.2
Alloy 3, AlCu9Si4Ni1	9.0	1.0	4.0	0.3	0.7	1.0	0.2

The melt was prepared in a SiC crucible using a resistance furnace by melting master alloys. The composition was checked using a spark spectrometer after each addition. Before casting, the melt was degassed with pure dry argon gas via an impeller for at least 30 min and the reduced pressure test was carried out to ensure a low level of hydrogen. With the melt at a temperature of 730°C the ingots were cast in a cylindrical iron mold with an internal diameter of 65 mm. Prior to casting the mold was pre-heated to a temperature between 240–280°C. Half of the cast billets were subjected to an industrial solution heat treatment at 510°C for 10 h. After solution heat treatment the parts were cooled at pressurized air to room temperature and an artificial ageing was done for 12 h at a temperature of 185°C. Metallographic samples were taken from the billet of each alloy and different condition. The behavior of the precipitation phases as a function of heat treatment was also examined using Differential scanning calorimetry (DSC) by the Perkin Elmer Netzsch-204 F1 instrument. The tested samples with thickness 2 mm, diameter 5mm (77–80 mg) were prepared by machining. Each sample was heated at a rate of 10 K/min from room Temperature to 650°C, holding for 5 min, and cooling at a rate of 40 K/min. The microstructure was analyzed by using light microscopy, scanning electron microscopy (SEM) and energy dispersive X-ray detection (EDS). For mechanical testing according to DIN 50125 – D08 mm, samples were machined parallel to the casting direction and at least five tensile samples were tested. Mechanical properties were evaluated in tension test at room temperature as well as elevated temperature (240°C). The presented results of mechanical properties are average values of at least five tension tests.

RESULTS AND DISCUSSION

Thermodynamic Calculation of Phase Equilibria in the AlCuSiMg System

Figure 1 shows a calculated isopleth in terms of temperature (°C) versus Ni and Mg for five component Al alloys Al-10 wt.% Cu, 4 wt.% Si, 1 wt.%Ni and 0.3 wt.% Mg. This calculation was made using a recently developed thermodynamic database TTAL8 for aluminum alloys. As it can be seen in Figure 1a, the solidus temperature (T_S) increases when 1 wt.% Ni is added due to the formation of high temperature stable Al_7Cu_4Ni phase. The ternary Al_7Cu_4Ni phase is in equilibrium with Al and Cu element can replace Ni in Al_3Ni_2 phase (Mondolofo, 1976). For alloys with Ni > 1 wt.% the solidus temperature is equal to the temperature of the reaction liq.+Al_3Ni_2+FCC$\rightarrow$$Al_3Ni_2$+FCC+$Al_7Cu_4Ni$ at 533°C. Generally Ni element in AlCu alloys reduces the rate of diffusion of Cu element in FCC Al-matrix due to the high potential formation of Al_7Cu_4Ni phase. Moreover the additions of Ni element to AlCu alloys produce some grain refinement during solidification (Mondolofo, 1976) and improve the mechanical properties at elevated temperature due to the formation of different high temperature stable Al_3Ni_2 or Al_7Cu_4Ni phases. Figure 1b shows an effect of Mg element on phase formation on AlCuMgSiNi alloys. The quaternary stable phase $Al_5Cu_2Mg_8Si_6$ appears only in high Si containing alloys. This phase would be formed from solidification reaction during equilibrium condition liq.+FCC+Si+$Al_2Cu$$\rightarrow$liq.+FCC+ $Al_5Cu_2Mg_8Si_6$+Al_2Cu with Mg> 0.8 wt.% at 515°C. The $Al_5Cu_2Mg_8Si_6$ phase is often present with Al_2Cu+Si and plays an important role in precipitation hardening and mechanical properties of AlCuSi based alloys after heat treatment.

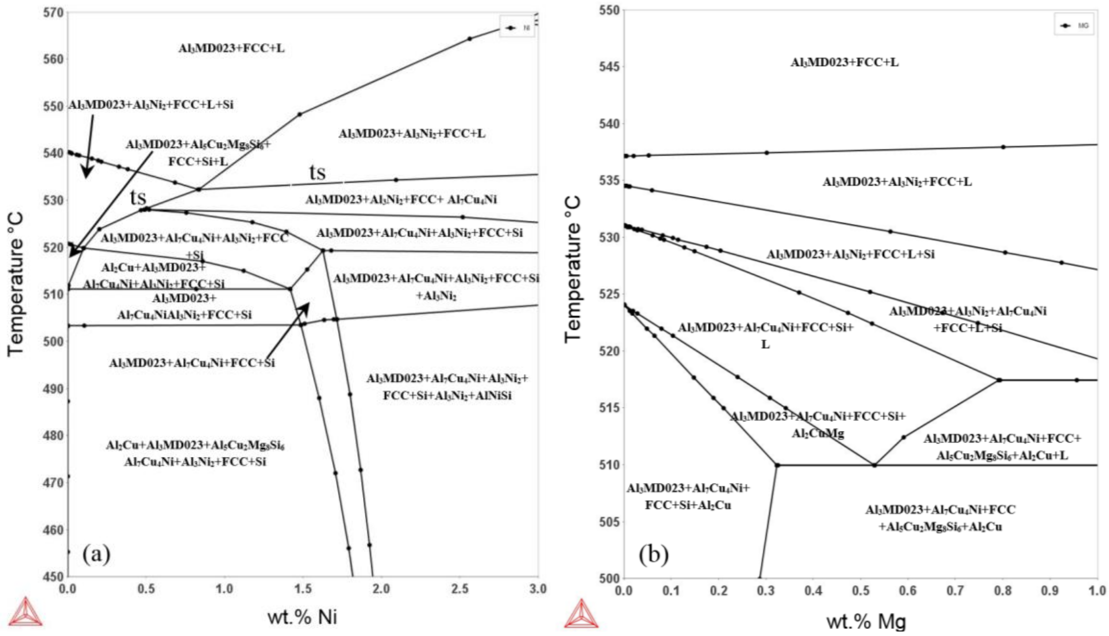

Figure 1. A calculated isopleth of AlCuMgSiNi phase diagram for 9 wt.% Cu, 4 wt.% Si, 1 wt.%Ni and 1 wt.% Mg a) effect of Ni element on solidification behavior and phase formation b) effect of Mg element on solidification behavior and phase formation

Microstructure

The as-cast microstructures of the alloys AlCu4Mg1, AlCu9Mg1 and AlCu9Si4Ni1 are shown in Figures 2a, b, and c, respectively. A typical microstructure of AlCu alloy includes Al-matrix phase, Al_2Cu- or $Al_2Cu(Mg)$- eutectic phase and particles of the intermetallic compounds, which were reported to be Fe-, Ni- and Mn-containing phases due to the secondary alloying elements, can be seen in Figure 2. Furthermore a microstructure of alloy 2 includes a high amount of Al_2Cu-, $Al_2Cu(Mg)$- and AlFeMnNi phase. The microstructure of alloy 3 existence a high surface fraction of Al_2Cu-, $Al_2Cu(Mg)$-, Al_7Cu_4Ni- and AlFeMnNi- phase as well as AlSi eutectic (Figure 2c).

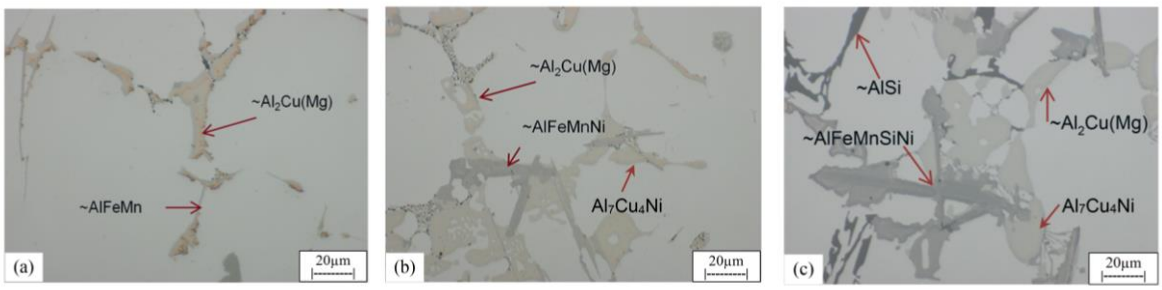

Figure 2. Optical microscope images of alloy in as cast state, (a) AlCu4Mg1, (b) AlCu9Mg1, (c) AlCu9Si4Ni1

Moreover from Figure 2 it is seen that the microstructure of alloy 3 consists of fine dendritic microstructure in comparison to 1 and 2. It was assumed that addition of Ni to AlCu alloying-system with another grain refinement element such as Ti and B resulted in changes slightly the morphology of the FCC Al-Matrix from coarse to fine dendrites microstructure (Mondolofo, 1976). The effect of heat treatment on microstructures specially for alloy 2 and 3 have been investigated using SEM (Figure 3) equipped with energy dispersive spectrometry (EDS)-analysis (see Table 1). Figure 3 and table 2 reveal the presence of different phases including Cu and Si as well as Ni containing phases at the grain boundary or in the FCC Al-matrix. Within the investigated composition ranges in spectrum 1 to 3 for each alloy, different phases such as ~Al_5(FeMnSi)3, ~Al_2CuMg and ~Al_7Cu_4Ni phases are found in the microstructures independent of heat treatment (Figure 3 a–d). It seems that the Cu in solid solution was reduced by the formation of Al_7Cu_4Ni intermetallic phases

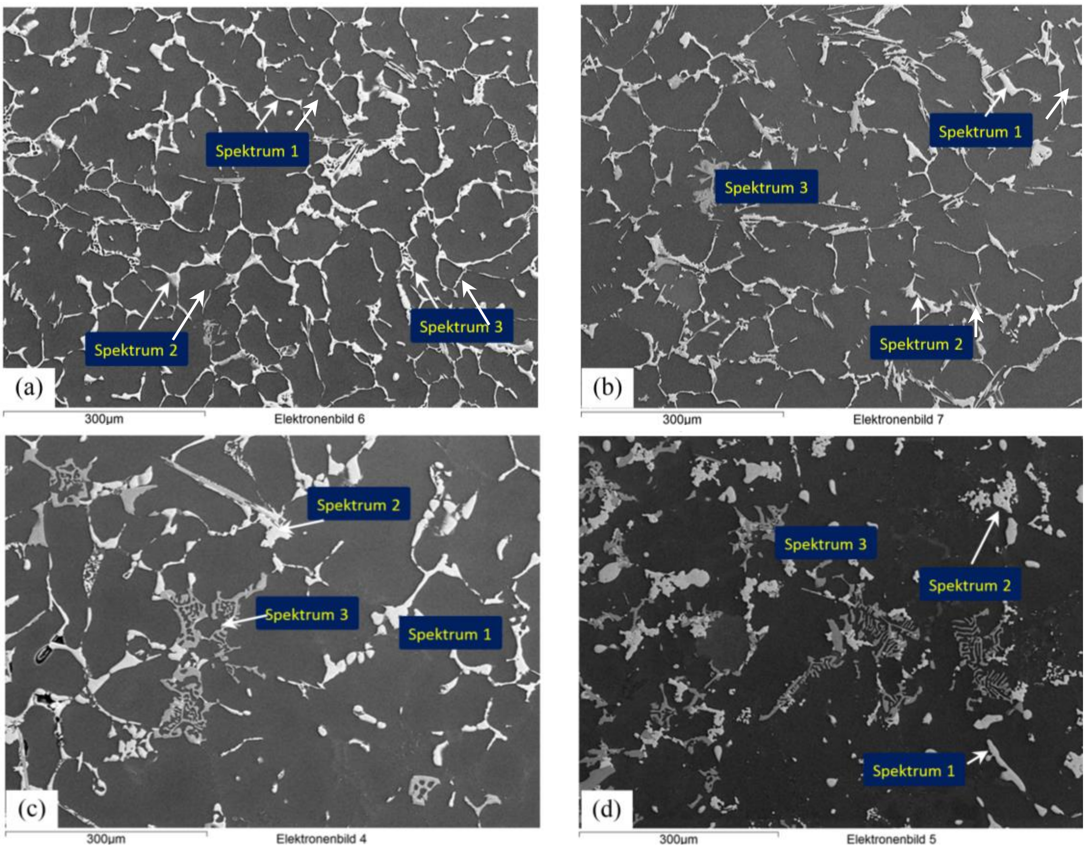

Figure 3. Backscattered SEM- micrograph of tested samples, (a) alloy 2- as cast, (b) alloy 2- after heat treatment (c) alloy 3- as cast, (d) alloy 3- after heat treatment

The further analyses of the Cu-rich intermetallic compounds showed that in the sample without heat treatment a significantly higher area fraction of Al_2CuMg- and Al_7Cu_4Ni phase with the blocky and fine platelet-like morphology was present at the grain boundary (Figure 3a and c) in comparison to heat treated alloy (Figure 3b and d). Generally it can be found that the amounts of binary and ternary eutectic microstructures in the alloy without heat treatment are more than in the alloy after heat treatment. Moreover, the homogenization at 510°C for 10 h dissolved a high amount of secondary elements, such as Cu, Mg, etc., into the Al-matrix. The Al_2CuMg phase with lower mass fraction [%] of Cu element and Mg element was particularly found in the alloy after homogenization (see table 2, comparison Spectrum 1 for each alloy depended on heat treatment).

Table 2. Compound compositions at grain boundaries in Alloy 2 (AlCu9Mg1) and alloy 3 (AlCu9Si4Ni1) detected by quantitive SEM/EDS, as cast and after heat treatment (HT)

Content		Spectrum	Cu [at%]	Mg [at%]	Si [at%]	Fe [at%]	Mn [at%]	Ni [at%]	Zr [at%]	Al [at%]
AlCu9Mg1	as cast	1	43.17	0.51	0.22	050	042	0.08	0.01	Bal
		2	39.25	0.56	0.45	0.61	0.47	3.80	1.12	Bal
		3	1.12	0.23	5.90	15.74	13.05	1.48	0.00	Bal
	after HT	1	20.15	0.25	0.18	0.78	0.51	5.17	0.02	Bal
		2	10.25	0.65	0.10	0.28	0.59	7.67	1.22	Bal
		3	2.28	2.41	7.34	10.02	8.10	1.07	0.09	Bal
AlCu9Si4Ni1	as cast	1	48.23	0.35	1.23	0.56	0.71	0.12	0.01	Bal
		2	45.26	0.75	2.18	0.16	0.41	7.09	1.26	Bal
		3	1.45	0.15	10.45	12.23	12.05	2.45	0.00	Bal
	after HT	1	19.13	0.15	3.34	0.79	1.12	13.75	0.09	Bal
		2	17.29	0.15	4.65	0.90	1.18	15.69	1.14	Bal
		3	1.98	0.05	8.98	18.11	14.78	2.89	0.02	Bal

The Al_7Cu_4Ni phase with lower mass fraction [%] of Cu was particularly found in the alloy after homogenization. It was assumed that Al_7Cu_4Ni phase would be chanched to Al_3Ni_2+Cu (FCC) after heat treatment (see table 2, comparison spectrum 2 for each alloy depended on heat treatment). But this treatment was not sufficient to redissolve this phase completely due to the high melting point of ternary phase Al_7Cu_4Ni (about 535°C).

Mechanical Properties

Table 3 shows the variation in the comparison of mechanical properties of different alloys as a function of heat treatment. Slight differences were found in the mechanical properties of these alloys in as-cast state and after heat treatment. The yield strength and ultimate tensile strength of three alloys decreases slightly at 240°C independent of sample state. The values of both ultimate tensile strength and yield strength at room temperature and elevated temperature (240°C) obtained for alloys AlCu9Si4Ni1 are higher in as cast or after heat treatment compared with alloy AlCu4Mg1 and AlCu9Mg1. This is due to the high Cu+Ni content in alloy AlCu9Si4Ni1 and increasing surface fraction of the hard enable phase Al2CuMg and Al_7Cu_4Ni (Mondolofo, 1976) in the Al-matrix specially in heat treated sample consequently. Normally the precipitation of Cu and Ni based intermetallic compounds block the movement of dislocation in the FCC Al-Matrix during mechanical testing, so that at the interface of the intermetallic compounds dislocations accumulate which leads to high yield strength and ultimate tensile strength during mechanical testing (Ogris, 2002). In the alloy 3 smaller elongation was indicated in as-cast and after heat treatment comparison to the alloys 2 and 3. This result can be attributed to the high surface fraction of AlSi-, Fe- and Mn-containing particles, which play an important role in decreasing the ductility in these alloys.

Table 3. Mechanical properties of the examined alloys at different test temperature, depended on samples condition (as cast, after heat treatment), averaged from five test samples

Alloy		As cast			510°C/10 h+185°C/12 h		
		$R_{p0,2}$ [MPa]	R_m [MPa]	A [%]	$R_{p0,2}$ [MPa]	R_m [MPa]	A [%]
AlCu4Mg1	25°C	160	199	1.20	209	262	1.5
	240°C	120	165	3.08	153	190	2.70
AlCu9Mg1	25°C	146	186	1.30	179	192	0.32
	240°C	141	162	0.5	152	182	1.10
AlCu9Si4Ni1	25°C	210	216	0.3	236	239	0.23
	240°C	168	170	0.31	185	204	0.60

Figure 4 (a, b) shows the behaviour of the DSC tareces for three alloys in as a cast and after heat treatment, heated with 10 K/min. The curves in Figure 4 (a) consist of three endothermic reactions referred to as I, II and IV and one exothermic reaction III. The endothermic peak I and II, could be contributed to the dissolution of Guinier-Preston zones (GP zones) and intermediate semi coherent θ´ phase. The exothermic process III could be ascribed to the formation of incoherent θ phase. The peak IV could be contributed to the dissolution of any phases. An alloy AlCu9Si4Ni1 shows especially a high volume of peak III. This is considered to the high amount of Cu, Ni and Si in this alloying system in contrast to alloy AlCu4Mg1 and AlCu9Si4Ni1 (Badini, 1995; Styles, 2012; Wang, 2007).

DSC Thermogram described in Figure 4 (b) related to all alloys heat treated by 510°C/10 h +185°C/12 h consist of three endothermic peaks (I, II and IV) and one exothermic peak III. The peak I and II which is not clear after heat treatment would be ascribed to formation of remaindered Guinier-Preston zones (GP zones) and intermediate semi coherent θ´ and Si-containing phase. The peak III related to formation of incoherent θ phase. Generally, the heat treatment causes the suppression in formation and dissolution of any process during the heating in contrast to the curves in as a cast state. The peak IV, especially for alloy AlCu9Si4Ni1 is displaced at increasingly high temperatures and required more heating to dissolution of hardening phases in the Al-matrix. In this case the hardening phases are still present in the matrix at high temperature, results in higher yield strength after heat treatment (Badini, 1995; Styles, 2012; Wang, 2007).

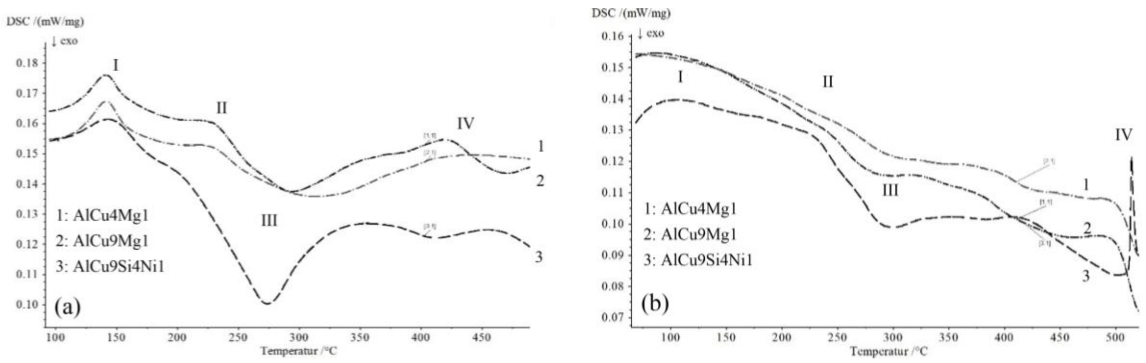

Figure 4. DSC curves of the samples heated with 10 K/min, a) alloys AlCu4Mg1, AlCu9Mg1 and AlCu9Si4Ni1as a cast b) alloys AlCu4Mg1, AlCu9Mg1 and AlCu9Si4Ni1 after heat treatment 510°C/10 h +185°C/12 h

CONCLUSIONS

The effect of alloying element and heat treatment on the mechanical properties of AlCuSi alloys were investigated. Conclusions drawn from this work are as follows.

- The heat treatment (510°C/10 h + 185°C/12 h) plays an important role in showing the effect of Cu+Ni on the mechanical properties of AlCuSi alloy due to formation of Cu containing phases in FCC Al-Matrix. Homogenization yielded the highest yield and ultimate tensile strengths of sample compared to the unhomogenized sample.
- Increasing the Cu content increases precipitation hardening and the elevated temperature strength but decreases the elongation.
- Si addition decreases ductility of AlCuSi casting alloys independent of any heat treatment.
- A higher yield strength at 240°C was found in alloys with more Cu content in comparison to the alloy with low Cu content of both the as-cast and heat treated condition due to the precipitation hardening effect of Cu in the FCC-Al-matrix especially under heat treatment condition.
- The optimization of heat treatment parameters is an important task to improve the mechanical properties of AlCuSi alloys as a function on alloying element.

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