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Effect of Cu in Al-Si Alloys with Phase Modelling

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Abstract

A comparison study on copper content and effect of homogenisation treatment was performed between two Al-Si alloys containing varying amount of copper. Observation of microstructure revealed the formation of several intermetallic phases due to the presence of copper. Image analysis on the acquired images confirmed the relative changes in percent phase fractions due to larger amount of copper since the total amount of intermetallic phases was increased. Also, higher copper content showed a positive response on measured hardness values. Homogenisation at 300^{o}C caused increased hardness up to 2 hour for age-hardening, which was decreased significantly after 4 hour of heating due to over-ageing. Solidification calculations by thermodynamic modelling approach showed that Al_2Cu and Al_3Ni_2 phases were predicted to be most stable at the homogenisation temperature and these two phases were responsible for the increased in hardness due to copper addition.

Keywords: *CALPHAD, Phase Modelling, Aluminium Alloys.*

1. Introduction

Aluminium is the most abundant metal in the earth's crust. It makes up about 8% by weight of the earth's solid surface [1]. Aluminium alloys with a wide range of properties are used in engineering structures. These properties can be largely modified by changing the alloy composition. Silicon, copper, zinc and magnesium are the most commonly used alloying elements in aluminium, which have sufficient solid solubility.

The effect of copper addition on the structural features and mechanical properties of Al-Si-Cu alloy have shown that copper addition increase the strength of these alloys [2-6]. Yield stress, hardness and micro-hardness increase with increase in copper content regardless of alloy composition [7]. Copper also improves machinabilitiy; however, castability, ductility and corrosion resistance are all decreased. The higher silicon alloys (e.g. Al–10Si–2Cu) are used for pressure die castings, whereas alloys with lower silicon and higher copper (e.g. Al–3Si–4Cu) are used for sand and permanent mould castings. In general, the Al–Si–Cu alloys are used for many of the applications of Al-Si alloys but where higher strength is needed. One example is the use of alloy 319 (Al–6Si–3.5Cu) for die cast (permanent mould) automotive engine blocks and cylinder heads in place of cast iron [8].

The as-cast microstructure of cast aluminium alloys normally displays significant segregation and super-saturation [8]. For this, homogenisation of aluminium alloys is often done to improve workability and mechanical properties by dissolving the non-equilibrium, brittle and inter-dendritic constituents, and by providing a more homogeneous structure.

Thermodynamic models have been developed for the calculation of various thermo-physical and physical properties with the aim of providing thermo-physical and physical properties for various types of multi-component alloys during solidification. Thermodynamic phase modelling approach is based on CALPHAD (CALculation of PHAse Diagrams) method [9], which is used for calculation of a wide range of materials properties for alloys and is particularly aimed at multi-component alloys. Using phase analysis modelling approach and microstructure analysis, it is possible to identify the phases present in the microstructure of an alloy, which has direct influences on the mechanical properties.

In this work, the effects of copper content and homogenisation treatment on microstructure and hardness values were studied using modelling, microstructural study and image analysis method.

2. Experimental

A master alloy was prepared by melting locally available aluminium pistons used for automobiles. Afterwards, copper wire was added at some predefined weight fractions into the master alloy to alter alloy composition. The raw materials were melt into a gas fired crucible pit furnace and were cast in a sand mould in the shape of a long rectangular bar. For changing the alloy composition, second casting was done in a permanent metal mould of circular cross-section (Length 170 mm, Diameter 17 mm). For both alloys, the melt was heated up to 850^{o}C and poured at 800^{o}C. Compositions of both alloys were measured by Optical Emission Spectrometer (Shimadzu PDA 700) and verified by wet chemical analysis methods. The average compositions found from OES analysis are shown in Table 1.

Table 1. Compositions of alloys used in experiment (in wt%)

Alloy	Al	Si	Cu	Mg	Fe	Ni	Mn
Master	86.09	7.99	**3.5**	0.76	0.70	0.51	0.34
Second	81.69	7.80	**8.46**	0.58	0.60	0.48	0.29

Homogenisation of the second alloy was done in a BlueM Electric furnace at 300^{o}C for 1, 2 and 4 hours. After homogenisation, the samples were quenched in water to retain the microstructure.
The hardness of the alloys at several steps was measured by Universal Testing Machine (Brinell hardness) and Rockwell Hardness Tester (in F scale). Then these values were converted into Rockwell B scale.
Microstructures of all samples were studied in unetched condition. All images were taken in Optica B-600 MET trinocular upright metallurgical microscope, using OpticaTM Vision Pro software. Images were taken at 500X magnification under the same RGB values and pixel resolutions for comparing. Image of graticule scale (1 div = 0.01 mm) was also taken at the same magnification to quantify the dimensions of microstructure.
Image analysis was accomplished using ImageJ version 1.43u to find out the percentages of different phases. At least 10 images of microstructures for each alloy were taken and analysed to find out the average values of the amounts of phase areas/fractions. Modelling of the alloys was done using thermodynamic modelling approach to predict the relationships among composition, temperature, phases and properties.

3. Results and Discussion

Microstructure observation

Fig. 1 shows the microstructure of as-cast master alloy and second alloy. In both images, presence of silicon and several intermetallic compounds in aluminium matrix are identified. Some of these phases were present as particles in the microstructure. Phases were identified based on previous works for these alloys [10-12].
The microstructure of the second alloy is somewhat different from the master alloy due to presence of more copper (Fig. 1(b)). Inter-particle spacing of the phases were small in this higher intermetallics containing alloy.

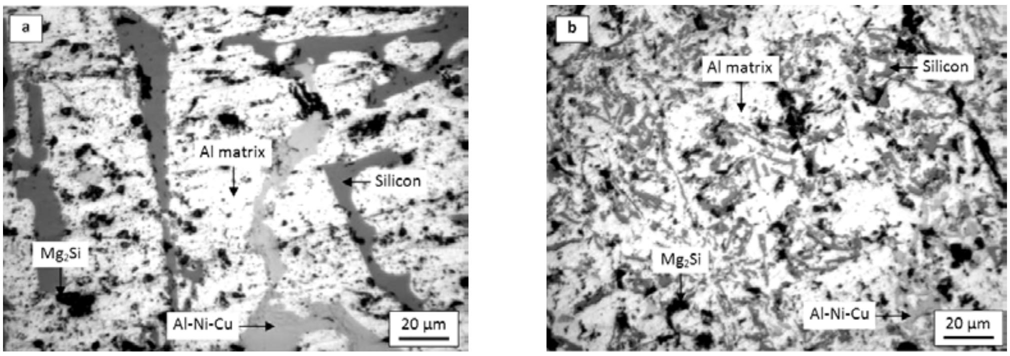

Fig 1. Microstructure of as-cast a) master alloy & b) second alloy

Fig. 2 shows the microstructure of the second alloy after homogenisation at 300^{o}C. After homogenisation of 1 hour, the fine particles of silicon and intermetallics are distributed around the dendritic pure aluminium matrix. With increased homogenisation time, the dendrites start to disappear and the particles grow in size due to higher atomic movement for longer time due to diffusion. After 4 hours of treatment, dendrites are removed completely, whereas the particles have grown all throughout the matrix.

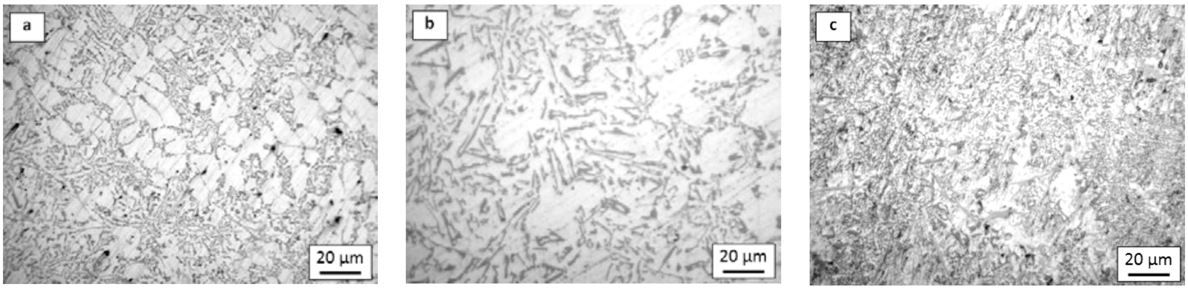

Fig 2. Microstructure of second alloy after heating at 300^{o}C for a) 1 hour, b) 2 hour & c) 4 hour

Image analysis

Image analysis results are shown in Fig. 3. From the achieved data, it can be seen that the amount of intermetallic compounds have increased significantly in the second alloy attributed to copper addition. The fraction of silicon did not increase in an appreciable amount since silicon does not form any primary phase with copper.

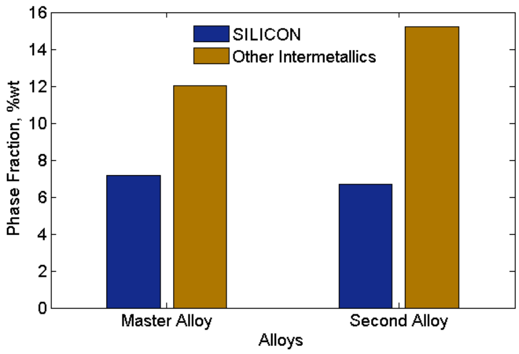

Fig. 3. ImageJ analysis result

Thermodynamic modeling

From the Step Temperature calculation predicted in thermodynamic modelling, the evolution and abolition of different phases in the master alloy are clearly observed in Fig. 4. It is evident that an Al-Ni phase (Al_3Ni_2) evolves at about 520^{o}C, grows up to 2 wt% and then totally abolishes at 400^{o}C. While this phase decreases in amount, another Al-Cu-Ni phase (Al_7Cu_4Ni) evolves at 490^{o}C and grows up to 4.40 wt% and then remains stable with lowering temperature. Therefore, those aluminium and nickel content in Al-Ni phase are transferred to the new Al-Cu-Ni phase.

Also, the alpha solid solution increases to 3.80 wt% after nucleating at 620^{o}C, and reduces to 2.50 wt% with lowering temperature. At the same period, an Al-Cu-Mg-Si phase ($Al_5Cu_2Mg_8Si_6$) continues to grow from 1.50 to 2.50 wt%.

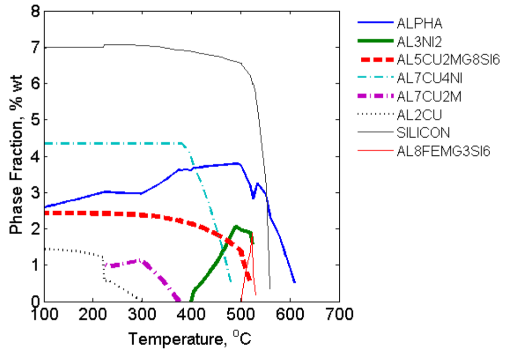

Fig. 4. Changes in intermetallic phases in master alloy

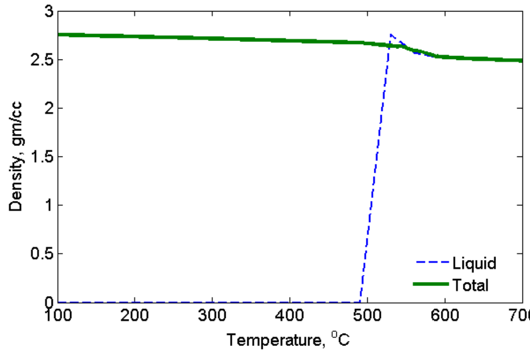

Fig. 5. Condition of mushy zone in master alloy

Using thermodynamics approach, solidification simulation in Scheil-Gulliver condition is also possible. Such modelling results are shown in Fig. 5 for density of the liquid and total system. The solidification of the master alloy starts at 620°C and becomes completely solid at 490°C.Aluminium alloys typically show considerable shrinkage and dendrite formation. The density of the liquid phase increases up to about 2.75 g/cm^3 due its enrichment with Cu and other elements.

From modelling of the second alloy, the formation of same phases as in the first alloy in step temperature calculation predicted is shown in Fig. 6. However, the amount of Al-Cu phases increased considerably due to increased copper content, compared to the master alloy. The transformation of Al-Ni phase to Al-Cu-Ni phase is seen for a very brief period here (between 520-515°C). In addition, the Al-Fe-Mg-Si phase does not form as occurred in master alloy.

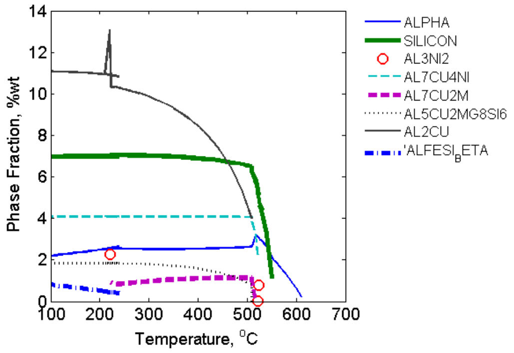

Fig. 6. Changes in intermetallic phases in second alloy

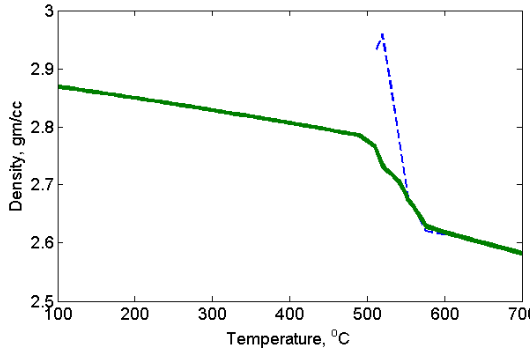

Fig. 7. Condition of mushy zone in second alloy

From solidification Scheil-Gulliver modelling, it can be seen that the alloy starts to solidify at 615°C and solidifies completely at 490°C. The density change observation in Fig. 7 shows that the liquid density increased like before with lowering temperature, but to a greater value of 2.95 g/cm^3, compared to 2.75 g/cm^3 for master alloy. The total density is also increased, which is an influence of copper content. From modelling, it was ascertained that copper content in liquid increases to 24 wt% in second alloy, compared to only 14 wt% for master alloy. This explains that the liquid enrichment is mostly caused by copper content.

Effects of phases in hardness

The hardness of the master alloy was found to be only 27.62 HRB, which increased significantly to 45.2 HRB by an increase in Cu content from 3.5 to 8.46 wt% as shown in Fig. 8.

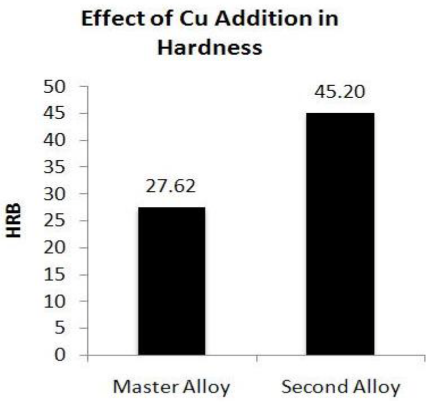

Fig. 8. Effect of cu addition in hardness

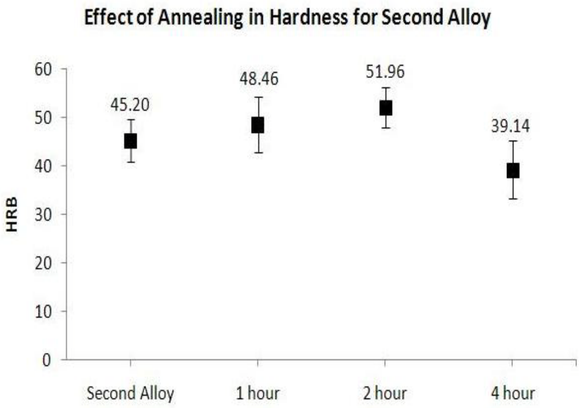

Fig. 9. Effect of homogenisation in hardness

Homogenisation of the second alloy developed gradual increase in hardness after heating at 300°C for 1 and 2 hours due to precipitation-hardening of the alloy. Nonetheless, it decreased considerably after heating for 4 hours as can be seen in Fig. 9. This is attributed to over-ageing.

The change in hardness by homogenisation is related to the precipitation of particular phases, which is confirmed by thermodynamic modelling. From single temperature calculation at homogenisation temperature (300°C) and room temperature (25°C), as shown in Fig. 10, it is evident that Al_3Ni_2 phase is stable at 300°C. Moreover, amount of Al_2Cu phase increased at 300°C. Since this microstructure is retained by quenching followed by homogenisation treatment, these two phases are apparently responsible for such increment of hardness of the second alloy.

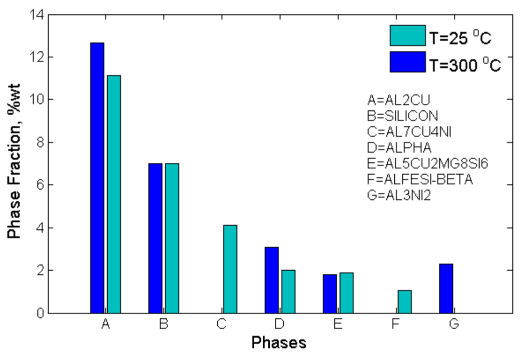

Fig. 10. Phases of second alloy at 300°C

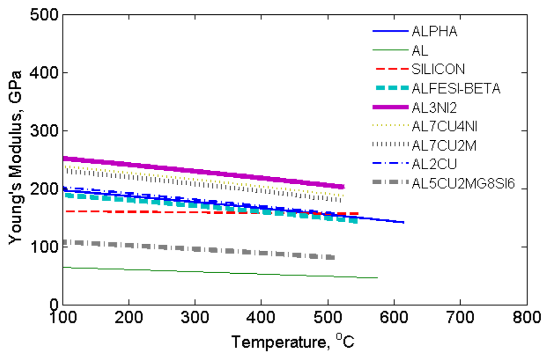

Fig. 11. Young's modulus of phases

Furthermore, up to 1 and 2 hours of homogenisation, Al_3Ni_2 and Al_2Cu phase continues to precipitate out of the matrix and results in precipitation-hardening phenomenon due to their fine size and coherency in the matrix, which provides resistance to dislocation motion. However, longer exposure to heat allows more diffusion to occur and particle coarsening of the phases reduces the Zener pinning effect of the particles and release of the dislocations which is considered as the reason for reduction of hardness of the second alloy.

This is further confirmed from property modelling of the phases of the second alloy (Fig. 11). The Al_3Ni_2 phase is predicted to provide maximum young's modulus amongst the phases. Therefore, presence of this phase is considered to be beneficial for strengthening apart from Al_2Cu phase alone.

4. Conclusion

a. The presence of copper lowers the solidification temperature significantly. Addition of copper increases the range of mushy zone, enriches the liquid and causes segregation and dendrite formation. Increasing copper causes higher hardness values. The microstructural study revealed the formation of several intermetallic phases. Modelling of these alloys confirmed the increased amount of intermetallic phases due to copper addition.
b. Homogenisation of the second alloy caused the removal of dendrites, and gradual increase in size of the particles and their growth throughout the matrix. The hardness values increased by precipitation-hardening process up to 2 hour homogenisation at 300^oC and lowered considerably after 4 hour homogenisation. With increase of homogenisation time, diffusion accelerates phase coarsening, which affects dislocation distribution adversely and results in lower hardness values observed.
c. Thermodynamic modelling shows that increase in copper content promotes formation of Al_2Cu and Al_3Ni_2 phases at 300^oC which apparently contributes to observed higher hardness which was confirmed by the higher Young's modulus of Al_3Ni_2 phase.

5. Acknowledgement

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6. References

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