Effect of Solution Treatment Temperature on The Microstructure and Mechanical Properties of Al-5.1Zn-1.9Mg Alloy Produced by Squeeze Casting

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Abstract. Aluminum alloys are widely used in aviation industries, especially for the body and wings of aircraft. The heat treatment for 7xxx series aluminum alloys is precipitation hardening which consists of solution treatment, quenching, and ageing. One key for successful ageing process is the amount of solute elements and vacancies dissolve in the matrix during solution treatment. Therefore, this research is aimed to study the effects of solution treatment temperature on the hardness and microstructure of Al-5.1Zn-1.9Mg (wt. %) alloy which produced by squeeze casting. Solution treatment temperatures were varied to 220, 420, and 490 °C for 60 minutes, followed by quenching. The samples were then stored at -10 °C to prevent natural ageing. Characterization included hardness testing and microstructural observation by using OM and SEM-EDS, XRD, and STA. Ageing was conducted at 130 °C for 48 h followed by hardness testing and microstructural observation. The results showed that increasing solution treatment temperature induced enhancement of second phase dissolution and the amount of trapped vacancies in the matrix. However dissolution of second phase was hardly detected at solution treatment temperature of 220 °C. It was shown by the volume fraction of the second phase found after homogenizing was 7.13 % and decreased to 7.06, 4.80, and 4.19 % after solution treatment at temperatures 220, 420, and 490 °C respectively. Therefore the increase in hardness after ageing at 130 °C for 48 hours was 11.54, 42.1, and 66.7 HRB for solution treatment temperatures of 220, 420, and 490 °C respectively. This can be concluded that increasing solution treatment temperature enhanced the hardness of alloy after ageing.

Keywords: Al-Zn-Mg, Solution Treatment, Precipitation Hardening, Fraction Volume

1. Introduction

Aluminum 7XXX series with zinc and magnesium as alloying element are commonly used in aviation industries due to a very high strength to weight ratio and their mechanical properties can be enhanced by heat treatment. Maximum solubility of zinc in aluminum is 83.1 wt. %, which will strengthen the alloy through solid solution [1]. Sofyan, et.al. [2] found that the increase in zinc also refines the dendrite structure in Al-xZn-6Mg composites. Magnesium addition in the Al-Zn alloy enhances formation of MgZn2 semi coherent precipitate and increases hardness [3]. Addition of other alloying elements, such as Cu, may even increase the hardness. However, when Zn and Mg content of the alloy is already high, addition of Cu will not give significant increase to the hardness [4]. The mechanical properties of these alloys increase by formation of precipitates which evenly disperse in the matrix through precipitation hardening and hinder the movement of dislocations [5].
The stages of precipitation hardening are solution treatment, quenching, and ageing. During solution treatment, matrix will be richer in alloying elements coming from dissolution of second phase particles [6]. The main purpose of solution treatment is to dissolve most of second phase particles, so that maximum precipitation hardening would be obtained during ageing. The solid solubility of atom increases with the rise of temperature, but at the same time, high temperature will create more vacancies that will be trapped during quenching. Therefore, solution treatment temperature should be carefully determined to obtain optimum hardness. Based on Figure 1, at the composition of Al-5.1Zn-1.9Mg (wt. %), the Mg5Al8 and Mg3Zn3Al2 second phase begin to dissolve at 277 °C (550 K). Therefore, conducting solution treatment at above 277 °C is expected to dissolve more solutes to the α-matrix. It is also supported by Li, et.al. [7], in Al-5.48Zn-2.02Mg (wt. %) alloy with solution treatment at 460 °C for 1 h, second phase already dissolved and the solubility increases with rise of temperature.

![Figure 1. Liquidus projection of the ternary diagram of Al-Zn-Mg [8]. Red line shows the composition of Al-5.1Zn-1.9Mg alloy.](image)

The number of vacancies at a certain temperature can be calculated by using equation (1) and (2) [5].

\[ N_D = N \times \exp\left(-\frac{Q_D}{kT}\right) \]  

\[ N = \rho \times N_A/A_r \]

Where \( Q_D \) is the activation energy, \( k \) is the Boltzmann constant (8.62 x 10⁻⁵ eV /atom K), \( T \) is temperature, \( \rho \) is density, \( N_A \) is the Avogadro number (6.02 x 10²³ atom/mol), and \( A_r \) is the relative atom. The calculation shows that the number of vacancies at 220, 420, and 490 °C is 0.13 x 10¹⁶, 21.23 x 10¹⁶, 67.17 x 10¹⁶ respectively. The exponential increase at 490 °C is potential to be harmful to the mechanical properties of the alloy.

Studies of precipitation hardening have been conducted by many researchers. Among all the parameters, the temperature and time of ageing process have been massively studied. However, few research investigated the effect of solution treatment temperature. Therefore, this paper investigated the effects of solution treatment temperatures of 220, 420, and 490 °C on the microstructures and hardness of Al-5.1Zn-1.9Mg (wt. %) alloy. It was intended to learn how solution treatment temperature affected the dissolution of second phases into the matrix and the amount of vacancy in aluminum alloy, that then influenced the precipitation processes.
2. Experimental Method
The alloy was made by squeeze casting process with pure Al, Zn and Mg ingots as starting materials. The ingots were melted in an electrical furnace at 850 °C and degassed by argon as well as stirred for 30 second for proper mixing. The metal mould with dimension of 17x17x1.5 mm³ was preheated to 300 °C, then the molten alloy was poured into mould. Semi solid alloy was then squeezed at 76 MPa for 10 min. The chemical composition of the as-cast alloy was shown in Table 1. The as-cast alloy was then homogenized at 400 °C for 4 h in a muffle furnace followed by air cooling. The samples were solution treated at 220, 420, 490 °C for 1 h, followed by water quenching and ageing at 130 °C for 48 h.

Table 1. The chemical composition of the Al-5.1Zn-1.9Mg (wt. %) alloy.

|    | Zn   | Mg   | Fe  | Cr  | Si  | Ni  | Al  |
|----|------|------|-----|-----|-----|-----|-----|
|    | 5.140| 1.938| 0.267| 0.048| 0.044| 0.027| Bal |

Microstructures were observed by using optical microscope and Scanning Electron Microscope (SEM). The samples were polished and etched by Keller’s reagent (2.5ml HNO₃ + 1 ml HCl + 1.5 ml HF + 95 ml distilled water). The as-homogenized and as-quenched microstructures were further quantitatively analyzed by using MagniSci. Brinell hardness test was performed according to ASTM E10 and five indentations were made for each measurement. In addition, STA test was conducted on STA 6000 Parkin Elmer at as-quenched samples to examine phase transformation, with scanning rate of 10 °C/min.

3. Results And Discussion

3.1 Microstructure and Hardness Analysis
Microstructures of Al-5.1Zn-1.9Mg alloy in various conditions are shown in Figure 2. All of them possess dendritic shape and black interdendritic phases. The black interdendritic phases consist of T (Mg₃Zn₃Al₂) and β (Mg₅Al₈) in accordance to ternary phase diagram in Figure 1. The as-cast microstructure, (Figure 2.(a)), has wide interdendritic area and measured to be 10.58 %, see Figure 3.

Figure 2. Microstructures of Al-5.1Zn-1.9 Mg: (a) as-cast, (b) as- homogenized, and solution treated at: (c) 220, (d) 420, (e) 490 °C.
Homogenization seemed to dissolve some of the interdendritic phase, that reduced the volume to 7.13 % (Figure 2(b)). This is in line with the purpose of homogenization to reduce the dendritic structure and segregation of alloying elements, as well as to uniform grain size [9]. It also led to a decrease in hardness from 53.5 HRB in as-cast to 47.25 HRB in as-homogenized (Figure 3).

Microstructure of as-quenched sample after solution treatment at 220 °C shows no significant different with that of as-homogenized condition. The volume fraction of the dendritic phase is 7.06 % (Figure 3). The hardness also shows the same number with as-homogenized, that is 42.96 HRB. This indicates that at solution treatment temperature of 220 °C, the second phase dissolution has not occurred as predicted by the liquidus projection of the ternary diagram of Al-Zn-Mg alloy in Figure 1.

Meanwhile, solution treatment at 420 °C rounded the dendritic structure and significantly decreased the interdendritic phase to 4.80 %, (Figure 2.(d)). It is also supported by the hardness number, which is out of range with HRB unit, so that it was tested using Rockwell E units and showed the value of 38.5 HRE. The low hardness maybe caused by the trapping of Zn and Mg atoms in the α-Al matrix that produced unstable state [5]. This indicates that at 420 °C the interdendritic phase already dissolves into the matrix. Further increase of solution treatment temperature to 490 °C reduced the second phase fraction to 4.19 % and hardness to 34.4 HRE (Figure 3) as well as the trapping of vacancies within the matrix [5]. More detailed observation by using SEM is provided in Figure 4. Reduction of second phase at higher solution treatment temperature is confirmed in this figure. This supports the notion that higher solution treatment temperature leads to enhanced dissolution of the second phase [6].

![Figure 3](image1.png)  
**Figure 3.** The effect of treatment on the percentage of interdendritic volumes and the hardness of Al-5.1Zn-1.9Mg alloy.

![Figure 4](image2.png)  
**Figure 4.** SEM images of Al-51Zn-1.9Mg alloy after solution treatment at temperature of : (a) 220, (b) 420, and (c) 490 °C followed by water quenching.
Figure 5 provides the microstructures of Al-5.1Zn-1.9Mg alloy after solution treatment at varied temperature followed by 130 °C for 48 h. Figure 5.(a) shows the ageing microstructure with solution treatment at 220 °C. The second phase, which is black, did not dissolve and still visible, so that supposed to be not many precipitates formed [8]. It is indicated by a slightly increase of hardness from 42.96 HRB in as-quenched to 54.5 HRB after 48 h ageing (Figure 6).

![Figure 5](image)

**Figure 5.** Microstructure of Al-51Zn-1.9Mg (wt. %) after ageing at 130 °C for 48 h: (a) solution treatment 220 °C, (b) solution treatment 420 °C, (c) solution treatment 490 °C.

![Figure 6](image)

**Figure 6.** The effect of solution treatment to the hardness of Al-5.1Zn-1.9Mg (wt. %) alloy after ageing at 130 °C.

The ageing microstructure with solution treatment at 420 °C is shown in Figure 4(b). Precipitates may have formed and characterized by significant increase in hardness to 42.1 HRB. However, the precipitates are not visible by optical microscopy because they are very small and evenly dispersed. Interestingly, samples that solution treated at 490 °C, achieved the highest hardness after ageing, compared to other samples. It is in contrast with their lowest as-quenched hardness. The solution treatment at 490 °C reduced the interdendritic volume to 4.19 % as well created 67.17x10^{16} vacancies. Both the solute atom and vacancies promote formation of precipitates that contribute to the highest hardness after ageing [4].

3.2 Simultaneous Thermal Analysis
Figure 7 illustrates the STA results on Al-5.1Zn-1.9Mg alloy after solution treatment at 220, 420, 490 °C, followed by quenching. The summary of temperatures of phase formation and dissolution is provided in Table 2 and Table 3. Figure 7.(a) shows that there is one endothermic peak at 286.6 °C, indicating the
dissolution \( \beta (\text{Mg}_5\text{Al}_8) \) and \( T (\text{Mg}_3\text{Zn}_3\text{Al}_2) \) as the only reaction and no precipitates formed. While at 420 and 490 °C (Figure 7.(b) and (c)), three exothermic and four endothermic peaks were detected, in connection with the formation of GP zone, \( \eta' \), \( \eta \) precipitates and the dissolution of GP zone, \( \eta' \), \( \eta \), as well as the second phase. In general, the peaks of 490 °C solution treated samples are slightly lower than those of 420 °C. This is because higher temperature of solution treatment created more vacancies, so that precipitation is easier to occur.

**Figure 7.** Heat flow and derivatives to temperature of Al-5.1Zn-1.9Mg (wt. %) alloy with solution treatment at: (a) 220, (b) 420, (c) 490 °C.

| Solution Treatment | GP zone Formation Temperature (°C) | \( \eta' \) | \( \eta \) |
|-------------------|-----------------------------------|---------|---------|
| 220 °C            | None                              |         |         |
| 420 °C            | 50.14                             | 153.98  | 273.90  |
| 490 °C            | 50.02                             | 120.02  | 253.52  |

| Solution Treatment | GP zone Dissolution Temperature (°C) | \( \eta' \) | \( \beta \& T \) Second Phases | \( \eta \) |
|-------------------|-------------------------------------|---------|---------------------|---------|
| 220 °C            | None                                | None    | 286.6               | None    |
| 420 °C            | 131.04                              | 230.11  | 291.74              | 340.10  |
| 490 °C            | 110.50                              | 230.46  | 287.84              | 341.11  |
According to Krishna et al.[10], in Al-4Zn-2Mg (% wt.) alloys, the phase forming intervals indicated by exothermic reaction are GP zone at temperature of 20-120 °C, η’ at 120-250 °C, and η at 150-300 °C. While the dissolution interval of each phase through endothermic reaction are GP zone at temperature of 50-150 °C, η’ at 200-250 °C, and η at 300-350 °C. All exothermic and endothermic peaks in Figure 7.(b) and (c) are in the same temperature intervals. Therefore, it suggests that the precipitation occurs in the studied alloy is GP zones → η’ → η (MgZn₂) [11].

4. Conclusion
The increase in solution treatment temperature on Al-5.1Zn-1.9Mg (wt. %) enhanced the dissolution of second phase β (Mg₅Al₈) and T (Mg₃Zn₁Al₂) and decreased the as-quenched hardness. It is shown by interdendritic phase volumes in the as-cast, as-homogenized, solution treated at 220, 420, 490 °C were 10.58, 7.13, 7.06, 4.80, and 4.19 %, respectively. While the hardnesses were 53.5 HRB, 47.25 HRB, 42.96 HRB, 38.5 HRE, 34.4 HRE, respectively. The solution treatment at 490 °C led to the highest hardness after ageing due to the highest dissolved solute atoms and the highest amount of vacancies trapped in the matrix that promoted formation of precipitates. Therefore vacancies in precipitation hardening has an important role as a place for precipitate to grow. The phase transformation occurred in the Al-5.1Zn-1.9Mg alloy during ageing was GP zone → η’ → η (MgZn₂).

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