

# FRACTIONAL RECRYSTALLIZATION KINETICS IN DIRECTLY COLD ROLLED Al-Mg, Al-Mg-Sc AND Al-Mg-Sc-Zr ALLOY

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**Abstract:** The evaluation of texture as a function of recrystallization has been characterized for directly cold rolled Al-6Mg, Al-6Mg-0.4Sc and Al-6Mg-0.4Sc-0.2Zr alloys. Samples were annealed isothermally at 400 °C for 1 to 240 minutes to allow recrystallization. Recrystallization kinetics of the alloys is analyzed from the micro-hardness variation. Isothermally annealed samples of aluminum alloys were also studied using JMAK type analysis to see if there exists any correlation between the methods. Recrystallization fraction behavior between two methods the scandium added alloys show the higher variation due to precipitation hardening and higher recrystallization behavior. The scandium and zirconium as a combined shows the more variation due to formation of  $Al_3(Sc, Zr)$  precipitate. From the microstructure it is also observed that the base Al-Mg alloy attained almost fully re-crystallized state after annealing at 400 °C for 60 minutes.

**Keywords:** Al-Mg alloys, annealing, fraction recrystallization, precipitate

## 1. INTRODUCTION

The 5xxx series Al-mg alloy are by the most popular amongst the aluminium based alloys and they possess the greatest technological importance. This alloy exhibits good recrystallization behavior and can find that the equality grain is formed [1]. After cold rolling the grains are elongated and non equaled in size. During heat treatment in different range of temperature the grain size gradually convert elongated to equiaxed. The use of scandium as an alloying element in aluminium has gained an increasing interest. The three principle effects that can be obtained by adding scandium to aluminium alloys are (i) grain refinement during casting, (ii) precipitation hardening from  $Al_3Sc$  particles and (iii) grain structure control from  $Al_3Sc$  dispersoids [2-5]. Addition of minor Sc and Zr to the Al-Mg alloys can effectively improve the recrystallization temperature and strength of the alloys due to the formation of  $L1_2$  structured  $Al_3(Sc, Zr)$  precipitates [6,7].

In the present study, recrystallization kinetics in directly cold rolled Al-Mg Alloy with scandium is studied via the methods of micro-hardness variation. Isothermal recrystallization kinetics can be represented by Johnson-Mehl-

Avrami-Kolmogorov (JMAK) type behavior. The recrystallization kinetics for the alloys in the present study is analyzed by assuming a JMAK type behavior for kinetics obtained from micro-hardness and obtaining parameters for recrystallization process.

## 2. EXPERIMENTAL

Melting was carried out in a resistance heating pot furnace under the suitable flux cover (degasser, borax etc.). Several heats were taken for developing base Aluminium-Magnesium alloy, Aluminium-Magnesium alloy containing scandium and with or without zirconium. In the process of preparation of the alloys the commercially pure aluminium (99.5% purity) was taken as the starting material. First the aluminium and aluminium-scandium master alloy (98 wt% Al + 2 wt% Sc) were melted in a clay-graphite crucible, then magnesium ribbon (99.7% purity) was added by dipping in to the molten metal. Zirconium were taken in the form of powder (99.98% purity) with in a cover of aluminium foil and were then added by plunging. The final temperature of the melt was always maintained at  $780 \pm 15^\circ C$  with the help of the electronic controller. Then the melt was allowed

to be homogenised under stirring at 700 °C. Casting was done in cast iron metal moulds preheated to 200 °C. Mould sizes was 12.5 x 51.0 x 200.0 in millimeter. All the alloys were analysed by wet chemical and spectrochemical methods simultaneously. The chemical compositions of the alloys are given in Table 1. Cold rolling of the cast alloys were carried out with a laboratory scale rolling mill of 10HP capacity at 75% reduction. The alloys were pieces into 10 x 12 x 50 mm and the deformation given was about 1.25 mm per pass. Samples for the studying recrystallization kinetics, 2.5 x 12 x 15 mm in size were obtained from the cold rolled sheet. The samples were isothermally annealed at 400°C for different times ranging from 1 to 240 minutes. Microhardness of the samples was measured with a Polyvermet microscope-cum microhardness tester, Reichert Jung Microduramet 4000 E. A Knoop indenter was pressed on to the sample using a cycle time of 15 seconds and loading rate of 10 gf/sec. The indentation tests were performed with 10gm load. Average results of fifteen tests are plotted. The optical metallography of the samples was carried out in the usual way. The specimens were polished finally with alumina and the etchant

**Table 1.** Chemical Composition of the Experimental alloys (wt%)

| Alloy | Mg   | Sc    | Zr    | Fe    | Si    | Al  |
|-------|------|-------|-------|-------|-------|-----|
| 1     | 6.10 | 0.000 | 0.000 | 0.382 | 0.380 | Bal |
| 2     | 5.97 | 0.400 | 0.000 | 0.314 | 0.335 | Bal |
| 3     | 5.85 | 0.400 | 0.185 | 0.335 | 0.345 | Bal |

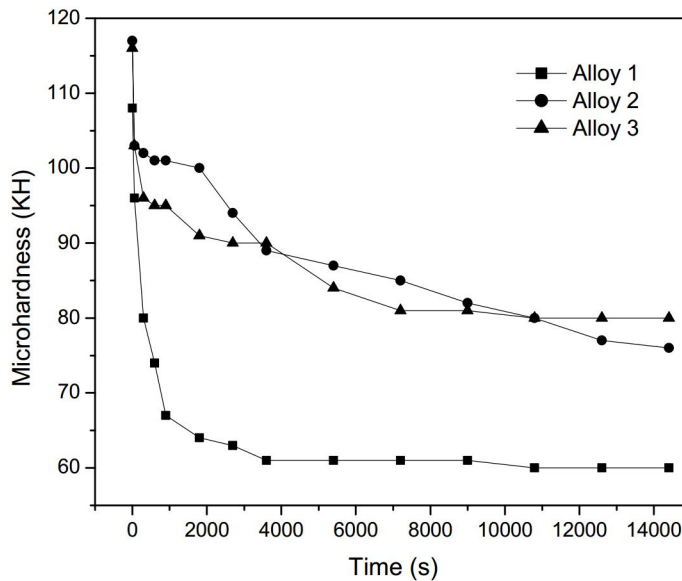
used was Keller’s reagent (HNO<sub>3</sub> – 2.5 cc, HCl – 1.5 cc, HF – 1.0 cc and H<sub>2</sub>O – 95.0 cc). The washed and dried samples were observed carefully in Versamet-II-Microscope at different magnification and some selected photomicrographs were taken.

Remarks:

Alloy 1 Al-6 wt% Mg

Alloy 2 Al-6 wt% Mg-0.4 wt% Sc

Alloy 3 Al-6 wt% Mg-0.4 wt% Sc-0.2 wt% Zr



**Fig. 1.** Isothermal annealing curve of the alloys, annealed at 400°C

### 3. RESULTS

#### 3. 1. Isothermal Annealing

Fig. 1 shows the isothermal annealing of the alloys at 400 °C. When the alloys are isothermally annealed at 400 °C, the rate and degree of initial softening is same for alloys 2-3. Alloy 1 shows a very fast and steep decrease in hardness followed by a constant value. The age hardening peaks are observed in alloys 2 and 3.

#### 3. 2. Recrystallization Kinetics From Microhardness Variation

The kinetics of recrystallization were determined from the microhardness values by considering the maximum and minimum values of microhardness, indicating deformed and completely recrystallized samples respectively. The maximum and minimum values for microhardness obtained in the present analysis are given in Table 2. The fraction recrystallized is obtained from the microhardness value by using the following formula:

$$X = \frac{H_{\max} - H_i}{H_{\max} - H_{\min}} \quad (1)$$

Where  $H_{\max}$  is maximum hardness corresponding to deformed sample ( $t = 0$ ),  $H_{\min}$  is minimum hardness corresponding to fully recrystallized sample and  $H_i$  is microhardness after a given annealing time [8]. Fully recrystallized sample got hold of the alloys annealed at 500 °C for one hour. Fig 2 shows the variation of microhardness and fraction recrystallized obtained from microhardness values for samples annealed at 400 °C. The base alloy 1 shows the maximum values of recrystallization but scandium added alloy 2 and 3 show the minimum. Alloy 3 combination of scandium and zirconium shows lowest recrystallization behaviour.

The kinetics of recrystallization can be represented in a mathematical form by using the JMAK relationship [9, 10]. The variation of fraction recrystallized with annealing time in JMAK relationship is given as

$$X = 1 - \exp[-(kt)^n] \quad (2)$$

Here  $n$  and  $k$  are the JMAK exponent and temperature dependent constant, respectively. This equation can be rearranged to a linear relationship by using a logarithmic expression.

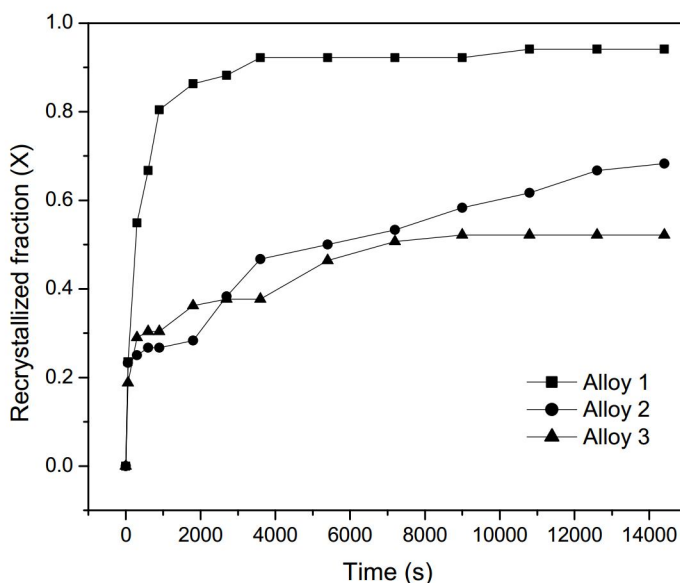


Fig. 2. Recrystallization kinetics obtained from microhardness data.

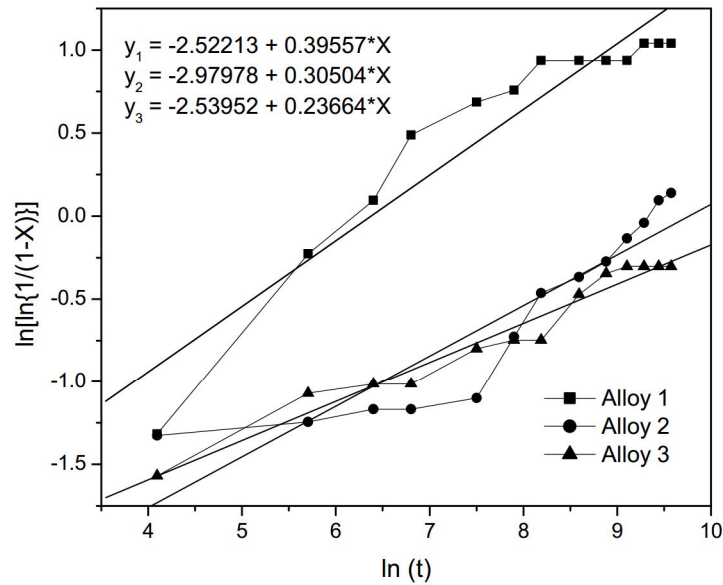


Fig. 3. Plot of  $\ln[\ln\{1/(1-X)\}]$  Vs.  $\ln(t)$ , showing a linear relationship with a slope equal to the JMAK exponent.

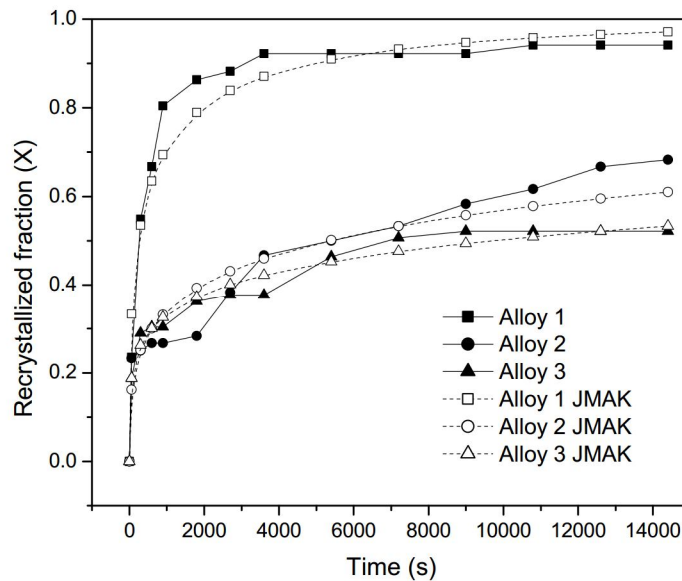


Fig. 4. Recrystallization kinetics for the alloys from Micro-hardness data and JMAK analysis.

$$\ln\left[\ln\left(\frac{1}{1-X}\right)\right] = n \ln(t) + n \ln(k) \quad (3)$$

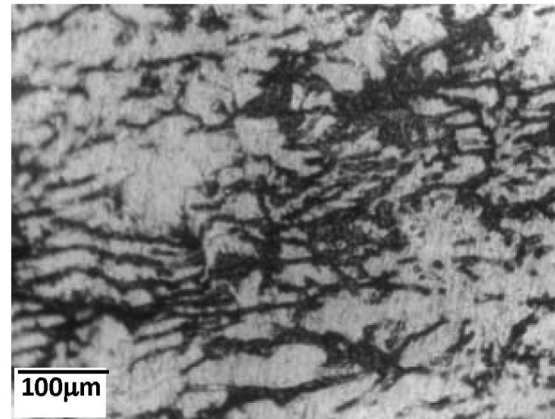
The slope of this linear expression will yield the exponent  $n$  and the parameter  $k$  can be

obtained from the ordinate as shown in Fig. 3.

The values of the JMAK exponent  $n$  and parameter  $k$  can be used to obtain recrystallization kinetics of the alloys annealed at 400 °C as shown in Fig. 4. Alloys 1-3 show the different slope for their different recrystallization behavior.

**Table 2.** Experimental value of maximum, minimum hardness and JMAK exponent of the alloys

| Alloy No. | $H_{max}$ | $H_{min}$ | n       | k        |
|-----------|-----------|-----------|---------|----------|
| Alloy 1   | 108       | 57        | 0.39557 | 0.001702 |
| Alloy 2   | 117       | 57        | 0.30504 | 0.000057 |
| Alloy 3   | 116       | 47        | 0.23664 | 0.000022 |



**Fig. 5.** Optical micrograph of 75% cold rolled alloy 1.

$$X = 1 - \exp[-(0.001702 \times t)^{0.39557}] \text{ for alloy 1} \quad (4)$$

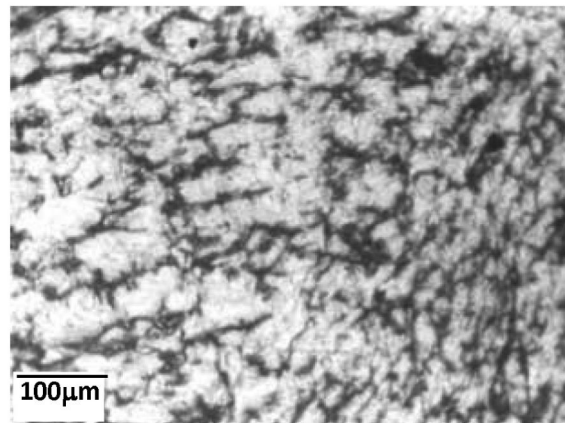
$$X = 1 - \exp[-(0.000057 \times t)^{0.30504}] \text{ for alloy 2} \quad (5)$$

$$X = 1 - \exp[-(0.000022 \times t)^{0.23664}] \text{ for alloy 3} \quad (6)$$

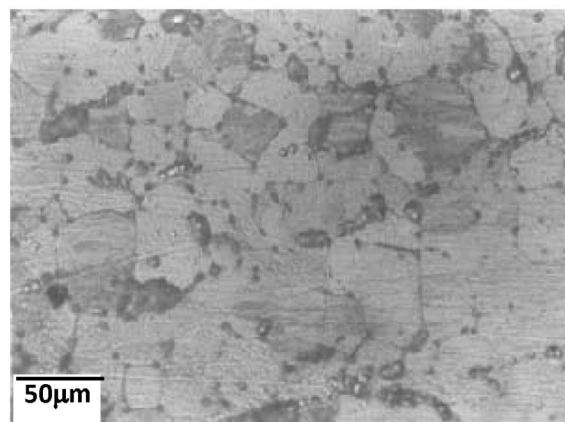
Recrystallization fraction between two methods, the base alloy 1 shows the minimum variation. Scandium added alloy 2 and 3 show the higher variation. Scandium and zirconium added alloy 3 shows the even more higher variation.

### 3. 3. Optical Micrographs

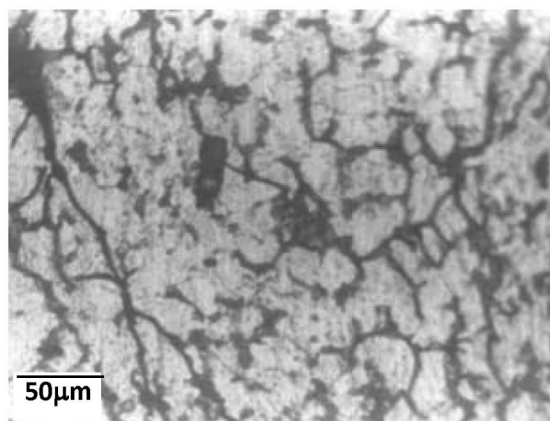
The cold worked alloy shows relatively coarse non-uniform grain structure. The overall appearance is columnar grains with second phase particles remaining aligned along the grain boundaries (Fig. 5). Fragmented dendrites, elongated along the direction of rolling, is observed in Fig. 6, showing the microstructure of cold worked alloy 3. In case of zirconium added alloy relatively coarsen grains are noted. If the alloys are annealed at 400 °C, the base alloy is seen to be recrystallised almost fully (Fig. 7). However, alloy 3 is recrystallised partially at the annealing treatment at 400 °C (Fig. 8).



**Fig. 6.** Optical micrograph of 75% cold rolled alloy 3.



**Fig. 7.** Optical micrograph of 75% cold rolled alloy 1, aged at 400 °C for 1 hour.



**Fig. 8.** Optical micrograph of 75% cold rolled alloy 3, aged at 400 °C for 1 hour.

#### 4. DISCUSSION

The initial softening of the cold worked alloys during isochronal ageing is thought to be due to rearrangement of dislocations at the ageing temperature. The age hardening of the alloys containing scandium is attributable to the formation of  $\text{Al}_3\text{Sc}$  precipitates. The maximum attainable hardness due to ageing the cold worked alloy has not exceeded the hardness values obtained due to cold working alone. This implies that the precipitation of  $\text{Al}_3\text{Sc}$  is not dislocation induced [3]. However peak temperature in zirconium bearing alloy is responsive to cold working. It is conjectured that  $\text{Al}_3\text{Zr}$  is formed at dislocations. The  $\text{Al}_3\text{Zr}$  being isomorphous with and soluble in  $\text{Al}_3\text{Sc}$ , the nucleation of  $\text{Al}_3\text{Sc}$  is facilitated indirectly by the presence of higher dislocation density. Moreover extensive cold working also generates large number of vacancies, which form vacancy-scandium atom complexes of high binding energy. The vacancy-solute atom complexes reduce the mobility and availability of solute atoms at low temperature to form G P zones. Hence hardening takes place only at a temperature high enough to decompose the complexes thereby making solute scandium atoms available for precipitate formation. Beyond peak hardness, over ageing effect due to coarsening of the precipitates is seen to have taken place. At higher ageing temperature there is ample scope for dislocation annihilation and this

softens the material.

The microhardness variation as well as fractional recrystallization includes the contribution of both recovery and recrystallization processes to the overall decrease in microhardness. However, the decrease in hardness is also due to precipitation coarsening of the alloy. It was reported earlier that precipitation coarsening of  $\text{Al}_3\text{Sc}$  occurs beyond 300 °C [11]. When adding scandium and zirconium into aluminium alloys simultaneously, part of Sc in the  $\text{Al}_3\text{Sc}$  (or  $\text{Al}_3\text{Zr}$ ) phase is substituted by Zr (or Sc). Thus  $\text{Al}_3(\text{Sc}, \text{Zr})$  complex particles are formed. These particles have higher thermal stability and are susceptible to coagulation to a lesser extent during heating. The higher thermal stability of  $\text{Al}_3(\text{Sc}, \text{Zr})$  particles makes some benefits for inhibiting recrystallization and reserving work-hardening. So hardness and strength of alloy basically maintain unchanged with the increase of stabilizing annealing time. Fig. 3 shows JMAK plot for recrystallization kinetics obtained from micro-hardness. Fig. 4 shows a plot for comparison of recrystallization kinetics as obtained from original micro-hardness data and from JMAK analysis. The overall kinetics behavior from the two methods of analysis is very similar. The higher difference in the curves for scandium added alloys can be attributed to the scale of analysis on each sample as well as precipitation hardening and recrystallization behaviour. The micro-hardness data represent an average behavior for recrystallization kinetics.

From the phase diagram of the alloy it is found that the present alloys would contain  $\alpha + \beta$  eutectic within the primary dendrites of  $\alpha$ . Here ' $\alpha$ ' is the aluminium rich solid solution and  $\beta$  is composed of intermetallics, primarily  $\text{Al}_8\text{Mg}_5$  along with aluminides of other metals like iron, chromium, zirconium, manganese, which are present in small quantities in the aluminium used for the present experimentation. The number of non-equilibrium segregation is dependent on the magnesium content and the concentration of other potential aluminide formers [11]. However scandium forms an anomalous supersaturated solid solution, which decomposes to form  $\text{Al}_3\text{Sc}$  [12]. Though general observations under optical

microscopy have not provided much information, the overall appearance of the microstructure resembles what are normally observed in cast aluminium alloy ingot [3]. The cold worked structures are comprised of elongated grains. The base alloy however has started recrystallising as it is known that recrystallisation of Al-6Mg alloy becomes completed at about 400 °C. However alloys 3 have dispersion of fine precipitates of Al<sub>3</sub>Sc. These precipitates are coherent with the matrix. It is reported that recrystallisation is almost impossible in aluminium alloys when such particles are already present [13]. The precipitates hinder the movement of sub-boundaries and grain boundaries. On increasing the temperature to 400 °C, the second phase constituent is almost dissolved in base alloy and there is nothing to hinder dislocation movement. As a result recrystallisation becomes complete. In alloys containing scandium the supersaturated solution decomposes to form Al<sub>3</sub>Sc at around 300 °C. These precipitates are known to be resistant to coarsening. There are reports saying that increasing the annealing temperature of Al-Mg-Sc alloy from 300 °C to 400 °C increases the size of Al<sub>3</sub>Sc precipitates from 4 nm to 13 nm. The precipitates of Al<sub>3</sub>Sc remain coherent with the matrix even when their size increases to 100 nm due to higher temperature of annealing [4]. In the present case however the precipitate size is around 15 nm when annealed at 400 °C. Therefore dislocation pinning force is very large. As a result recrystallisation is not possible. On the contrary alloy 3 with zirconium has shown onset of recrystallisation at 400 °C. In fact it has been reported earlier that kinetics of Al<sub>3</sub>(Sc<sub>x</sub>Zr<sub>1-x</sub>) coarsening is faster. Moreover Al<sub>3</sub>Zr precipitation is dislocation induced. Since Al<sub>3</sub>Sc forms isomorphously with Al<sub>3</sub>Zr, the precipitates grow pretty faster due to solute transfer by pipe diffusion. Thus loss in coherency of these precipitates occurs earlier. This is why alloy 3 shows sign of feeble recrystallisation in its microstructures when annealed at 400 °C.

## 5. CONCLUSION

Al-Mg alloy shows the small difference between two methods. Scandium added alloys show the

larger difference between two methods due to precipitation of Al<sub>3</sub>Sc, which age harden the alloys as well as recrystallization inhibitor at higher temperature. Sc and Zr added simultaneously in Al-Mg alloy to form Al<sub>3</sub>(Sc, Zr) particles, the recrystallization temperature of the alloys is enhanced by more stronger.

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