Proceedings of the 12th International Conference on
Aluminium Alloys, September 5-9, 2010, Yokohama, Japan

pp. 401-406

An Ageing Model for Al-Mg-Si Alloys Assuming the Needle-Shaped Morphology for Precipitates

Abbas Bahrami[1,2], Alexis Miroux[1] and Jilt Sietsma[2]

[1] Materials innovation institute (M2i), Mekelweg 2, 2628 CD, Delft, the Netherlands

[2] Materials Science and Engineering Department, Delft University of Technology, Mekelweg 2, 2628 CD, Delft, the Netherlands

In the present study an age-hardening model for Al-Mg-Si alloys has been developed considering cylindrical morphology with constant aspect ratio for precipitates of constant stoichiometry Mg_2Si. Although the shape of the precipitates has been recognized as an important factor controlling the mechanical properties of Al-Mg-Si alloys, the vast majority of precipitation models assume spherical precipitates. In the developed model, it is assumed that precipitation during underageing is controlled by simultaneous nucleation and growth and then after peak-age, it becomes coarsening-controlled. The transition from nucleation-growth regime to the coarsening regime happens when the concentration of the alloying element in the matrix becomes equal to the equilibrium concentration. The developed microstructural models are then combined with a precipitation-strengthening model to predict the evolution of yield strength of Al-Mg-Si alloys during ageing. The predictions of the model are in a good agreement with experimental data.

Keywords: *Al-Mg-Si alloys, Age-hardening model, Morphology.*

1. Introduction

Simulations of precipitation reactions and precipitation strengthening during ageing have gained considerable interest during the past decades [1-11]. The idea of combining precipitation reaction models with precipitation strengthening models was first introduced by Shercliff and Ashby [12-13]. They developed "a mathematical relation between process variables (alloy composition, and ageing temperature and time) and the alloy strength based on physical principles (thermodynamics, kinetics theory, and dislocation mechanics) [13] ". The first age-hardening model was successfully applied to 2000 and 6000 series aluminum alloys. Since then, a lot of attempts have been made to develop new age hardening models for different applications; i.e. isothermal ageing of naturally aged [2], pre-aged [1], and pre-deformed alloys [14], non-isothermal ageing [3], precipitation reactions during ageing [15], and multi-stage ageing [16,17]. For the sake of simplicity, the various previously developed age-hardening models use the simple assumption of spherical particles in a metal matrix. Nevertheless, the presence of the elongated needle-like β'' and rods of β' precipitates phase both oriented in <001>Al directions is thought to be the main source of hardening [18-20]. The objective of the present paper is to develop an ageing model which is able to predict the evolution of radius and length of precipitate as well as their number density and volume fraction during isothermal ageing. The precipitates are assumed to be cylindrical with constant aspect ratio. The microstructural reactions are divided into two parts: (i) Simultaneous nucleation and growth during underageing and (ii) coarsening during overageing. In order to model the simultaneous nucleation and growth of precipitates, the ageing time is divided into a series of smaller steps wherein the variation in the mean concentration of alloying elements in the matrix is assumed to be negligible. During each time step a new group of precipitates are added to the system while previously formed particles keep growing. In the end of the time step, the mean concentration of alloying element in the matrix is updated. This continues until the peak-age point where the coarsening starts. The beginning of coarsening is when the concentration of alloying element in the matrix decreases to the interface equilibrium value. The microstructural outputs of the model are then used predict the yield strength of the Al-Mg-Si alloys.

2. Age-hardening model

Precipitation is a phenomenon where the initial supersaturated alloy is decomposed into matrix and a new phase which is an agglomeration of solute atoms. The precipitation is traditionally categorized into three stages: Nucleation, growth, and coarsening. In this model, simultaneous nucleation and growth is assumed to happen first as long as there is enough solute remained in the matrix, and as soon as the mean concentration of alloying element in the matrix is reached to the equilibrium value, the coarsening starts. In the microstructure model, it is also assumed that there is no precipitation sequence and metastable β'' and β' precipitates are in meta-equilibrium with the matrix right from the beginning of ageing. These assumptions have been already successfully applied to modeling the precipitation hardening in Al–Mg–Si alloys [3-5]. In order to simplify the problem, it is assumed that

- All the precipitates have cylindrical morphology
- The aspect ratio of precipitates is constant during ageing
- The stoichiometry of precipitates is Mg_2Si from the beginning
- The interfacial energies at the tip and at the rim of precipitates are identical
- Copper is assumed to remain in solid solution
- The kinetics is controlled by the diffusion of Mg in the matrix

Figure 1 shows the schematic representation of the assumed cylindrical morphology, having the aspect ratio (A), which is defined as h/r (h is the half length and r is the radius of precipitates).

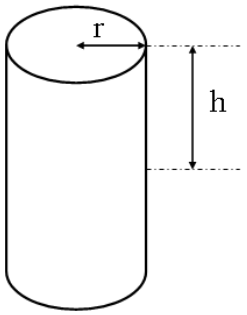


Fig. 1: Schematic representation of the assumed cylindrical morphology for precipitates

Assuming that precipitates nucleate with the cylindrical morphology, the change in the Gibbs free energy of the system due to nucleation can be written as

$$\Delta G_{nucl} = -2\pi r^2 h \Delta G_v + 4\pi r h \gamma + 2\pi r^2 \gamma. \tag{1}$$

where ΔG_v is the driving force per mole of solute atom to transform to precipitate from supersaturated solid solution and γ is the interfacial energy between precipitate and the matrix. Using the definition of aspect ratio, critical radius is calculated as

$$r_{cr} = \frac{2}{3}\left(\frac{2A+1}{A}\right)\frac{\gamma}{\Delta G_v}. \tag{2}$$

Putting the value of ΔG_v ($\approx \frac{RT}{v_m}\ln\left(\frac{C_m}{C_e}\right)$ for the case of a quasi-binary system) into Eq. (2) yields

$$r_{cr} = \frac{2}{3}\frac{v_m \gamma}{RT}\left(\frac{2A+1}{A}\right)\left(\ln\left(\frac{C_m}{C_e}\right)\right)^{-1}. \tag{3}$$

where C_m is the mean concentration of alloying element in the matrix, C_e is the equilibrium concentration of alloying element, and v_m is the molar volume of precipitate. Provided that the incubation time and elastic coherency strains around the nucleated particles can be neglected the nucleation rate J is conveniently expressed as [7]

$$J = J_0 \exp(-\frac{\Delta G^*}{RT} - \frac{Q_d}{RT}), \tag{4}$$

where J_0 is a pre-exponential term, ΔG^* the energy barrier and Q_d the activation energy for diffusion of the controlling alloying element. Based on the classic theory of nucleation, Myhr et al. [16] proposed an approximate expression to calculate ΔG^*. This equation is given below

$$\Delta G^*_{het} = \frac{(A_0)^3}{(RT)^2[\ln(C_m / C_e)]^2}. \tag{5}$$

where A_0 is a temperature-dependent constant. Peripheral and longitudinal growth rates of a needle-like precipitate of radius r and half-length h are determined by the composition gradient outside the precipitate, the diffusivity of solute atoms, and the aspect ratio of precipitate. The diffusion-controlled thickening and lengthening of a precipitate approximately obey the following rules [21]:

$$r = \frac{2}{3}\left(\frac{C_m - C_r}{C_p - C_r}\right)^{1/2}\left(\frac{D}{\pi A}t\right)^{1/2}. \tag{6}$$

$$h = \frac{2}{3}\left(\frac{C_m - C_r}{C_p - C_r}\right)^{1/2}\left(\frac{DA}{\pi}t\right)^{1/2}. \tag{7}$$

where C_r is the equilibrium interface concentration around the precipitates. C_r is given by [22]

$$C_r = C_e\left\{1 + (1 + \frac{1}{A})\frac{\gamma V_m}{RTr}\frac{1 - C_e}{C_p}\right\}. \tag{8}$$

As it is mentioned earlier, an iterative method is used to solve the set equation (6), (7), and (8). The ageing time during underageing is divided into a series of smaller time ranges (Δt) in such a way that the ageing time in the i^{th} step (t_i) is given by

$$t_i = t_{i-1} + i\Delta t. \tag{9}$$

If the Δt is assumed to be very small, one can rationally assume that the change in supersaturaion during time period Δt is negligible. Therefore, the increment in the radius of precipitate is calculated as

$$\Delta r = \frac{1}{3}\left(\frac{C_m - C_r}{C_p - C_r}\right)^{1/2}\left(\frac{D}{\pi A}\right)^{1/2} t^{-1/2}\Delta t. \tag{10}$$

And, consequently the radius of precipitates in the i^{th} step is written as

$$r_i = r_{i-1} + \frac{1}{3}\left(\frac{C_m - C_r}{C_p - C_r}\right)^{1/2}\left(\frac{D}{\pi A}\right)^{1/2} t_i^{-1/2}\Delta t. \tag{11}$$

At each time step, while previously-formed precipitates are growing according to equation (11), a new population of precipitates of radius r_{cr} nucleates. At the end of each time step, the mean concentration of alloying element in the matrix is updated using the below equation.

$$C_m = C_0 - C_p \sum_j 2\pi r_j^2 h_j N_j \,. \tag{12}$$

where N_j is the number of j^{th} group of precipitates with radius r_j and half-length h_j. The ageing time would increase step-by-step as long as the calculated mean concentration of alloying element (C_m) is higher than the highest equilibrium interface concentration (C_r). The time when C_m becomes equal to C_r is assumed to be the end of nucleation-growth regime and beginning of coarsening. The driving force for coarsening is provided by the difference between the size-dependent interfacial concentrations of alloying element and the remaining concentration of alloying elements in the matrix. The matrix concentration after peak-age and during coarsening is $C_{r_{peak}}$. So, the supersaturation can be written as [22]

$$\Omega = \frac{C_{r_{peak}} - C_r}{C_p - C_e} \approx \frac{C_{r_{peak}} - C_r}{C_p} \,. \tag{13}$$

Putting the values of C_r and $C_{r_{peak}}$ from Eq. (8) into Eq. (13), and the obtained equation into growth equation yields

$$r^3 = r_{peak-age}^3 + \frac{2}{3}\frac{D}{\pi}\frac{\gamma V_m}{RT}\frac{1}{A}\left(1+\frac{1}{A}\right)\frac{C_e(1-C_e)}{C_p^{\,2}}\left(t - t_{peak-age}\right) . \tag{14}$$

where $r_{peak-age}$ and $t_{peak-age}$ are peak-age radius (radius of precipitates at peak-age point) and time (the time when coarsening starts) respectively.

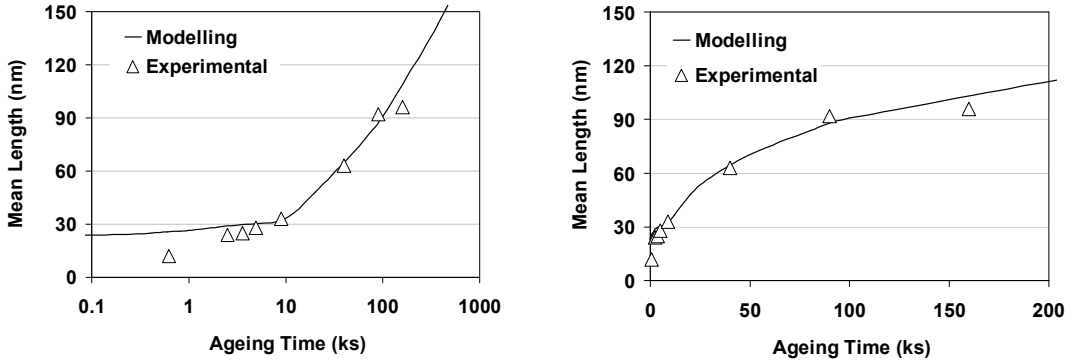
Strengthening model is a framework in which the overall strength of the artificially aged alloy can be obtained by the addition of the intrinsic strength of aluminum, the solid solution, and precipitates strength [4]. The contribution of precipitates to the total yield strength is given by [23]

$$\begin{cases} \sigma_{ppt} = 2BGbM\left(\dfrac{f_v}{\sqrt{3}\pi}\right)^{1/2} \dfrac{r_{peak-age}^{\frac{3(m-1)}{2}}}{r_{trans}^{\frac{3m}{2}}} r^{1/2} & r < r_{peak-age} \quad (a) \\ \sigma_{ppt} = 2BGbM\left(\dfrac{f_{peak}}{\sqrt{3}\pi}\right)^{1/2} \dfrac{r^{(\frac{3m}{2}-1)}}{r_{trans}^{\frac{3m}{2}}} & r_{peak-age} < r < r_{trans} \quad (b) \\ \sigma_{ppt} = 2BGbM\left(\dfrac{f_{peak}}{2\pi}\right)^{1/2} \dfrac{1}{r} & r_{trans} < r \quad (c) \end{cases} . \tag{15}$$

where f_{peak} is the volume fraction of precipitates at peak-age, B is a constant close to 0.5, b is the Burgers vector, M is the Taylor factor, m is a constant close to 0.6 [23], and r_{trans} is the radius above which precipitates are non-shearable (equal to 3.5 nm) [23]. The way how to calculate the contributions alloying elements in the matrix to the yield strength is explained in the literature [3].

3. Application of the model and discussion

The developed model is applied to isothermal ageing of the alloy AA6061 (1.12 wt% Mg, 0.57 wt% Si, 0.25 wt% Cu). Figure 2 shows predictions of precipitate length in the alloy AA6061 aged at 190 °C. The aspect ratio is chosen equal to 10 to get the right value at peak-age (10 ks).



a b

Fig. 2: Prediction of the length of precipitate in the alloy AA6061 aged at 190 °C in the a) Log-scale and b) linear scale

As it is seen, there is an overestimation of the modelling results in the underage regime. The precipitation sequence in this alloy is believed to be [23]

$$\text{SSSS} \rightarrow \text{(Si and Mg clusters)} \rightarrow \text{Mg/Si co-clusters} \rightarrow \text{GP-zones} \rightarrow \beta'' \rightarrow \beta' \rightarrow \beta$$

where SSSS is the supersaturated solute solution, β'' is a needle-like precipitate, β' is a rod-like precipitate, and β is the equilibrium phase in the precipitation sequence. In the early stage of ageing GP-zones have spherical morphology. Therefore, the assumption of constant aspect ratio through the ageing is not an entirely correct physically-based assumption. Figure 3 shows the prediction of the yield strength of the alloy AA6061 together with experimental data at ageing temperature 170 °C. As it is seen there is a reasonably good agreement between experimental data and model predictions.

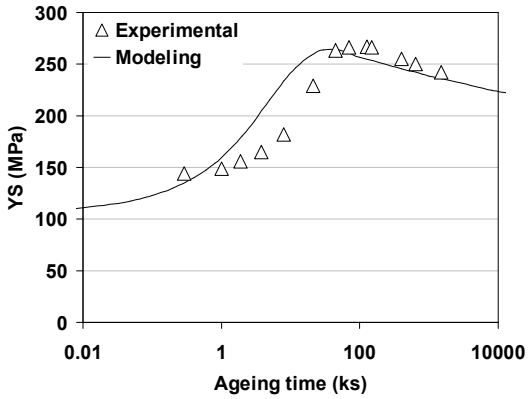


Fig. 3: Prediction of the yield strength of the alloy AA6061 aged at 170 °C.

4. Summary

A physically-based age-hardening model is developed for Al-Mg-Si alloys assuming cylindrical morphology for precipitates. In the model it is assumed that the aspect ratio of precipitate is constant

during ageing, inferring that precipitates nucleate with cylindrical morphology. The model, applied to the isothermal ageing of the alloy AA6061, is fitted in such a way that it gives the right prediction for the length of precipitate at peak-age. The modelling results show an overestimation of the length of precipitate in the underage regime compared to the experimental data. This is due the assumption of constant aspect ratio through the whole ageing. The model also predicts the yield strength of the alloy AA6061 reasonably well.

Acknowledgments

This research was carried out under Project No. MC4.05213 in the framework of the Research Program of the Materials Innovation Institute M2i (www.m2i.nl).

References

[1] S. Esmaeili and D.J. Lloyd,: Acta Mater. 53 (2005) 5257.
[2] S. Esmaeili, D.J. Lloyd and W.J. Poole,: Acta Mater. 51 (2003) 2243.
[3] O.R. Myhr, Ø. Grong and S. J. Andersen,: Acta Mater. 49 (2001) 65.
[4] O.R. Myhr and Grong,: Acta Metall. Mater. 39 (1991) 2693.
[5] O.R. Myhr and Grong,: Acta Metall. Mater. 39 (1991) 2703.
[6] H.R. Schercliff and M.F. Ashby,: Mater. Sci. Tech. 7 (1991) 85.
[7] O.R. Myhr and Ø. Grong,: Acta Mater. 48 (2000) 1605–1615.
[8] B.I. Bjorneklett, Ø. Grong, O.R. Myhr and A.O. Kluken,: Acta Mater. 1359-6454 (98) 6257.
[9] W. Sha,: Mater Design 5 (2007) 1527.
[10] W.J. Poole, H. Shercliff and T. Castillo,: Mater. Sci. Tech. 13 (1997) 897.
[11] H.I. Aaronson, K.R. Kinsman and K.C. Russell,: Scripta Mater. 4 (1970) 101.
[12] H.R. Shercliff and M.F. Ashby,: Acta Metall. Mater. 38, No. 10 (1990) 1789.
[13] H.R. Shercliff and M.F. Ashby,: Acta Metall. Mater. 38, No. 10 (1990) 1803.
[14] A. Deschamps and Y. Brechet,: Acta Mater. 47 (1999) 293.
[15] D.H. Bratland, Ø. Grong, H. Shercliff, O.R. Myhr and S.T. TjØtta,: Acta Mater. 45 (1997) 1.
[16] O.R. Myhr, Ø. Grong, H.G. Faejr and C.D. Marioara,: Acta Mater. 52 (2004) 4997.
[17] Ø. Grong and H.R. Schercliff,: Progress in Mater. Sci. 47(2002) 163.
[18] W.F. Miao and D.E. Laughlin,: Scripta Mater., Vol. 40, No. 7 (1999) 873.
[19] J.F. Nie, B.C.Muddle and I.J. Polmear,: Mater. Sci. Forum Vols. 217-222 (1996) 1257.
[20] K.G. Russell and F. Ashby,: Acta Meter. 18 (1970) 891.
[21] G. Liu, G.J. Zhang, X.D. Ding, J. Sun and K.H. Chen,: Mater. Sci. Eng. A344 (2003) 113.
[22] M. Ferrante and R.D. Doherty,: Acta Met. 27 (1979) 1603.
[23] S. Esmaeili,: Precipitation hardening behavior of AA6111, PhD thesis, Feb. 2002, pp. 132.