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The kinetics of the discontinuous precipitation and dissolution in Mg-rich Al alloys

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The morphology and growth kinetics of the discontinuous precipitation and dissolution reactions in supersaturated Mg-Al solid solutions containing 7.3, 9.1 and 10.9 at % Al have been investigated by optical and scanning electron microscopy and X-ray measurements. The volume fraction of regions transformed by the discontinuous precipitation reaction, the reaction front velocity, the interlamellar spacing and the average composition of the solute-depleted lamellae were determined as a function of the temperature. For the first time, the kinetics of the discontinuous dissolution reaction has been studied in the Mg-Al system. It has been shown that the transport of the solute atoms during both reactions is governed by grain boundary diffusion. © *1999 Kluwer Academic Publishers*

1. Introduction

Discontinuous precipitation (DP) is a heterogeneous reaction which leads to the formation of a lamellar structure behind a moving grain boundary. The moving grain boundary, called reaction front (RF), acts as a short circuit path for the diffusion of the solute atoms [1]. The lamellar structure consists of the new precipitated phase and the solute-depleted matrix phase which are not in complete equilibrium regarding the chemical composition.

Discontinuous dissolution is the reverse reaction in regard to DP. It occurs in some systems near the solvus line where the volume diffusion process is relatively slow compared with the grain boundary diffusion process [1]. The DP reaction occurs in Mg-rich solid solutions of the Mg-Al system [2–6] resulting in the formation of a lamellar two-phase structure: the precipitated $Al_{12}Mg_{17}$ phase and the depleted solid solution [7]. The occurrence of the DD process was only mentioned for this system [8]; however, no example of this phenomenon was presented.

In this paper, morphological and kinetical studies of the DP and DD reactions were undertaken in three Mg-Al solid solutions containing 7.3, 9.1 and 10.9 at % Al. It will be shown that DD occurs in a large temperature range below and above the solvus temperature of the Mg-7.3 at % Al alloy. The Arrhenius parameters of the grain boundary chemical diffusion are determined for both reactions using the equation of Petermann and Hornbogen [9].

2. Experimental

The Mg-Al alloys were prepared by vacuum induction melting of the elements of high purity (3N5) and casting into 11 mm diameter rods. They were then homogenized at 703 K for 48 days in Duran glass capsules and water quenched. Samples of 6 mm thickness were cut off from the rods by spark erosion, and a final homogenization was applied at 703 K for 48 h, followed by a water quenching. The resultant average grain size was about 200 μm.

For the study of the DP reaction the samples were annealed between 425 and 650 K for time periods between 1 min and 72 h. The investigation of the DD reaction was performed only in the Mg-7.3 at % Al alloy. In this case the samples were aged at 498 K for 3.5 h which ensured 30% of the whole sample to be covered by discontinuous precipitates. The seam width and the interlamellar spacing are then 80 and 0.75 μm, respectively. After this heat treatment, the dissolution heat treatment was applied at temperatures ranging from 585 to 665 K.

A standard metallographic technique was used for the sample preparation including wet grinding, prepolishing and "Minimet" polishing with 6 and 1 μm diamond paste, using a "Nylon" polishing cloth. Prior to the optical and scanning electron microscopy the samples were etched with 3% nital.

Quantitative metallographic methods were applied to determine the velocity of the DP and DD reactions. In the first case, the measurements of the so-called seam widths were performed after various times of ageing.

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In the second case, the widths of the receding distances were measured. At each temperature 60 widths were measured, averaged and normalized by using the factor $\pi/4$ according to the method proposed by Lück [10]. Finally, the reaction front velocity of DP and DD was taken as the slope of the cell width vs the annealing time.

Lück's method was also used for the determination of the thickness of the $Al_{12}Mg_{17}$ lamellae as an average from 15 measurements performed for each ageing temperature in the steady state period of growth, directly on an optical microscope with a special monitor enabling to magnify the image up to several thousands to one. In the necessary cases the thickness was determined from scanning electron microscope pictures.

The volume fraction of the regions transformed by the DP reaction was determined using a Kontron image analyser for isothermal ageing at 498 K. The lattice parameters of the depleted phase were determined using a 4-circle goniometer diffractometer (CuK_α radiation, 0.125–0.5 degrees/min scanning speed). The 6 peak positions in the range $2\theta = 45$–75° were substituted in an adequate lattice parameter's refinement software where θ is the Bragg angle of reflection. The resulting a and c values were correlated with the lattice parameter-composition data of Raynor [11] in order to obtain the average Al concentration.

3. Experimental results and discussion

3.1. Morphology

Ageing in the temperature range 425–650 K produced DP as observed earlier in Mg-Al alloys [3, 4, 12]. Fig. 1a shows a typical microstructure of cells spreading out from their initial grain boundary position towards the adjacent grain areas, leaving behind an alternate mixture of the depleted solid solution and the new phase $Al_{12}Mg_{17}$ which is an intermetallic compound with the α-Mn (A12) structure [7]. In addition to a stable type of parallel growing lamellae (Fig. 1b), with a more or less constant spacing, other morphologies can be found in the Mg-Al system (Fig. 1c). This results from the tendency of the lamellae to undergo the fragmentation and subsequent spheroidization process.

"Ghost" images left in the positions where the $Al_{12}Mg_{17}$ lamellae had previously existed are shown in Fig. 2 after annealing the Mg-7.3 at % Al alloy at 645 and 625 K. These images show convincingly the discontinuous mode of the dissolution and it makes possible to measure the velocity of the backward migrated RF. However, in the areas where the fragmentation and spheroidization of the $Al_{12}Mg_{17}$ lamellae has already occurred, the continuous dissolution process can be favoured.

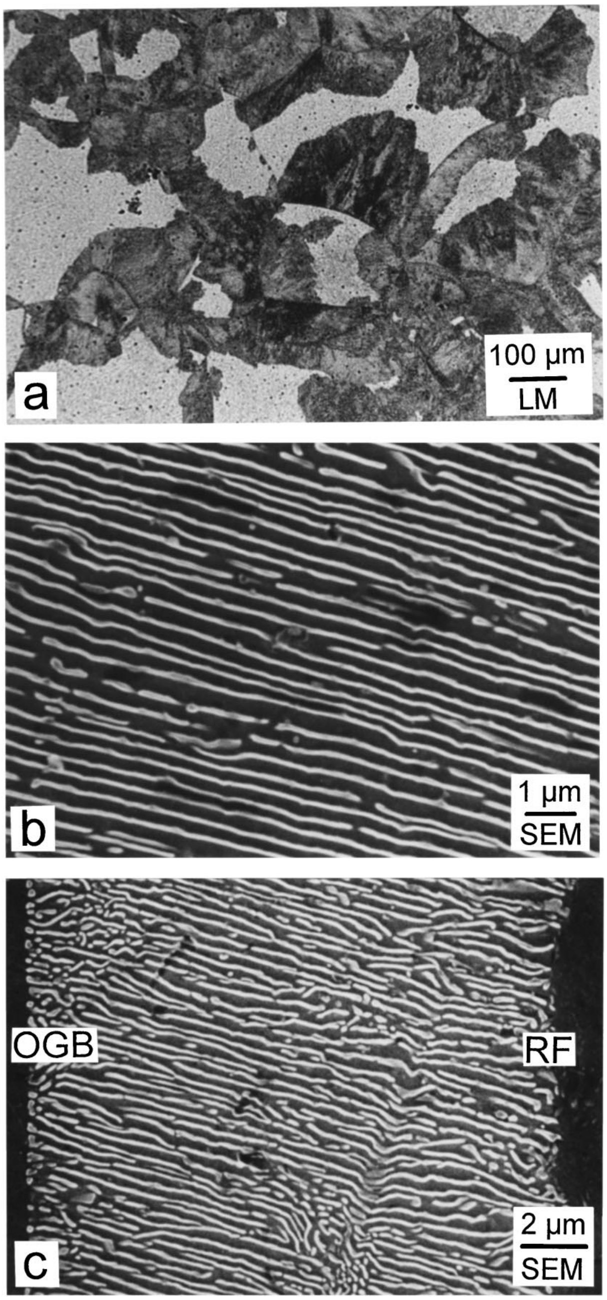

Figure 1 Morphology of the DP cells: (a) Mg-9.1 at % Al annealed at 475 K for 3 h, (b) regular lamellar structure in Mg-9.1 at % Al annealed at 425 K for 16 h, and (c) irregular lamellar structure in Mg-10.9 at % Al annealed at 498 K for 15 min RF-reaction front; OGB-original position of the grain boundary.

3.2. The material volume transformed

The volume fraction, X, of the regions transformed by DP after ageing at 498 K was measured through quantitative image analysis, basing on the principle that the surface fraction is equal to the volume fraction [13]. The obtained results were analysed according to the Johnson-Mehl equation [14]:

$$X = 1 - \exp\left(-\left(\frac{t}{\tau}\right)^n\right) \qquad (1)$$

where t is the annealing time, τ is a time constant and n is a coefficient characteristic for the transformation process. The experimental data plotted on a loglog $(1/1 - X)$ vs $\log t$ scale show a linear behaviour for the major part of the DP process (Fig. 3). The values of the parameter n calculated from the slope of the lines in Fig. 3 are equal to 0.85, 0.95 and 1.0 for the three Mg-Al alloys containing 7.3, 9.1 and 10.9 at % Al, respectively. These values are very close to $n = 1$. According

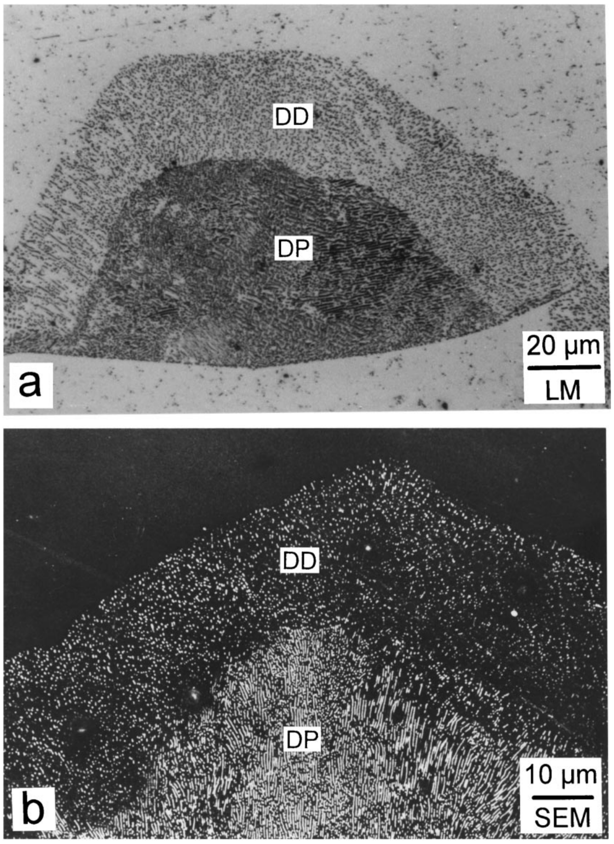

Figure 2 Morphology of the discontinuous dissolution (DD) reaction in Mg-7.3 at % Al showing the "ghost" images in the dissolved area. Dissolution heat treatment at 625 K for 7 min. DP-discontinuous precipitation, OGB-original position of the grain boundary.

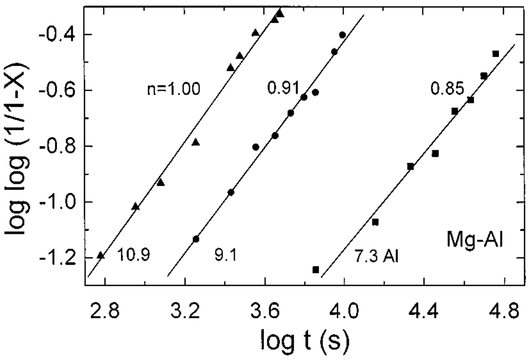

Figure 3 Evolution of log log $(1/1-X)$ vs. log t where X is the volume of the material transformed by the DP reaction and t is the ageing time at 498 K for the Mg-Al alloys containing 7.3, 9.1 and 10.9 at % Al.

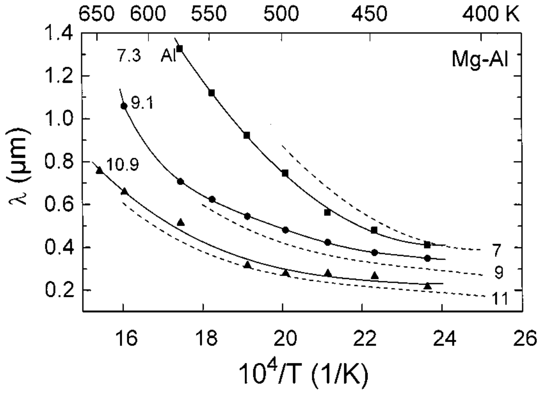

Figure 4 Interlamellar spacing vs. reciprocal temperature for the three Mg-Al alloys studied. The data of Frebel and Behler [3] are given for comparison (dashed lines).

to Christian [15] this corresponds to a process of grain boundary nucleation after saturation.

3.3. Interlamellar spacing and growth rate

Fig. 4 illustrates the change of the interlamellar spacing, λ, as a function of the reciprocal temperature. In agreement with previous work [3], the interlamellar spacing increases with increasing temperature and decreases with an increase of the initial composition.

Fig. 5a–c shows the time dependence of the seam width for the DP reaction at three different ageing temperatures while Fig. 5d shows the "ghost image" width as a function of the dissolution time for three temperatures for the Mg-7.3 at % Al alloy after prior ageing at 498 K for 3.5 h. A good fit of the respective set of data points within the period of time studied allows to determine the velocity, v, of the DP and DD reactions applying a suitable regression analysis.

Fig. 6 summarizes the growth rate data as a function of the reciprocal temperature. As can be seen the velocity for the DP reaction increases with increasing ageing temperature which is a characteristic behaviour of any diffusion-controlled reaction. This increase is less pronounced at higher temperatures due to the decrease of the driving force for the DP reaction. It should be also noted that the values of the velocity of the DP reaction increases roughly 5 times with an increase of the solute content in the alloy from 7.3 to 10.9 at % Al. However, the values of the velocity of the DD reaction are not an extension of the tendency observed for DP reaction. At the DD temperature of 585 K, the velocity of DD reaction is almost 5 times smaller than for the highest investigated DP temperature. This temperature is close to the critical one for the DD reaction which was found to be 580 K according to Sulonen's empirical formula [16]; "The equilibrium solute concentration at the critical temperature is the arithmetic mean of the solute content in the alloy and the average solute content in the $Al_{12}Mg_{17}$ lamellae". Therefore, it is supposed that the total driving force for the DD reaction available close the critical temperature does not allow a fast reaction front movement. No DD reaction was observed above 665 K due to a domination of another mode of dissolution, called continuous dissolution governed by volume diffusion, which does not involve any grain boundary movement.

3.4. Average solute concentration in the lamellar precipitates

The average solute content x_{av} within the $Al_{12}Mg_{17}$ lamellae was determined by X-ray measurements (Fig. 7). It was not possible to find a definitive tendency between the x_{av} value and the solute content in the alloy. Therefore, the solid points given in Fig. 7 represent an average value for the three Al contents in the alloys investigated. The obtained results are in

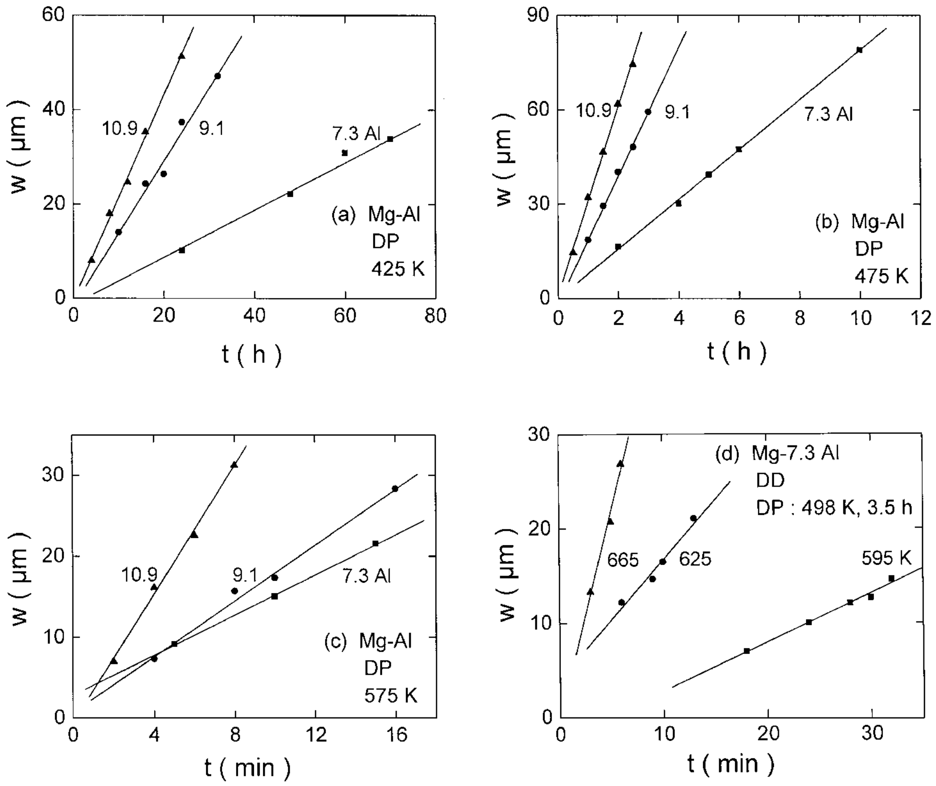

Figure 5 Examples of the DP width w as a function of the annealing time at three different temperatures for the three Mg-Al alloys studied (a–c) and of the DD "ghost" image width as a function of the dissolution time for Mg-7.3 at % Al prior aged at 498 K for 3.5 h and then annealed at 595, 625 and 665 K (d).

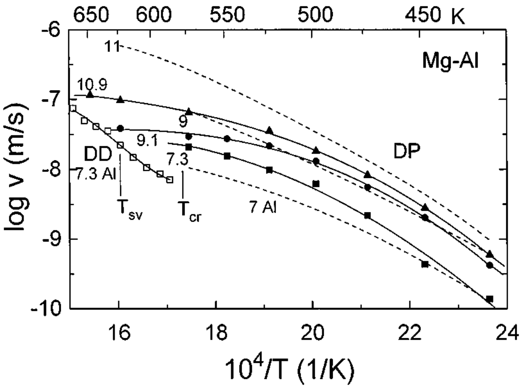

Figure 6 Velocity of DP and DD reactions vs. reciprocal of temperature for the three Mg-Al alloys studied. The data of Frebel and Behler [3] are given for comparison (dashed lines). T_{sv} is the temperature which corresponds to the limit of the Al solubility in (Mg) for a Mg-7.3 at % Al alloy. T_{cr} is the critical temperature of the DD reaction (see definition in the text).

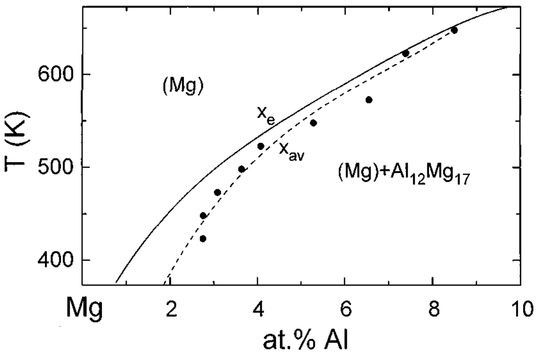

Figure 7 Equilibrium solvus concentration after Massalski [11] for the Mg-Al system with the values of the average Al content left in the (Mg) lamellae. The x_{av} values of Frebel and Behler [3] for the DP reaction are also given (dashed line).

a good agreement with the experimental data reported by Frebel and Behler [3], who also did not find any distinguished relationship between the solute content in the alloy and the average solute content in the lamellar precipitates.

3.5. Driving force and grain boundary diffusivity

The driving force for DP was calculated using the following equation:

$$\Delta G_{\mathrm{DP}} = -RT\left[x_o \ln\frac{a_o^{\mathrm{Al}}}{a_e^{\mathrm{Al}}} + (1-x_o)\ln\frac{a_o^{\mathrm{Mg}}}{a_e^{\mathrm{Mg}}}\right] \times \left[1-\left(\frac{x_{\mathrm{av}}-x_e}{x_o-x_e}\right)^2\right] + \left(\frac{2\sigma V_m}{\lambda}\right) \qquad (2)$$

The first term represents the chemical free energy available for the growth of DP. The multiplicative concentration term means that only a fraction of the total free enthalpy is available due to the incomplete equilibrium of the solute depleted lamellae ($x_{\mathrm{av}} > x_e$). Here x_o is the solute content in the alloy, x_e is the equilibrium concentration within the (Mg) lamella, and a_o and a_e are the corresponding thermodynamic activities of the components Al and Mg. The second term is associated with the free enthalpy needed for the creation of the interphase boundaries in the lamellar microstructure. Here, σ is the interfacial free energy per unit area which is assumed to be 290 mJ/m^2 [3], and V_m is the molar volume of the cellular aggregate. The thermodynamic activities of Mg and Al necessary for the calculation were taken from the paper of Behler and Frebel [3].

The respective Al concentration was taken from Fig. 7 and the interlamellar spacing from Fig. 4. The molar volume was calculated as $V_m = 1.365 \times 10^{-5}$ m^3/mol.

The free energy changes accompanying the DD reaction were calculated using the method of Chuang [17]:

$$\Delta G_{\mathrm{DD}} = \frac{T_{\mathrm{DD}}}{T_{\mathrm{DP}}}\Delta G_{\mathrm{DP}} - \left(\frac{T_{\mathrm{DD}}}{T_{\mathrm{DP}}} + 1\right)\frac{2\sigma V_m}{\lambda} \qquad (3)$$

where T_{DD} and T_{DP} are the annealing temperatures for DD and DP, respectively, and ΔG_{DP} is the driving force of the DP reaction at T_{DP}.

However, Equation 3 is valid for an ideal solid solution which could not be the case for the Mg-7.3 at % Al alloy. Therefore, an alternative calculation of the chemical part of the driving force for the DD reaction was performed based on the free energy data for the $\mathrm{Al_{12}Mg_{17}}$ and (Mg) phases extracted from the thermodynamic evaluation of the Al-Mg system given by Lüdecke and Hack [18]. The the free energy of the two-phase mixture, $\Delta G_{(\mathrm{Mg})+\mathrm{I}}$ was calculated for the dissolution temperature using the following equation resulting from the lever rule:

$$\Delta G_{(\mathrm{Mg})+\mathrm{I}} = \left[\frac{x_{\mathrm{I}} - x_o}{x_{\mathrm{I}} - x_{\mathrm{av}}}\right]\Delta G_{(\mathrm{Mg})} + \left[\frac{x_o - x_{\mathrm{av}}}{x_{\mathrm{I}} - x_{\mathrm{av}}}\right]\Delta G_{\mathrm{I}} \qquad (4)$$

where x_{I} is the Al concentration in the intermetallic compound. The free energy of the two phases (Mg) and $(\mathrm{Al_{17}Mg_{12}})$, $\Delta G_{(\mathrm{Mg})}$ and ΔG_{I}, was obtained from the thermodynamic functions delivered by Lüdecke and Hack [18]. The chemical part of the driving force for the DD reaction, $\Delta G^{\mathrm{C}}_{\mathrm{DD}}$, was then determined by substracting $\Delta G_{(\mathrm{Mg})+\mathrm{I}}$ from the free energy of the (Mg) phase at the final concentration which is assumed to be x_0, and finally, ΔG_{DD} is obtained by adding the interfacial free energy ($-2\sigma V_m/\lambda$, with $\lambda = 0.75\ \mu$m). The calculation was performed only for the Mg-7.3 at % Al alloy for which both DP and DD experiments were made. Surprisingly, the calculated ΔG_{DD} values were within a relative error of 10% in agreement with the values from Equation 3. Therefore, the driving force values presented in Fig. 8 were calculated from Equation 3 for different annealing temperatures. As can be seen from this figure the changes in the driving force for DD are not sensitive to an increase of the dissolution temperature, being still around 100 J/mol. This value corresponds to the total driving force of the DP reaction at 475 K. However, it should be emphasized that the calculation of ΔG_{DD} assumed a full homogenization of the solid solution created after DD reaction. This is not the case in the Mg-Al system where strong etching experiments revealed fluctuations of the Al concentration left in the post-dissolution area. As a simulation procedure showed [19] the maximum (up to 20 wt % Al) of the fluctuation was at the previous position of the $\mathrm{Al_{12}Mg_{17}}$ lamellae, while the minimum (4–6 wt % Al) was at a distance relevant to the middle of the (Mg) lamellae. Therefore, the driving force presented in Fig. 8 must be divided by a factor 3 to account for the solute fluctuations in the newly formed $(\mathrm{Mg})_\sim$ solid solution (see upper curve for ΔG_{DD}).

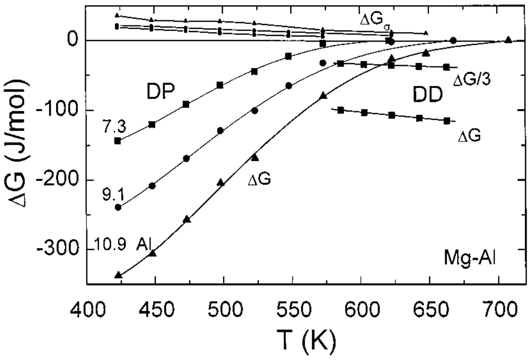

Figure 8 Gibbs free energy terms as a function of the temperature for the DP reaction in the Mg-Al alloys studied and for the DD reaction in Mg-7.3 at % Al aged previously at 498 K. ΔG_σ is the surface free energy associated with the lamellar interfaces and ΔG is the total driving force for the DP and DD reactions.

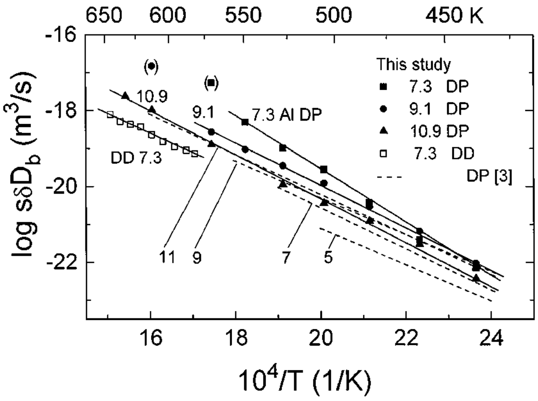

Figure 9 Arrhenius plot of the grain boundary diffusivity for the DP reaction in three Mg-Al alloys and for the DD reaction in Mg-7.3 at % Al aged previously at 498 K. The data of Frebel and Behler [3] obtained for the DP reaction are given for comparison. The data points in brackets () were not taken into account in the fitting procedure.

3.6. Grain boundary diffusivity and activation energy

For the consistency the grain boundary diffusivities for the DP and DD reactions were evaluated using the equation of Petermann and Hornbogen [8]:

$$s\delta D_b = \frac{RT}{-8\Delta G}\lambda^2 v \qquad (5)$$

Here, D_b is the grain boundary diffusion coefficient, δ is the grain boundary width, s is the segregation factor, ΔG is the total driving force for the DP or DD reactions, R is the gas constant, and T is the absolute temperature of the process. Fig. 9 represents the Arrhenius plot of the calculated $s\delta D_b$ data as a function of the reciprocal temperature. There is no definitive tendency between the changes of the diffusivity in spite of the increase of the solute content from 7.3 to 10.9 at % Al. All the changes in the $s\delta D_b$ values are within the values which are well below one order of magnitude, especially at low ageing temperatures. It seems to be a feature of the Mg-Al system because Frebel and Behler [3] found the same tendency for Mg alloys containing 5–11 at % Al. Here, the changes of the diffusivity were even more

TABLE I Arrhenius parameters for the DP and DD reactions in Mg-Al alloys

Reaction	Alloy	$(s\delta D_b)_o$ (m^3/s)	Q_b (kJ/mol)
DP	Mg-7.3 at % Al	8.46×10^{-6}	138.9
DP	Mg-9.1 at % Al	1.15×10^{-9}	105.3
DP	Mg-10.9 at % Al	1.53×10^{-9}	109.7
DD	Mg-7.3 at % Al	2.15×10^{-11}	100.6

limited, up to 1/3 order of magnitude. Such a behaviour was also observed in other systems like Fe-Zn [20, 21].

Unfortunately, there are no available data concerning the grain boundary diffusion process along stationary grain boundaries in this system. However, as can be seen from Fig. 9 the diffusivity values obtained in this study are in good agreement with those determined by Frebel and Behler [3] who used the Turnbull [22] and Sundquist [23] model of the DP reaction. The data obtained by Porter and Edington [4] for a Mg-9 at % Al alloy are slightly lower but the overall tendency is also preserved. It should be mentioned that Porter and Edington measured the Al concentration profiles across the (Mg) lamellae using the high spatial electron energy loss spectroscopy microanalysis technique. Then, the $s\delta D_b$ values were extracted using Cahn's [24] equation and the local values of the interlamellar spacing and the growth rate. They are slightly less than the corresponding diffusivity values for the DP reaction in Mg-7.3 wt % Al which reflects the influence of the relatively slow velocity of the cell receding during the DD reaction.

The values of the pre-exponential factor, $(s\delta D)_o$, and the activation energy, Q_b, evaluated from the Arrhenius plot in Fig. 9 are collected in Table I. The values of the activation energy are between 100 and 138 kJ/mol. The only available data concerning the volume diffusion of Al in Mg are those of Moreau *et al.* [25]. They determined the Arrhenius parameters of the diffusion of Al in Mg as: $D_o = 12 \times 10^{-4}$ m^2/s and $Q = 144$ kJ/mol. Using those parameters the volume diffusion coefficient is within 10^{-19}–10^{-15} m^2/s for the temperature range 475–625 K. These values are approximately 7 orders of magnitude less than the diffusion coefficient D_b at the migrating reaction front of the DP and DD at the corresponding temperatures (assuming $s = 1$ and $\delta = 0.5$ nm). Therefore, it can be concluded that the DP and DD reactions in the Mg-Al system are governed by the diffusion of the Al atoms along the grain boundaries.

4. Conclusions

The investigation performed in the Mg alloys containing 7.3, 9.1 and 10.9 at % Al revealed the occurrence of discontinuous precipitation in the temperature range from 425 to 650 K. The reaction is characterized by the formation of a lamellar structure of the solute-depleted parent phase (Mg) and the precipitate intermetallic compound $Al_{12}Mg_{17}$ behind the moving reaction front.

Subsequent annealing of such a structure performed for the Mg-7.3 at % Al alloy showed the occurrence of the discontinuous dissolution reaction in the temperature range from 585 to 665 K. The DD process resulted in the formation of an inhomogeneous solid solution manifested by the presence of the so-called "ghost" images in the post-dissolution area.

The volume fraction of regions transformed by the DP reaction, the reaction front velocity, the interlamellar spacing and the average composition of the solute-depleted (Mg) lamellae determined as a function of the temperature and solute content were found to show a behaviour typical for the discontinuous precipitation reaction. The grain boundary diffusivities calculated using the Petermann-Hornbogen equation were found to be in the range from 10^{-17} to 10^{-23} m^3/s. These values convincingly showed that the transport of the solute atoms during the DP and DD reactions in Mg-Al alloys is governed by grain boundary diffusion.

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