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A unified description of solid solution creep strengthening in Al-Mg alloys

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Abstract

It is proposed that the creep strengthening of aluminium due to the addition of Mg atoms in solid solution and the variation of the stress exponent, n , with the stress (from $n \approx 5$ to $n \approx 3$) is due to a unique microstructural feature, that is, the stress variation of the total to mobile dislocation density ratio. To support this idea, creep data recorded from the literature of pure Al-Mg alloys and of pure aluminium have been analyzed in the frame of the Strength Difference Method, SDM. A strengthening proportional to the applied stress is found. On this basis, a model which considers a change of the dislocation density/velocity due to the presence of the Mg atoms in solid solution and the solute drag and climb forces for dislocation motion was assumed. The new model, which also takes into account published data of the dislocation density measured at different applied stress, describes naturally the curvature of experimental Al-Mg creep data, associated traditionally with the change in deformation mechanism from dislocation glide controlled ($n=3$) to dislocation climb controlled ($n=3$) mechanism. The model does not undermine the relevance of aluminum self diffusion for dislocation climb process (vacancy diffusion) as the creep controlling mechanism in this solid solution alloy. (203).

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1. Introduction

The creep behavior of solid solution Al-Mg alloys at intermediate temperatures ($0.3-0.6 T_m$, where T_m is the melting temperature) has been extensively investigated with the aim of understanding the effect of the presence of foreign atoms on the high temperature performance of metallic alloys [1-8]. Usually, the creep behavior of crystalline solids has been studied in the frame of the power law relation between steady state creep rate, $\dot{\epsilon}$, and the applied stress, σ , *i.e.*,

$$\dot{\epsilon} = C \sigma^n \quad (1)$$

(where n is the stress exponent and C a microstructure and temperature dependent constant) and the Arrhenius type dependence of $\dot{\epsilon}$ with temperature, T , *i.e.*,

$$\dot{\epsilon} = C' \exp\left(\frac{Q_c}{RT}\right) \quad (2)$$

(where Q_c is the activation energy for creep, R the universal gas constant, and C' a microstructure and stress dependent constant). For pure aluminum, the stress exponent n is typically about five and Q_c has been identified with the activation energy of self diffusion of Al atoms, equal to 142 kJ/mol [9], which accounts for the relevance of vacancy diffusion in the process of dislocation climb mechanism as the rate controlling process for creep. For the case of Al-Mg alloys, however, n varies in the range of 3-5 [3,10] and values of Q_c in the range of 138-170 kJ/mol have been reported [11]. These differences have been a subject of debate for more than 40 years and as a result, specific “deformations mechanisms”, associated with the interaction between solute atoms of Mg and moving dislocations have been proposed depending on the testing conditions. In summary, it is interpreted that $n=3$ indicates a dislocation viscous glide (solute drag) rate control, whereas $n=5$ indicates a dislocation climb controlled mechanism [12].

However, there is not yet full agreement to explain the transition from $n=3$ to $n=5$. Some authors [13] find that sub-grain formation occurs at high stresses, or H regime, when recovery is rate controlling and the stress exponent is approximately five. A sub-grain size proportional to the inverse of the applied stress is obtained. These sub-grains do not form at medium stresses, M regime, where $n=3$, and dislocation viscous glide is considered to be rate controlling [13]. Other authors, however, defend that the presence or not of sub-grains merely reflects the condition prior to testing rather than that developed during creep [14]. Studies of the low stress interval, L regime, with $n\sim 5$, are hardly ever found in the literature [15].

Despite all the above findings, little progress to understand the “real” strengthening of these alloys with respect to pure Al has been made. To deepen this strengthening, the so called stress reduction method [16] was proposed. The main drawback of this method resides in that it requires some fitting factors which are not based on microstructure related parameters.

The purpose of the present work is to analyze the creep strengthening of Al-Mg alloys with respect pure Al using experimental data reported in the literature. This study has been conducted in the frame of the strength difference method, SDM, proposed by these authors recently [17-19]. This method has been successfully applied to understand the contribution of the load transfer mechanism on the high temperature behavior of discontinuously reinforced metal matrix composites [17]. Also, it was very useful to account for the contribution of the dispersion of Al_2O_3 particles developed during powder metallurgy on the high temperature behavior of 6061Al alloys [18]. Similarly, the SDM will be used here as the base to analyze the contribution of Mg atoms addition in aluminum in the improved creep behavior of solid solution Al-Mg alloys making use of experimental data published in the literature.

2.- Data on the creep of Al-Mg alloys and pure Al

Data of the creep behavior (steady state creep rate as a function of the stress) of Al-Mg alloys and of pure aluminum (between 99.9% and 99.999% purity), both obtained by ingot metallurgy procedures, have been reported in different investigations [20, 21]. Specifically, data of the Al-Mg alloys have been taken from the work of Oikawa *et al* [3]. In these alloy, the Mg contents (in wt.%) are: 0.5, 1.0₂ and 3.0 (wt.%). Data of pure Al have been taken from Straub *et al* [21]. In the plot of Figure 1, the creep data of $\dot{\epsilon}$ vs. σ (in a log-log scale) at 600 K of these three alloys [3] and of pure Al (99.99%) [21] are shown. The analysis of creep data at 600K is conducted here because no published data of the creep of pure Al and pure Al-Mg alloys at other temperature have been found.

The n value for the Al-Mg alloys varies in the range of 3-5. These values are summarized in Table 1 for the different testing conditions. The trend of the strain rate-stress behavior for the three alloys is very similar. At a low stress (L regime) it is $n\sim 5$, in the intermediate stress regime (M regime) $n\sim 3$, and at a high stress (H regime) a value of $n\sim 5$ is again found, irrespective of Mg content. These data have been traditionally analyzed under two different views. Firstly, the data can be fitted for every stage where $n\sim 3$ or $n\sim 5$. As mentioned above, these values are typically associated with a dislocation glide and a dislocation climb controlled creep mechanisms, respectively [22]. Secondly, the whole data range is fitted under a power law relation with a unique n value (average) as it is shown in Table 1. In this case, none of these values correspond to pure dislocation glide controlled or pure dislocation climb controlled mechanisms.

On the other hand, for pure Al, it is generally assumed that $n\sim 5$. It can be seen from the original work of Straub and Blum [21] (see the plot of figure 5 in ref. [21]), however, that a variation of n with temperature exists and that some deviations (change in slope)

at a fixed temperature are also appreciated. A similar behavior is seen in the original data of Servi and Grant [20], also for pure Al, (see their plots of figures 4 and 6).

The creep behavior of pure metals and alloys is, as above mentioned, commonly analyzed in the frame of equations (1) and (2), and specific n values are associated to a given “deformation mechanism”. Such values and mechanisms have been tabulated, for example, in the work of Walser and Sherby [23] (see Table 1 in [23]). The progressive variation of n with temperature and/or stress which reveals the real data, however, is an indication of the complexity of understanding this behavior even in pure metals. According to Table 1 of ref. [23], a combined effect of several mechanisms depending on testing conditions must occur. Furthermore, and most important, little emphasis has been devoted to the real creep strength increase (at any given $\dot{\epsilon}$ and temperature) of the alloy with respect the pure metal, as it is seen from the creep strength difference shown in Figure 1.

3.- Analysis and discussion

From Figure 1 it is seen that the creep resistance (applied stress needed for the alloy to deform plastically at a given $\dot{\epsilon}$) of the three Mg alloys at 600 K is clearly higher than that of pure Al. Using the SDM, applied in the same manner as in previous investigations [17-19], the plot of Figure 2 is obtained. In this plot, the strength increment, $\Delta\sigma$, at any given $\dot{\epsilon}$ is shown for the three Al-Mg alloys as a function of σ . Figure 2a is for the whole range of applied stress and Figure 2b is a detail of the low applied stress regime. As can be seen, $\Delta\sigma$ is proportional to σ . Also, the slope $\Delta\sigma/\sigma$ increases with the amount of Mg atoms, and no clear “transition” of $\Delta\sigma/\sigma$ with σ is appreciated.

The trend shown in figure 2 can be analyzed on the basis of well known relations and existing models. As the starting point, the well known and accepted Orowan’s relation,

can be used. It is known that the presence of solid solution atoms can obstruct dislocation motion during deformation of Al [24]. As a consequence, the dislocation density, ρ , and/or their velocity, \vec{v} , can be modified. Accordingly, it is proposed here that the creep strength increase of solid solutions Al-Mg alloys with respect pure Al is attributed mainly to two contributions:

- 1.- The variation of the dislocation density with respect to pure Al.
- 2.- An additional force between solute atoms and dislocations which modify their velocity.

Assuming constant dislocation density for any given creep testing conditions, Orowan's equation reads, $\dot{\epsilon} = \rho_m b \vec{v}$, where b is the Burgers vector and ρ_m the mobile dislocation density. Therefore, denoting as (ρ_m, v^{Al}) and (ρ_m, v) the above terms for pure Al and the Al-Mg alloys, respectively, it is readily obtained that,

$$\rho_m^{Al} = \rho_m \frac{\vec{v}}{\vec{v}_{Al}} \quad \text{or} \quad \rho_m - \rho_m^{Al} = \rho \left(1 - \frac{\vec{v}}{\vec{v}_{Al}} \right) \quad (3)$$

at any given $\dot{\epsilon}$. This term can be interpreted as the increment of the mobile dislocation density due to the addition of Mg atoms in Al. Using now the Taylor's relationship between the total dislocation density, ρ_{tot} , and creep applied stress, σ , for pure Al (σ_{Al}) and Al-Mg alloys, (σ), we have,

$$\sigma^2 - \sigma_{Al}^2 = (\sigma - \sigma_{Al})(\sigma + \sigma_{Al}) = \sigma^2 \left(1 - \frac{v}{v_{Al}} \right)$$

and rearranging terms,

$$\Delta\sigma \approx \left[\frac{\sigma}{\sigma + \sigma_{Al}} \left(1 - \frac{v}{v_{Al}} \right) \right] \sigma \quad (4)$$

Equation (4) reveals proportionality between $\Delta\sigma$ and σ . This is qualitatively in agreement with the trend obtained experimentally using the SDM shown in figure 2. It

also accounts for the multiplicative effect of applied stress and dislocation velocity on $\Delta\sigma$ and supports the proposed effect of solute atoms on dislocation density/velocity. The presence of solute atoms causes an additional force on dislocations. Consequently, a decrease in their velocity and, hence, an increase of the dislocation density should result. In summary, as dislocations motion becomes more difficult, Frank-Read sources are further activated leading to an increase of the dislocation density and an increase of the creep stress for plastic flow at a given strain rate.

From Taylor's relationship we have a correlation between ρ_{tot} and σ ,

$$\rho_{tot} = \left(\frac{\sigma}{\alpha G b M} \right)^2 \quad (5)$$

Where α is a constant, G is the shear modulus, and M the Taylor factor.

On the other hand, the ratio of mobile to total dislocation density, ρ_m / ρ_{tot} , in Al-Mg alloys has been investigated by Orlova [5] by transmission electron microscopy, TEM, and found that as the applied stress increases ρ_m / ρ_{tot} also increases. The experimental data of Orlova is reproduced in figure 3 together with the fit obtained. This fit follows the expression (dotted line in figure 3):

$$\frac{\rho_m}{\rho_{tot}} = A \sigma^2 + B \sigma + C \quad (6)$$

where A , B , and C are constants (Table II).

Using equations (5) and (6) it is obtained,

$$\rho_m = \left(\frac{1}{\alpha G b M} \right)^2 \sigma^2 (A \sigma^2 + B \sigma + C) \quad (7)$$

At this point, it should be noted that dislocation velocity depends on whether dislocation motion on its gliding plane is a viscous glide controlled or a climb controlled process. On the following both cases will be treated separately.

Glide controlled:

For the case of dislocation viscous glide controlled mechanism, dislocation velocity results from its interaction with the solute atoms: A process of solute drag takes place during dislocation motion. This process has been studied in [25] where it was proposed that the drag force needed to move the dislocation at a given \vec{v} is expressed as [26]:

$$F_{drag} = B_d v = \frac{c_0 \beta^2 \ln(r_2 / r_1)}{\Omega D_{sol} k T} v; \quad \beta = \frac{G b^4}{3} \epsilon_a \quad (8)$$

where c_0 is the solute concentration at an infinite distance from the dislocation, β is the size interaction parameter, ϵ_a is the misfit factor, r_2 and r_1 the outer and inner cut-off radii, D_{sol} the diffusion coefficient of the solute into the aluminum matrix, Ω the atomic volume, k the Boltzmann's constant and G the Shear Modulus. If we consider that the resulting force on the dislocation, F , is due to σ , \vec{v} is described as:

$$\vec{v} = \frac{F}{B_d} = \frac{b \sigma}{2 B_d} \quad (9)$$

Therefore, substituting equations (7), and (9) in the Orowan's relationship, it is found that when dislocation motion is viscous glide controlled, the strain rate, $\dot{\epsilon}_{glide}^{sd}$, is expressed as:

$$\dot{\epsilon}_{glide}^{sd} = \frac{\Omega k T}{2 c_0 \ln(r_2 / r_1) (\alpha G \beta M)^2} \sigma^3 (A \sigma^2 + B \sigma + C) D_{sol} \quad (10)$$

Climb controlled:

According to the Takeuchi & Argon's model [26] the solute drag force for dislocation climb is also given by equation (8). During a dislocation climb process, however, an extra force is needed to move the dislocation away from the gliding plane. This force arises from the vacancy motion (vacancy gradients) around climbing dislocations [27].

This extra force is given by the expression:

$$F_{c\text{limb}} = B_c \nu = \frac{b^2 k T \ln(c^* / c^*_0)}{2\pi\Omega D_L} \nu \quad (11)$$

where c^* is the vacancy concentration around the dislocation, c^*_0 is the vacancy concentration at an infinite distance from the dislocation, and D_L is the lattice self diffusion coefficient in aluminum. Therefore, the force needed to move the dislocation on its gliding plane at $\vec{\nu}$ is:

$$F = F_{drag} + F_{c\text{limb}} = (B_d + B_c) \nu$$

and the dislocation velocity resulting from an externally applied stress is described by:

$$\nu = \frac{b\sigma}{2(B_d + B_c)} \quad (12)$$

Substituting now equations (7) and (12) in the Orowan's relationship and taking into account equations (8) and (11), it is found that when dislocation motion (on its gliding plane) is a climb controlled process, the strain rate, $\dot{\epsilon}_{c\text{limb}}^{sd}$, is expressed as,

$$\dot{\epsilon}_{c\text{limb}}^{sd} = \frac{\pi\Omega k T}{(\alpha G M)^2} \sigma^3 (A \sigma^2 + B \sigma + C) \left[\frac{D_{sol} D_L}{2\pi c_0 \beta^2 \ln(r_2 / r_1) D_L + (bkT)^2 \ln(c / c^*_0) D_{sol}} \right] \quad (13).$$

Substituting values from the literature, Table III, it is found that $2\pi c_0 \beta^2 \ln(r_2 / r_1) D_L \ll \ll (bkT)^2 \ln(c / c^*_0) D_{sol}$ and equation (13) is reduced to:

$$\dot{\epsilon}_{climb}^{sd} = \frac{\pi \Omega}{(\alpha G b M)^2 k T \ln(c / c^*_0)} \sigma^3 (A \sigma^2 + B \sigma + C) D_L \quad (14)$$

If $\dot{\epsilon}_{glide}^{sd}$ and $\dot{\epsilon}_{climb}^{sd}$ are compared (equations (10) and (14)) at 600K, it is found that $\dot{\epsilon}_{climb}^{sd} / \dot{\epsilon}_{glide}^{sd} = 0.24$, see Table II. Creep obviously occurs by dislocation motion involving viscous glide and climb processes, both taking place in an inter-dependent manner. Therefore, creep of the Al-1%Mg must be described by equation (14), which accounts for the creep rate involving the slowest process.

The prediction of this new model is summarized in figure 4, where comparison with the experimental data [3] and the prediction derived from Takeuchi & Argon's model [26] is made. It is remarkable the very good predictive capability of the new model.

This result is also consistent with the assumption that self diffusion of Al atoms (though vacancy migration) is the underlying mechanism that controls creep of these Al-Mg alloy, as revealed by the plot of figure 5. In this figure, the diffusion coefficient compensated strain rate as a function of Young's modulus compensated stress in a double logarithmic scale is shown for pure Al-1%Mg alloy (from data of ref. [11]) at different temperatures (570, 600 and 640 K). As can be seen, an excellent correlation is obtained when D_L is used. The correlation, however, diverges when D_{sol} is used to compensate $\dot{\epsilon}$. For comparison, data of pure Al (from [20]) is also included.

Furthermore, it is worth mentioning, that equation (14) describes, the variation on stress exponent, n , between 3 and 5 found experimentally. This change in "mechanism" is, from the point of view of the present analysis, a consequence of the variation of

dislocation density ratio (total to mobile) with applied stress, as obtained experimentally by Orlová [5], figure 3 and equation (7). Unfortunately, data at other temperatures have not been found in the literature to extend the present analysis.

It is finally noteworthy that the term c_0 does not appear in equation (14). This suggests, in principle, that the same strain rate would be obtained regardless the amount of Mg atoms in the alloy. This term is, again, manifested through the stress term, appearing from the variation of the dislocation density values reported by Orlová [5] and the fit revealed by equation (7). Other stress values (or fitting parameters of equation (7)) would result at other temperatures or in alloys with different amount of Mg atoms. As above mentioned, no data in the literature is reported to give further support to this model. The dependence of dislocation density with stress shown in figure 3 is at present difficult to understand and needs of further investigation.

The present unified view of the creep behavior of Al-1%Mg solid solution alloy is not in contradiction with the microstructures reported in the $n \approx 5$ and $n \approx 3$ regions by different authors; namely, a sub-grains structure or a random array of dislocations, respectively [5,13,14]. The model is irrespective of the resulting structure developed with strain as it only consider the total to mobile dislocation density ratio; only dislocation motion, obstructed by the presence of Mg atoms, explains creep deformation and creep strengthening. This view is, somewhat, in agreement with Horiuchi and Otsuka's [14] analysis of the creep of Al-Mg solid solution alloys, who conclude that the high temperature deformation of solid solution alloys is not necessarily controlled by the viscous motion of dislocations regardless that a value of $n=3$ is obtained.

4.- Summary

A thorough analysis has been made using the creep data of pure Al, pure Al-Mg alloys and of Al-Mg alloys with varying dislocation densities due to different applied stresses available in the literature. The Strength Difference Method, SDM, which permits obtaining the experimental creep strengthening of Al-Mg alloy with respect to pure Al, has been used. The SDM reveals proportionality between the strength increment and the applied stress. The trend found gives support to the model proposed, which is based on the change of dislocation density and dislocation velocity. The model makes use of the equation derived by Takeuchi and Argon [26] for dislocation velocity in a solute cloud. It takes also into account the solute drag and climb forces for dislocation motion on its gliding plane. For the latter, the local force arising from vacancy motion to move the dislocation away from its gliding plane is considered. As a result, a very reasonable correlation with experimental data of the creep behavior of pure Al-1wt%Mg alloy at 600 K is obtained. The change in n obtained experimentally is associated with a variation of the mobile to total dislocation density ratio with applied stress, rather than to a change in deformation mechanism (dislocation glide controlled to dislocation climb controlled). The relevance of the solute drag force for dislocation motion, however, does not undermine the fact that aluminum lattice self diffusion, through vacancy motion (vacancy gradients), is the mechanisms that controls the creep behavior of Al-Mg alloys.

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Material	n(L)	n(M)	n(H)	n(average)
Pure Al [21]				4.9
Al-0,5%Mg [3]	4.7	3.8	-	3.9
Al-1%Mg [3]	4.9	3.3	4.2	3.8
Al-3%Mg [3]	4.6	3.2	-	3.3

Table I. Stress exponent values, n, in the power law creep for the materials investigated. For the Al-Mg alloys, n values corresponding to the L, M and H stress regimes are compared to the average value obtained for the whole range of applied stress.

Parameter	Value (Units)
<i>A</i>	-0.0011
<i>B</i>	0.043
<i>C</i>	-0.042
α	0.3
<i>G</i>	21500 (MPa)
<i>b</i>	$2.86 \cdot 10^{-10}$ (m)
<i>M</i>	3.06
<i>c</i> ₀	0.005/0.01/0.03 (%)
$\ln(r_2/r_1)$	6.9-9.2
$\ln(c^*/c_o^*)$	10^{-1} - 10^{-2}
ϵ_a	0.0232
β^2	$1.2 \cdot 10^{-30}$
Ω	$1.2 \cdot 10^{-29}$ (m ³)
<i>D</i> _{sol}	$6.2 \cdot 10^{-6} e^{(-115000/RT)}$ (m ² s ⁻¹)
<i>D</i> _L	$1.7 \cdot 10^{-4} e^{(-142000/RT)}$ (m ² s ⁻¹)
<i>R</i>	8.314 (J/K mol)
<i>k</i>	$1.38 \cdot 10^{-23}$ (J/K)
<i>T</i>	600(K)

Table II. Parameters used to calculate the strain rate using equations (10), (13) and (14).

Figure captions:

Figure 1. Minimum creep rate as a function of the applied stress at 600K of Al-Mg alloys [3]. The creep data of pure aluminum [21] at 600K are also shown. Stress exponent for each interval of applied stress is indicated in parenthesis.

Figure 2. a) Creep strength increment of the Al-Mg alloys with respect to pure aluminum. b) Detail of the low stress range indicated by discontinuous square in figure 2a). The strength increment has been calculated using the Stress Difference Method.

Figure 3. Experimental data, measured by TEM [5], of the ratio between mobile and total dislocation densities for the Al-1%Mg alloy. Dotted line is the resulting polynomial fit (fitting values are reported in Table II).

Figure 4. Strain rate prediction from equation (14) and experimental values from figure 1 for the Al-1%Mg alloy. For comparison, the prediction obtained with the Argon-Takeuchi's model is also shown.

Figure 5. Creep strain rate normalized by the aluminum self diffusion coefficient vs. applied stress normalized by the Young's Modulus. For comparison creep strain rate normalized by the solute (Mg) diffusion coefficient are shown.

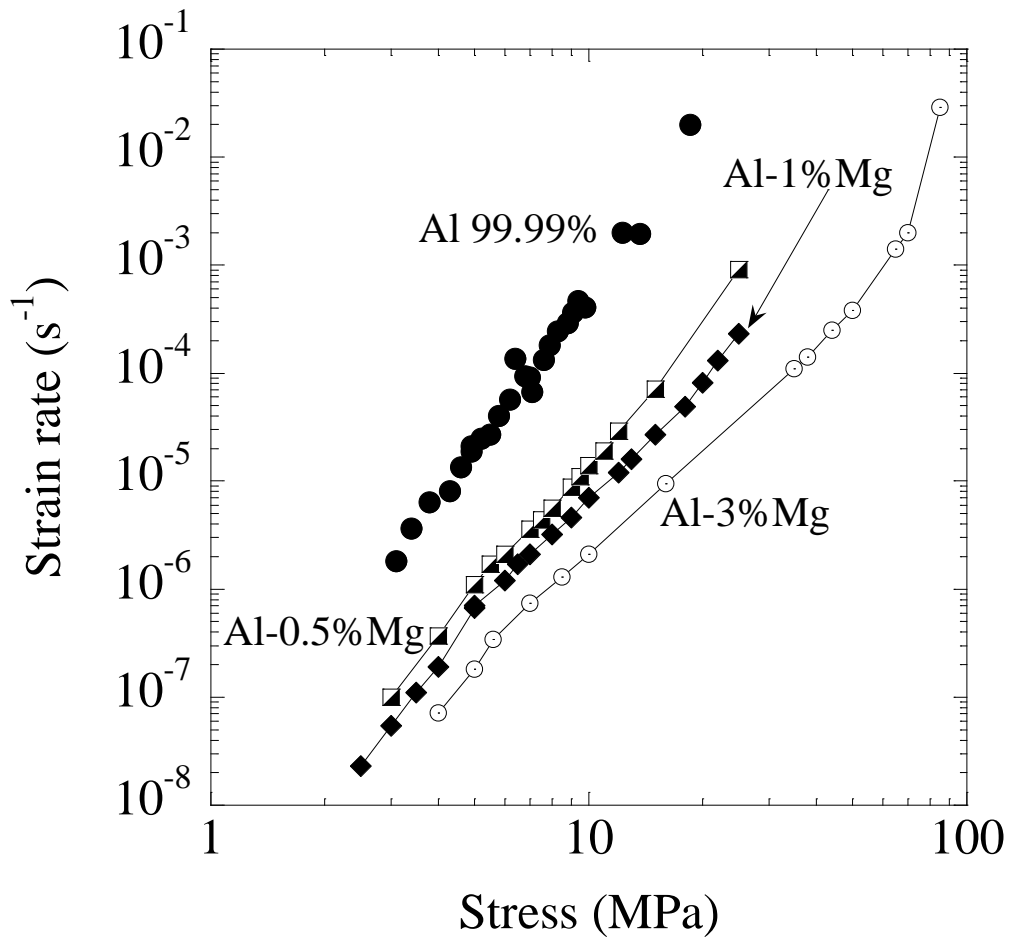


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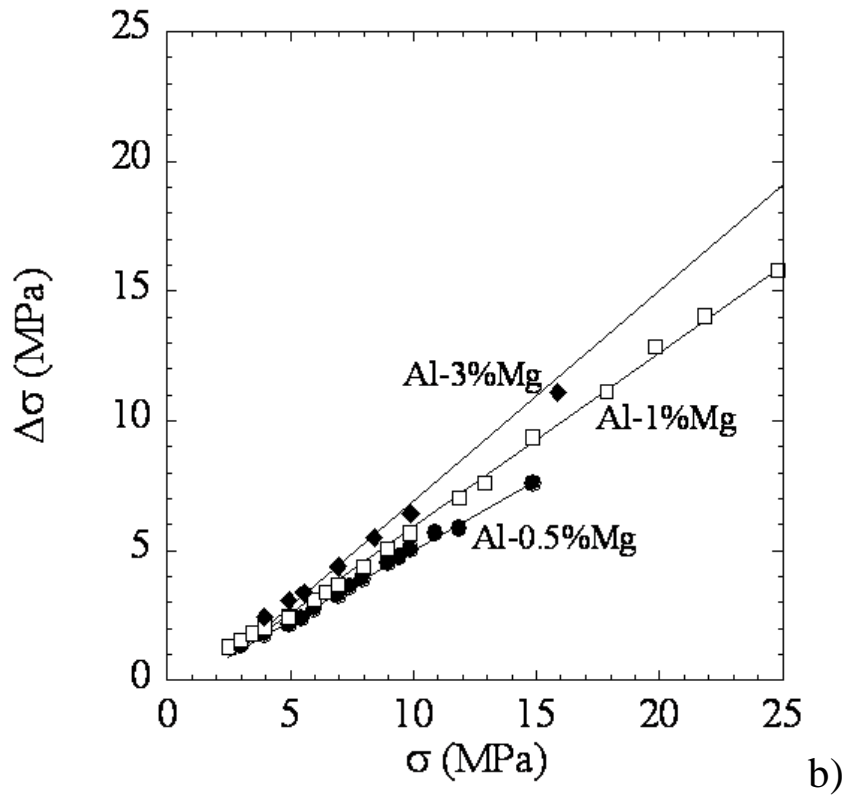
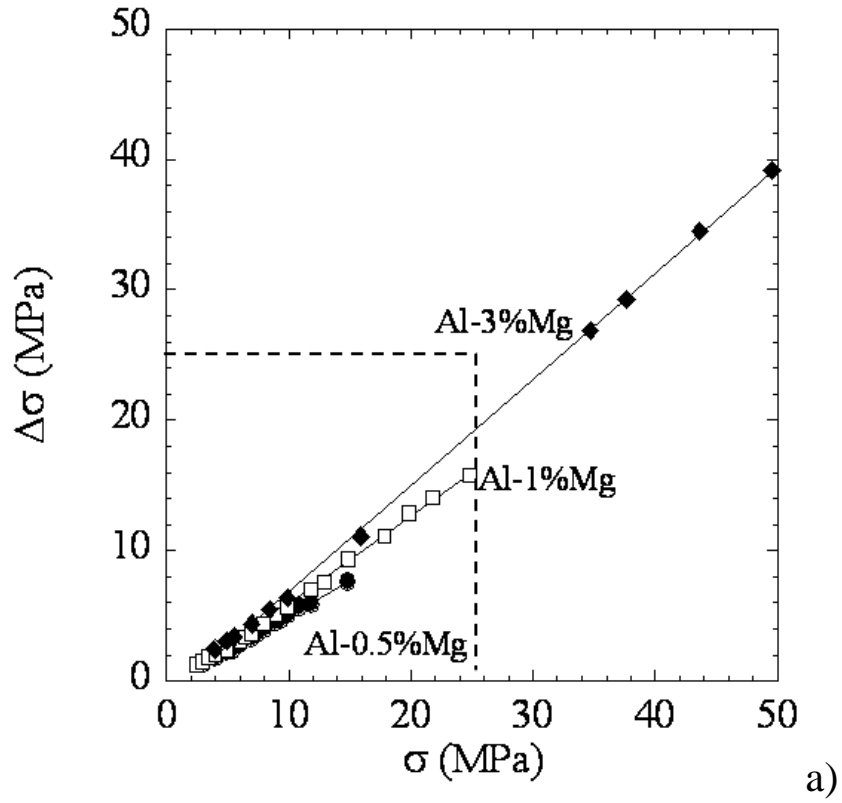


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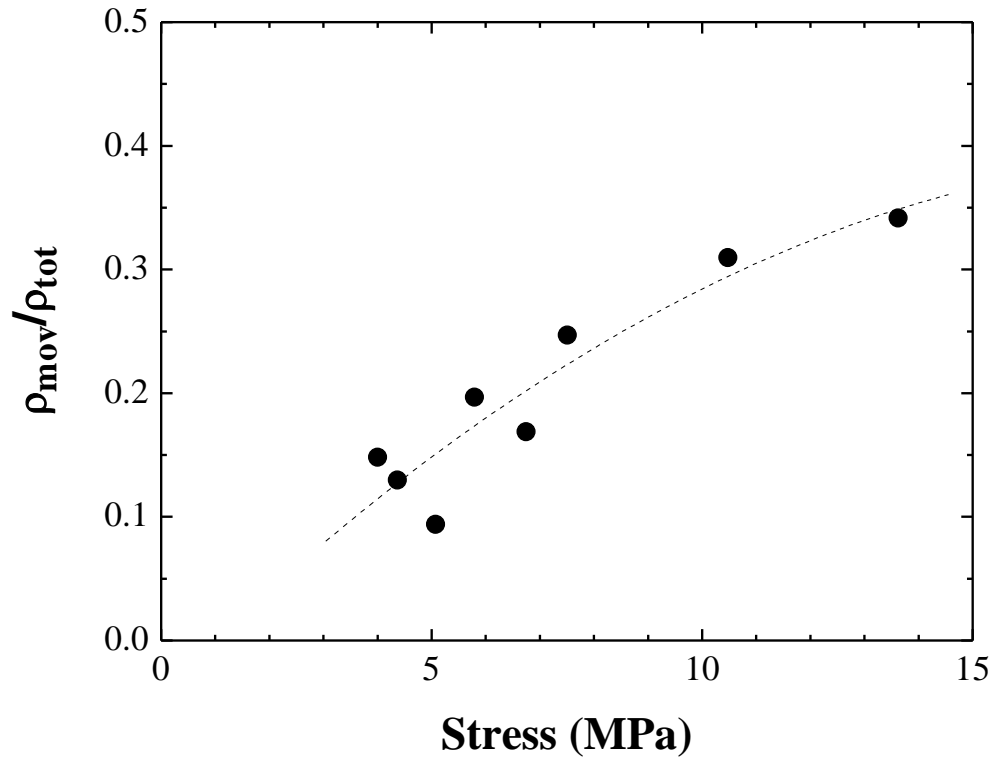


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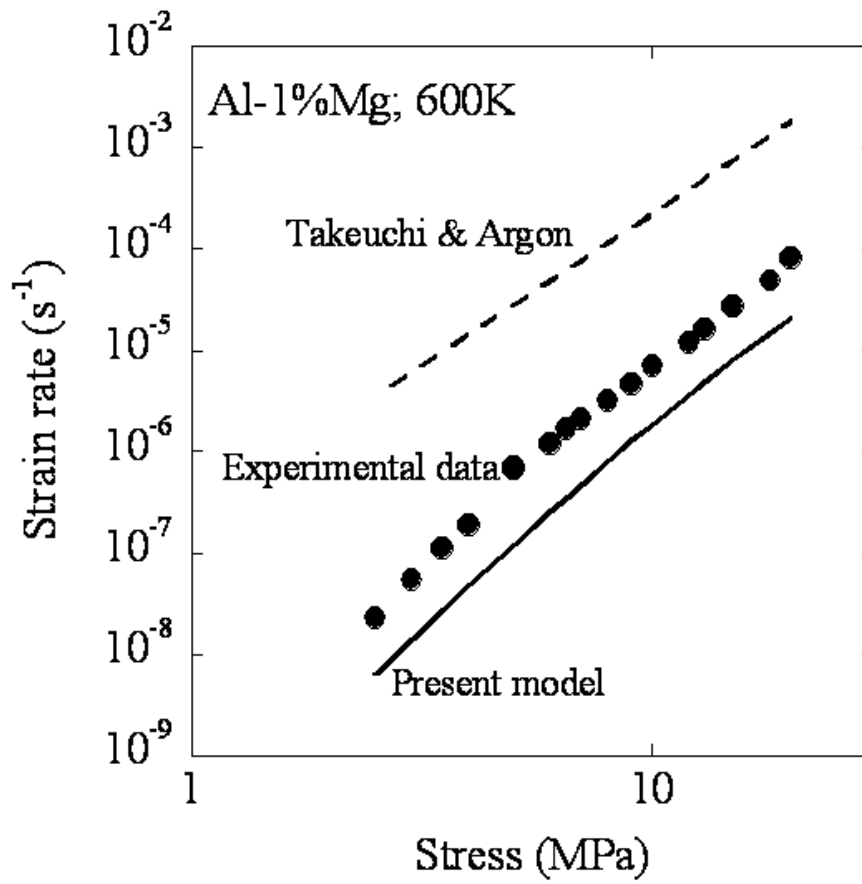


Figure 4. Strain rate prediction from equation (14) and experimental values from figure 1 for the Al-1%Mg alloy. For comparison, the prediction obtained with the Argon-Takeuchi's model is also shown.

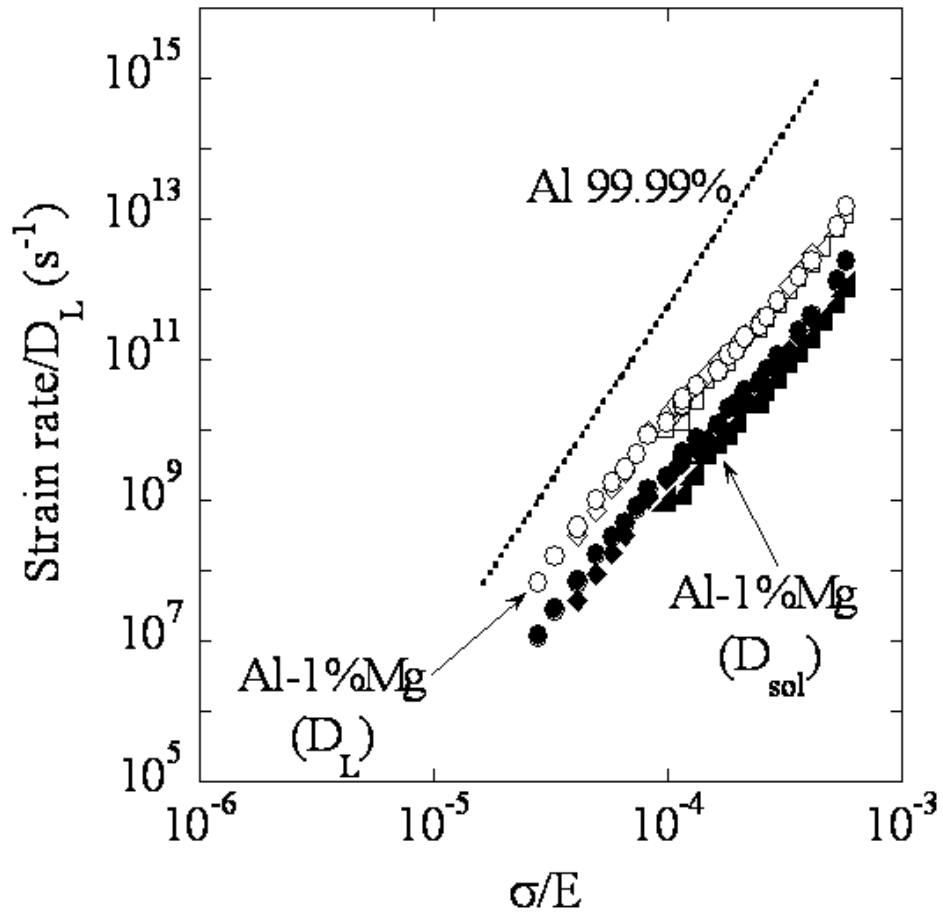


Figure 5. Creep strain rate normalized by the aluminum self diffusion coefficient vs. applied stress normalized by the Young's Modulus. For comparison creep strain rate normalized by the solute (Mg) diffusion coefficient are shown.