



## Thermally-activated flow in nominally binary Al–Mg alloys



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### ABSTRACT

The temperature-dependent flow behavior in nominally binary Al–Mg alloys measured recently (Jobba et al., 2015; Niewczas et al., 2015) is interpreted in the context of a parameter-free solute strengthening model. The recent measurements show consistently higher strengths as compared to literature data on true binary Al–Mg alloys, which is attributed to the presence of Fe in the nominally binary Al–Mg. Using the Fe concentration as a single fitting parameter, the model predictions for the newer materials when treated as Al–Mg–(Fe) alloys agree with experiments to the same degree as obtained for the true binary Al–Mg. The model then predicts the activation volume in good agreement with experimental trends.

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In interpreting experimental data for strengths of Al alloys, multiple mechanisms operating simultaneously are often invoked because the application of simple or ad hoc theories does not explain observed trends. For instance, Hall–Petch effects [2], anomalous athermal stresses [4], solute clustering [4,5], and/or unphysical dislocation/solute interactions [6–8], have been invoked to justify deviations between various solute strengthening theories [9,10,6–8] and experimental data. However, such reasonable attempts to rationalize experimental data obfuscate the relevant underlying mechanisms. Here we re-examine recent data on a set of nominally binary Al–Mg alloys first reported by Jobba et al. [1] and then further analyzed by Niewczas et al. [2]. We show that the finite-temperature flow behavior of the alloys in these works can be explained by the inclusion of a low concentration of Fe without the need to invoke any other additional mechanisms.

The solute strengthening model was developed in Refs. [11,12] and only key points are summarized here. When moving through a random field of solutes with concentration  $c$ , an initially straight dislocation can lower its energy by bowing into regions containing favorable solute configurations and bowing away from regions with unfavorable solute configurations. The segments thus reside in favorable solute configurations and require stress- and thermally-driven activation to escape and move to the next favorable environment. The characteristic energy barrier  $\Delta E_b$  for pinned segments is

$$\Delta E_b = 1.22 \left( \frac{w_c^2 \Gamma \Delta \tilde{E}_p^2(w_c)}{b} \right)^{\frac{1}{3}} c^{\frac{1}{3}}, \quad (1)$$

where  $\Gamma$  is the line tension of the dislocation and  $b$  is the Burgers vector magnitude. The quantity  $\Delta \tilde{E}_p(w_c)$  is related to the standard deviation of the overall solute/dislocation interactions as the dislocation segment moves a distance  $w_c$  through the random solute field of concentration  $c$ , and  $w_c$  is the characteristic roughening amplitude emerging from the theory. The characteristic stress required to move the dislocation segment over the energy barrier at zero temperature is

$$\tau_{y0} = 1.01 \left( \frac{\Delta \tilde{E}_p^4(w_c)}{\Gamma b^5 w_c^5} \right)^{\frac{1}{3}} c^{\frac{2}{3}}. \quad (2)$$

If  $w_c$  is the same for different solutes, then the energy barrier and zero temperature flow stress for the alloy with  $q$  different solute types in solution is

$$\Delta E_b = \left[ \sum_q (\Delta E_b^q)^3 \right]^{\frac{1}{3}}, \quad \tau_{y0} = \left[ \sum_q (\tau_{y0}^q)^3 \right]^{\frac{1}{3}}, \quad (3)$$

where the superscript  $q$  is a quantity for solute  $q$ . For Al,  $w_c$  is nearly independent of solute type and so Eq. (3) can be applied to binary Al–Mg and nominally binary Al–Mg–(Fe) [12].

All quantities in Eqs. (1) and (2) are derived or material properties. First-principles calculations provide the key solute/dislocation interaction energies needed in the theory. The coefficients of the concentration scaling for the energy barrier and

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zero-temperature yield stress in Eqs. (1) and (2) are shown in Table 1 for Mg and Mn. In these results, the dislocation line tension  $\Gamma$  for Al is taken as 0.25 eV/Å, as derived from atomistic studies [13]; this value is lower than used in Ref. [12] but the same as that used in Ref. [11]. Also shown are the coefficients for Fe, which cannot be predicted by the theory and are instead obtained from experimental flow stress measurements on binary Al–Fe by Diak and Saimoto [14].

At finite temperatures, dislocation motion is thermally activated. At high stresses/low temperatures, the stress-dependent energy barrier  $\Delta E(\tau)$  is

$$\Delta E(\tau) = \Delta E_b \left[ 1 - \frac{\tau}{\tau_{y0}} \right]^{3/2} \quad (4)$$

At low stress or high temperatures, the energy barrier scales logarithmically with the applied stress [8,15],

$$\Delta E(\tau) = 0.51 \Delta E_b \ln \left( \frac{\tau_{y0}}{\tau} \right). \quad (5)$$

At quasistatic loading rates, transition state theory [16,17] then connects the energy barrier  $\Delta E(\tau)$  to the strain rate  $\dot{\epsilon}$  and temperature  $T$  as

$$\dot{\epsilon}(\tau, T) = \dot{\epsilon}_0 \exp \left( -\frac{\Delta E(\tau)}{kT} \right), \quad (6)$$

where  $k$  is the Boltzmann constant,  $\dot{\epsilon}_0 = \rho b d v_0$ ,  $\rho$  is the dislocation density per unit area,  $d$  is the flight distance over which the dislocation moves after each escape and  $v_0$  is the attempt frequency. Eq. (6) can be inverted to yield the finite temperature flow stress  $\tau_y$  for a given strain rate  $\dot{\epsilon}$  as

$$\tau_y(\dot{\epsilon}, T) = \begin{cases} \tau_{y0} \left[ 1 - \left( \frac{kT}{\Delta E_b} \ln \frac{\dot{\epsilon}_0}{\dot{\epsilon}} \right)^{2/3} \right] & \frac{\tau_y}{\tau_{y0}} \geq 0.5 \\ \tau_{y0} \exp \left( -\frac{1}{0.51} \frac{kT}{\Delta E_b} \ln \frac{\dot{\epsilon}_0}{\dot{\epsilon}} \right) & \frac{\tau_y}{\tau_{y0}} < 0.5 \end{cases} \quad (7)$$

The high and low stress (or low and high temperature) expressions above agree very well over the intermediate range of stress  $0.3 < \tau/\tau_{y0} < 0.6$  [12]. Here, as in all previous applications of this solute strengthening model, we use  $\dot{\epsilon}_0 = 10^4 \text{ s}^{-1}$  [12,15,18]; the precise value has a small effect on the predictions.

A measurable quantity of importance in thermally-activated flow is the apparent activation volume  $V(\dot{\epsilon}, \tau)$ , defined as

$$V(\dot{\epsilon}, T) = -\frac{\partial \Delta E(\tau)}{\partial \tau}. \quad (9)$$

The activation volume is related to the area swept by the dislocation during the thermal activation process, and so is a sensitive measure of the length scales involved in the actual activation process. In the low-temperature regime,

$$V(\dot{\epsilon}, T) = \frac{3}{2} \frac{\Delta E_b}{\tau_{y0}} \left( \frac{kT}{\Delta E_b} \ln \frac{\dot{\epsilon}_0}{\dot{\epsilon}} \right)^{1/3}. \quad (10)$$

and the theory satisfies the “stress equivalency principle” [19].

**Table 1**

Computed  $T = 0$  yield stress  $\tau_{y0}$  and energy barrier  $\Delta E_b$  for Mg, Mn, and Fe solutes in Al, normalized by the appropriate solute concentration factor. Parameters in bold for Fe solutes are back-calculated from experimental data [14]. Note that these parameters differ from Ref. [12] due to the use of a more accurate line tension.

Solute	$\tau_{y0}/c^{2/3}$ (MPa)	$\Delta E_b/c^{1/3}$ (eV)
Mg	427	3.25
Mn	807	6.62
Fe	15,047	25.34

We first demonstrate the quantitative predictions of the model by comparing to measured flow stresses in Al–Mg and Al–Mn binary alloys [14,5,20,21]. Diak et al. have verified negligible concentrations of other elements, and in particular the Fe concentrations are below 3 ppm [14]. The single-crystals Al–Mg materials of Asada et al. were fabricated from 99.999% and so are also expected to have negligible Fe. The critically resolved yield stresses of single crystal Al alloys containing 1.1% and 3.3% Mg were measured by Asada et al. at a strain rate  $\dot{\epsilon} = 4.2 \times 10^{-4} \text{ s}^{-1}$  and various temperatures; we consider measurements between 78 K and 300 K to avoid other high-T phenomena (e.g. dynamic strain aging [3] and solute drag [22–24]). Diak et al. measured the flow stresses  $\sigma_y$  in uniaxial tension of two polycrystalline binary Al–Mg alloys at 78 K [14,5] and of three binary Al–Mn alloys at 78–263 K [21], with  $\dot{\epsilon} = 5 \times 10^{-5} \text{ s}^{-1}$ . In making predictions, we convert critical resolved shear stresses  $\tau_y$  to equivalent uniaxial yield stresses  $\sigma_y$  by multiplying  $\tau_y$  by the Taylor factor  $M = 3.06$  [25] to put all results on the same footing.

The experimentally-measured yield stresses are shown versus the predicted yield stresses in Fig. 1(a) for the temperature range 78 K–300 K. The model predictions agree well with experiments for the polycrystalline materials, and show a moderate overprediction for the single-crystal materials at lower temperatures. All predictions are within 30% with most predictions being within 15%. The deviations suggest that the model might overestimate the zero temperature yield stress  $\tau_{y0}$  while underestimating the energy barrier  $\Delta E_b$ . However, the model’s parameters were derived from first-principles and are not adjustable. The polycrystalline results differ from earlier results [12] due to the use of the line tension derived from atomistic simulations [13], and this value of  $\Gamma$  actually makes the predictions slightly worse relative to the experiments.

In more recent studies, Jobba et al. have measured the yield stress of various Al–Mg alloys with concentrations ranging from 0.5% to 4.11% at 4 K, 78 K and 298 K at a strain rate  $\dot{\epsilon} = 1.6 \times 10^{-4}$  [1,2]. We make comparisons between theory and experiments at 78 K and 298 K since dynamic effects occur at temperatures near 0 K [16]. These measured yield stresses are much larger than those found by Diak et al. or Asada et al. Similarly, the measured yield stresses are generally larger than those predicted by the model, especially when  $\tau_y$  is small, as seen in Fig. 1(b). Jobba et al. also report rather high yield stresses for nominally pure Al ( $\sim 12$ –18 MPa). The larger strengths measured by Jobba et al. and Niewacz et al. would be inconsistent with a reduction in Mg in solution relative to the measured overall content, so here we use the stated Mg content as equal to the Mg concentration in solution. The similarity in measured strength between the  $\sim 3\%$  Mg alloy and  $\sim 4\%$  Mg alloy suggests, however, that the Mg in solution in the  $\sim 4\%$  Mg alloy could in fact be closer to  $\sim 3\%$ . The discrepancy between the previous literature results, which agree reasonably with the theory, and those reported in Refs. [2,1], which are rather larger, can be rationalized by the presence of dilute Fe solutes in the nominal binary Al–Mg alloys of Jobba et al. They reported that the as-fabricated alloys contain  $\sim 6 \times 10^{-4}$  Fe solute, of which an unknown amount is in solid solution. Here, we assume that there is some common concentration  $c_{\text{Fe}}$  of Fe in all of the materials studied by Jobba et al. and use  $c_{\text{Fe}}$  as a single fitting parameter for the yield stress.

For alloys containing both Mg and Fe solutes, the 0 K yield stress  $\tau_{y0}^{\text{eff}}$  and energy barrier  $\Delta E_b^{\text{eff}}$  are obtained from the alloy law of Eq. (3). We obtain  $c_{\text{Fe}}$  by minimizing the total relative difference between theory and experiment given by over all experimental data points at both 78 K and 298 K. This yields an Fe concentration of  $c_{\text{Fe}}$  of 20 ppm and predictions of the model using this  $c_{\text{Fe}}$  for all the Al–Mg–(Fe) alloys are shown in Fig. 1(c). The fitted value of

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