



# Cluster strengthening in aluminium alloys

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We consider the elastic effect of clusters and propose a size misfit strengthening model. Our approach is similar to solute strengthening, where the size misfit of clusters is assumed to be the sum of elementary atomic misfits. The proposed model is compared to the coherency strengthening model, and the results indicate that our model is applicable to clusters  $\sim < 1$  nm in diameter. The prediction of the model is reasonably consistent with the reported experiments of aluminium alloys.

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Solute clustering is observed widely in many Al alloys [1–6], and contributes to the strength of alloys, even dilute alloys [7,8]. Cluster hardening was originally proposed in research on aged Al–Cu–Mg alloys [2,9,10], which concluded that the rapid early hardening during ageing is due to solute clustering rather than Guinier–Preston–Bagaryatsky zone formation. Recent application of atom-probe tomography (APT) provides more quantitative evidence of solute clustering related to hardening [4,6]. Ringer et al. qualitatively interpreted cluster hardening as “an exaggerated form of solid solution strengthening”, i.e. arising from solute–dislocation interaction involving a chemical mechanism [2,11]. Recently, researchers have attempted to quantitatively explain the hardening effect of clusters. Areal glide simulation [12] suggests that the spatial distribution of weak obstacles (e.g. solutes) does not affect the strength. The strengthening effect of clusters observed experimentally must be due to chemical and/or elastic effects, which change the obstacle strength. A simplified model—originally proposed for precipitates (or Guinier–Preston (GP) zones) [13]—has been adapted for modulus hardening of co-clusters (containing various elements) [4]. It is applicable to large clusters but oversimplified for small ones. An analytical model for cluster strengthening due to short-range order has been proposed for solute dimers in Al–Cu–Mg and Al–Mg–Si [4,5]. The order

energy was estimated by differential scanning calorimetry (DSC). Marceau et al. [14] characterized the size distribution of clusters and estimated the contribution of cluster strengthening by using an areal glide model, where the strength of each cluster is assumed to be proportional to its radius on the glide plane. Proville et al. [15] simulated the interactions between solute pairs and the Shockley partial dislocations in the core region using molecular dynamics, and proposed a hardening model for Ni–Al, which has a lower stacking fault energy than the Al alloys considered here.

Previous theoretical investigations are often limited to solute dimers or large clusters, and the elastic effect has not been considered. In the present work, a model considering the elastic effect of small clusters is proposed to estimate the contribution of clusters to the yield stress.

A misfit strain field arises due to the size misfit of solute in matrix, which is the main strengthening effect of solutes. Clusters would cause a similar strain field as solutes. An approach similar to solute strengthening theory is proposed to estimate the strength contribution due to the size misfit of clusters. According to elasticity theory, the interaction energy of a substitutional atom with an edge dislocation is [16]:

$$\Delta E = 3 \frac{1 - \nu}{1 + \nu} \sigma_h \Delta V. \quad (1)$$

The parameter  $\sigma_h$  is the hydrostatic stress induced by an edge dislocation,  $\nu$  is the Poisson's ratio of Al, and  $\Delta V$  is the volume misfit of a solute, i.e. the volume changed by the substitution for a host atom.  $\Delta V = 3\Omega\delta$ ,

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where  $\Omega$  is the atomic volume of the host lattice, and  $\delta$  is the size misfit parameter (or lattice misfit). The interaction force on the dislocation in the glide direction  $y$  is:

$$f = -\frac{d\Delta E}{dy} = -3\frac{1-\nu}{1+\nu}\Delta V\frac{d\sigma_h}{dy}. \quad (2)$$

The maximum interaction force between a dislocation and a solute atom,  $f_m$ , is achieved at a certain relative position of the solute and the dislocation. An approximate equation of  $f_m$  is given by  $|f_m/2T_L| \approx 0.181\delta$  [17], if the contribution from modulus misfit is neglected.  $T_L$  is the line tension of an edge dislocation, assumed to be constant here ( $T_L = Gb^2/2$ , where  $G$  is the shear modulus of the host lattice and  $b$  is the Burgers vector). Solid-solution hardening is usually discussed in terms of either Fleischer–Friedel theory or Mott–Labusch theory. If clusters are treated as point obstacles and exist in a low density, the Fleischer–Friedel model can be used. The increment of the critical shear stress in this model,  $\Delta\tau_F$ , is [16]:

$$\Delta\tau_F = (2T_L)^{-1/2}b^{-2}f_m^{3/2}c_p^{1/2}, \quad (3)$$

where  $c_p$  is the planar concentration of obstacles, which is proportional to the total solute concentration  $c_0$ . Thus, Eq. (3) has only two variables, i.e.  $f_m$  and  $c_0$ , and  $f_m$  is proportional to  $\Delta V$ , since  $\Delta V$  is a constant as deduced from Eqs. (1) and (2). Eq. (3) can be simplified as

$$\begin{aligned} \Delta\tau_F &= Af_m^{3/2}c_0^{1/2} = A_1\Delta V^{3/2}c_0^{1/2} \text{ or } \Delta\tau_F \\ &= 0.12b(G\delta)^{3/2}(c_0/2T_L)^{1/2}, \end{aligned} \quad (4)$$

where  $A$  and  $A_1$  are constants. The equations above are valid for solute strengthening at 0 K, and will be adapted for clusters in the present work. At first, the monodispersion of clusters is assumed for simplicity, i.e. each cluster is identical in composition and size, containing the same number of atoms,  $i$ . The polydispersion of clusters will be discussed later. The small clusters are treated as point obstacles similar to mono atoms, since the size of a small cluster is of the same order as the Burgers vector. The volume misfit of a small cluster is assumed to be approximately the sum of misfits of all the atoms in the cluster, i.e.  $\Delta V_{cl} \approx \sum_i \Delta V_i$ . If the cluster contains like solute atoms, the volume misfit of a small cluster  $i$  is approximately proportional to the number of atoms  $i$ , i.e.  $\Delta V_{cl} \approx i \cdot \Delta V$ . Thus, the maximum resistive force of an  $i$ -cluster is approximately:

$$f_{cl,m} \approx i \cdot f_m. \quad (5)$$

The concentration  $c_0$  in Eq. (4) is replaced by the cluster concentration, which equals  $c_0/i$ , when all the solutes form  $i$ -clusters. Eqs. (4) and (5) give:

$$\Delta\tau_{cl,F} = Af_m^{3/2}c_0^{1/2}i = i\Delta\tau_F. \quad (6)$$

Eq. (6) suggests that the strengthening effect increases linearly with the number of atoms per cluster, as a result of the relative weight of the powers of  $f_m$  and  $c_0$  in Eq. (3).

The stress contribution will be reduced significantly at elevated temperatures due to the atomic thermal motion. The temperature dependence can be described by [18]:

$$\Delta\tau(T) = \Delta\tau_0 \exp\left(-\frac{kT}{0.51E_b} \ln \frac{\dot{\epsilon}_0}{\dot{\epsilon}}\right), \quad E_b \approx A_2\Delta V^{2/3}c^{1/3}, \quad (7)$$

where  $\Delta\tau_0$  is the increase in critical shear stress at 0 K,  $E_b$  is the energy barrier, and  $A_2$  and  $\dot{\epsilon}_0$  are constants. The thermal activation energy is adapted for clusters as:

$$E_{cl,b} \approx A_2\Delta V_i^{2/3}c_i^{1/3} = i^{1/3}E_b. \quad (8)$$

The increase in cluster size increases the thermal activation energy, leading to reduced thermal effect and more retained yield stress at elevated temperatures.

The strengthening of polydisperse clusters can be calculated in a similar approach as for monodisperse clusters, using the superposition law for multicomponent hardening. The concentration  $c_i$  is defined as the atomic concentration of solute atoms that enter each  $i$ -cluster, including the monatomic solutes  $i = 1$ . The concentration of  $i$ -clusters then equals  $c_i/i$ . The total solute atomic concentration, including the atoms in the small clusters, is the sum of the solute atoms in all the clusters  $c_i$ , i.e.  $c_0 = \sum_i c_i$ ,  $i = 1, 2, 3, \dots$ . The cluster distribution can be measured using atom probe tomography [14]. For the time being, the distribution of atoms in clusters,  $c_i$ , is simply assumed to fit a Poisson distribution. The mean value of  $i$  determines the Poisson distribution of  $c_i$ . The superposition law is widely described as  $\Delta\tau_{cl}^q = \sum_i \Delta\tau_i^q$ ,  $1 \leq q \leq 2$  [19]. This equation gives the upper and lower limits of superposition when  $q = 1$  and  $q = 2$  respectively. Labusch [20] obtained  $q = 1.5$  for obstacles of the same number density but different strengths [21], which is often used in literature [22]. Recently, the superposition law has been re-examined by 2-D areal glide simulations [23], which indicates that  $q = 1.5$ – $2$  could fit most of the cases. The ratio of cluster strengthening to monatomic solute strengthening is plotted as a function of the mean number of atoms per cluster in Figure 1 with  $q = 1.5$  and  $2$ . The strengthening due to polydispersed clusters is proportional to the mean number of atoms per cluster in a linear law, similar to that of monodispersed clusters. The superposition power  $q$  influences the calculation, as shown in Figure 1. However, the strengthening caused by polydisperse clusters is no less than the strengthening by monodisperse clusters of the same mean size.

One basic assumption in the present model is that the volume misfit of a cluster increases linearly with the number of atoms per cluster. Research on the lattice parameter in cubic intermetallics and solid solutions suggests that the volume of intermetallics equals approximately the sum of solute atomic volumes up to

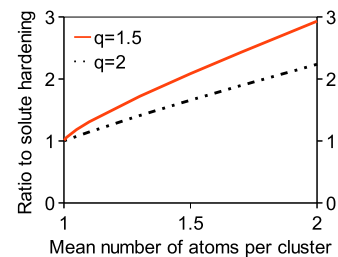


Fig. 1. The ratio of cluster strengthening to mono-atom solute strengthening as a function of the mean number of atoms per cluster when  $q = 1.5$  and  $2$ .

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