

## Letter

## Inverse Ripening and Rearrangement of Precipitates under Chemomechanical Coupling



Reza Darvishi Kamachali\*, Christian Schwarze

Interdisciplinary Centre for Advanced Materials Simulation (ICAMS), Ruhr-University Bochum, 44801 Bochum, Germany

## ARTICLE INFO

## Article history:

Received 21 November 2016

Received in revised form 10 January 2017

Accepted 16 January 2017

## Keywords:

Phase transformation

Coherent precipitation

Ripening

Elasticity

## ABSTRACT

A coupling between diffusional and mechanical relaxation raised from composition-dependent elastic constants, and its effects on the evolution of precipitates with finite misfit strain are investigated. Inverse ripening has been observed where smaller precipitate grows at the expense of a larger one. This occurs due to fluxes generated under elastically-strained solute gradients around precipitates that scales with  $(\frac{R}{r})^6$  where  $R$  and  $r$  are the precipitate radius and the radial coordinate, respectively. Both isotropic and anisotropic dependency of elastic constants on the composition were considered. The latter leads to the emergence of new patterns of elastic anisotropy and rearrangement of precipitates in the matrix.

© 2017 Elsevier B.V. All rights reserved.

The effect of internal stress on the kinetics of precipitation and distribution of precipitates in solid state is a long-standing problem with many applications in metallurgy and materials science [1,2]. The internal stress is generated as a result of lattice mismatch between the precipitate and matrix. Although classical theory of ripening does not include the effect of stress [3], there have been extensive studies which take the elasticity into account (see the review paper by Fratzl et al. [4] and references therein). The elastic energy produced during precipitation influences different aspects of precipitation via diffusion: A spatial flux of atoms is expected under the gradients of elastic energy,  $-\nabla \frac{\partial f^{\text{el}}}{\partial c}$  with  $f^{\text{el}} = \frac{1}{2} [c^{\text{ij}} - \epsilon^{\text{ij}}] C^{\text{ijkl}} [c^{\text{kl}} - \epsilon^{\text{kl}}]$ , that adds to the fluxes under concentration gradients. During phase separation, elastic heterogeneity between two phases has been shown to have significant effects on the morphology of the evolving phases [5,6] and kinetics of their transformation [7]. The space dependency of elastic properties is also discussed in some literature. In particular, mechanically-driven fluxes originating from size mismatch of alloying elements (Vegard's law) have been widely investigated [8–11]. In this study, we focus on the less investigated case of local coupling between stiffness of materials and local composition varying at a transformation front. This coupling exists due to the fact that the atomic bonds between solute and solvent atoms are often of different strength. Thus, the local elastic constants of the solution should be a function of local composition,  $C^{\text{ijkl}}(c(x, t))$ . In this case, the

mechanical relaxation can be achieved by means of diffusion, leading to local softening/stiffening of the materials. Although the possibility of this effect has been introduced long ago [10,12], it was largely neglected until recently, when we discussed its influence on the precipitation in NiTi shape memory alloys [13]. Furthermore, Steinbach and co-workers have shown that a 'strained equilibrium' may exist [14], i.e. stabilized concentration gradients will be produced in the neighbourhood of a single precipitate, if elastic constants of the matrix are composition dependent. In this work we investigate the effect of 'chemomechanical' coupling due to composition-dependent elastic constants on the ripening of precipitates. New mechanisms of inverse ripening and rearrangement of the precipitates are discovered in the presence of this coupling.

We study precipitation and ripening of  $\delta'$  ( $\text{Al}_3\text{Li}$ ) in aluminium-8 at.% lithium system, at 473 K, but the effect is general and can be extended to a large class of materials. The precipitate holds a volumetric transformation strain about –1% and has a coherent interface energy of  $0.014 \text{ J m}^{-2}$  [15]. Both precipitate and aluminium matrix are cubic crystals with nearly isotropic stiffness with Zener indexes of 0.77 and 1.29, respectively [16]. Ab-initio studies [17] and experimental measurements [18] reveal that the elastic constants of aluminium–lithium solution depend on the lithium solute content, anisotropically. In accordance with these observations, a linear relation

$$C^{\text{ijkl}}(c) = C_0^{\text{ijkl}}(1 + \kappa^{\text{ijkl}} c) \quad (1)$$

is considered for small composition variation in the matrix. Whilst  $C^{11}$  and  $C^{44}$  increase (positive coupling factor),  $C^{12}$  decreases (negative coupling factor) with increasing the solute content. Thus

\* Corresponding author.

E-mail address: [reza.darvishi@rub.de](mailto:reza.darvishi@rub.de) (R. Darvishi Kamachali).

$\kappa^{ijkl}$  (at.%<sup>-1</sup>) is the anisotropic coupling matrix with  $\kappa^{11} = \kappa^{22} = \kappa^{33} = 0.005$ ,  $\kappa^{12} = \kappa^{13} = \kappa^{23} = -0.023$  and  $\kappa^{44} = \kappa^{55} = \kappa^{66} = 0.052$ . For comparison, two ‘isotropic’ cases of coupling with  $\kappa^{ijkl} = \kappa = 0.01$  and  $0.05$  at.%<sup>-1</sup> were also considered in this study.  $C_0^{ijkl}$  are elastic constants of pure aluminium and  $c = c(x, t)$  is the local lithium concentration. Since the elastic constants depend on the composition, thus the elastic anisotropy of the matrix also becomes a function of composition that will be discussed in the following. We treat the precipitate as stoichiometric phase. The total flux of the lithium atoms in the matrix is given by

$$\mathbf{J} = -D\nabla c - \frac{1}{2}M\nabla \left[ (\epsilon^{ij} - \epsilon^{*ij}) \kappa^{ijkl} C_0^{ijkl} (\epsilon^{kl} - \epsilon^{*kl}) \right] \quad (2)$$

where  $D = 1.2 \times 10^{-18} \text{ m}^2 \text{ s}^{-1}$  is diffusion coefficient at 473 K [19],  $M = \frac{D}{\mathcal{R}T}$  is atomic mobility with  $\mathcal{R}$  the gas constant,  $\epsilon^{ij}$  are the total strains and  $\epsilon^{*ij}$  are the eigenstrains. Eq. (1) is used in evaluation of mechanical equilibrium ( $\nabla^j \frac{\partial f^{\text{el}}}{\partial \epsilon^{ij}} = 0$ ) at each time step and Eq. (2) gives temporal evolution of composition ( $\dot{c} + \nabla \cdot \mathbf{J} = 0$ ). These two equations are temporally coupled that results in the chemomechanical cross-coupling effect. For interface kinetics, we use the phase-field method as detailed in [20,21]. We apply a finite difference scheme to solve phase-field and diffusion equations. An iterative spectral solver with periodic boundary conditions is employed to obtain the mechanical equilibrium [22]. Cubic simulation boxes with 64 grid cells are considered with a grid spacing of 1 nm and time stepping of 0.25 s. A linearised phase diagram is assumed where the chemical driving force is proportional to the deviation from equilibrium matrix composition,  $\Delta G^{\text{ch}} = m \Delta S (c - c_\infty)$  where  $m = 60 \text{ K at.\%}^{-1}$  is the slope of the curve in the T–c phase diagram,  $\Delta S = -9.7315 \times 10^5 \text{ J K}^{-1} \text{ m}^{-3}$  is the entropy of formation of the precipitate phase [23], and  $c_\infty$  is the equilibrium composition at a flat interface. The precipitates grow from nuclei with negligible size. Thus, any presumption of size and shape is avoided. Elasticity and coupling accompany the transformation from the beginning. Across the interface, Reuss homogenization scheme is applied. The elastic energy, diffusion potentials and driving forces have been derived and computed, accordingly. More details of the simulation set-up and derivations will be given elsewhere soon [24]. The interface kinetics reproduced by the phase-field equation follows the Gibbs-Thomson relation

$$\frac{V}{M_b} = -\sigma K + \Delta G^{\text{ch}} + \Delta G^{\text{el}} \quad (3)$$

where  $V$  is interface velocity,  $M_b$  is interface mobility,  $\sigma$  is interface energy,  $K$  is interface curvature and  $\Delta G^{\text{ch}}$  and  $\Delta G^{\text{el}}$  are driving forces. In general, the chemical driving force is in favour of precipitation from a super-saturated matrix, whilst the elasticity can suppress or accelerate the growth depending on the global mechanical equilibrium. In the current simulations, since the volume fraction of the precipitate is much less than 50%, elasticity (along surface tension) suppresses its growth and  $\Delta G^{\text{ch}}$  is the only positive driving force.

Fig. 1 summarizes the results of single precipitate simulations. The black dots in Fig. 1(a) show growth due to chemical driving force, neglecting elasticity, where the volume fraction of the precipitate is only influenced by its curvature. The elastic contribution, once included, suppresses growth and equilibrium volume fraction (red<sup>1</sup> dots). Considering chemomechanical coupling, the effects of elasticity are enforced in both regards. Fig. 1(b) shows the concentration profile at equilibrium  $t = 10,000$  s. In the presence of chemomechanical coupling, a concentration gradient is stabilized around the precipitate, whilst without the coupling those gradients in the

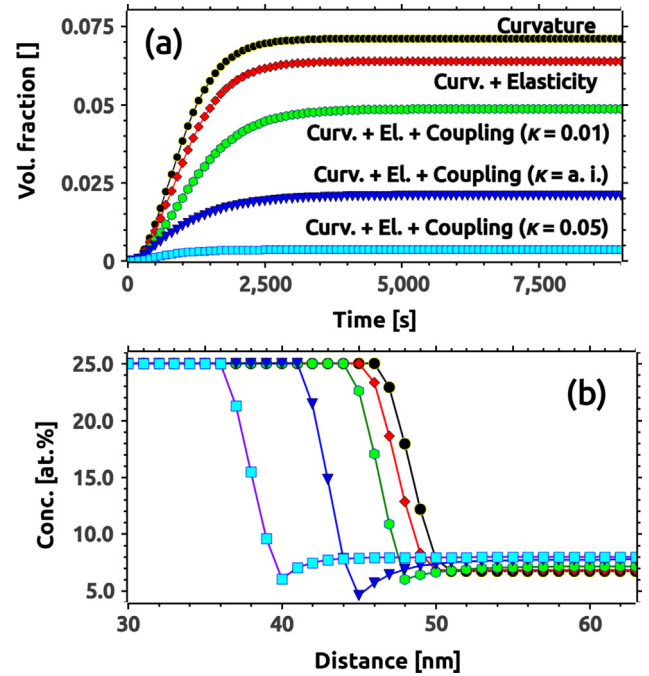


Fig. 1. (a) Evolution of a single precipitate and (b) concentration profiles from the centre of the precipitate ( $t = 10,000$  s) are shown.

matrix vanish. This is a result of non-vanishing elastically-driven fluxes around the precipitate which balance with the Fick's diffusion (Eq. (2)). The analytical solution of Eq. (2) will be discussed shortly. Note that elasticity without the coupling corrects equilibrium composition at the interface but does not influence solute distribution. The chemomechanical coupling also corrects the equilibrium composition of the matrix raising its concentration, as shown in Fig. 1(b), by pushing the solute atoms into the matrix.

Fig. 2 shows the results of simulations for a pair of precipitates with different coupling situations. The precipitates nucleate at different time steps that gives the initial size difference in the growth stage. The conventional theory of ripening [3] predicts that precipitates compete with respect to their size. This is indeed observed in our simulations if there is no chemomechanical coupling. Fig. 2(a) compares evolution of precipitates with and without elasticity. Compared to normal ripening, elasticity results in deceleration of the growth and acceleration of the ripening in the later stages. The cross-coupling between diffusion and mechanical relaxation, however, changes the ripening behaviour, entirely: As soon as the matrix is depleted and the precipitates ‘feel’ each other via diffusion of the solute atoms, the larger precipitate shrinks until the two precipitates equilibrate with respect to their size (Figs. 2(b) and (c)). This is called ‘inverse ripening’ in the current letter in contrast to conventional ripening in which larger precipitates grow at the expense of smaller ones. Similar behaviour has been observed for  $\kappa = 0.05$  at.%<sup>-1</sup>.

Two mechanical effects on the kinetics of transformation are (i) the direct offset of elastic energy between precipitate and matrix phases (third term in Eq. (3)) and (ii) the indirect modification of solute flux (entering second term in Eq. (3)). Whilst the first effect is always present upon elastic interaction, the latter is only a consequence of the chemomechanical coupling introduced in this study. The results show that the interfacial energy offset cannot lead to inverse ripening. In fact, elasticity even accelerates the ripening process when there is no chemomechanical interaction (Fig. 2(a)). These observations acknowledge the significance of

<sup>1</sup> For interpretation of color in Fig. 1, the reader is referred to the web version of this article.

Download English Version:

<https://daneshyari.com/en/article/5453289>

Download Persian Version:

<https://daneshyari.com/article/5453289>

[Daneshyari.com](https://daneshyari.com)