Comparison of coarsening behaviour in non-conserved and volume-conserved isotropic two-phase grain structures

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Abstract

Microstructure evolution in non-conserved and volume-conserved isotropic two-phase grain structures, with equal interfacial energy for all interfaces and equal volume fractions of the two-phases, is compared based on large-scale phase-field simulations. Two important observations are as follows: (1) the growth mechanism has a larger effect on the grain size distributions of the two alloys than on their grain topology distributions; (2) the growth rate and normalized grain size relation of the grains per topological class are much more scattered for the conserved system than the non-conserved system.

Keywords: Grain growth, Bulk diffusion, Simulations, Phase field model

Two-phase polycrystalline materials can offer better material properties, like high strength, toughness and corrosion resistance over single-phase polycrystalline materials. A two-phase microstructure, with α and β phases may contain α/α , β/β and α/β interfaces. Cahn [1] analyzed the stability of various microstructural features for two-dimensional (2D) non-conserved two-phase (NCTP) systems. It was found that the energetic ratios $\sigma_\alpha/\sigma_{\alpha/\beta}$ and $\sigma_\beta/\sigma_{\alpha/\beta}$, with σ_α , σ_β and $\sigma_{\alpha/\beta}$ the energies of the α/α , β/β and α/β interfaces respectively, control the stability, spatial distribution and grain shapes of the 2 phases in a two-phase system. This was later confirmed by Holm et al. [2] with Monte Carlo-Potts simulations for the 2D NCTP systems. Furthermore, Cahn [1] extended the von Neumann-Mullins law [3, 4], which describes an exact relation between the rate of change of grain area (dA/dt) of an individual grain and its number of sides (F) in the 2D isotropic single-phase systems, to the 2D isotropic NCTP systems.

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Cahn [1] also suggested that a further extension of the von Neumann-Mullins law to 2D volume-conserved two-phase (VCTP) systems would be a formidable mathematical problem. Fan and Chen [5, 6] performed simulations for 2D VCTP and speculated that the modified von Neumann-Mullins law proposed by Cahn [1] may apply to 2D two-phase systems in general [6]. Holm et al. [2] also suggested that the microstructures of the NCTP and VCTP systems in 2D and 3D with equal volume fraction ($V_\alpha = V_\beta = 0.5$) will be structurally equivalent, as the triple junction stability criteria in 2D and triple line stability criteria in 3D for the NCTP and VCTP alloys are the same. However, in a previous study [7], it was observed that various microstructural features in the NCTP and VCTP systems are influenced by the different growth controlling mechanisms and growth kinetics. The goals of the present work are: (1) to compare microstructure evolution; (2) to test the applicability of the von Neumann-Mullins law in 2D; and (3) to compare volumetric growth rates in 3D for isotropic NCTP and VCTP systems with $V_\alpha = V_\beta = 0.5$. Furthermore, $\sigma_\alpha = \sigma_\beta = \sigma_{\alpha/\beta}$, was chosen so that the growth kinetics in the NCTP alloy and single-phase system is identical.

A two-phase phase-field model was employed to carry out large-scale simulations for the NCTP and VCTP systems [7]. Further details on simulations can be found in [7], where the effect of the volume fractions on volume-conserved isotropic two-phase coarsening was studied. The 50/50 alloy in [7] is renamed as VCTP alloy in the present study. Some microstructural features of NCTP and VCTP systems were discussed in brief and the summary of this discussion in [7] are as follows: (1) a growth exponent $m \sim 2$ for the NCTP alloy and $m \sim 3$ for the VCTP alloy was found in the 2D and 3D simulations; (2) All four types of triple junctions (TJs) namely $\alpha\alpha\alpha$, $\alpha\alpha\beta$, $\alpha\beta\beta$, and $\beta\beta\beta$ TJs are present in equal volume fraction in the NCTP alloy, whereas there are less $\alpha\alpha\alpha$ and $\beta\beta\beta$ TJs than $\alpha\alpha\beta$ and $\alpha\beta\beta$ TJs in the VCTP alloy in 2D simulations; and (3) In the 2D VCTP alloy, most of the grains with 3 and 4 sides had a grain size comparable to the mean grain size $\langle R \rangle$, whereas, in the 2D NCTP alloy, most of the grains with 3 and 4 sides had a grain size smaller than the $\langle R \rangle$. The grain size distributions (GSDs) and grain topology distributions (GTDs) of the total grain structure ($\alpha + \beta$ phase grains) for the NCTP and

VCTP alloys were only discussed very briefly and not compared in [7].

Thus, in the present study, further analysis related to the GSDs and GTDs was carried out first. The GSDs obtained for the NCTP alloys are found to be significantly wider than those obtained for the VCTP alloys in both the 2D and 3D simulations as shown in Figs. 1a and 1b. For the 2D simulations, the shape of the GSD for the NCTP alloy is close to that obtained previously from grain growth simulations for a single-phase system [8–10]. Similarly for the 3D simulations, the GSD for the NCTP alloy matches that obtained from previous single-phase grain growth simulations [8, 11, 12]. The GSD obtained for the VCTP alloy in 3D simulations matches the GSD obtained for solid particle coarsening in a liquid matrix with a volume fraction of the solid particles of 0.78 [13]. The Weibull distribution (with $\beta = 3.035$) gave a good fit to the GSD of the VCTP alloy, whereas the Rios distribution (with $\nu = 3.2$) gave a good fit to that of the NCTP alloy [14]. The GTDs are shown in Figs. 1c and 1d. For 2D simulations, the GTD is peaked around $F = 6$ for the VCTP alloy, whereas it is peaked between $F = 5$ and $F = 6$ for the NCTP alloy, which is in agreement with previous studies [8, 15] for single-phase grain structures in 2D. In the 3D simulations, the GTDs obtained for the NCTP and VCTP alloys are similar except towards the tail (at high F) where the VCTP alloy has fewer grains with $F \geq 30$ than the NCTP alloy. The GTDs obtained from the 3D simulations match those obtained in previous experimental [16, 17] and simulation [8, 11, 12] studies. Thus, the difference in growth mechanism and growth kinetics in the NCTP and VCTP alloys has a stronger influence on the GSDs than on the GTDs.

The evolution of individual grains in the isotropic 2D single-phase system is exactly described by the von Neumann-Mullins law [3, 4], which is given as

$$\frac{dA}{dt} = -m\sigma\frac{\pi}{3}(6 - F), \quad (1)$$

where A is the area of a given grain, t is time, m is grain boundary mobility, σ is grain boundary energy and F is the number of sides of the considered grain. According to Eq. (1), the rate of change of the area of an individual grain depends only on its number of sides F . Thus, all grains with $F > 6$ will grow, while all grains with $F < 6$ will

shrink and all grains with $F = 6$ will not change size and remain stable. Eq. (1) was extended by Cahn [1] for two-dimensional non-conserved isotropic two-phase systems. It was also shown [1] that the modified von Neumann-Mullins law reduced to Eq. (1) when $\sigma_\alpha = \sigma_\beta = \sigma_{\alpha/\beta}$ (which is equivalent to the NCTP alloy in the present study). In Figs. 2a and 2b, the rates of area change dA/dt of all the grains, as obtained from the 2D simulations, are plotted against their normalized grain sizes ($R/\langle R \rangle$) for the NCTP and VCTP alloys. For the NCTP alloy, the data points obtained for grains of a same topological class are located along a horizontal line (see fig. 2a), indicating that the dA/dt is similar for all grains from the same topological class independent of the grain size. However, for the VCTP alloy, there is a considerable scatter on the data points obtained for grains of a same topological class (see Fig. 2b). This contrast between 2D NCTP and VCTP alloys is again seen in Figs. 2e and 2f where the average and standard deviation of the dA/dt data for a given F is plotted. It is clear that the standard deviation within a given topological class is much larger in the VCTP alloy than the NCTP alloy. These results show that, for the 2D VCTP alloy, the von Neumann-Mullins law (1) is only valid for the average of dA/dt as a function of F and not for the growth rate of the individual grains. Our findings for the 2D NCTP system are consistent with previous observations from 2D single-phase systems [10]. It is important to note that the small variation among the growth rates of the individual grains of a given topological class for the NCTP alloy seen in the simulations is due to numerical inaccuracies. As pointed out in [10], whenever a shrinking grain has a size comparable to the diffuse interface width in a phase-field model, its kinetics are affected. This may have some influence on the kinetics of the neighbouring grains, explaining the slightly larger variations in growth rates as compared to [18]. The variations for a given topological class for 2D NCTP are similar to those in the Kim et al. [10] study.

In Fig. 3, we show the evolution of the area and number of sides of 2 individual grains as a function of time for the VCTP alloy. Grain A has $F = 6$ sides and grain B has $F = 7$ sides at $234 \times 10^3 s$. The area of grain A is slightly smaller than that of grain B at $234 \times 10^3 s$. As time progresses, grain B shrinks and finally disappears due to growth

of a larger neighbouring grain, whereas the size of grain A is almost constant during the considered time. The topological evolution of grains A and B is peculiar compared to the behaviour of individual grains during coarsening in single-phase and NCTP systems. While shrinking, grain B first loses two sides, to become a 5-sided grain, then gains 1 side, to become again a 6-sided grain while still shrinking, and finally again loses 1 side to become a 5-sided grain before disappearing. In the meantime, Grain A first loses its two sides, becoming a 4-sided grain and later again gains 2 sides; however, even as a 4-sided grain, the area of grain A is almost constant, while a 4-sided grain would shrink and disappear quickly in the NCTP system. This shows again that the growth kinetics of the individual grains in a VCTP system is not only determined by its number of neighbours, as is the case in the NCTP systems, but also by the properties (like phase type and grain size) of the neighbouring grains. It also explains the large scatter on the growth rate data in the 2D VCTP alloy (see Figs. 2b and 2f). The fact that a smaller grain can grow in the presence of larger grains and another relatively large grain can shrink in a VCTP system were also observed experimentally by Boettinger et al. [19] for coarsening of a high-volume fraction of solid grains distributed in a liquid matrix (see Fig.3 in [19]).

In 3D, the volumetric growth rate as function of number of sides, also known as the 3D von Neumann relation, can be written as [10, 20, 21]

$$V^{-1/3} \frac{dV}{dt} = 2m\sigma G(F), \quad (2)$$

where, $G(F)$ is dimensionless growth rate, which is a function of the number of sides F only, and the other parameters as given for Eq. (1). Due to the extra degree of freedom for 3D systems, unlike the von Neumann-Mullins law (1) for 2D isotropic single-phase systems, Eq. (2) is not exact. Thus, the $dV/dt - R/\langle R \rangle$ relation in Fig. 2c for the NCTP, obtained from the 3D simulations, shows a weak clustering of grains belonging to a same topological class. However, for the 3D VCTP alloy, the $dV/dt - R/\langle R \rangle$ relation in Fig. 2d shows a large scatter within each topological class compared to the data for the 3D NCTP alloy. This contrast between 3D NCTP and VCTP alloys is again seen in Figs. 2g and 2h. The average growth rate of the 3D NCTP and VCTP system are

similar but the standard deviation for average growth rate of a given topological class is much larger in the 3D VCTP alloy than the 3D NCTP alloy. For the 3D NCTP system, the standard deviation on the average growth rate within a given topological class is of a similar magnitude as those obtained in previous simulation studies of single-phase systems [10, 22, 23]. The average growth rate (dV/dt) is zero at $F \approx 15$ for both, the NCTP and VCTP alloy, which is in agreement with previous findings from grain growth simulations for single-phase systems [22, 24]. Previous experimental [17] and simulation [10, 11] results for the average volumetric growth rate are also in agreement with our results.

The Aboav-Weaire relation [25, 26] relates the number of sides F of a grain to the average number of sides of the nearest neighbours $m(F)$. For 2D single-phase systems, the Aboav-Weaire relation is given as: $m(F) = 6 - a + (6 \cdot a + \mu)/F$, where μ is the second moment of F and a is a constant. The Aboav-Weaire relation obtained in this study for the NCTP and VCTP alloys in 2D are shown in Figs. 4a and 4b. The parameters a and μ were found to be, 1.18 and 1.7 respectively for the 2D NCTP system and 1.0 and 2.0 respectively for the 2D VCTP system. Similarly the Aboav-Weaire relation for 3D grain structures is given as: $m(F) = \bar{F} - 1 + (\bar{F} + \mu)/F$, where $\bar{F}$ is the average of F . The parameters $\bar{F}$ and μ were found to be, 14.71 and 9.80 respectively for the 3D NCTP system and 15.33 and 9.08 respectively for the 3D VCTP system. The results from Fig. 4 suggest that the Aboav-Weaire relation holds for NCTP and VCTP systems in 2D and 3D. There is thus no significant effect of growth mechanism or properties of neighbouring grains on the Aboav-Weaire relation.

In the NCTP systems, for all interfaces, the individual kinetics depend only on the curvature of the interface, whereas, in the VCTP alloys, there are 2 types of interfaces, with different kinetics. There are interfaces between 2 grains of a same type; their kinetics are determined only by the curvature of the interface, as in the NCTP alloys. There are also interfaces between 2 grains of a different type and with a different composition; the movement of these boundaries requires long range diffusion, and the kinetics of such an interface are determined both by its curvature and the grain arrangement in its neigh-

bourhood. This explains the larger spread on the growth rate data within a topological class for VCTP alloys than NCTP alloys.

In summary, we have shown that several microstructural features and the GSDs of isotropic NCTP and VCTP alloys with $V_\alpha = V_\beta = 0.5$ and $\sigma_\alpha = \sigma_\beta = \sigma_{\alpha/\beta}$ differ due to a different growth mechanism and different growth kinetics, despite similar triple junction or triple line criteria (in 2D and 3D respectively). In the 2D NCTP system, the growth behaviour of the individual grains is uniquely related to its topological class, while in the 2D VCTP system, it is also affected by characteristics of the neighbouring grains. Similarly, the results for the 3D VCTP system show large scatter relative to the 3D NCTP system. The existing analytical growth models should thus be revised for conserved systems.

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