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# Pinning effect of spheroid second-phase particles on grain growth studied by three-dimensional phase-field simulations

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ABSTRACT

For the first time, the pinning effect of small spheroid particles with aspect ratios 1, 2 and 3 on grain growth in polycrystalline structures was studied by three-dimensional phase-field simulations. This was possible by using an efficient parallel sparse bounding box algorithm. Simulation results for different volume fractions of the second-phase particles show that distributions of particles with higher aspect ratios have a stronger pinning effect, since grain boundaries have the tendency to align with the largest cross-section of the particles. The number of particles at triple, quadruple or higher order junctions increases with volume fraction, and with aspect ratio. Nevertheless, the final grain size has a volume fraction dependence of the form $1/f_V^{0.93}$, following theoretical predictions assuming random intersections between grain boundaries and particles. A generalised Zener relation with a prefactor depending on the aspect ratio is proposed.



## 1. Introduction

The microstructure of materials is often composed of multiple grains with different crystallographic orientations. Under certain conditions, the larger grains will start to grow and the smaller grains will shrink. This phenomenon, called grain growth, is thus characterised by an increase of the mean grain size. Small second-phase particles, such as precipitates and insoluble inclusions, exert a strong pinning effect, also called Zener pinning, on the grain boundaries. They restrain the mobility of grain boundaries and eventually inhibit grain growth, limiting the final mean grain size of the microstructure. Insight into the pinning effect of particles is of great technological importance, since for many applications a tailored grain size is required to obtain materials with the desired properties. Examples are the addition of a small amount of alloying elements to High Strength Low Alloyed (HSLA) steels and Ni-based super alloys in order to obtain materials with a small grain size and high strength [1–3], the use of precipitates in thin films to induce abnormal grain growth in order to obtain films with a large grain size for high electrical conductivity and reduced electromigration damage [4–6], and, recently also the use of particles (such as precipitates, carbon nanotubes and organic or amorphous particles) to stabilize nanocrystalline materials [7–10].

In most analytical studies [11–14] on Zener pinning it is assumed that normal grain growth is arrested when a critical mean grain radius $\langle R\rangle_{\text{lim}}$ is reached. Most often a relation of the form

$$\frac{\langle R\rangle_{\text{lim}}}{r} = K\frac{1}{f_V^b} \tag{1}$$

is obtained, where $r$ is the radius of the second-phase particles and $f_V$ the volume fraction. The values of the parameters $K$ and $b$ vary among the different studies, depending on which assumptions are made on the shape and properties of the particles and boundaries, and on the assumptions made on the position of the particles with respect to the boundaries.

The pinning force exerted by one particle on a grain boundary can be analytically calculated based on the position of the boundary, the shape of the particle and the properties of the particle–matrix interface and grain boundary [11,14–16]. The calculation of the total pinning force of a distribution of multiple particles is more complex. The number of particles that lie at a grain boundary, as well as the geometry of the grain boundary at and near each grain boundary-particle intersection, has to be known. However, this appears to be extremely difficult to describe analytically. In this respect, computer simulations turn out to be helpful. They are a practical tool, not only to determine the number and the geometry of boundary-particle intersections, but also to study the role of different characteristics of the second-phase particles separately. Together with existing analytical theories and experimental findings, they provide valuable insights.

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The pinning effect of second-phase particles has mostly been studied by two-dimensional computer simulations using Monte Carlo Potts models [17–19], front-tracking-type models [20–22] and phase-field models [23–30]. It is found that for two-dimensional systems, relation (1) is obeyed, with $b = 0.5$, and most particles are in contact with a grain boundary in the pinned microstructure. Only few studies consider three-dimensional systems. Simulations for three-dimensional systems [19,26,27] show that the fraction of particles in contact with a grain boundary is significantly lower than in two-dimensional systems. Furthermore, fitting of relation (1) to the results obtained from three-dimensional simulations gives values for $b$ and $K$ that are very different from those obtained for two-dimensional simulations. In [19], $b = 1.02$ and $K = 0.728$ are obtained; in [22], the value $b = 1.0$ is extracted. In contrast, in the work of [26], the parameter values $b = 0.870$ and $K = 1.42$ are found. The study in [27] shows that even for columnar grain structures, the pinning effect is very different from that observed in two-dimensional simulations. In two dimensions, the grain boundaries are basically lines interacting with particles, which split into two different boundary segments when they meet a particle. In contrast, in three dimensions, the grain boundaries are surfaces that remain a single entity when meeting a particle. In order to balance the interfacial tensions at a particle-boundary intersection, the grain boundary assumes a so-called dimple shape. The extra curvature thus created contributes to the driving force of grain growth [31]. Furthermore, in two dimensions, the pinning force of one particle is maximal when the grain boundary meets the particle at an angle which is twice as large as is the case in three dimensions. Therefore, the pinning effect in two-dimensional systems is in general much stronger than in three-dimensional systems [27]. It is thus important that predictive computer models reflect the three-dimensional nature of Zener pinning for bulk material systems as well as for thin films.

Although the values obtained for the coefficients in the Zener relation in the different simulation studies seem to vary considerably, it was shown in [26,28] that the limiting mean grain sizes obtained with different simulation techniques, in fact, agree quite well. Experimentally determined limiting mean grain sizes are however in general much smaller than those predicted by simulations. Based on experimental data, it was also concluded that the Zener relation should have different parameter values for respectively low and high volume fractions [13,14]. Such a transition between two regimes has not been observed in computer simulations. These discrepancies between experimental and theoretical results indicate that one or several essential aspects were not considered in previous computer simulations. A possible hypothesis is the effect of particle shape. Except for [29], all the above mentioned studies only performed simulations of grain growth in the presence of spherical particles. In reality, however, particles are generally not spherical. Experimental microstructures show that, even in the case where the particle–matrix interface is incoherent and has properties that are independent of orientation, the particle shape deviates from spherical because of inhomogeneities in the surrounding matrix during formation of the particle [32]. For example, particles formed on a former boundary are lance-shaped, the solute flux for particles formed near another particle or a grain boundary is not spherically symmetric, particles may change shape in an anisotropic way during deformation processes. It has been shown that the pinning force of a single particle is strongly dependent on the geometry at the grain boundary-particle sections and the particle shape [11,15,19,33]. The effect of particle shape seems to be even far more important than the effect of the anisotropy of the particle–matrix interfacial energy [11,15]. The advantage of phase-field simulations is that simulations can be performed and the effect of particle dispersions can be computed without making prior assumptions on the number of particles interacting with the grain boundaries and their orientations with respect to the boundary.

According to [34], the maximal pinning force $F_Z$ of an ellipsoid particle is, for the case where the boundary intersects the particle perpendicular to its major axis,

$$F_Z = F_Z^S \frac{2}{(1 + r_a) r_a^{1/3}} \tag{2}$$

and, for the case where the boundary intersects the particle along a plane containing the major axis,

$$F_Z = \frac{F_Z^S}{\pi} \frac{(1 + 2.14\ r_a)}{r_a^{1/3}}, \quad r_a \geqslant 1, \tag{3}$$

where $r_a$ is the aspect ratio of the particle and $F_Z^S$ the drag from a spherical particle of equal volume. The last equation shows that if the boundary intersects the particle along a plane containing the major axis, its pinning force is considerably larger than that of a spherical particle with equal volume, even for small aspect ratios. If the boundary intersects the particle perpendicular to the major axis, the pinning force of the particle is however much smaller than that of a spherical particle. As a consequence, the effect of particle shape on the overall pinning effect of a particle distribution depends on the possibility that the boundary-particle intersection contains the major axis. In [15], it was analytically calculated that within well-defined conditions, particle dispersions of mono-orientation can be very effective in pinning migrating boundaries. This was confirmed by the results of two-dimensional phase-field simulations in [29], which show that a dispersion of mono-oriented ellipse-shaped particles with a high aspect ratio is more effective in pinning than a dispersion of circular particles of the same size.

Based on the work of [15,34], it is generally assumed that dispersions of ellipsoid particles have a stronger pinning effect than dispersion of spherical particles, although to our knowledge, the effect of random dispersions of ellipsoid particles on grain growth has not been verified by three-dimensional mesoscale simulations yet, mainly because of computational limitations. To gain more insight in the dependence of the pinning force of a particle distribution on the shape of the particles, we performed three-dimensional phase-field simulations of grain growth in systems with spheroid particles for different aspect ratios and volume fractions of the particles. Phase-field modelling is a versatile tool for simulating microstructural evolution phenomena. It allows one to predict the evolution of complex morphologies with different thermodynamic driving forces. In [35], a phase-field model is used to model the microstructure of a single-phase material by a set of non-conserved phase-field variables that distinguish the different crystallographic orientations of the grains. Inside a grain, one phase-field variable takes the non-zero equilibrium value 1, while the other variables assume values close to zero. Across the grain boundaries, the corresponding phase-field variables vary continuously to their equilibrium value in the neighbouring grains. In this work, we employ an extension of this model for grain growth in the presence of incoherent second-phase particles with constant properties presented in [24,25,27]. The effect of aspect ratio and volume fraction was studied for incoherent spheroid particles with aspect ratios 1, 2 and 3. A parallelised bounding box algorithm [28] is applied to make three-dimensional simulations with a large amount of grains feasible. The purpose of this work is to provide new information that might lead to a generalisation of the classical Zener type pinning theories to the effect of spheroid particles on grain growth.