# Simulation of the kinetics of grain-boundary nucleated phase transformations

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**Abstract.** The kinetics of phase transformations for which nucleation occurs on parent-microstructure grain boundaries, and the resulting microstructures, were investigated by means of geometric simulations. The influences of parent microstructure grain-boundary area density, parent grain-size distribution and parent→product kinetics were analysed. Additionally, the simulated kinetics were compared with predictions from two kinetic models, namely a model proposed for spatially random nucleation and a model proposed for grain-boundary nucleation. It was found that the simulated transformed fraction as function of time lies in between the two model predictions for all investigated parent microstructures and parent→product kinetics.

## Introduction

For nucleation-and-growth phase transformations, the assumption of homogeneous (i.e. spatially random) nucleation, even though often used in modelling, does not hold in many cases. Nucleation often occurs heterogeneously on container walls or on microstructural features of the parent phase like inclusions or defects, such as grain boundaries. This should be reflected in the modelling of phase transformations, since mean-field kinetic models are usually derived assuming homogeneous nucleation. This paper is concerned with an investigation of the kinetics of phase transformations and the resulting microstructure, by means of geometric simulations, if nucleation occurs on grain boundaries of the parent phase.

First, kinetic models for random nucleation and for grain-boundary nucleation are briefly discussed, then the simulation method is described and finally, the influence of the main factors on the kinetics and resulting microstructure are discussed.

## Kinetic Models

Nucleation-and-growth phase transformations with random, bulk nucleation can be modelled with mean-field kinetic models which go back to works by Johnson, Mehl, Avrami and Kolmogorov [1] (see also [2] for a recent review). These "JMAK-like" models all have in common that the assumption of a spatially random arrangement of nuclei allows to relate the real transformed (volume) fraction of a specimen, $f$, with the hypothetical, so-called extended fraction, $f^{\mathrm{ex}}$, which is defined as the volume fraction of all growing grains if they could grow without impingement (i.e. if they could grow "through" each other). Thus:

$$\mathrm{d}f = (1-f)\mathrm{d}f^{\mathrm{ex}}. \tag{1}$$

The calculation of the extended transformed fraction can be done straightforwardly if specific models for nucleation and growth are adopted; i.e. the nucleation rate per unit volume of untransformed material, $\dot{N}_V$, and the growth rate $v$ must be known. Additionally, the dimensionality, $d$, of the growth and the shape of the growing grains (expressed by the shape

factor $g$) need to be known, as well as whether linear or parabolic growth prevails [expressed by the exponent $m$, which equals 1 for linear (e.g. interface-controlled) and 2 for parabolic (e.g. diffusion-controlled) growth]:

$$f^{\mathrm{ex}}(t) = \int_0^t \dot{N}_V(\tau) g \left( \int_\tau^t v(t')\mathrm{d}t' \right)^{d/m} \mathrm{d}\tau, \tag{2}$$

where $\dot{N}_V(\tau)$ refers to the grains nucleated at time $\tau$, which grow with velocity $v$ from time $\tau$ until the current time $t$. Eq. 1 cannot be used if nucleation occurs on grain boundaries. Cahn [3] proposed a model for nucleation on grain boundaries by approximating grain boundaries as planes which are randomly distributed in space. Since nucleation is supposed to occur randomly on these planes, the impingement of growing grains *on planes* can be described, in analogy to Eq. 1, by

$$\mathrm{d}O = (1 - O)\mathrm{d}O^{\mathrm{ex}}, \tag{3}$$

where $O$ denotes the transformed area fraction on a plane (parallel to or on a grain boundary), and $O^{\mathrm{ex}}$ the corresponding extended area fraction. Likewise, since planes are randomly distributed, the impingement of material growing *from different planes* can be described by Eq. 1. For such a model of grain-boundary nucleation, the nucleation rate needs to be expressed per unit area of grain boundary, $\dot{N}_O$; the grain boundary area per unit volume is denoted by $S_V^{\mathrm{GB}}$. For a detailed description of the model, see Refs. [3, 4]. In the case of linear growth of spherical product grains, Cahn's model is given by

$$f(t) = 1 - \exp\left\{ -S_V^{\mathrm{GB}} \int_{-\infty}^{+\infty} \left[ 1 - \exp\left( - \int_0^t \pi(v^2(t-\tau)^2 - y^2)\dot{N}_O(\tau)\mathrm{d}\tau \right) \right] \mathrm{d}y \right\}. \tag{4}$$

The assumption of randomly distributed planes in this grain-boundary nucleation model may not be a valid approximation of a realistic grain-boundary network.

**Simulation Method**

The method to simulate the kinetics of phase transformations and the resulting microstructure which is used in this work can be called geometric simulation method [5]. The corresponding algorithm discretises time in time steps and space in voxels (on a three-dimensional, cubic grid); see [6]. It needs input values for the nucleation and growth rate prevailing in the simulated specimen. For each time step, the number of nuclei that should emerge according to the nucleation rate is determined and as many nucleation positions are chosen; either randomly in the whole specimen or from a list of available nucleation sites (see below). For each nucleus, be it formed in the current or in a previous time step, the radius that the grain, originating from this nucleus, would have without interaction with other grains is determined by integrating the growth rate from the time of nucleation until the current time. Within a sphere of this radius around each nucleation position, it is checked for each voxel if it has already been transformed. If this is the case, transformation cannot take place. If the voxel is untransformed yet, it is transformed now and is assigned to the considered nucleus/grain. This algorithm thus corresponds to the assumption of hard impingement.

If nucleation doesn't occur randomly in the whole specimen, a list of permitted nucleation sites needs to be generated. In the simulations compatible with the assumptions of Cahn's model for grain-boundary nucleation, this is realised by generating randomly a certain number

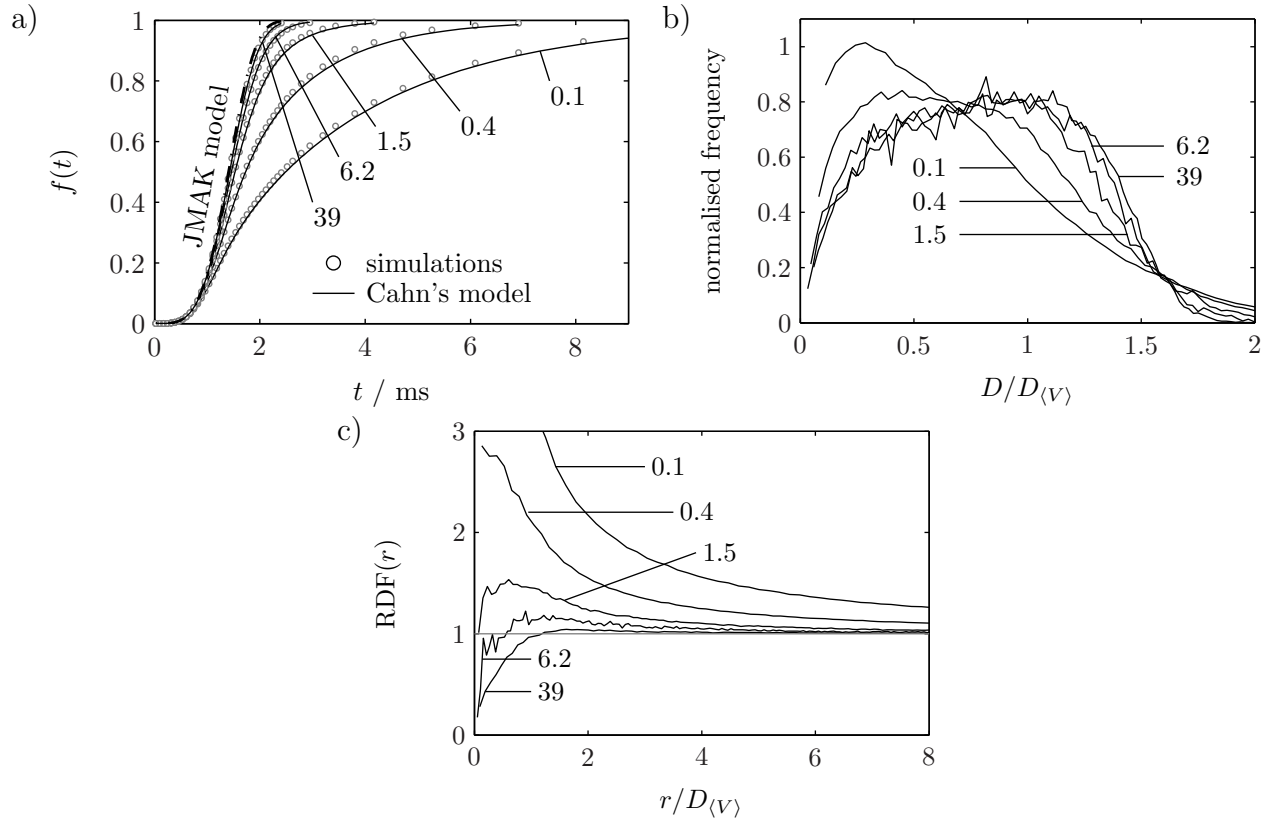


Fig. 1: The results of simulations with random nucleation on randomly distributed planes for varied ratio $S_V^{\mathrm{GB}}/\dot{N}_O$ (values indicated in the figures). a) The transformed fraction as a function of time (symbols) together with the predictions of Cahn's model (solid lines) and a JMAK-like model (dash-dot line). b) The resulting grain-size distributions. c) The radial distribution function of nucleation positions.

of planes cutting the simulation cube and by determining the voxels that are cut by each plane. For simulations using more realistic grain-boundary arrangements, two simulations are performed: First, a simulation with nuclei randomly distributed in the bulk is done and the microstructure (denoted "parent microstructure") resulting from this simulation is then used in a second simulation to investigate the effect of grain-boundary nucleation. A voxel of the parent microstructure is defined to be a grain-boundary site and hence a valid nucleation position in the second simulation ("parent→product transformation") if it has exactly two different grains in its neighbourhood (of 26 surrounding voxels).

In all simulations, interface-controlled (i.e. linear) growth is assumed. Nucleation is assumed to occur continuously in the untransformed volume (with one exception, where pre-existing nuclei are assumed). The temperature is either held constant during the transformation (i.e. isothermal annealing) or increased at a constant rate (i.e. isochronal annealing). Arrhenius-dependencies are assumed for the nucleation and growth rates with the activation energies $Q_{\mathrm{N}}$ and $Q_{\mathrm{G}}$ for nucleation and growth, respectively.

## Simulation Results

**Influence of the GB area density.** The influence of the grain boundary area density is revealed by the results shown in Fig. 1 a) together with the predictions of Cahn's model (Eqs. 1, 3 and 4) and a JMAK-like model (based on spatially random nucleation; Eqs. 1 and 2) for the corresponding input parameters. Since $S_V^{\mathrm{GB}}$ and $\dot{N}_O$ have opposing effects on the kinetics, the

ratio $S_V^{\mathrm{GB}}/\dot{N}_O$ was varied while keeping the value of $S_V^{\mathrm{GB}} \times \dot{N}_O = \dot{N}_V$ constant. This ensures that the JMAK-like model predicts the same kinetics for all simulations. For all input parameters, the simulations agree well with the predictions of Cahn's model.

For high values of the ratio $S_V^{\mathrm{GB}}/\dot{N}_O$, the simulated phase transformation kinetics (as well as the prediction of Cahn's model) become identical to the prediction of the JMAK-like model; the smaller the ratio $S_V^{\mathrm{GB}}/\dot{N}_O$ is, the slower the transformation and the larger the deviation from the prediction of the JMAK-like model becomes. The grain-size distributions resulting for the same set of simulations are shown in Fig. 1 b). The smaller the ratio $S_V^{\mathrm{GB}}/\dot{N}_O$ is, the broader the product grain-size distribution gets and the more its maximum moves towards small grain sizes.

**Influence of the parent microstructure.** Departing from various parent microstructures, the transformation was carried out using the same parent→product kinetics (isothermal transformations). By adjusting the length scale of the simulated parent microstructure, it was ensured that all simulations pertain to the same grain boundary area density.

The grain-size distributions of all employed parent microstructures are shown in Fig. 2 a), including the (extreme) grain-size distribution that results if the space between randomly distributed planes is filled up and defined as grains. The resulting simulated kinetics are shown in Fig. 2 b), together with the predictions by Cahn's model and the JMAK-like model. All simulated kinetics lie in between these two extremes. Only the results of the simulation with nucleation on grain boundaries of a parent microstructure, the grain-size distribution of which is very broad and has its maximum at small grain sizes, comes close to the prediction by Cahn's model. Generally, the broader the parent grain-size distribution is, the more the simulated kinetics deviates from the prediction of the JMAK-like model.

The grain-size distributions of the product microstructures are shown in Fig. 2 c). They differ distinctly from the grain-size distribution that is obtained for a simulation with random nucleation in the bulk. The shape of the parent grain-size distribution doesn't have a strong influence on the product grain-size distribution.

**Influence of the parent→product kinetics.** The results of the simulations for varied parent→product kinetics (parent microstructure generated by an isothermal transformation) are shown in a plot of the simulated transformed fraction versus the transformed fraction according to the JMAK-like model (Fig. 3 a). Thus, any deviation from the diagonal in Fig. 3 a) means a deviation from the kinetics according to the JMAK-like model. The normalised cumulative number of nuclei/grains, as generated in the simulation, has been plotted as function of the simulated transformed fraction in Fig. 3 b). By comparing Figs. 3 a) and 3 b) it follows that the simulated kinetics deviate most from JMAK if the nucleation predominantly or entirely takes place at the beginning of the transformation.

## Discussion

The radial distribution function of nucleation sites, $RDF(r)$, represents the probability of finding one nucleus at a certain distance, $r$, from another nucleus. It equals one for all $r$ for a random arrangement of nuclei (ignoring the size of a nucleus). The $RDF$s for the simulations presented in Fig. 1 are shown in Fig. 1 c). It can be seen that even for high ratios $S_V^{\mathrm{GB}}/\dot{N}_O$, where the simulated kinetics coincide with the JMAK-like model, the $RDF$ does not equal one for all $r$. This can be understood as follows. In the simulations, a constant nucleation rate per untransformed unit volume is assumed throughout the transformation. However, after the specimen has partially transformed, nuclei can only form in untransformed regions and hence can only occur *at a certain minimum distance* from the, already existing, nuclei. Therefore, a *negative* correlation between nucleation positions occurs, indicated by an $RDF$ smaller than

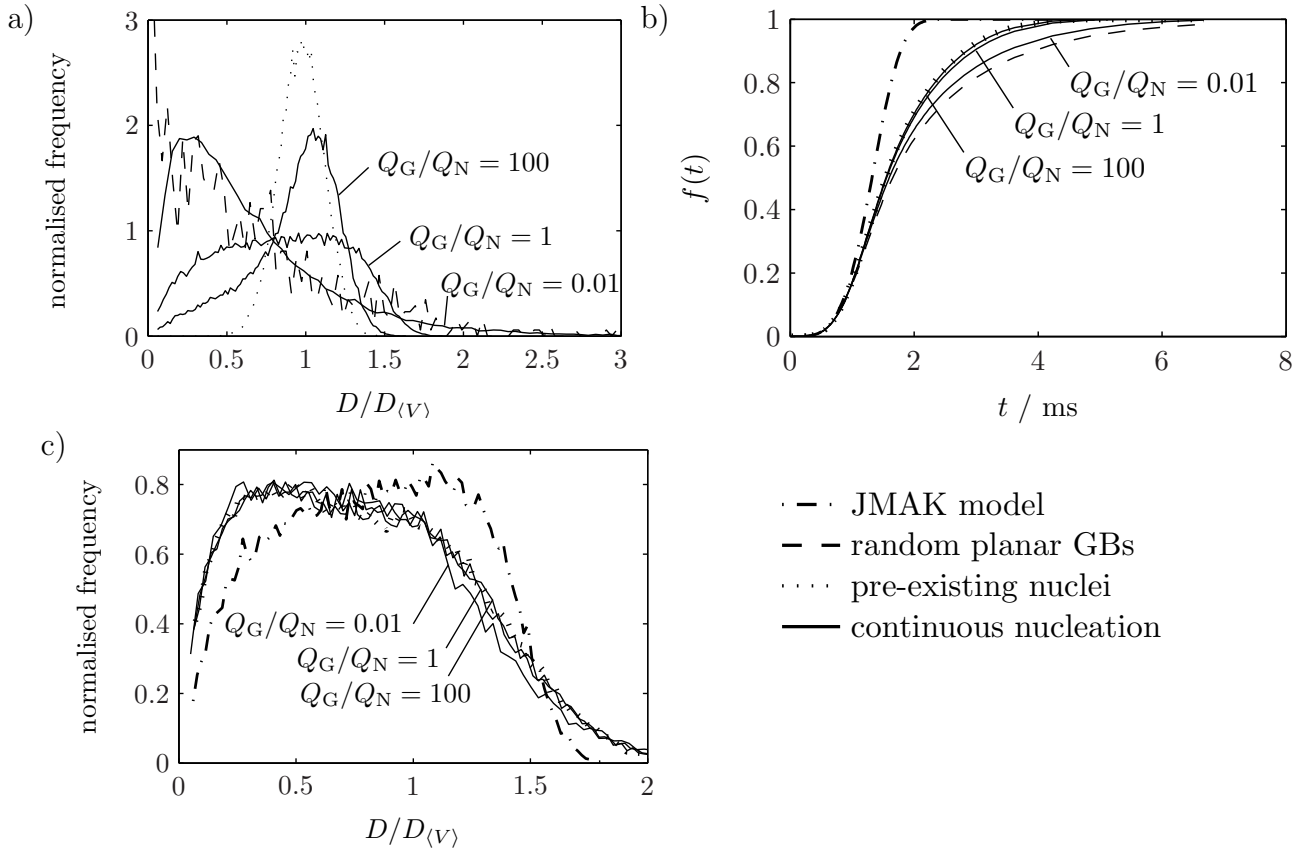


Fig. 2: a) The grain-size distributions of parent microstructures for subsequent simulations. All were derived using the assumption of spatially random nucleation, but varying the parent kinetics. b) The resulting simulated parent→product kinetics. For all calculations, the same constant nucleation and growth rates were assumed. c) The resulting grain-size distributions of the product microstructure.

one at small distances between pairs of nuclei. A truly random arrangement can only be realised by adopting pre-existing nuclei (i.e. all nuclei are present before the onset of tranformation). For decreasing ratios of $S_V^{\mathrm{GB}}/\dot{N}_O$ (and the same nucleation rate $\dot{N}_V$, see above), the same number of nuclei must form on a more and more restricted grain-boundary area and therefore nuclei are more likely to appear close to each other. This *positive* correlation of nucleation positions is exhibited in Fig. 1 c) and is the reason for the retardation of the transformation kinetics as observed (Fig. 1 a): The closer to each other nuclei form, the sooner they impinge which leads to slow transformation kinetics and a high number of small grains.

A similar reasoning can be used to explain that transformations nucleating on parent microstructures with a broad grain-size distribution with many small grains are relatively slow (Fig. 2): Small parent grains lead to many nuclei which are close to each other and impinge early in the transformation. Moreover, for such parent microstructures, the last part of the transformation is further slowed down because there are a number of very large parent grains (which can be transformed only by product grains nucleated at grain boundaries of these grains).

If a substantial part of the nucleation occurs late during the transformation, the negative correlation of nucleation positions, as a consequence of the necessity that nucleation can only occur in untransformed regions, and the positive correlation of nucleation positions, due to their confinement to the remaining, unoccupied parent grain-boundary area, compensate each other partly, so the deviation from JMAK-like kinetics in such a case is not as strong as for pre-existing nuclei (see Fig. 3).

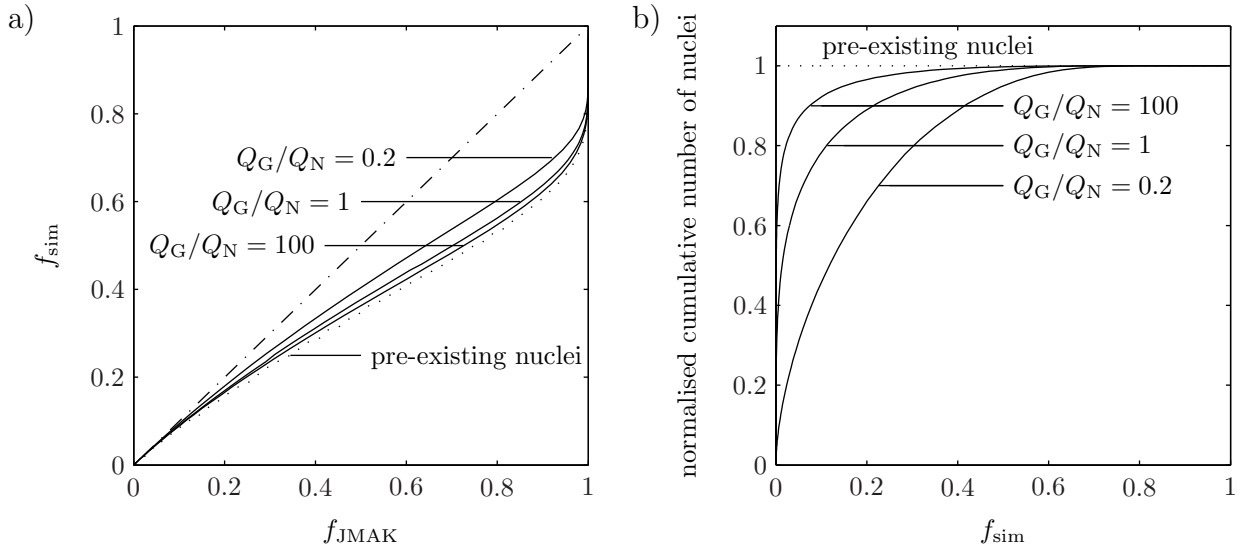


Fig. 3: The results of simulations using the same product microstructure (derived using a constant nucleation and growth rate) and final number of nuclei, but varying the parent→product kinetics. a) Simulated transformed fraction as function of the transformed fraction predikted by the JMAK-like model and b) normalised cumulative number of nuclei/grains during the simulation as function of simulated transformed fraction.

## Conclusions

The kinetics of grain-boundary nucleated phase transformations and the resulting grain-size distributions were analysed by means of gemometrical simulations and compared with a JMAK-like model (assuming spatially random nucleation) and Cahn's model for grain-boundary nucleation. If the grain size of the parent microstructure is smaller than the grain size of the product microstructure (i.e. the ratio $S_V^{\mathrm{GB}}/\dot{N}_O$ is large), the kinetics and resulting microstructure is identical to results obtained assuming spatially random nucleation. If $S_V^{\mathrm{GB}}/\dot{N}_O$ is small, JMAK-like models and Cahn's model for grain-boundary nucleated transformations give upper and lower bounds for the development of the transformed fraction; both models cannot describe the simulated kinetics correctly. The transformation kinetics is most strongly influenced by grain-boundary nucleation if *i*) the parent microstructure has a broad grain-size distribution with many small grains and *ii*) if nucleation takes place predominantly at the beginning of the transformation.

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