


# Simultaneous and sequential transformations with nucleation at preferred sites

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## Abstract

The advent of new materials may increase transformations that involve two or more product phases. This transformation involving two or more product phases may occur simultaneously or sequentially. It is well-known that heterogeneous nucleation has advantages over homogeneous nucleation. Several sites may become preferred because they favor heterogeneous nucleation. The interface of precipitates and the grain boundaries are one of the preferred locations for nucleation. We model simultaneous and sequential transformations nucleated at the preferred sites. This work employs both computer simulation and exact analytical solutions. Two product phases, 1 and 2, were considered. The product phase 1 nucleated at the matrix/particle interface and the product phase 2 nucleated at the grain boundaries. The analytical model showed an excellent agreement with simulation data. For each case, computer simulation obtained the microstructural evolution. We discussed the effect of nucleation at the preferred sites on the microstructure.

**Keywords:** Phase transformations; Recrystallization; Particle Stimulated Nucleation (PSN); Computer simulation.

## 1 Introduction

Many phase transformations occur, in which a matrix phase transforms into just one product or phase. In the recrystallization context, the recrystallization of a deformed matrix into a product without deformation. However, the continuous advent of new materials may increase transformations that involve two or more product phases. When the matrix phase transforms into two or more product phases, this transformation can be simultaneous or sequential. This transformation will be simultaneous when the product phases start simultaneously and sequentially when the product phases start at different times. An analytical model that can treat simultaneous and sequential transformations was developed by Rios and Villa [1]. Many computer simulations of simultaneous and sequential transformation were studied by Alves et al. [2].

The diffusional phase transformations are also known as transformations by nucleation and growth. Therefore, one of the most critical steps in phase transformations is nucleation. It is well-known that heterogeneous nucleation typically has a kinetic advantage over homogeneous nucleation. Several sites may become preferred in this context, as they favor heterogeneous nucleation. The surface of precipitates and the grain boundaries are preferred sites. The nucleation at these two preferred sites was observed by Goetz [3] in

dynamic recrystallization. The nucleation on second phase particles has likewise been observed at particles that were formed inside the weld metal [4-6] or in the recrystallization with the occurrence of the Particle Stimulated Nucleation (PSN) [7-10]. Recently, Alves et al. [11] developed an analytical model to treat the transformations that nucleate at the interface of spherical particles. Their model is also valid for PSN.

This work employs analytical and computational methods to simulate simultaneous and sequential transformations that have nucleated at the grain boundaries and the interface of spherical particles. We considered the transformation of a parent polycrystalline matrix with spherical particles into two product phases: phase 1 and phase 2. The product phase 1 nucleated at the matrix/particle interface and the product phase 2 nucleated at the grain boundaries. We discuss the effect of the nucleation at the parent matrix's preferred sites, which influences the microstructure and materials properties.

## 2 Methodology

The Causal Cone method [12] was adopted in the simulations. The simulations employed a cubic matrix

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of 300 x 300 x 300 cubic cells with periodic boundary conditions. The length of each cell was approximately  $0.33\mu\text{m}$ . Therefore, the matrix was  $100\mu\text{m}$  long in each direction. As was mentioned above, a parent polycrystalline matrix with spherical second phase particles was used as a base matrix for the transformation. Figure 1 shows this base matrix. The mean diameter of the spherical particles was  $5\mu\text{m}$  and 153 particles filled 1% of the matrix volume. Within the cubic matrix, 3376 nuclei were nucleated at the matrix/particles interface. Therefore, the mean grain size at the end of the transformation was  $10\mu\text{m}$ , approximately. Alves et al. [11] developed the methodology to design this polycrystalline matrix.

The polycrystalline matrix transforms into two distinct product phases, called phase 1 and phase 2. This phase transformation occurs simultaneously or sequentially. Both product phases had the same initial number of nuclei,  $N_1 = N_2$  and the same growth velocity,  $G_1 = G_2$ . Phase 1 nucleated at the matrix/particle interface and phase 2 nucleated at the grain boundaries. The nucleation was by site saturation and the growth velocities were constant in both cases. The available volume for the transformation is the free volume of the polycrystalline matrix, i.e., the regions out of the particles. There was no growth inside the second phase particles. In the sequential case, the incubation time for phase 2 was  $V_{I1} = 0.1$ .

A homogeneous Poisson Point Process defined the nucleation of both product phases [13,14]. Thus nuclei are stochastically independent. Less precisely, all nuclei were uniform randomly distributed in space.

Based on this stochastic independence, Rios and Villa introduced the superposition principle [1]. Thus, any model that was initially developed for the transformation involving just one product phase can describe the situation in which two or more product phases occur simultaneously or sequentially.

For the cases treated in this work, the product phase 1 nucleated at the matrix/particle interface; thus, it could be treated by the Alves et al. [11] model. On the other hand, JMAK's formalism [15-18] described the product phase 2 nucleated at the grain boundaries. This behavior is in agreement with previous work [19]. It is worthy of note that the equations were not shown here for brevity's sake. Instead, the reader is referred to [1,11,15-17] for more details about the models.

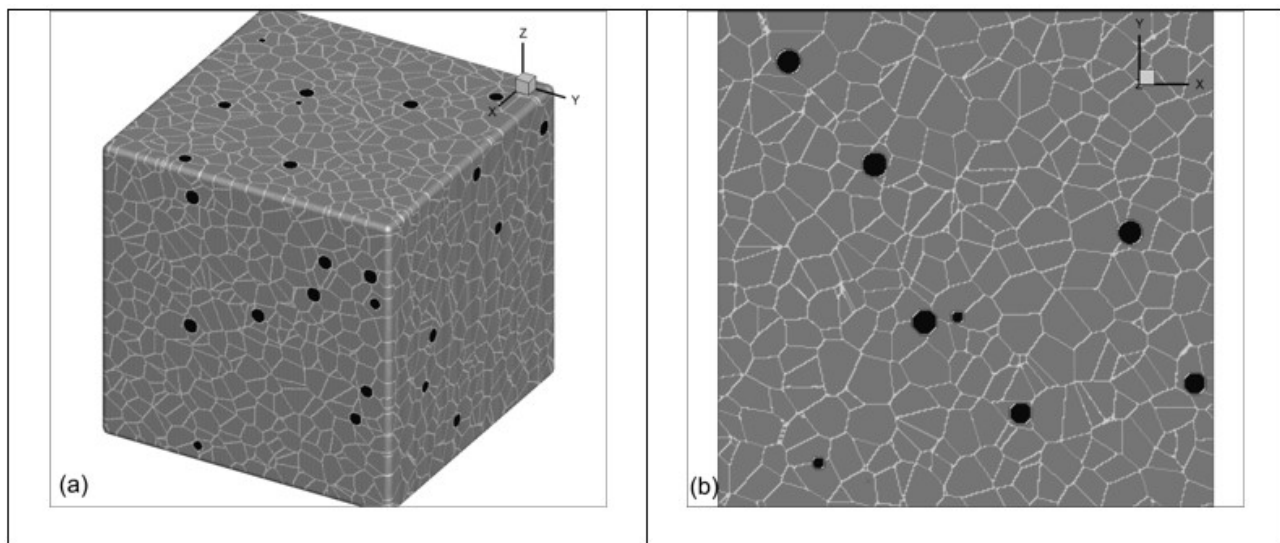
### 3 Results and discussion

The computational data were compared with the analytical methods mentioned above. One compares them utilizing the kinetics curves, mean volume fraction,  $V_V$ , against time, and the microstructural path, mean surface area between recrystallized and non-recrystallized material per unit of volume,  $S_V$ , versus  $V_V$ . It is important to note that in all cases, the coefficient of determination,  $R^2$ , was invariably greater than 0.99. For each case, one shows the simulated microstructure evolution. In all simulated microstructures, the product phase 1 is in light grey, phase 2 is depicted in dark grey, and the second phase particles are black.

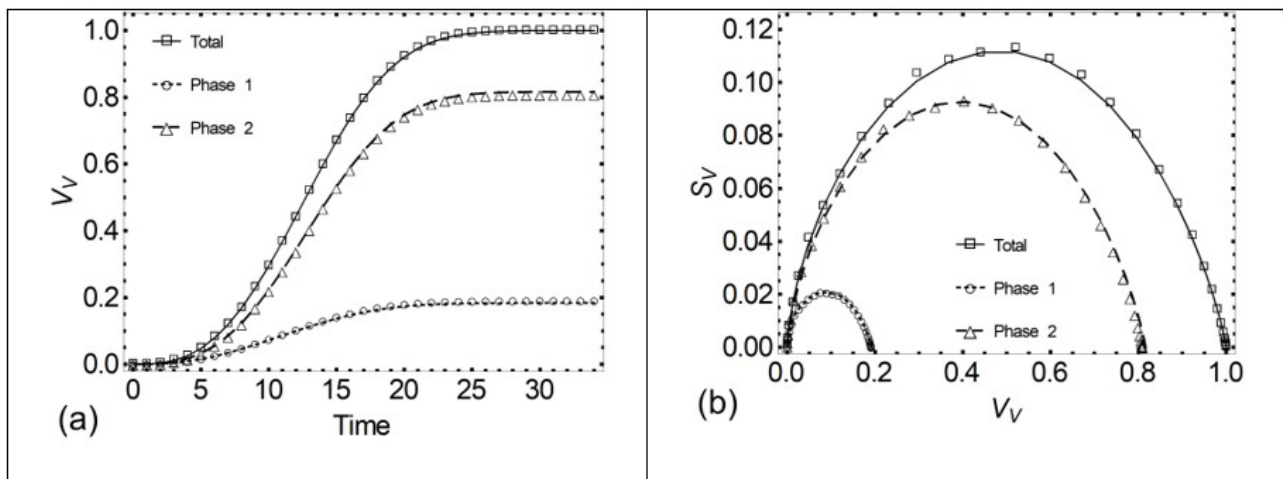
#### 3.1 Simultaneous transformations

The simultaneous transformation with the number of nuclei  $N_1 = N_2 = 1688$  takes place. The sum of both product phase nuclei was equal to the total number of nuclei of the previous polycrystalline matrix, shown in Figure 1. Therefore, the achieved mean grain size was about  $10\mu\text{m}$ . Figure 2 shows the kinetic curves and the microstructural path.

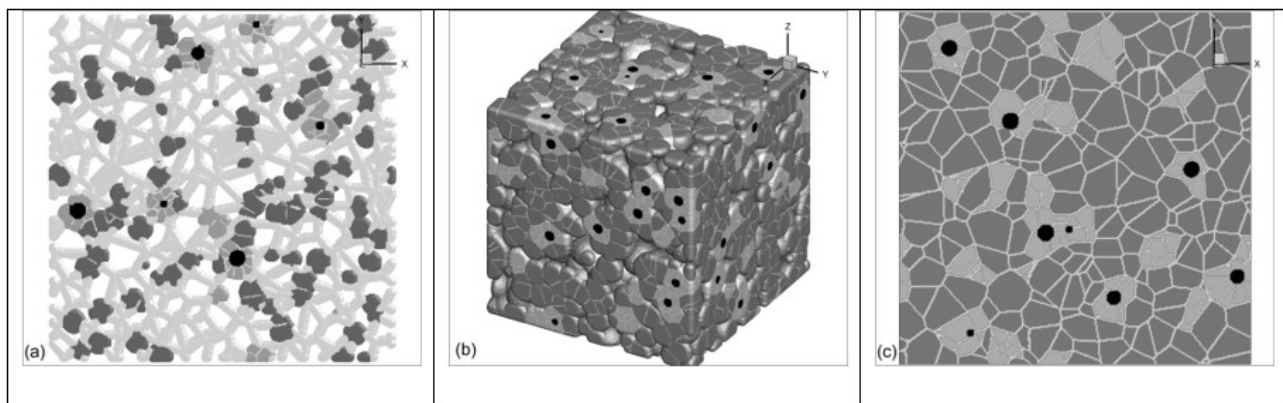
As one can see, there was an excellent agreement between the computer simulation results and the analytical model. Figure 3 shows the simulated microstructure evolution.



**Figure 1.** Representation of the parent polycrystalline matrix before transforming into the two product phases. (a) 3d; (b) 2d section.



**Figure 2.** Simultaneous transformation with  $N_1 = N_2 = 1688$  and  $G_1 = G_2$ . Phase 1 nucleated at the matrix/particle interface and phase 2 nucleated at the grain boundaries. Dots represented the data from the simulations, and lines represented the analytical results. Phase 1 was analytically modeled by the Alves et al. [11] model. Phase 2 was analyzed using the JMAK [15-17] model. (a)  $V_V$  versus time; (b) Microstructural path.



**Figure 3.** Simulated microstructure of the simultaneous transformation shown in Figure 2. (a)  $V_V = 0.1$  showing the nuclei position. (b)  $V_V = 0.5$ . (c) 2d cross-section of the fully transformed matrix.

In Figure 3a, the grains of phase 1 are close to the particles, and the grains of phase 2 are close to the grain boundaries (the tessellation in Figure 3a). This emphasizes the preferred sites for nucleation. From Figure 3a, for  $V_{V1} = 0.1$ , it is difficult to distinguish the grain size of each product phase; this may highlight that both phases nucleated simultaneously.

Figure 2a exhibits unusual behavior. Although both phases nucleated together and have the same parameters, they don't equally contribute to the transformation. Rios and Villa [1] observed that in a simultaneous transformation with the same parameters, at the end of the transformation, one expects  $V_{V1} = V_{V2} = 0.5$ . In our case, each phase's distinct contribution occurred because they nucleated at different sites. Phase 1 nucleated at the matrix/particle interface, and the particles affected its growth. There was no growth inside the second phase particles, as mentioned before. Therefore, phase 1 grains can't keep their spherical shape at the beginning of the transformation due to an early

impingement with the same phase nucleated at the same particle. This simulation employed 153 particles with 1688 nuclei for phase 1. The mean number of nuclei per particle is greater than 10. The higher the mean number of nuclei per particle, the greater the probability that the nuclei forming at the matrix/particle interface impinge, as reported by Alves et al. [11]. One can easily observe this early impingement in Figure 3a. Figure 3a shows that the grains of phase 1 quickly envelop the particles and grow "squeezed" against each other.

Phase 2 nucleated at the grain boundaries. Therefore, phase 2 has a lower probability of finding a neighbor grain at the beginning of the transformation than the grains of phase 1. Hence, phase 2 suffers less from the impingement than phase 1. This fact can be evidenced by the larger grain size of phase 2, see Figure 3c. Phase 1 impinged both particles, and the grains nucleated at the same particle. By contrast, Figure 2a shows that phase 2 dominates the transformation.

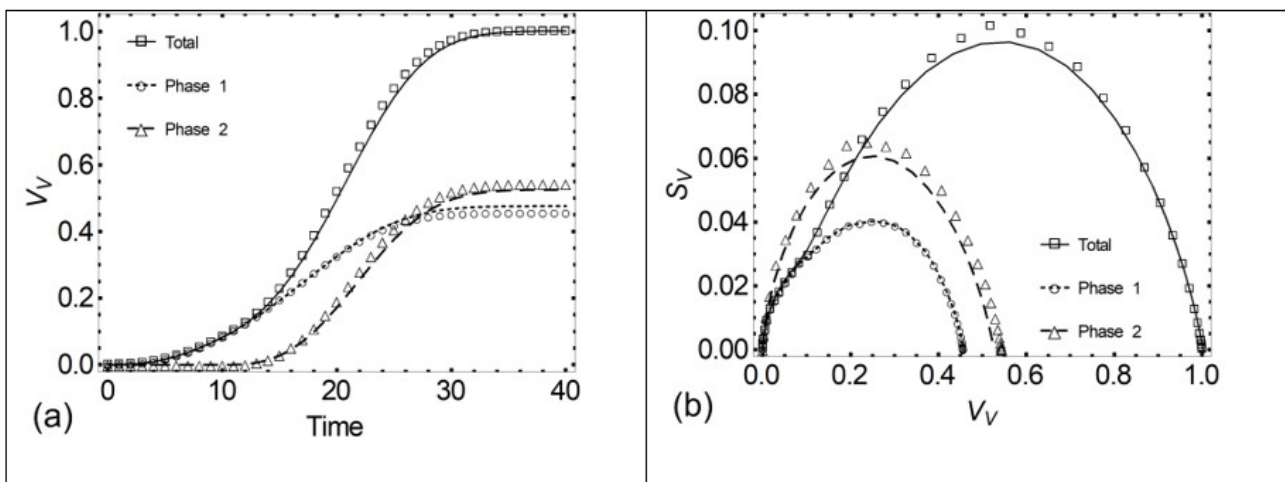
Figure 2b shows the microstructural path. As one can see, the same impingement effect was identified in  $S_V$ .  $S_{V1} < S_{V2}$  since part of the grains of phase 1 was in contact with the particles.

### 3.2 Sequential transformations

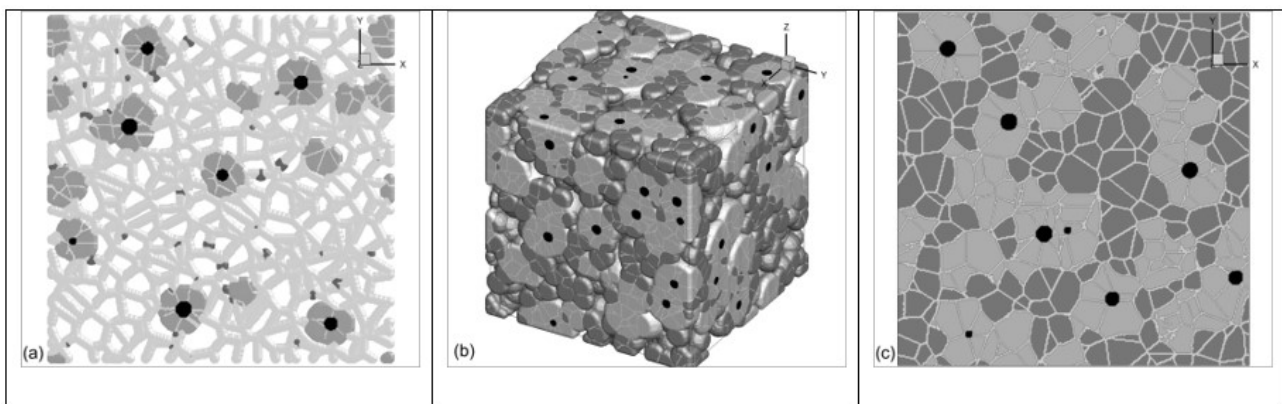
This section considers the sequential transformation with the same parameters as the previous section, which treated the simultaneous transformation. Therefore, one considered the number of nuclei  $N_1 = N_2 = 1688$  and  $G_1 = G_2$ . The only difference was the incubation time for phase 2. Phase 2 nucleates later, when  $V_{V1} = 0.1$ . Figure 4 shows the kinetic curves and the microstructural path for this sequential transformation. As in the previous case, one can see that there was also an excellent agreement between the computer simulation results and the analytical model. A different behavior than the previous behavior observed by Rios and Villa [1] was seen here too. Rios and Villa [1] observed that

when a sequential transformation, in which the parameters of both product phases are the same and the nuclei of these product phases are uniform randomly distributed, takes place, a slight advantage for phase 1, say the nucleation of phase 2 when  $V_{V1} = 0.1$ , causes a predominance of phase 1. In fact, their results showed that in these conditions phase 1 filled about 80% of matrix volume in the end of the transformation. In our case, see Figure 4, the critical difference is that the nucleation takes place at the preferred sites. The phase 1 nucleated at the matrix/particle interface and phase 2 nucleated at the grain boundaries. As was mentioned in the previous section, phase 1 impinges more than phase 2. Owing to the effect of impingement, phase 2 reaches the volume fraction of phase 1, even though it nucleated later than phase 1.

Figure 5 shows the simulated microstructure evolution. One can see that islands of phase 1 grains involved the second phase particles. Figure 5a empathizes the nucleation



**Figure 4.** Sequential transformation with  $N_1 = N_2 = 1688$  and  $G_1 = G_2$ . Phase 1 nucleated at the matrix/particle interface and phase 2 nucleated at the grain boundaries when  $V_{V1} = 0.1$ . Dots represented the data from the simulations, and lines represented the analytical results. Phase 1 was analytically modeled by the Alves et al. [11] model. Phase 2 was analyzed using the JMAK [15-17] model. (a)  $V_V$  versus time; (b) Microstructural path.



**Figure 5.** Simulated microstructure of the sequential transformation shown in Figure 4. (a)  $V_V = 0.1$  showing the nuclei position. (b)  $V_V = 0.5$ . (c) 2d cross-section of the fully transformed matrix.

of phase 1 at the matrix/particle interface and the nucleation of phase 2 at the grain boundaries when  $V_{V1} = 0.1$ .

It should be noted that the shape and grain size of the previous polycrystalline matrix may affect the transformations in which the nucleation takes place at the grain boundaries. This effect is outside the scope of this work. However, it has been studied previously [19].

#### 4 Conclusion

Computer simulation and analytical models were employed to investigate simultaneous and sequential transformations that nucleated at preferred sites. One parent polycrystalline matrix with spherical second phase particles transforms into two product phases, 1 and 2, simultaneously or sequentially. Phase 1 nucleated at the matrix/particle interface and phase 2 nucleated at the grain boundaries. Our results permitted us to conclude that:

- All computer simulations showed an excellent agreement with the analytical models. No curve fitting was done in this work. In all cases, the coefficient of determination,  $R^2$ , between the simulation results and the analytical results was invariably greater than 0.99. The simulated microstructure evolution was obtained for each case. This is the main advantage of computer simulation and may help the researchers better understand and infer the characteristics of simultaneous and sequential transformations;
- The simulated microstructures showed that nucleation at each preferred site caused a significant change in

the microstructure, which may affect the material properties. Each preferred site for nucleation has a very distinct effect on transformation behavior. This was also evidenced by the kinetic curves;

- Phase 1, nucleated at the matrix/particle interface, had its growth more affected by impingement. This impingement effect resulted in a significant predominance of phase 2 in simultaneous transformation. In the sequential transformation, phase 2 could still reach the volume fraction of phase 1, even though phase 2 nucleated later;
- Phase 2, nucleated at the grain boundaries, had its growth less affected by impingement than phase 1. The grain boundary area is larger than the particle's surface area. Therefore, phase 2 has less probability of finding a neighbor grain than phase 1. Thus, the impingement has a more negligible effect on phase 2 than on phase 1.

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