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On the Zener limit of grain growth through 2D Monte Carlo simulation

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ABSTRACT

Extensive 2D Monte Carlo simulations were carried out on a square lattice to verify the Zener limit in the presence of inert second phase particles. Matrices of various sizes (from 100 to 10,000) were first run to stagnation to investigate the effect of matrix size on the limiting grain size, scaling constant and the fraction of second phase particles interacting with the grain boundaries (ϕ). The optimum matrix size selected was then run to stagnation for a wide range of particle fractions, at different *Q-States*, to arrive at limiting grain size and other parameters. Larger matrices ($\geqslant$1000) were found to be more suitable than smaller matrices to carry out simulation studies, while there was negligible effect on limiting grain size upon any variation in the *Q-States*. Based on our studies of particle-pinned regimes, a new relationship between the Zener limit and the fraction of second phase particles lying on the grain boundaries, i.e. $R(lim) \propto 1/\exp(\phi)$, is proposed, while the limiting grain size observed obeys a square root dependence on the particle fraction, agreeing largely with the earlier simulation results in 2D.

1. Introduction

It has been more than six decades since Cyril Stanley Smith published a seminal paper [1] which outlined the fundamental principles of interpretation of microstructures, in terms of limited grain sizes, due to interaction between second phase particles and moving grain boundaries. The paper included a paragraph taken from a letter written by Zener to Smith referring to grain growth in particle-containing materials which has since then become a subject of several analyses and further refinements. Zener proposed that the driving pressure for grain growth due to grain boundary curvature would be counteracted by the pinning pressure exerted by the second phase particles on the boundary. As a result, normal grain growth would be completely inhibited with the grain size reaching a critical maximum grain radius ($R(lim)$) given by:

$$R(lim) = \frac{4r}{3f} \tag{1}$$

where $R(lim)$ is called the limiting grain size (or the Zener limit), r is the mean radius of the pinning particles and f is the volume fraction of second phase particles present in the matrix. This equation is known as Smith–Zener equation and is rewritten in a more generic form as:

$$R(lim) = k f^{\frac{r}{m}} \tag{2}$$

where k is a dimensionless scaling constant and m is an index for f. Eq. (2) becomes the Smith–Zener Eq. (1) when $k = 4/3$ and $m = 1$.

In the process of deriving the theoretical limit for particle-pinned maximum average grain size, Zener made several assumptions regarding the particle-grain boundary interaction and particle distribution. Ever since Zener and Smith, their limiting grain size equation has been investigated thoroughly, both theoretically and experimentally. Several modifications have been suggested by subsequent scientists who have carried out experiments on a wide variety of metals containing different shapes, sizes, distributions and volume fractions of second phase particles. Similarly the Zener limit has also been verified, over the years, through both 2D as well as 3D simulations [2–11]. The results of a few simulations, investigating the Zener limit, have been summarized in Table 1.

Almost all 2D simulation results (see Table 1) [2–9] have indicated that the limiting grain size scales inversely with the square root of the impurity fraction, while most 3D simulation results [4,7,10] with the exception of Miodownik et al. [11], have suggested a cube root dependence of the Zener limit on the impurity fraction.

Looking from the 2D simulation perspective, which is the focus of this paper, it can be observed that almost all results have offered similar modifications to the Zener limit and vary among them, only with respect to the scaling constant (k), as given in Eq. (2). The fraction of second phase particles lying on the grain boundaries (ϕ)

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Table 1
Modifications to the Zener limit over the years through 2D & 3D computer simulations.

Sl. no.	Refs.	Equation for limiting grain size	Comments
1.	Smith [1]	$R(lim) = 1.33\ r/f$	Theoretical result assuming randomly distributed mono-sized spherical particles.
2.	Srolovitz et al. [2]	$R(lim) \approx r/f^{0.5}$	A 2D simulation result working on a 150 × 150 triangular lattice.
3.	Doherty et al. [3]	$R(lim) = 1.7\ r/f^{0.5}$	Result of 2D Monte Carlo simulation with a non-random particle distribution.
4.	Hazzledine and Oldershaw [4]	$R(lim) = 1.8\ r/f^{0.5}$	A 2D computer simulation result.
5.	Hassold et al. [5]	$R(lim) = 0.6\ r/f^{0.55}$	Result of a 2D MC simulation on a 200 × 200 triangular lattice where $f \leqslant 0.1$.
6.	Gao et al. [6]	$R(lim) = 0.59\ r/f^{0.52}$	A 2D simulation on a 500 × 500 triangular lattice considering the degree of contact between the particles and the grain boundaries.
7.	Kad and Hazzledine [7]	$R(lim) = 1.41\ r/f^{0.5}$	Result of a 2D computer simulation in the presence of second phase particles of various shapes such as spheres, needles and plates.
8.	Soucail et al. [8]	$R(lim) \propto \{1/(f)^{0.5}\}$	Result of 2D simulation where f is the fraction of second phase particles actually in contact with the grain boundaries.
9.	Huang et al. [9]	$R(lim) \propto 1/f^{0.5}$	A 2D simulation on a 200 × 200 lattice.
10.	Anderson et al. [10]	$R(lim) = 4.5 \pm 0.8\ r/f^{0.31\pm0.02}$	The first effort on 3D simulation of the Zener limit yielded a cube root dependence on the particle fraction, while working on hexagonal lattice of 100^3 size, with f varying from 0.005 to 0.16.
11	Hazzledine and Oldershaw [4]	$R(lim) \propto r/f^{0.33}$	A 3D simulation result when $f > 0.01$.
12.	Kad and Hazzledine [7]	$R(lim) \propto 1/f^{0.3}$	3D Potts model simulation on a 235 × 235 × 235 size fcc lattice pinned by particles of different shapes such as spheres, needles and plates.
13.	Miodownik et al. [11]	$R(lim) = 0.728\ r/f^{1.02}$	Result of 3D simulation on 200^3 & 400^3 matrix size square lattices, with simulation temperature $kT = 1$, particle size of 27 pixels, and $0.025 < f < 0.15$.

was brought into the Smith–Zener equation, probably for the first time, by Soucail et al. [8]. They proposed that the limiting grain size scaled with $1/\sqrt{\phi}$ although Srolovitz et al. [2] and Doherty et al. [3] had earlier discussed briefly the effect of ϕ on the limiting grain size. It is also noticed that most simulations have been run on matrices of lower sizes ($\leqslant$500) with notable exceptions being Kad and Hazzledine [7] and Soucail et al. [8] who have worked on matrices of sizes up to 2000 × 2000. To avoid finite system size effects and generate good statistics for the pinned grain size, the pinned size ($R(lim)$) must be less than one third of the matrix size [11]. When smaller matrices are deployed with low surface fractions of second phase particles and run to stagnation, they grow to large grain sizes and tend to violate the minimum size requirements. The present work has explored matrices of various sizes in order to select the optimum size, before investigating the Zener limit.

2. Monte Carlo Potts model simulation method

The Potts model based on Metropolis algorithm and periodic boundary condition, which is now a well established technique for the Monte Carlo simulation of grain growth [2,12,13], was employed in the present study. A square lattice with eight nearest neighbors was considered since a review of the literature indicated not so substantial work on this type in comparison with the hexagonal lattice. The effect of lattice temperature was ignored and the simulations were run at steady state, i.e. at $T = 0$.

A continuum microstructure is bitmapped onto a two-dimensional lattice initially taking the form of a square matrix populated with random numbers ranging from 1 to Q, where Q represents the number of grain orientations. A matrix element is chosen at random and is compared with its nearest neighbors to compute the number of unlike neighbors. The chosen element is then flipped to a new random number ranging from 1–Q, and the number of unlike neighbors is computed once again. If the difference between the unlike neighbors of the flipped and the original elements (ΔE) is either zero or negative, the flipped element replaces the original element in the matrix, else the original element is retained. N^2 such iterations form one Monte Carlo Step (*MCS*), where N is the matrix size and *MCS* the measure of time. The second phase particles are introduced into the matrix as random static elements which do not participate in the coalescence of grains, rather preventing it.

3. Hardware and software

All simulations were run on a Pentium i-5 processor with a 16 GB RAM built on an ASUS Motherboard B3 Model. A highly parallel version of the code that was capable of high speeds was evolved on Java platform. However, the whole simulation process took more than 2 weeks of computer time, with the stagnation of 10,000 × 10,000 size matrix alone taking 147 h of non-stop CPU time.

The code that was developed to record the limiting grain size was done in such a way that the simulations would stop automatically once the Hamiltonian energy value, as calculated from the Metropolis algorithm, would not change continuously for a run of 5000 *MCS*. The limiting grain size was calculated only once i.e. when the simulation ended, since computing it in between would have taken away considerable CPU time. Therefore, the reduction of Hamiltonian, which indirectly indicates grain growth, was taken as a measure to signal grain growth stagnation, with the help of its own stagnation. The second phase particles were all given a uniform size of one pixel and made static by giving a *Q-State* value of *zero*.

4. Results and discussions

The effect of matrix size on the limiting grain size and other parameters was first investigated by running various matrices (of sizes ranging from 100–10,000) to stagnation considering constant sample values of *Q-state* as 64 and fraction of second phase particles (f) as 0.1. For all matrix sizes less than 1000, all the parameters that were computed were averaged over three trials. The various parameters considered were:-the limiting grain size ($R(lim)$)-calculated as *sqrt* (A/π) where A is the mean area of all grains at stagnation, the largest grain size at stagnation ($R(max)$), scaling constant (k) assuming $m = 0.5$, and lastly the fraction of second phase particles actually lying on the grain boundaries (ϕ). ϕ was computed considering the inert particles which lay on the edges of each grain and computed as a fraction of f.

Figs. 1a–d show the variations of the grain growth parameters against increasing matrix sizes. The largest grain size at stagnation as seen in Fig. 1b is about 2–3 times the corresponding stagnating averaging grain size as seen in Fig. 1a, indicating normal grain growth. More importantly, it can be observed that at higher matrix sizes ($N \geqslant 1000$), all the parameters seem to be stable, although

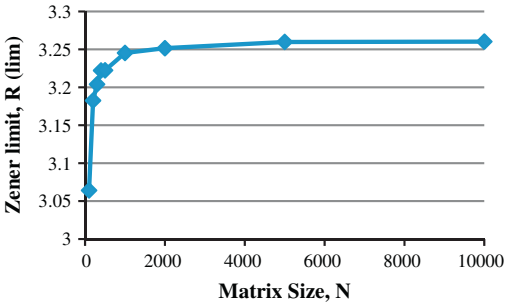

Fig. 1a. Limiting grain size (in pixels), ($R(lim)$) vs. N.

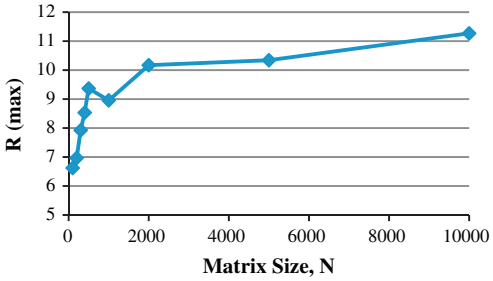

Fig. 1b. Largest grain size (in pixels), R(max) vs. N.

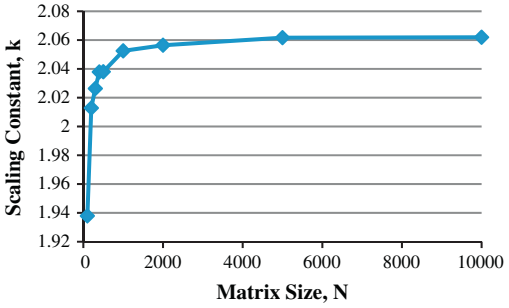

Fig. 1c. Scaling constant, k vs. N.

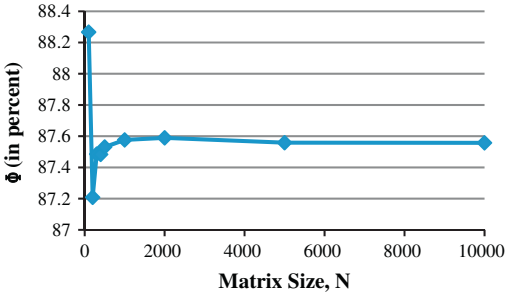

Fig. 1d. Fraction of f interacting with grain boundaries, (ϕ) vs. N.

the variations at lower matrix sizes seem to be nominal. However, collective observations suggest that it would be better to select a larger matrix ($N \geqslant 1000$) to carry out Monte Carlo simulation of grain growth, which would remove any adverse size effects. This observation is consistent with the results obtained by Kad and Hazzledine [7] who suggested that a minimum matrix size of 500 is desirable for better simulation results.

The selected optimum size ($N = 1000$) was then run to stagnation for three different *Q-States* (32, 64 and 127) for a host of

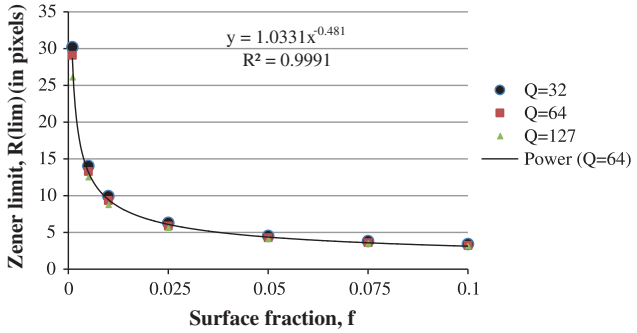

Fig. 2. The Zener limit, ($R(lim)$) vs. f, for various *Q-States*.

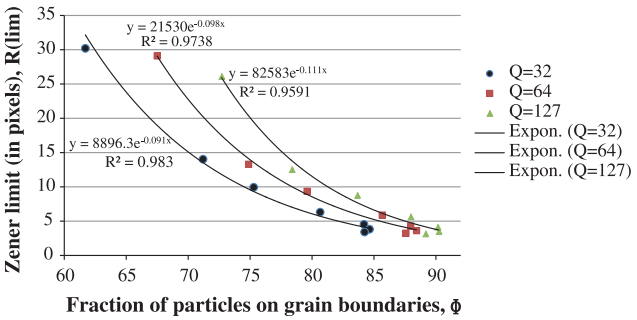

Fig. 3. The Zener limit, ($R(lim)$) vs. ϕ, for different *Q-States*.

surface fractions of second phase particles ranging from $f = 0.001$ to 0.1. From Fig. 2, it is evident that varying *Q-States* has little effect on the limiting grain size. It is also very well established from Fig. 2 that the $R(lim)$ scales inversely as the square root of f. Observing the near-perfect fit that the limiting grain size values contribute to the power equation and the excellent correlation ($R^2 = 0.9991$) between the two parameters, it is suggested that the Zener limit, under 2D investigation, may be given a new modification as follows:

$$R(lim) = \frac{1.0331r}{f^{0.481}} \tag{3}$$

Approximating the value of k to be unity, the proposed modified Zener limit can be rewritten as,

$$R(lim) = \frac{r}{f^{0.48}} \tag{4}$$

The proposed new value of the Zener limit comes closest to the value suggested by Srolovitz et al. [2], as can be observed from Table 1. The only difference, however, is that Srolovitz et al. considered a triangular lattice while the present work considered a square lattice. Most 2D simulations to date, including ours, have produced consistent results over the years, unlike 3D simulations, which have thrown Zener limits scaling with f ranging from $1/\sqrt[8]{f}$ [4,7,10] to $1/f$ [11]. This is due to the fundamental difference in the way particles remove curvature completely in 2D but not in 3D, and hence the inconsistency [13].

Fig. 3 shows the variation of limiting grain size against ϕ, corresponding to various surface fractions f, which increase from left to right. We can clearly see from Fig. 3 that the Zener limit scales inversely but somewhat exponentially with ϕ, as demonstrated with reasonable correlations. This is contrary to Soucail et al. [8] who suggested that the Zener limit varies as $1/\sqrt{\phi}$. The proposed exponential relationships are again in contrast to the experimental results by Stearns and Harmer [14] who have reported the relation between $R(lim)$ and ϕ to be linear. Our work suggests that at higher values of f, there is an exponential increase in the number of parti-

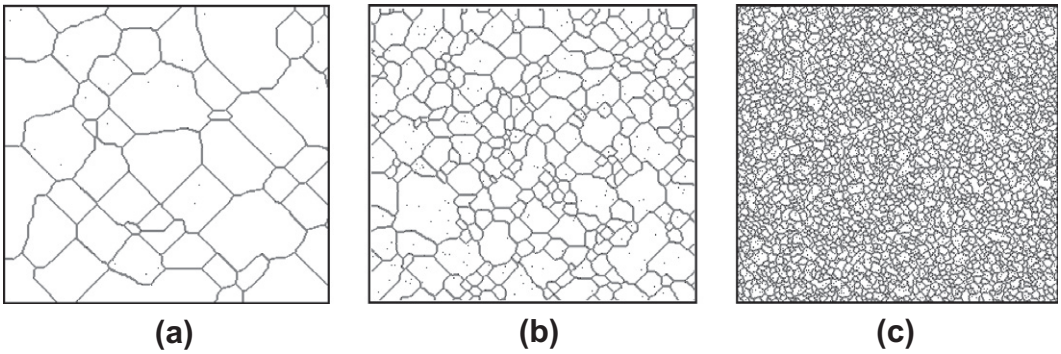

Fig. 4. Pinned microstructures at: (a) $N = 1000$, $Q = 64$, $f = 0.001$, *MCS* (at stagnation) = 506,076, (b) $N = 1000$, $Q = 64$, $f = 0.01$, *MCS* (at stagnation) = 159,151, and (c) $N = 1000$, $Q = 64$, $f = 0.1$, *MCS* (at stagnation) = 52,226.

cles interacting with the grain boundaries, than at lower values of f. This could be because higher values of f lead to lower values of $R(lim)$ and hence higher grain boundary area. This leads to a higher incidence of particles on the grain boundaries, at a rate which is more than linear since the variation of $R(lim)$ vs. f is also non-linear.

Fig. 4a–c show a few simulated microstructures of the pinned regimes with relevant details. It is clear from the pictures that the average grain size decreases with increase in surface fraction of second phase particles. The number of Monte Carlo steps required to bring about stagnation decreases with increasing particle fraction because of enhanced grain boundary pinning at higher values of f.

5. Conclusions

1. It was found that a matrix of a minimum size of 1000 is desirable for better results while carrying out Monte Carlo simulation of grain growth in two dimensions.
2. A new modification is suggested to the Smith–Zener equation under 2D simulation investigation, for values of surface fractions, $0.001 \leqslant f \leqslant 0.1$, i.e. $R(lim) = \frac{r}{f^{0.48}}$
3. The Zener limit scales inversely and exponentially with the fraction of second phase particles interacting with the grain boundaries, $(R(lim) \propto 1/\exp(\phi))$.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.commatsci.2012.02.013.

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