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Microstructural Descriptors and Cellular Automata Simulation of the Effects of Non-random Nuclei Location on Recrystallization in Two Dimensions

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The effect of non-random nuclei location and the efficiency of microstructural descriptors in assessing such a situation are studied. Cellular automata simulation of recrystallization in two dimensions is carried out to simulate microstrutural evolution for nuclei distribution ranging from a periodic arrangement to clusters of nuclei. The simulation results are compared in detail with microstrutural descriptors normally used to follow transformation evolution. It is shown that the contiguity is particularly relevant to detect microstructural deviations from randomness. This work focuses on recrystallization but its results are applicable to any nucleation and growth transformation.

Keywords: *microstructure, kinetics, recrystallization, computer simulation, cellular automata*

1. Introduction

Microstructural evolution during recrystallization as well as during other nucleation and growth reactions is normally studied by means of microstructural descriptors obtained by quantitative metallography measurements carried out on a planar section. The most fundamental descriptor is the volume fraction, that constituted the basis of the early formal theories of Johnson-Mehl[1], Avrami[2-4] and Kolmogorov[5], the JMAK theory. In this theory, nucleation is assumed to take place on sites randomly located in the matrix. This assumption is important because randomness allows one to use an exact geometrical relationship to take care of the impingement. When JMAK theory is applied the question that is always asked is whether or not the nuclei are randomly dispersed. The standard answer is that if the equations based on the assumption of random nuclei are obeyed then the assumptions are likely to be valid. Later, the area per unit of volume between recrystallized regions and non-recrystallized matrix was introduced by Cahn and Hagel[6] and by DeHoff[7]. DeHoff was the first to propose the concept of microstructural path. He proposed that recrystallization follows a path in the S_V *vs.* V_V space. The microstructural path method, MPM, has been subsequently developed and extensively employed[8], notably by Vandermeer and coworkers in many excellent papers[9-11]. More recently, the interface area per unit of volume between recrystallized grains[8,10,11] has been measured. Vandermeer and coworkers[10-12] have shown that using these measurements the mean intercept length of transformed grains[10,13] and the contiguity[11,12] can be obtained and these can be useful descriptors which are particularly sensitive to deviations from randomness. There is more than one way that the location of nucleation sites can deviate from randomness. One possibility is that the nuclei tend to a periodic arrangement. Price[14] has treated this case. Another possibility is the occurrence of clusters of nuclei on grain edges and faces[15]. Also, nucleation may be more concentrated in certain parts of bulk volume than in others[16,17]. In recent papers[18-21], computer simulation has been used to investigated the kinetics in conditions that depart from randomness.

In a previous paper[22], cellular automata simulation of recrystallization in two dimensions was compared in detail with mathematically exact analytical theories considering both kinetic and geometrical aspects. Very good agreement was observed between the cellular automata simulation and the theoretical results. The simulation allows precise data to be generated, without the often substantial experimental errors which are unavoidable in recrystallization data. This allows one to focus on the geometrical issues of the transformation and apply the theoretical results with confidence in real materials.

In this work, site-saturated recrystallization is simulated by using cellular automata[18-26] in two dimensions in order to investigate the effect of nuclei distribution on the kinetics. Rios et al.[21] carried out a preliminary study of the effect of periodically located nuclei on the kinetics. Here, that work is expanded and simulations are carried out for nuclei located randomly, periodically and clustered within certain areas of the microstructure. The results are assessed by means of microstructural path descriptors[11,22]. Although this work has recrystallization as main focus its results are general, valid for any nucleation and growth transformation complying with the assumptions of the simulation.

2. Description of the Simulation

Cellular automata methodology was used to simulate recrystallization. The implementation followed that of Hesselbarth and Göbel[21-23,26] using the von Neumann neighborhood criterion. The matrix consisted of a square lattice with 812 x 812 cells and 784 nuclei. One cell was considered to have unit area and consequently the side of a single cell had unit length. The units of all quantities reported here follow from this. The number of nuclei per unit of area, N_A, was equal to 1/841. The matrix size and number of nuclei were chosen for reasons described elsewhere[21,22,26]. The nucleation was site-saturated: all nuclei appeared at $t = 0$. In previous work[22] the nuclei were randomly located in the matrix. In this work in addition to the random nuclei distribution, the nuclei were arranged periodically[21] and in clusters. For the periodic arrangement they were located in the center of "boxes" of 29 x 29 cells. There were in total 784 "boxes". For clustering the simulation procedure was as follows. Nucleation was allowed to occur randomly within a number of randomly selected boxes from the set of 784 boxes mentioned above. First one nucleus

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was randomly placed in each of the 784 boxes, then 392 boxes were randomly chosen and two nuclei were randomly located in each box, then 196 boxes were randomly selected with four nuclei per box and so on. The minimum number of boxes was 49 with 16 nuclei inside. The simulation produced a sequence of matrices as a function of time. Time is discrete in CA, it takes integer values starting from $t = 0$. One time unit corresponds to the interval between two consecutive matrix updates[23,26]. From the simulated matrices, all the desired quantities could be extracted. Hesselbarth and Göbel[23] give a more detailed account of two-dimensional cellular automata in general. Oliveira[26] gives further details of the present simulation.

3. Simulation Results and Discussion

In what follows simulation results are presented and qualitatively discussed. In the next section selected cases will be quantitatively discussed.

3.1. Nucleation and growth conditions

As mentioned in section 2, the nucleation was site-saturated: all nuclei appeared at $t = 0$. The behavior of the interface velocity, v, was studied in detail in Rios et al.[22] for randomly located nuclei. With regard to growth, the interface velocity was calculated from the growth of a single grain by means of the Cahn and Hagel[7] equation:

$$v = \frac{da}{ldt} = 0.5 \text{ units of length/unit of time} \quad (1)$$

For the simulation the velocity was calculated from the global parameters: area fraction, A_A, and Interfacial area length between recrystallized grains and matrix, L_A, units are omitted from now on throughout the paper:

$$v = \frac{dA_A}{L_A dt} = 0.51 \pm 0.01 \quad (2)$$

The interface velocity during the simulation remained very close to 0.51 that is practically the same value expected from the theoretical prediction, $v = 0.5$, for a single grain. Therefore, the growth occurs with constant interface velocity[22]. The interface velocity was calculated for the simulations carried out here using Equation 2 and the same result was obtained in spite of the non-randomness of nuclei location. The only exception was the simulation using nuclei periodically located where there was a slight increase in v at the later stages of the simulation with v reaching 0.55 for $A_A = 0.95$. Beyond that it increased more rapidly, reaching 0.60 for $A_A = 0.99$. Consequently, at the very end, $A_A > 0.95$, the simulation of the periodic nuclei could not maintain a strictly constant interface velocity.

On the whole, these results show that interfacial velocity remained constant during the simulation and was not affected by non-randomness. This is what one would theoretically expect thus showing the soundness of the simulation method in the present case.

3.2. Area fraction, A_A

Figure 1 shows A_A as a function of time, simulated by CA. The full line corresponds to randomly located nuclei and as shownd in previous work is in good agreement with the theoretical analytical expression:

$$A_A = 1 - \exp(-2N_A t^2) \quad (3)$$

The theoretical expression is not shown in Figure 1, in order not to overload the figure.

Figure 1 clearly shows the effect of a periodic arrangement and of clustering on the kinetics. The transformation is faster for a periodic nuclei arrangement and slower for clusters. Clustering slows down the transformation. The reason for this is the impingement. Overall growth of one region is slowed down when it meets another growing region even though the interfacial velocity remains constant. Periodically arranged nuclei grow without impingement for longer times than randomly arranged nuclei so that the overall kinetics is faster. On the other hand clustered nuclei impinge early and their overall kinetics is slower. The more severe the clustering is the slower the transformation is.

It is worthy of note that clustering does not seem to significantly change the overall shape of area fraction *vs.* time curve. Figure 2 shows the same data, replotted in a classical way. This kind of plot tends to accentuate the small area fraction, $A_A < 0.1$, behavior. For random nuclei the line is straight, clustering makes the lines to curve. Still, for area fractions between 0.1 and 0.9, where most of experimental data are normally measured the lines are not far from straight lines. Fitting these curves is a more precise way to quantify this and will be done later in this paper.

3.3. Interfacial area length between recrystallized grains and matrix, L_A

Figure 3 shows L_A as a function of time, simulated by CA. The full line corresponds to randomly located nuclei and as shown in

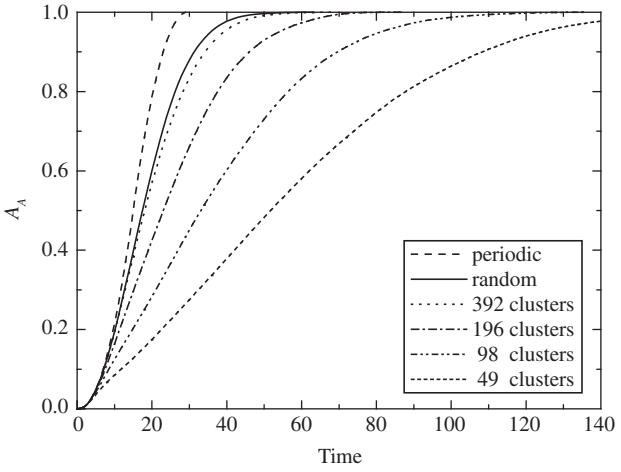

Figure 1. Area fraction against time, simulated by CA. The reaction is faster for a periodic nuclei arrangement and slower for clusters. The number of nuclei per unit of area is the same in all simulations.

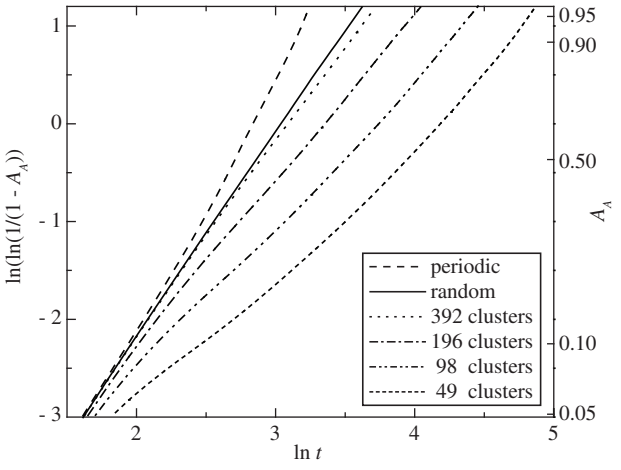

Figure 2. Area fraction against time, simulated by CA. Data from Figure 1 is replotted in the classical way. For random nuclei the line is straight, clustering makes the lines to curve.

previous work is in good agreement with the theoretical analytical expression, not shown in Figure 2:

$$L_A = 8N_A t \exp(-2N_A t^2) \quad (4)$$

Figure 4 shows L_A now plotted as a function area fraction, simulated by CA. The full line corresponds to randomly located nuclei and as show in previous work is in good agreement with the theoretical analytical expression, not shown in Figure 3:

$$L_A = \sqrt{32N_A}\,(1 - A_A)\sqrt{\ln\left(\frac{1}{1 - A_A}\right)} \quad (5)$$

The maxima observed in Figuras 3 and 4 can be explained as follows. At the early stages of the reaction the grains grow without impingement and therefore L_A increases. As impingement starts part of L_A is "transformed" into interfacial area between recrystallized grains, L_R, and therefore L_A starts to decrease. Eventually, when the matrix is fully transformed, only interfacial area between recrystallized grains remains and L_A is equal to zero. From this explanation one can infer that the maximum is higher for periodic that for random nuclei for the same reason that its kinetics are faster: impingement occurs later in the periodic arrangement than for random arrangement. Therefore L_A for periodic nuclei can become larger than for random nuclei. The maximum value of L_A decreases with clustering for identical motive: impingement occurs earlier for clusters and does not allow L_A to increase as much as it does for the random arrangement. Figure 4 shows that a peak develops at the low area fraction range for the most severe clustering conditions. This peak corresponds to an early impingement of the grains within the clusters, as will be seen in more detail later in this paper.

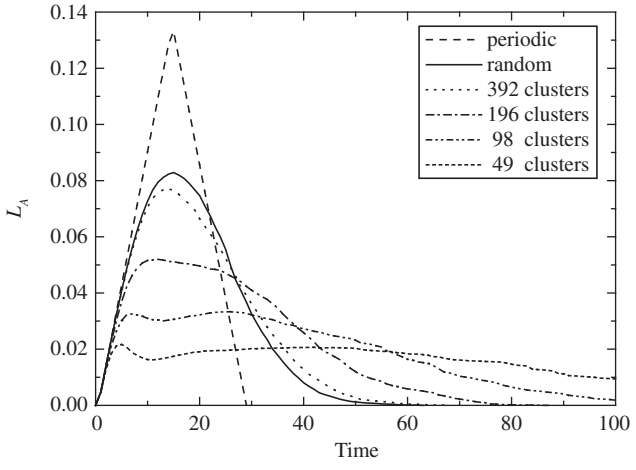

Figure 3. Interfacial area length between recrystallized grains and matrix, L_A, against time, simulated by CA. The maximum in L_A is more pronounced for a periodic nuclei arrangement and less pronounced for clusters.

3.4. The contiguity parameter, C_R

An important quantity is the contiguity parameter, C_R. The contiguity is the ratio of immobile interface length to the total interface length and is quite sensitive to deviations of nuclei location from randomness[11,12]. It can be defined as:

$$C_R = \frac{2L_R}{L_A + 2L_R} \quad (6)$$

where L_R is the interfacial area length between recrystallized grains.

Figure 5 shows C_R as a function of area fraction, simulated by CA. The full line corresponds to randomly located nuclei.

A comparison of Figure 5 with Figures 1-4 shows that C_R is a quite good parameter to evaluate deviations from randomness as advocated by Vandermeer[11]. The C_R *vs.* A_A curve, solid line in Figure 5, for randomly located nuclei divide the (A_A, C_R) plane in two distinct regions. Below the random curve lies the region in which nuclei deviate from randomness tending to a periodic arrangement. Above the random curve lies the region in which the nuclei tend to cluster. So by comparing the experimental or simulated data with the random curve a good indication of departures from randomness becomes evident. In the present simulations, C_R emerges as the most reliable microstructural descriptor as far as deviations from randomness are concerned.

In the next section a detailed analysis is carried out showing the uncertainties associated with trying to infer information using only the usual descriptors: A_A and L_A.

4. Detailed Analysis

The well-known generalized form of JMAK kinetics is, see for example, Rios and Padilha[8]:

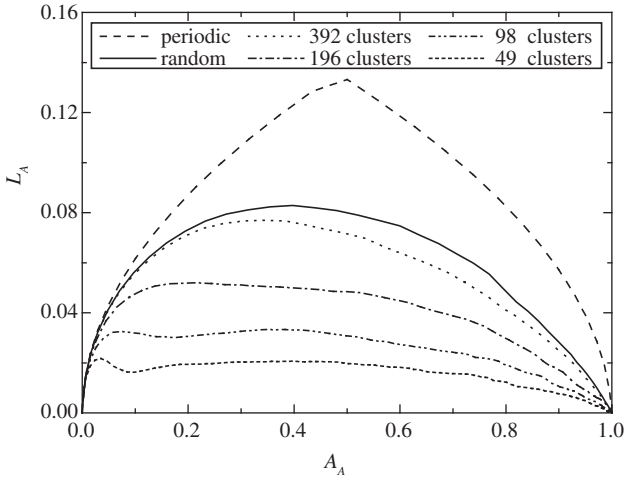

Figure 4. Interfacial area length between recrystallized grains and matrix, L_A, against area fraction, simulated by CA. The maximum in L_A is more pronounced for a periodic nuclei arrangement and less pronounced for clusters.

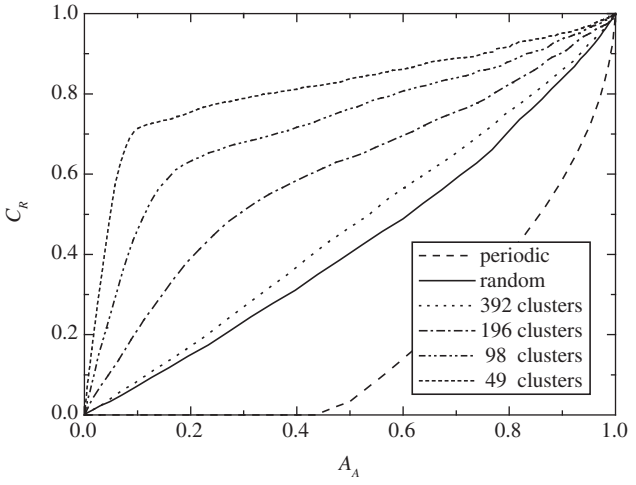

Figure 5. Contiguity, C_R against area fraction, simulated by CA.

$$A_A = 1 - \exp(-Kt^n) \quad (7)$$

The exact expression for randomly located nuclei is given by Equation 3. Equation 3 which can be generalized for a time dependent velocity, G:

$$G = \frac{G_0}{t^k} \quad (8)$$

where G_0 and k are constants. G was used instead of v to emphasize that it is an "apparent" interface velocity. Equation 8 can be used to generalize Equation 3:

$$A_A = 1 - \exp\left(-8N_A\left(\frac{G_0}{1-k}\right)^2 t^{2(1-k)}\right) \quad (9)$$

For $k = 0$, $G_0 = v = 0.5$ and Equation 9 reduces to Equation 3. This formalism is valid for site-saturated reactions when $k \neq 1$. Figure 6 shows the results of three simulations: periodic, random and 98 clusters.

Equation 3 is plotted along the random simulation and Equation 9 is force fitted to the periodic and cluster kinetics. Fitting produces correlation coefficients of $R^2 = 0.999$, apparently indicating very good fit. For periodic nuclei this results in a time dependent apparent velocity that increases with time:

$$G_{per} \propto t^{0.36} \quad (10)$$

For nuclei cluster, the apparent velocity decreases with time:

$$G_{clu} \propto t^{-0.21} \quad (11)$$

Equations 10 and 11 show that an analysis solely based on volume fraction could give an erroneous time dependent velocity whereas in all simulations the interface velocity remains constant. For periodic nuclei it could predict that the boundary velocity increases with time. More interesting is the result for the cluster. It predicts that the velocity decreases with time. This is result is quite interesting because it is believed that the velocity decreases with time during recrystallization. Two main causes are invoked for this[16,28]:

a) Concurrent recovery: this would result in a decrease in the stored energy during recrystallization and a consequent decrease in grain boundary velocity; and
b) Existence of deformation gradients in the microstructure: the idea here is that the recrystallization would begin in regions of higher stored region and grow into regions of progressively lower stored energy. This would cause a decrease in grain boundary velocity.

Stüwe et al.[29] is an example of a model that takes recovery into account whereas Rios[30] is an example of a deformation gradient model.

The present analysis suggests yet a third possibility: interpreting a non-random microstructure using formalism similar to Equations 7 and 9, essentially valid for randomly located nuclei, can lead to misleading time dependent apparent velocity as in Equations 10-11. In short, Equation 7 is quite flexible and will give good fit but any data inferred from the fitting parameters is uncertain, unless one can be sure that the nuclei distribution is random. The apparent decrease in growth rate observed here is not real because the interface velocity is kept constant during the simulation. It is purely an artifact arising as a result of the limitations of the mathematical formalism used to analyze the data. This does not mean that the effects mentioned above cannot lead to a decrease in interface velocity in real materials but that, time dependencies resulting from best-fitting Equation 7 must be taken with care. More reliable is of course to determine the interface velocity by means of the Cahn and Hagel equation, Equation 2. Unfortunately, this latter method can also be subjected to error[8] owing to the unavoidable experimental scatter and normally small number of points of real datasets[8,31].

An indication of randomness is often sought by plotting the microstructural path: L_A *vs.* A_A and using Equation 5 to fit it. Good fit is often taken as an indication that the reaction is site saturated and that the nuclei distribution is not far from randomness thus validating the above approach. Figure 7 shows this plot for three simulations: periodic, random and 98 clusters. Equation 5 is plotted along the random simulation. Equation 5 is force fitted to the periodic and cluster kinetics by allowing N_A to vary in Equation 5. From force fitted curves an "apparent" number of nuclei per unit of area can be calculated.

For periodic nuclei, the apparent number of nuclei per unit of area, $N_{Aper} \cong 2.4 \times 10^{-3}$, is about twice the value used in the simulation, $N_A \cong 1.2 \times 10^{-3}$. The correlation coefficient was $R^2 = 0.89$. The agreement was not very good as shown in Figure 7.

For clustered nuclei, the apparent number of nuclei per unit of area $N_{Aper} \cong 1.9 \times 10^{-4}$, almost an order of magnitude smaller than the simulation value. The correlation coefficient was $R^2 = 0.94$. The agreement was not good but much better than for the periodic nuclei.

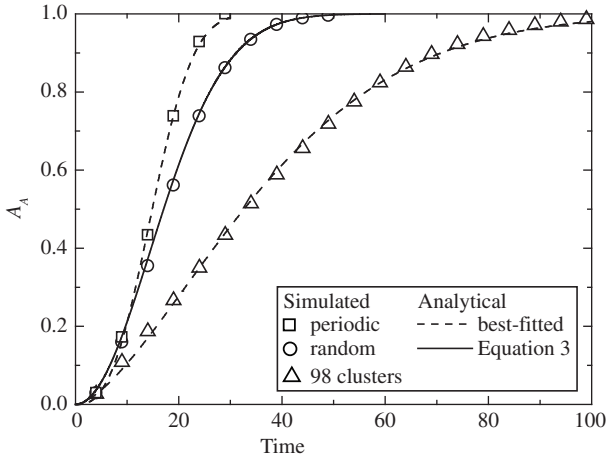

Figure 6. Area fraction against time, simulated by CA. Simulation data are force fitted by JMAK kinetics.

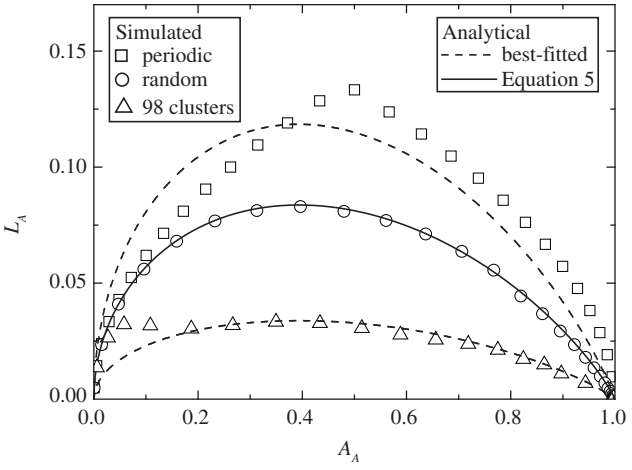

Figure 7. Interfacial area length between recrystallized grains and matrix, L_A, against area fraction, simulated by CA. Simulation data are force fitted by Equation 5, allowing N_A to vary.

Actually, most of the disagreement was caused by the peak observed for an area fraction about 0.05. Beyond $A_A \approx 0.10$ the agreement was good.

Therefore, the bad fit between the microstructural path expression, Equation 5, and data may provide some indication that the nuclei are randomly located. However, one must consider that recrystallization data normally consist of a small number of experimental points, of the order of ten points, and that experimental errors are rarely less than ± 10%. Under these circumstances correlation coefficients are not usually high[8,28,31]. Consequently, drawing conclusions from fitting of a small number of experimental points subject to large errors can be highly problematic. One cannot always be sure that the low correlation coefficient comes from experimental errors or from a true deviation from randomness.

The peak that develops in the low area fraction region might be an indication that there is clustering. An extreme case is shown in Figure 8 where the microstructural path for the most severe clustering condition simulated, 49 boxes is shown.

Figure 8 shows that for severe clustering a clear peak develops. The curves were calculated by using two values of number of nuclei per unit of area. One value for the transformation within the cluster, N_{Aw}, that is, supposing that in the beginning the reaction proceeds as if it occurred in a region containing $N_{Aw} = 16/841 = 16N_A$ nuclei instead of the average value of $N_A = 1/841$. This gives the dotted line in Figure 8. In the later stages, after impingement within the cluster has taken place, the transformation occurs, outside the clusters, as if each of the 49 boxes behaved as a single region, giving $N_{Ao} = 1/(16 \times 841) = 1/(16N_A)$ nuclei, a value sixteen times smaller than the average value $N_A = 1/841$. It can be seen that, for severe clustering, the agreement between the curve generated with N_{Ao} and the simulation is good for $A_A > \approx 0.10$. In practice one might easily miss the peak because one rarely has a large number of experimental measurements below ≈ 0.10.

The detailed analysis carried out in this section reinforces the need to use the contiguity that was shown in the previous section to be highly sensitive to non-randomness. The methodology used in this section is often employed in real cases but the fitted parameters could result in erroneous velocities when nuclei are not randomly located.

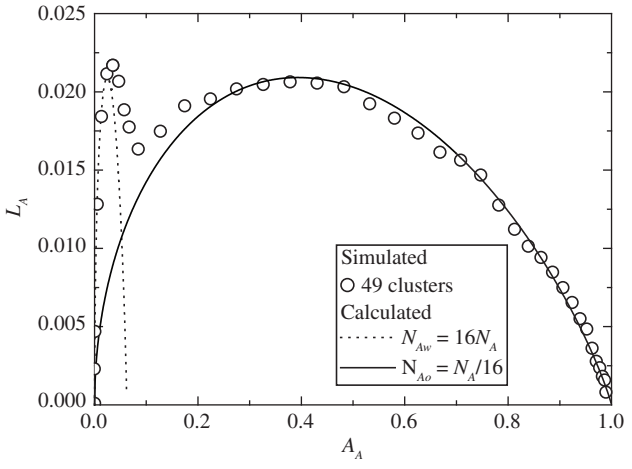

Figure 8. Interfacial area length between recrystallized grains and matrix, L_A, against area fraction, simulated by CA. Curves are calculated using with two different values of number of nuclei per unit of area, as explained in the text.

5. Summary and Conclusions

Cellular automata simulation of two dimensional recrystallization has been carried out to investigate the effect of non-randomness on nuclei location on microstructural evolution. The results were compared with several microstructural descriptors. The main conclusions are:

a) Conventional kinetic analysis based solely on the area fraction against time data could generate misleading time dependent velocity. For instance, when nuclei are located in clusters this analysis may erroneously suggest that the velocity decreases with time. Notice that in the simulation the interface velocity remained constant in all cases;

b) A microstructural path based on the L_A against A_A plot was more sensitive to deviations from non-randomness using the precise data provided by the simulation. However, when applied to real experimental data which normally consists of a limited number of data points subject to significant experimental errors one might have difficulties to detect non-randomness; and

c) It is clear from the simulations that the contiguity, C_R, is more sensitive to non-randomness than the other measurements considered: area fraction, A_A, interface length between recrystallized grains and the matrix, L_A, and interface length between recrystallized grains, L_R. Therefore, a more reliable data analysis should include the contiguity, as suggested by Vandermeer[11], in order to establish to what extent non-randomness influences microstructural evolution.

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