Original Article

Stereological analysis of the microstructure of pure iron with random nucleation

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A R T I C L E   I N F O

Article history:
Received 14 October 2013
Accepted 30 August 2014
Available online 13 October 2014

Keywords:
Cellular automata
Recrystallization
Stereology
Pure iron

A B S T R A C T

Microstructural characterization of metallic materials is of paramount importance in the qualification and quantification of their desired final properties. Metallic materials have often been characterized by means of optical and electronic microscopy. Results from these techniques are in most of the cases based on 2D analysis. Stereological methods are then employed to obtain 3D information. However, such methods are based on assumptions and approximations of the real material structure. Therefore, it is essential to know the limitations of these methods. A further complication arises when one wishes to compare real materials with computer simulation results and stereological analytical techniques. In this paper, methods normally applied to real microstructure are applied to microstructures simulated by cellular automata (CA) simulation. Experimental results from microstructural characterization of polycrystalline pure iron were used as the starting point for the simulation. Consequently, we could apply analytical formulae of stereology to experimental data from pure iron and to computer generated microstructures from cellular automata simulation. Comparison of analytical formulae, experimental results and computer simulation provided useful insights on limits of applicability and on the meaning of the stereological analysis.

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1. Introduction

Recrystallization and grain growth are fundamental issues of Materials Science. Therefore, continuous progress is always required in understanding those phenomena in order to achieve better control and consequently increasing better final properties of polycrystalline materials. In this regard, detailed knowledge of the 3D microstructure of the polycrystal is paramount. Traditional metallographic techniques invariably carry out measurements on a planar section [1,2]. From those measurements, one cannot obtain all 3D microstructural parameters. For example, intrinsic 3D quantities, such as the volume of the individual grains, the number of

* Technical contribution presented at the 66th ABM International Annual Congress, São Paulo, Brazil, July 18th to 22nd, 2011.
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grains per unit of volume or the Gaussian curvature per unit cannot be obtained from classical 2D quantitative metallo-
graphic techniques.

Recently, it has become apparent that there is a transition from 2D to 3D microstructural characterization. Specifically,
today one has 3D computer simulation, 3D analytical theories and 3D microstructural characterization techniques. This can
be seen in the widespread number of papers, conference sessions and even entire conferences dedicated to the topic of 3D
Materials Science.

Still, for a long time simple techniques will persist and one may use the 3D developments to improve the simpler 2D techniques. For example, some assumptions may now be tested against a 3D computer simulation. In this way, one may develop useful approximations that may be very helpful for the Engineer to obtain a reasonably accurate result for a fraction of the cost and the time of a full 3D experiment. Following this reasoning, the present work utilizes experimental measures, analytical methods and computer simulations to revisit earlier results and test new approximate expressions that relate 2D measurements to 3D quantities by means of approximate expressions.

2. Methods and materials

2.1. Experimental methods and materials

Samples of polycrystalline pure iron were used. The composition of the pure iron in ppm was: C-41, Mn-940, P-15, S-205
Si-160, Al-20, N-80, Ti-10, Cu-30, Cr-100, Mo-20, Nb-10, V-10, B-4, O-165. The sample was cold rolled up to 80% then annealed
550 °C for 3600 s (1 h) in a quartz tube containing Ar. Specimens were cut from this sample taking care to avoid deformation
during cutting. The specimens were then ground and polished. A final polishing step consisting of polishing in a col-
loidal silica solution was employed. After this, the specimens were etched with Nital 3% solution for observation under the
optical microscope. Stereological measurements were carried out in a Nikon Eclipse optical microscope with image analy-
sis software NIS-Elements D 3.0. The number of grains per unit of area, \( N_A \), and the number of grains per unit of length of
test line, \( N_l \), were measured using standard techniques [3].

2.2. Computer simulation

A 3D cellular automata computer simulation was carried out. A cubic mesh with 300 \( \times \) 300 \( \times \) 300 cubic cells, henceforward
also referred to as the “matrix”, was employed. A von Neumann neighborhood was adopted. Simulation details can be
found in Ref. [4–9]. The simulation employed a 22,300 nuclei uniform randomly located within the matrix at the start of the transformation. This number of nuclei was chosen according to reasons explained in a previous work [10]. The simulation was carried out in a dual processor Intel Xeon workstation with eight cores in each processor and 48GB of memory. The computer program was written in Fortran 2003 and compiled in an Intel FORTRAN compiler using OPEN MP 3.0 directives for shared memory parallelization. Each simulation run took about 10,000 s. In order to assign units to this simulation a length to the edge of a cubic cell has to be chosen. We chose this length in such a way to make the number of nuclei per unit of volume equal to that calculated for pure iron using DeHoff’s formula. This was done because DeHoff’s result was consid-
ered the better approximation in that case. Details are given below and results are in Tables 1 and 2, later in this paper. This choice gave a cubic cell with an edge length equal to about 0.5578 \( \mu \)m.

2.3. Analytical expressions

The analytical expressions employed in this study either derived above or obtained from publications are summarized in
Table 1.

In addition to \( N_l \), \( N_A \), and \( N_V \) obtained by DeHoff [2] and for Voronoi polyhedral [1] we derived expressions for these quantities supposing that the grains could be considered as cubes or spheres.

For spherical polycrystalline grains, using \( S = 4\pi R^2 \), \( V = \frac{4}{3}\pi R^3 \) and the caliper, \( D = 2R \), gives, \( S_V \), the grain boundary area by unit of volume

\[
S_V = \frac{S}{V} = \frac{3}{2R} \tag{1}
\]

Deriving expressions for \( N_V \) is straightforward

\[
N_V = \frac{1}{V} = \frac{3}{4\pi R^3} \tag{2}
\]

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<tbody>
<tr>
<td>( N_l ) (mm(^{-1})) from ( N_A )</td>
<td>67.3</td>
<td>67.0</td>
<td>69.2</td>
<td>62.4</td>
<td>70.4</td>
</tr>
<tr>
<td>( N_A ) (( \times 10^{-2} ) mm(^{-2})) from ( N_l )</td>
<td>33.0</td>
<td>33.3</td>
<td>31.2</td>
<td>38.5</td>
<td>30.2</td>
</tr>
</tbody>
</table>
Eq. (1) together with the well-known expression [3]

\[ N_A = N_V D \]  \hspace{1cm} (3)

give other expressions involving \( N_L \) and \( N_A \) may be derived

\[ N_V = \frac{\sqrt{6\pi}}{6} N_A^{3/2} \]  \hspace{1cm} (4)

\[ N_V = \frac{16}{9\pi} N_L^3 \]  \hspace{1cm} (5)

\[ N_A = \frac{8N_L^2}{3\pi} \]  \hspace{1cm} (6)

\[ N_L = \frac{1}{4} \sqrt{6\pi N_A} \]  \hspace{1cm} (7)

For the cube, we may use an identical reasoning as above, recalling that the mean area and mean volume of the cube are \( a^2 \) and \( a^3 \) respectively, where \( a \) is the edge length of the cube. Moreover, from stereology [3], \( \lambda = 4V/S \), where \( V \) is volume and \( S \) the area, for cubic grains \( \lambda = 1/N_L = 3a/2 \). As a result, for cubic grains

\[ N_V = \frac{2\sqrt{6\pi}}{9} N_A^{3/2} \]  \hspace{1cm} (8)

\[ N_V = \frac{8}{27} N_L^3 \]  \hspace{1cm} (9)

\[ N_L = \frac{1}{2} \sqrt{6\pi N_A} \]  \hspace{1cm} (10)

\[ N_A = \frac{2}{3} N_L^2 \]  \hspace{1cm} (11)

3. Results and discussion

3.1. Measurements of \( N_L \) and \( N_A \) in pure iron compared with approximate methods

Fig. 1 shows an optical micrograph of fully recrystallized pure iron. \( N_L \) and \( N_A \) were measured for the pure iron. These experimental values were employed to assess the theoretical expressions obtained by DeHoff [2], for the Voronoi polyhedra [1] and for spherical grains, Eqs. (6) and (7) and for the cubic grains Eqs. (10) and (11). In order to do this the experimental value of \( N_L \) was inserted into the theoretical equation to calculate \( N_A \) and the experimental value of \( N_A \) was inserted into the equations to calculate \( N_L \). The results obtained were compared with the experimental values. We considered that the best theoretical equation was the one that would give the best agreement of calculated and experimental values. The results of these calculations are summarized in Table 2. Table 2 shows that DeHoff’s approximated expression gives the best results. For example, DeHoff’s values of \( N_L \) calculated for experimentally measured \( N_A \) is very close to the experimental value of \( N_L \). The same agreement was observed for values of \( N_A \) calculated for experimentally measured \( N_L \) as shown in Table 2. Of the other expressions, Voronoi showed better agreement than the cube, which on its turn gave a better agreement than the sphere approximation.

In view of these results, we considered DeHoff’s approximation to be the best choice to calculate \( N_V \) for pure iron. Therefore, the values of \( N_V \) used for pure iron in what follows were those calculated using DeHoff’s approximation.

3.2. Values of \( N_L \) and \( N_A \) obtained by computer simulation compared with approximate methods

Fig. 2 shows a cellular automata computer simulation of the growth of nuclei uniform randomly located within the matrix with a constant velocity up to a fully transformed “microstructure”. Fig. 2a shows a tridimensional view whereas Fig. 2b shows a planar section of Fig. 2a. The values measured from the planar section were 76.2 \( \text{mm}^{-1} \) for \( N_L \) and 3824 \( \text{mm}^{-2} \) for \( N_A \).

Table 3 shows values of \( N_L \) and \( N_A \) obtained by cellular automata computer simulation compared with approximate methods. In this situation, one would expect cube to be a good approximation and that is indeed the case. The simulation method adopted: all nuclei already present at \( t=0 \), without any further nucleation, and all grains growing with a constant velocity in cellular automata simulation produce a cube shaped growing volume as illustrated in Fig. 3b. This point is discussed further in the next section.

3.3. Measurements of \( N_L \) and \( N_A \) in pure iron compared with values obtained by computer simulation

Table 4 presents a summary of the quantities measured in pure iron along with those obtained by cellular automata simulation using \( N_V \) of the simulation equal to that calculated for pure iron by DeHoff’s approximation. Therefore, in Table 4 for pure iron and the simulation values are the same. There is some discrepancy between the values of \( N_L \) and \( N_A \). As said above, the cube not DeHoff’s approximation gave the best agreement with the simulation owing to the cube shape of the growing regions of the simulation.

Difficulties of comparing recrystallization evolution in a single crystal pure iron with cellular automata simulation were discussed in depth by Salazar et al. [5]. The difference
Fig. 2 – (a) Tridimensional view of a fully transformed computer simulation of the nucleation and growth reaction. (b) Planar section obtained from the 3D "microstructure" depicted in (a).

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<tr>
<th>Table 3 – Values of N\textsubscript{L} and N\textsubscript{A} obtained by cellular automata computer simulation compared with calculations from approximate methods.</th>
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<td>--------------------------</td>
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<tr>
<td>N\textsubscript{L} (mm\textsuperscript{-1})</td>
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<tr>
<td>N\textsubscript{A} (\times 10\textsuperscript{-2} mm\textsuperscript{-2})</td>
</tr>
<tr>
<td>*N\textsubscript{V} (\times 10\textsuperscript{-4} mm\textsuperscript{-3}) from N\textsubscript{L}</td>
</tr>
<tr>
<td>*N\textsubscript{V} (\times 10\textsuperscript{-4} mm\textsuperscript{-3}) from N\textsubscript{A}</td>
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\* Values of N\textsubscript{V} in the column “Cellular automata” were those used in the simulation. In the other columns, values of N\textsubscript{V} obtained from N\textsubscript{L} and N\textsubscript{V} from N\textsubscript{A} were calculated using the analytical formulae of Table 1.

in the shape of the growing simulation region (cube) and the shape of the growing recrystallized grain in the case examined by Salazar et al. [5], spherical, was responsible for this difference. This is even more so because of the “stepped” nature of the moving boundary of the simulation. The consequence is that a simulation using the same N\textsubscript{V} as that of the phase transformation or recrystallization should give a larger interfacial area density per unit of volume, as observed here. In practice, for applications of cellular automata simulation to recrystallization [5], this difference is not problematic because it can be accounted for by a shape factor or correction factor that is independent of time [5].

Fig. 3 – Growth of a single isolated region with a constant velocity: (a) Spherical growth and (b) Growth during a cellular automata simulation.
as it can be seen here, absolute values of interfacial areas of simulated and real microstructures do not agree so well. The simulation may be corrected to agree with the experiment because the disagreement is systematic not a random error and may be accounted by a time and volume fraction independent correction factor [5].

4. Conclusions

- Stereological measurements carried out in pure iron, \( N_l \) and \( N_A \), were compared with four possible approximations to the (mean) grain. The grain shape proposed by DeHoff gave the best result. The point of this good agreement is that if one only had one experimental measurement of, say \( N_l \), DeHoff’s equation could be used to estimate \( N_A \).
- Stereological measurements, \( N_l \) and \( N_A \), were also carried out in a cellular automata simulated microstructure, \( N_V \) was known for the simulation. These measurements were compared with four possible approximations of the grain shape. In this case, the cube approximation gave the best result.

Conflicts of interest

The authors declare no conflict of interests.

Acknowledgements

This work was supported by Conselho Nacional de Desenvolvimento Científico e Tecnológico, CNPq, Fundação de Amparo à Pesquisa do Estado do Rio de Janeiro, FAPERJ and Coordenação de Aperfeiçoamento de Pessoal de Nível Superior, CAPES. Daniel Souto de Souza is grateful to Universidade Federal Fluminense (UFF) for his “scientific initiation” studentship. Help from Dr. Simone Oliveira is gratefully acknowledged.

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