THEORY OF METALS

Grain Growth in a Microstructure with Bimodal Grain Size Distribution

V. Yu. Novikov*

Treptower Str., 74D, Hamburg, 22147 Germany
*e-mail: v.novikov@gmx.de

Abstract—Grain growth in the textureless material characterized by bimodal grain size distribution has been investigated. Grain growth in a pure material can eliminate the second maximum in the distribution, despite the presence of abnormally coarse crystallites in the initial structure. This is true when the number of coarse crystallites in the initial state is small and can be explained by normal grain growth in the fine-grained matrix. The size effect of matrix grains on the second maximum in the distribution under these conditions has been analyzed. The second maximum has been shown to remain and even increase due to the inhibition of boundary migration.

INTRODUCTION

Controlling the microstructure of a material is one of the main tools for influencing its properties. As a rule, technologists tend to form a homogeneous structure in single-phase alloys; however, in some cases, they create inhomogeneities, for example, by various manipulations on the surface layers of the product. In this regard, some publications on single-phase materials with a bimodal structure draw attention [1−3]. In this case, we are talking about a global structural inhomogeneity rather than a local one, i.e., the inhomogeneity over the entire cross section of a product. A bimodal structure increases the plasticity of materials with very fine grains (see, for example, [4, 5]). However, insufficient attention is paid to changes in the created structure that are possible during both heat treatment of products and their operation. These changes can be caused, first of all, by grain growth.

According to modern understanding, grain growth in the case of a double-hump grain size distribution (GSD) is supposed to lead to the formation of a structure with coarse grains and the disappearance of the fine-grained matrix, i.e., to the disappearance of the first maximum in the GSD. Moreover, this seems natural, since the grain growth process develops through the absorption of finer grains by their immediate coarser neighbors [6, 7]. Along with that, one often cites the work [8], which used the Monte Carlo method to show that a coarse crystallite introduced into a fine-grained structure slows down its growth with time and finally becomes a part of the standard GSD. These results are quite unexpected having in mind of numerous experimental data. For example, the data on abnormal grain growth give the evidence that coarse crystallites do not become a part of the normal GSD in the fine-grained matrix, and, on the contrary, intensively absorb fine grains. Grain growth in materials with a bimodal structure seems to remain unstudied, insofar as the author is aware. This work aims to fill this gap both for the progress of new techniques for microstructure control and the further development of the grain growth theory.

DESCRIPTION OF THE MODEL AND INITIAL DATA

The model [9, 10] that describes the grain growth by the migration of grain boundaries toward their curvature centers and can be used to investigate changes in the GSD over time, was used for numerical simulations [9, 10]. In contrast to the Monte Carlo method (see, e.g., [8]) or the phase-field method ([11]), we can study the behavior of infinitely large ensembles of grains using this model. Therefore, the model [9, 10] that provides higher statistical reliability of the results is used in this work to study the GSD evolution.

To find changes in a particular GSD, the model calculates the boundary displacements between growing and absorbed grains belonging to the size classes i and j, where $i > j$:

$$\Delta d_{ij} = \gamma M \left(1/D_j - 1/D_i - Z \right) \Delta t, \qquad (1)$$

where γ and M are the energy and the mobility of their boundary, D_j and D_i are the diameters of the absorbed

Table 1. Characteristics of the initial structure of polycrystals

Polycrystal	N_2	N_2/N_1	D_{max}/D_1	D_1, μm
P1	0.5×10^3	$\sim 0.65 \times 10^{-5}$	~6.2	~4.3
P2	0.2×10^5	$\sim 0.26 \times 10^{-3}$	~6.2	~4.3
M	–	–	~3.9*	~4.3*

* Since the GSD in the M polycrystal is single-hump, $D_1 = D$.

and growing grains, Z is the coefficient characterizing the braking force, and Δt is the time step. The boundary displacements are recalculated into GSD changes, taking into account the average probability of the contact between the growing grain and its nearest neighbors of different sizes and the condition under which the system volume is constant. In this work, we assume that $\gamma = 500$ erg/cm² and $M = 2 \times 10^{-11}$ cm⁴/(erg s) [12] are the same for all grain boundaries and are time-independent.

We investigated the grain growth in three-dimensional polycrystals with a bimodal GSD. The total number of grains in the initial state was $\sim 0.8 \times 10^8$, the average size (diameter) of matrix grains $D_1 \approx 4.3$ μm, the size of coarse crystallites $D_2 \approx 6.2 D_1$, their relative

Fig. 1. GSD in the P1 and P2 polycrystals after the second or first maxima have disappeared, respectively ($t = 30$ min).

number $N_2/N_1 = 0.4 \times 10^{-5} - 0.5 \times 10^{-3}$, where N_2 and N_1 are the numbers of abnormally coarse and matrix grains, respectively. The matrix grain size distribution was close to lognormal, with the maximum grain size of $\sim 3.9 D_1$. Grains of different sizes were considered to be randomly spaced. We took the disappearance of the minimum between maxima in the GSD as the evidence that the bimodality does not exist anymore. The bottom of the maximum is assumed to extend along the size axis from the left or right GSD boundary to the minimum. The kinetics of the grain growth and the GSD evolution were studied at different initial N_2. They were studied by determining the changes in the average D and maximum D_{max} grain sizes. The average D_1 and D_2 grain sizes, the N_1 and N_2 numbers of grains, and their relative volumes were found for each maximum.

RESULTS AND DISCUSSION

The simulation of grain growth without a retarding force (i.e., when $Z = 0$ in Eq. (1)) showed that the double-hump GSD transforms into a single-hump GSD over time. If the initial number of abnormally coarse crystallites is greater than $\sim 0.1 \times 10^5 \approx 0.13 \times 10^{-3} N_1$ (this corresponds to their volume fraction of ~2%), the second maximum, as expected, increases, and the first maximum disappears. Moreover, an increase in N_2 reduces the lifetime of the first maximum. At lower initial N_2 values, on the contrary, the second maximum disappears, i.e., coarse crystallites, despite their size advantage, become a part of the GSD of the matrix during grain growth. The lifetime of the second maximum decreases with decreasing N_2. To understand the causes for this, let us compare the simulation results for polycrystals with opposite behavior of maxima, namely, P1, where the first maximum is retained, and P2, where the second maximum is concerved. The M polycrystal, the microstructure of which is similar to that of the matrix in P1 and P2, was used as a reference. Table 1 describes the polycrystals. Figure 1 illustrates their GSD after the maxima have disappeared.

Figure 2 shows microstructural parameter variations that occur in these polycrystals over time. A comparison of Figs. 2a and 2c indicates that the average grain size D in P1 and M polycrystals changes almost in the same way, whereas D_{max} in P1 grows much faster than in M. A comparison of Figs. 2b and 2c shows that the behavior of D in P2 and M polycrystals, on the contrary, is quite different, obviously, due to the absorption of matrix grains.

The D_{max} in the P2 polycrystal rises higher than in M. A comparison of Figs. 2a and 2b shows that D_2 in P1 increases faster than D_1, whereas the same is observed in P2 only at the initial stages, and, further, their rates of change are practically the same. In addition, one can see that D_2, D, and D_{max} grow slower in
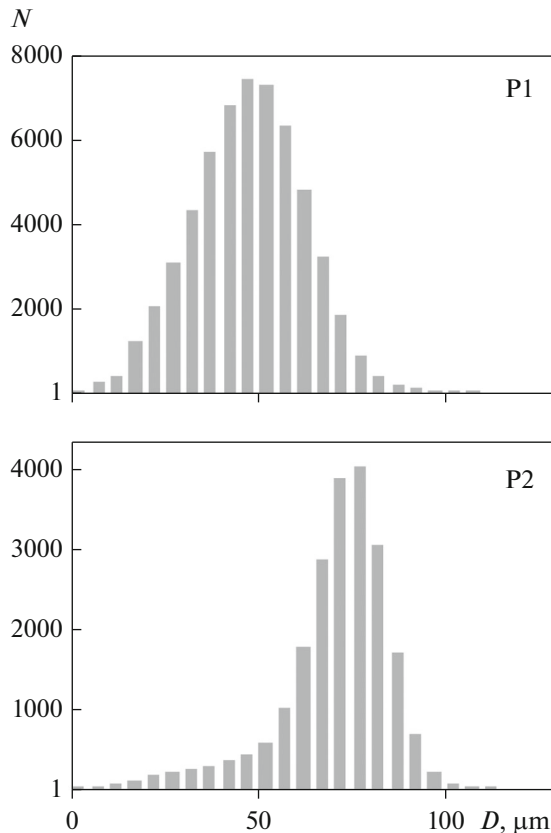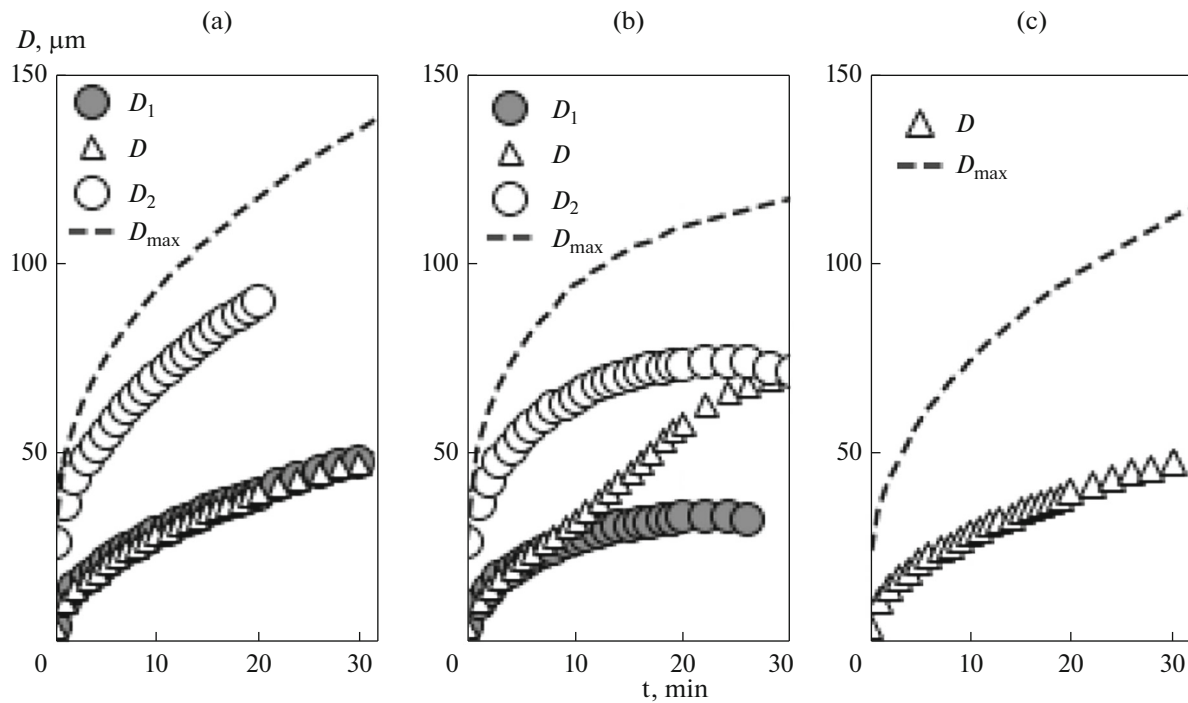
Fig. 2. Grain growth kinetics in the (a) P1 and (b) P2 polycrystals with double-hump GSDs, as well as in the (c) M polycrystal with the GSD similar to the matrix GSD in P1 and P2.

the P2 polycrystal than they do in P1. Finally, the D_{max} value grows faster than the average grain size D_1 of the matrix in the P1 polycrystal. All these details are discussed below.

Additional information on the evolution of the microstructure is presented in Fig. 3. We see that the number of matrix grains decreases by ~3 orders of magnitude in the M polycrystal, where there was initially no second GSD maximum. Similarly, N_1 also decreases in P1 for the time of the maximum existence. The latter seems to be related to a small number of coarse crystallites in the initial structure. The number of matrix grains in the P2 polycrystal, however, decreases by ~5 orders of magnitude. The different behavior of P1 and P2 appears to be caused by the fact that the number of coarse crystallites that absorb the matrix is significantly greater in P2 than in P1. In addition, Fig. 3 shows that N_1 decreases in two stages: first rapidly and then noticeably slower. At the first stage in P1 and P2 polycrystals they are almost the same, but at the second stage they differ significantly, namely, N_1 in P1 changes in the same way as in M, whereas in P2 it falls much faster. The above fact suggests that the different behavior of N_1 at the second stage in P1 and P2 can be connected with the different behavior of the second GSD maximum. We used the changes in N_1 at the second stage to identify the processes occurring at this stage.

Therefore, the possible growth processes under the analyzed conditions should be mentioned: (1) normal grain growth in the matrix, (2) increase in the size of

coarse crystallites due to matrix absorption, and (3) consumption of coarse crystallites by one another. The first two are discussed below without much attention to the third, since it develops extremely slowly, because of the very low curvature of the boundaries between coarse grains. An increase in the average grain
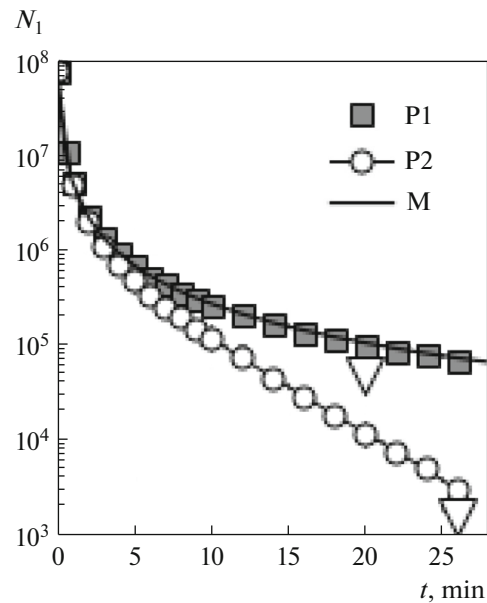

Fig. 3. Change in N_1 in the P1, P2, and M polycrystals during grain growth. The triangles indicate the moment when one of the maxima disappears.

size during normal growth is the result of the interaction in a large group of grains, where they can be absorbed by some of their neighbors and absorb others simultaneously. The growth of abnormally coarse crystallites in the same group is a so-called one-way process; i.e., they grow absorbing the matrix grains (since the interaction with the neighboring similar coarse crystallites, as was mentioned above, is very weak). It is important to keep in mind that the growth rate depends on how much the size of the absorbed neighbors differs from the size of the growing grain. During normal grain growth, the growing and absorbed grains have close sizes whereas the size of coarse grains growing in the matrix is significantly larger than the size of the absorbed neighbors. Therefore, the size of coarse crystallites is expected to grow faster than the average matrix grain size. As a result, in the first case, N_1 decreases more slowly than it does in the second case. We can, therefore, consider that the absorption of matrix grains by abnormally coarse crystallites is the main reason for N_1 changes at the second stage in the P2 polycrystal. Regarding P1, the fact that the behavior of N_1 at the second stage in the polycrystal is close to that of N_1 in M (see Fig. 3) brings us to the conclusion that normal growth seems to play the main role here. Thus, the different behavior of GSD maxima during the grain growth in the P1 and P2 polycrystals depends on the development of normal growth in the fine-grained matrix.

One should keep in mind that normal growth in the matrix can develop freely only in those parts of the structure that are not occupied by coarse crystallites. Since the size of these regions is proportional to $(1/N_2)^{1/3}$, where N_2 is the number of coarse crystallites, then the smaller N_2 is, the more pronounced the normal grain growth in the matrix is supposed to be. Judging by the values of N_2 for the studied polycrystals (see Table 1), this size in P2 is ~53 μm, while in P1 it is ~3.5 times greater. Therefore, normal grain growth in the P1 matrix can develop much longer than in P2, despite its absorption by coarse crystallites.

This analysis explains the results presented in Fig. 2. The nearly complete agreement between D and N_1 in the P1 and M polycrystals is associated with the easy development of normal growth in the P1 matrix. The fact that the normal grain growth in the P2 polycrystalline matrix is slower, and D and D_{max} are smaller than those in the M polycrystal, can be explained by the following. The number of coarse crystallites in P2 is greater, and they quickly absorb the matrix, which increases the probability of their contact with each other and thus reduces their growth rate. Therefore, D_2 grows slower in P2 and reaches a smaller value than that in P1. The more rapid D_{max} growth, as compared with the average D_1 matrix grain size in the P1 polycrystal, is explained above.

The different GSD evolution in the P1 and P2 polycrystals seem to be caused by the changes in the partial GSD of coarse crystallites and the GSD of matrix grains, which happen during the grain growth. On the one hand, the width of the first one increases during the growth due to an increase in D_{max}. The minimum size of coarse crystallites is not expected to decrease significantly, since it differs only slightly from D_2. On the other hand, normal grain growth in the matrix is supposed to increase the GSD width and, consequently, increase the number of grains in the size range between the maximums of the common GSBs. Absorption of the matrix by abnormally coarse crystallites reduces the number of grains in it and changes its GSD. The latter implies that the number of matrix grains with sizes smaller than D_1 or close to it reduces. The result of the above evolution of the partial GSD depends on how fast the matrix grains are absorbed. If they are absorbed slowly (i.e., at low N_2, for example, in the P1 polycrystal), they have time to fill the minimum in the GSD of the polycrystal near D_2. If they are absorbed quickly (at high N_2, for example, in P2), they fill the minimum near D_1. The existence of minimum in the common GSD is a sign of the existence of two maxima (see above). This fact means that in the first case, the second maximum disappears, while in the second case, the first one disappears. In addition, in the first case, coarse crystallites can themselves contribute to the disappearance of the second maximum, because they reduce its height during their growth.

According to the above explanation, the behavior of the second GSD maximum in a pure material with low initial N_2 depends on the rate of normal grain growth in the matrix. If this rate is increased, for example, by decreasing the initial D_1 value, this maximum disappears faster. Our results confirm that the lifetime of the second maximum in polycrystals with a lower D_1 decreases sharply. However, if the rate of grain growth is decreased, the lifetime of the second maximum increases. Figure 4 confirms this fact. Even a weak deceleration of grain growth in the P1 polycrystal (see Fig. 4a) significantly increases the lifetime of the second maximum, as follows from Fig. 4b. Moreover, a small increase in the coefficient of growth retardation was found to increase the volume fraction of the grains forming this maximum by at least a factor of ten (see Fig. 4b).

In connection with the above, let us turn to the abnormal grain growth in anisotropic transformer steel, which is known to be associated with the stabilization of the fine-grained matrix by fine second-phase precipitations. The relative number of crystallites growing in this material during abnormal growth is small ($\leq 10^{-4}$). Therefore, we cannot reject the possibility that the second GSD maximum can disappear due to grain growth in the absence of the disperse phase, as is shown in Fig. 4b for the P1 polycrystal. It seems reasonable to assume that the second phase particles in transformer steel not only stabilize the fine grain matrix, but also contribute to conservation of
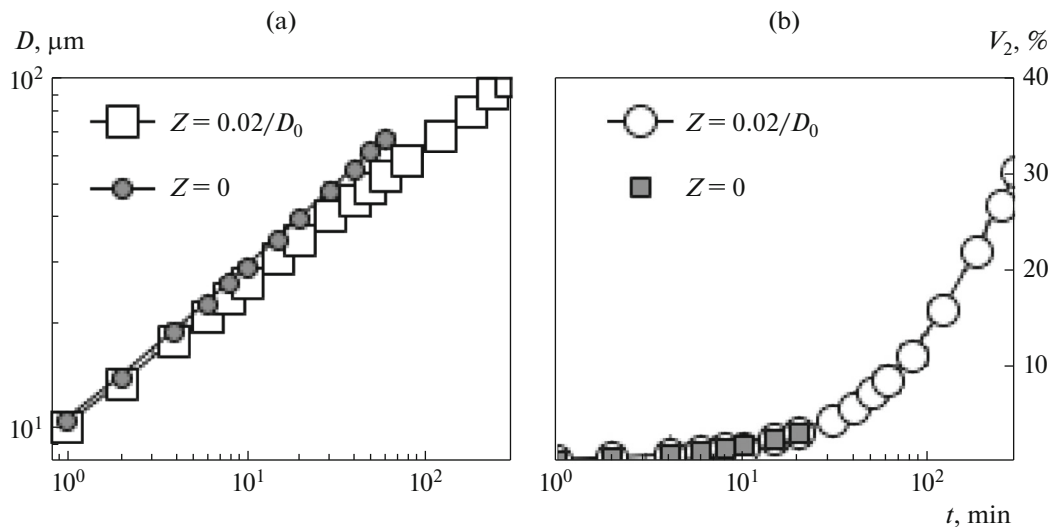
Fig. 4. Effect of the Z retarding force on (a) the kinetics of grain growth and (b) the change in the relative grain volume of the second maximum V_2 in the P1 polycrystal. D_0 is the average grain size in the initial structure.

the second GSD maximum formed by $\{110\}\langle001\rangle$ crystallites.

CONCLUSIONS

(1) Grain growth induced by boundary curvature in a texture-free material with abnormally coarse crystallites was investigated by numerical simulation.

(2) Grain growth in a pure material with a bimodal structure causes only one of two maxima to remain. This fact depends on the number of coarse crystallites in the initial structure.

(3) The second maximum disappears when the number of coarse crystallites is small. The development of normal grain growth in the fine-grained matrix is responsible for its disappearance.

(4) When the matrix grain size is decreased, the second maximum disappears more rapidly.

(5) A decrease in the grain growth rate causes the second distribution maximum to retain and be more pronounced.

REFERENCES

1. Y. Wang, M. Chen, F. Zhou, and E. Ma, "High tensile ductility in a nanostructured metal," Nature **419**, 912–915 (2002).

2. M. Zha, H-M. Zhang, Z-Y. Yu, X.-H. Zhang, X.-T. Meng, H.-Y. Wang, and Q.-C. Jiang, "Bimodal microstructure—A feasible strategy for high-strength and ductile metallic materials," J. Mater. Sci. Technol. **34**, 257–264 (2018).

3. H. W. Höppel, M. Korn, R. Lapovok, and H. Mughrabi, "Bimodal grain size distribution in UFG materials produced by SPD: Their evolution and effect on mechanical properties," J. Phys.: Conf. Ser. **240**, 012147 (2010).

4. D. Witkin, Z. Lee, R. Rodriguez, S. Nutt, and E. Lavernia, "Al−Mg alloy engineered with bimodal grain size for high strength and increased ductility," Scr. Mater. **49**, 297−302 (2003).

5. G. Niu, H. Wu, D. Chang, N. Gong, and D. Tang, "Study of microstructure and properties of bimodal structured ultrafine-grained ferrite steel," Metals **7**, 316 (2017).

6. M. Hillert, "On the theory of normal and abnormal grain growth," Acta Metall. **13**, 227−238 (1965).

7. V. Novikov, *Grain Growth and Control of Microstructure and Texture in Polycrystalline Materials* (CRC Press, Boca Raton, 1997).

8. D. J. Srolovitz, G. S. Grest, and M. P. Anderson, "Computer simulation of grain growth−V. Abnormal grain growth," Acta Metall. **33**, 2233−2247 (1985).

9. V. Yu. Novikov, "Computer simulation of normal grain growth," Acta Metall. **26**, 1739−1744 (1978).

10. V. Yu. Novikov, "On conditions for secondary recrystallization in a material with random orientation," Acta Metall. **29**, 883−887 (1981).

11. Y. Liu, M. Militzer, and M. Perez, "Phase field modeling of abnormal grain growth," Materials **12**, 4048 (2019).

12. G. Gottstein and L. S. Shvindlerman, *Grain Boundary Migration in Metals: Thermodynamics, Kinetics, Applications* (CRC Press, Boca Raton, 2010).

Translated by T. Gapontseva