

Modeling of grain growth kinetics in complexly alloyed austenite

A. A. Vasilyev[†], S. F. Sokolov, D. F. Sokolov, N. G. Kolbasnikov

[†]vasilyev_aa@mail.ru

Peter the Great Saint Petersburg Polytechnic University, 29 Polytekhnicheskaya St., St. Petersburg, 195251, Russia

A quantitative model is presented to describe the kinetics of grain growth in complexly alloyed austenite. The model assumes that the activation energy of grain growth is proportional to the activation energy of bulk self-diffusion, which is calculated as a function of the chemical composition of the solid solution using the previously obtained formula. The empirical parameters of the model are determined on the basis of experimental data on the kinetics of isothermal grain growth in steels with the chemical composition varying in a wide range: C (0.05 ÷ 0.32), Mn (0.30 ÷ 1.88), Si (0.01 ÷ 0.29), Ni (0.0 ÷ 4.0), Cr (0.0 ÷ 2.0), Mo (0.0 ÷ 0.5), Nb (0.00 ÷ 0.05) available in the literature. The model allows one to obtain good agreement with the experiment for the considered steels in which the minimum (~ 79.7 kJ/mol) and maximum (~ 243.7 kJ/mol) values of the activation energy of grain growth differ by 3 times. The average absolute value of the relative error in calculating the grain size is about 11% that is comparable to the measurement error. Taking into account the influence of the chemical composition on the activation energy of grain growth, implemented in the developed model, it is possible to obtain agreement with the experiment without accounting for the solid-solution pinning of moving boundaries (the solute drag effect) requires a large number of additional empirical parameters (two exponential parameters for each alloying element). This result deserves further consideration from the physical viewpoint and verification on both simple carbon steels and steels with various quantities of Mn, Mo and Nb, which, according to literature, exert the strongest solute drag effect.

Keywords: austenite, grain growth, alloying, modeling.

1. Introduction

The normal grain growth exerts a notable effect on the austenite microstructure during hot rolling and strongly influences the microstructures of the heat affected zone in weld joints of steels. Owing to the practical significance of this phenomenon, it has attracted attention in many original works [1–17] and in monographs [18,19]. Grain growth is conventionally expressed [1–7] by the equation:

$$D^n - D_0^n = A \exp\left(-\frac{Q_{GG}}{RT}\right)t, \quad (1)$$

where D is the grain size at time t , D_0 is the initial size, Q_{GG} is the apparent activation energy of the process fitted to experimental data, A and n are the auxiliary parameters. According to various applications of Eq. (1), to fit the experimental results, n commonly exceeds 2 corresponding to parabolic growth originally predicted in the classical work [20] for pure metals. Higher values of n are ascribed to the growth retardation due to both phase precipitations (Zener's pinning [18]) and solute atoms of alloying elements or impurities (solute drag [21–23]). The corresponding activation energies has no certain physical meaning and strongly vary depending on the composition of the steel and the temperature range under study [2]. Other works [8–17] apply a more physically motivated approach where differential equations for the grain growth allow for the pinning of migrating boundaries by particles. However,

when evaluating the mobility of boundaries, the apparent activation energy Q_{GG} of their migration (activation energy of growth) is still treated as a fitting parameter independent of the chemical composition. It should be noted that all current models of this kind satisfactorily predict the austenite grain growth only with steel compositions previously used in their calibration.

According to the physics of diffusional rearrangements of atoms during the boundary migration of growing grains, the related activation energy should be comparable with the activation energy of grain boundary self-diffusion that, in turn, correlates with a similar characteristic for the bulk self-diffusion (activation energy of self-diffusion, AESD), which is higher by about two times. It is notable that known results obtained by the radioactive tracer technique, which have been compiled by the authors [24], undoubtedly indicate that AESD in solid solutions of austenite depends on their chemical compositions. An empirical expression proposed in the last quoted paper enables accurate calculation of AESD in terms of quantities of alloying elements (C, Mn, Si, Ni, Cr, Mo, Nb, V, Ti) most important for up-to-date steels. According to this expression, the addition of Mn, Si, Mo, Nb or Ti may result in an essential increase of AESD whereas Ni, Cr, V and, especially, C reduce this term. With the mentioned proportionality of activation energy of the grain boundary self-diffusion to AESD kept in mind, it is advisable to employ the former with allowance for its dependence on the chemical composition in modeling the

grain growth kinetics. This approach is the first to discard numerous auxiliary parameters conventionally related to the effect of the chemical composition on the solute drag mechanism [21–23].

The present paper formulates a model for grain growth in austenite depending on the elemental composition of the solid solution. Wide literature data have been used to determine the empirical model parameters relevant to the kinetics of the process in steels of various compositions.

2. Model description

The material energy reduction due to diminishing the whole boundary area is considered for a thermodynamic driving force of the grain growth. In case of pure metals or solid solutions the effective pressure P_G moving the boundaries takes the form [18]:

$$P_G = \alpha_G \frac{\gamma_{GB}}{D}, \quad (2)$$

where D is the average grain size (volumetric diameter), γ_{GB} is the specific energy of grain boundaries, α_G is the coefficient of the order of unity. Allowing for the influence of the chemical composition on mobility of boundaries in alloyed austenite, the boundary velocity V_G is expressed by:

$$V_G \equiv V_G(T, Y_{AE}) = M_{GB}(T, Y_{AE}) P_G, \quad (3)$$

where $M_{GB}(T, Y_{AE})$ specifies mobility, and $Y_{AE} = \{y_C, y_{Mn}, y_{Si}, \dots\}$ represents a set of average fractions of the sites of substitution and interstitial sublattices occupied, respectively, by the atoms of substitution alloying elements and carbon. Mobility term is expressed with allowance for entropy:

$$M_{GB}(T, Y_{AE}) = M_0 \exp\left(\frac{S_{GG}(Y_{AE})}{R}\right) \exp\left(-\frac{Q_{GG}(Y_{AE})}{RT}\right), \quad (4)$$

where $Q_{GG}(Y_{AE})$, $S_{GG}(Y_{AE})$ are activation energy and entropy of the grain growth, respectively, and M_0 is the empirical parameter, R is the gas constant and T is the absolute temperature. According to [24], the activation entropy proportional to the corresponding energy will be treated with the fitting parameter β_{GG} , i.e. $S_{GG}(Y_{AE}) = \beta_{GG} Q_{GG}(Y_{AE})$.

The present model presumes that $Q_{GG}(Y_{AE}) = \alpha_{GG} Q_{SD}(Y_{AE})$, where $Q_{SD}(Y_{AE})$ is AESD in the solid solution of complexly alloyed austenite and α_{GG} is the empirical parameter. According to [24], the dependence of AESD on the chemical composition of the solid solution takes on the form:

$$\begin{aligned} Q_{SD}(Y_{AE}) = & 311691 - 278242(1 - \exp(-0.394 y_C)) + \\ & + 88752 y_{Mn}^{0.31} + 22801 y_{Si} - 6490 y_{Cr} + \\ & + 84864 y_{Mo}^{0.65} - 38575 y_{Ni}^{0.3} - 7298 y_V + \\ & + 132594 y_{Nb}^{0.263} + 82128 y_{Ti}^{0.401} \text{ (J/mol)} \end{aligned} \quad (5)$$

and the rate of grain growth is expressed by:

$$\frac{dD(t)}{dt} = 2V_G = K_G(T, Y_{AE}) \frac{1}{D(t)}, \quad (6)$$

where function $K_G(T, Y_{AE})$ corresponds to Eqs. (2) – (4). At a constant temperature Eq. (6) is easily integrated and leads to

the classic parabolic dependence [20]:

$$D^2(t) - D_0^2 = 2K_G(T, Y_{AE})t, \quad (7)$$

where D_0 is the initial grain size.

In reality, however, the grain growth kinetics in austenite is complicated due to the retarding influence of various small particles of carbides, nitrides or carbonitrides, which remain in steel up to high temperatures. This effect (Zener's pinning) is taken into account in terms of the effective moving pressure [19]:

$$P_G^* = P_G - P_Z, \quad (8)$$

where P_Z is Zener's pressure:

$$P_Z = \alpha_Z \gamma_{GB} \sum_i \frac{f_{pi}}{R_{pi}}, \quad (9)$$

expressed with volume fraction f_{pi} and the average radius R_{pi} for i -th type of particles, factor α_Z is of the order of unity. With (8) and (9) kept in mind, equation:

$$\frac{dD(t)}{dt} = K_G^*(T, Y_{AE}) \left[\frac{1}{D(t)} - \alpha_Z \sum_i \frac{f_{pi}}{R_{pi}} \right], \quad (10)$$

expresses the grain growth rate, where $\alpha_Z^* = \alpha_Z / \alpha_G$, and $K_G^*(T) = \alpha_G \gamma_{GB} K_G(T)$.

Yet, it is difficult to make use of Eq. (10) since even in case of the single type of the barrier particles accurate determination of their dimensions and volume fraction [17] is rarely possible. That is why, to allow for Zener's pressure in empirical models, a virtually ultimate grain size D_{lim} is introduced:

$$D_{lim} = \frac{1}{\alpha_Z} \left(\sum_i \frac{f_{pi}}{R_{pi}} \right)^{-1}, \quad (11)$$

which value should be fitted to relevant experimental data. Making use of such an expedient, we express the growth rate, depending on chemical composition, as follows:

$$\begin{aligned} \frac{dD(t)}{dt} = & M_0^* \exp\left(\frac{\beta_{GG}^* Q_{SD}(Y_{AE})}{R}\right) \times \\ & \times \exp\left(-\frac{\alpha_{GG} Q_{SD}(Y_{AE})}{RT}\right) \left[\frac{1}{D(t)} - \frac{1}{D_{lim}} \right]. \end{aligned} \quad (12)$$

Apart from D_{lim} , Eq. (12) employs only three fitting parameters (M_0^* , α_{GG} , $\beta_{GG}^* = \beta_{GG} \alpha_{GG}$) determined in calibration on numerous steels of various chemical composition. It is notable that allowance for reduction of Q_{SD} by certain alloying elements, mentioned in the introduction, enables the model to predict their accelerating effect on the grain growth.

3. Model calibration, calculation results and their discussion

The model has been calibrated with data on the isothermal grain growth kinetics for two industrial (S1 (DQSK), S2 (A36) [9,25]) and 8 laboratory (S3–S10 [3,11]) steels whose chemical compositions are listed in Table 1 (for brevity sake, they are shown with only two decimal digits).

Table 1. Chemical compositions (wt.%) of steels used to calibrate the model. Q_{SD} is the activation energy of bulk self-diffusion calculated with Eq. (5) provided that all alloying elements are in solid solution (its minimum and maximum values are marked in bold).

Steel	C	Mn	Si	Ni	Cr	Mo	Nb	Al	N	Q_{SD} , J/mol	Reference
S1	0.17	0.74	0.01	-	-	-	-	0.04	0.005	159483	[9, 25]
S2	0.04	0.30	0.01	-	-	-	-	0.04	0.005	177402	
S3	0.08	0.49	0.26	-	-	0.49	-	-	-	394163	[3]
S4	0.21	1.47	0.27	-	1.0	0.49	-	-	-	366351	
S5	0.32	1.01	0.29	-	2.0	0.50	-	-	-	322450	
S6	0.09	0.95	0.27	2.0	1.0	0.50	-	-	-	361565	
S7	0.19	0.52	0.27	2.0	2.0	0.49	-	-	-	306470	
S8	0.20	1.04	0.26	4.0	-	0.49	-	-	-	309090	
S9	0.32	1.51	0.27	2.0	-	0.50	-	-	-	292282	
S10	0.05	1.88	0.04	-	-	0.49	0.048	0.05	0.004	487472	[11]

The austenite grain size in steels S1 and S2 has been evaluated using optical microscopy. To this end, the specimens were quenched to get final structures which enable appropriate chemical etching to reveal the boundaries of the previous austenite grains. Apart from fully martensitic states, particular structures with ferritic contours decorating the sought boundaries were employed. Both the experimental procedures and the image treatment are described in detail elsewhere [9, 25]. According to these works, the relative error of thus determined grain size does not exceed 10%. A similar approach [3] used for steels S3 to S9 evaluates the former grain size with about the same error. It is worth noting that the average grain diameters (equivalent area diameters, EQAD) evaluated on planar sections have been multiplied by about 1.2, following [25], in order to get actual volumetric diameters. The data on the grain growth kinetics in steel S10 have been obtained by the “laser ultrasonics” technique [11], previously verified by conventional metallographic analysis.

As follows from the employed data on the kinetics of the grain growth in austenite of the considered steels at 1000 to 1200°C, this process develops under the retarding effect of undissolved particles. Thus, in Al-killed industrial steels S1 и S2 particles of AlN [9] do matter whereas in S10 boundary migration is affected by both AlN and Nb (C,N) particles that corresponds to a variation of the temperature-dependent parameter n in Eq. (1) in the range 3.5 to 5 [11]. In case of steels S3–S9, the parameter n exceeds 4 that also the evidence of a strong barrier effect of the particles. However, there are no relevant data in [3] to clear up their type.

Values of D_{lim} substituted in Eq. (12) to implement the modeling have been found on experimental plots $D(t)$ of the grain growth as virtual asymptotes for greater time. Values of the remaining parameters of the model, ensuring the minimal average relative error in calculating the grain size,

were found on the basis of experimental data only for the highest temperatures (1150 and 1200°C). Here, 10 of 31 data sets were employed with D_{lim} set to 350 μm .

Calibration involved two steps where the first one was aimed to find a set of values for combined parameter $M_0^*(Y_{AE}) = M_0^* \exp(\beta_{GG}^* Q_{SD}(Y_{AE})/R)$, as well as optimal α_{GG} , that proved to be close to 0.5 as expected. The second step making use of $\{M_0^*(Y_{AE})\}$ set has determined $M_0^* = 4.8 \times 10^2 \text{ m}^3 \text{N}^{-1} \text{s}^{-1}$ and $\beta_{GG}^* = 3.91 \times 10^{-4} \text{ K}^{-1}$. Fig. 1 represents the corresponding predictions for the kinetics of the grain growth compared to relevant experimental data for some of the considered steels. As these plots evidence, the model complies with the experiment not only at high but also at relatively low temperatures.

Fig. 2 providing a comparison of the predicted to the actual grain sizes for all considered steels and temperatures confirms the model performance; indeed, the average magnitude of the relative error does not exceed 11% comparable to the measurement errors. An essential variation of the chemical compositions where the model works also should be noted; corresponding activation energies of the grain growth vary in the range where the highest (243.7 kJ/mol, S10) and lowest (79.7 kJ/mol, S1) values differ by about three times (Table 1).

The model allows for the influence of the chemical composition of the solid solution on the mobility of the boundary in a novel way. Unlike the classical solute drag theory [21] or the up-to-date “statistical solute-pinning theory” [22, 23], the considered proportionality of the growth activation energy to the activation energy of the bulk self-diffusion enables the model to get rid of numerous auxiliary parameters specific for particular alloying elements. It is remarkable that even with such a simplification, the model shows good results on steels with high contents of Mn, Mo and Nb, to which the most pronounced solute drag is commonly attributed to [11].

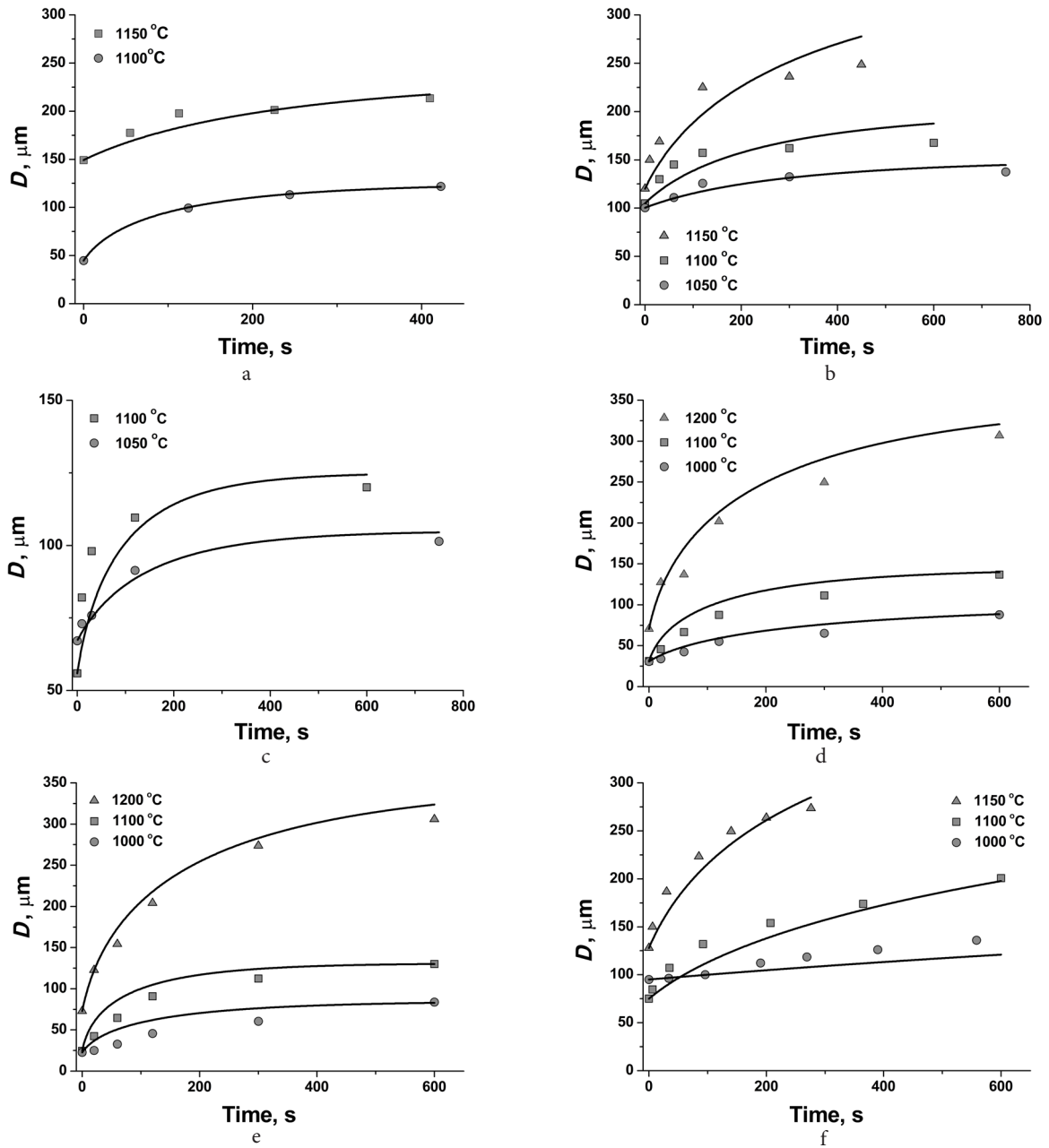


Fig. 1. Curves for grain growth kinetics according to the model and respective experimental data [3, 9, 11, 25] on steels S1 (a), S2 (b, c), S4 (d), S8 (e), S10 (f).

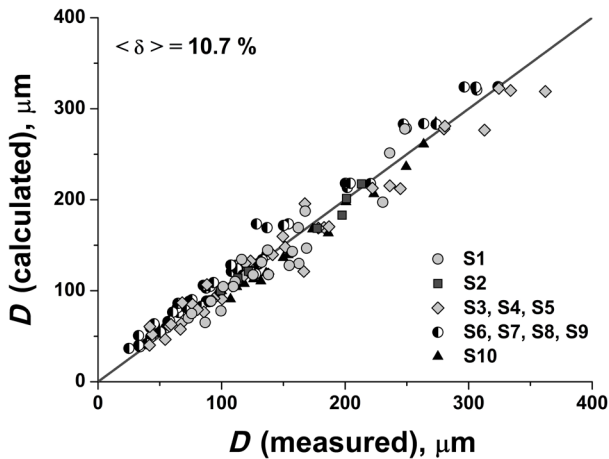


Fig. 2. Comparison of model predictions to actual grain sizes; $\langle \delta \rangle$ is a magnitude of the average relative error.

4. Conclusions

A simple quantitative model for the grain growth kinetics in austenite has been formulated that is based on the proportionality of the activation energy of the process to the activation energy of the bulk self-diffusion, the latter being dependent on the chemical composition of the solid solution according to the previously established empirical expression. Empirical parameters of the model are fitted to relevant literature data for steels of various compositions.

The corresponding predictions comply well with the experiments on various steel compositions where the highest (243.7 kJ/mol) and lowest (79.7 kJ/mol) values of the calculated activation energy differ by about three times. The average magnitude of the relative error in calculating the grain size does not exceed 11% comparable to the measurement errors.

Taking into account the influence of the chemical composition on the activation energy of grain growth, implemented in the developed model, it is possible to obtain agreement with the experiment without accounting for the solid-solution pinning of moving boundaries (the solute drag effect) requires a large number of additional empirical parameters. This result deserves further consideration from the physical viewpoint and verification on both simple carbon steels and steels with various quantities of Mn, Mo and Nb which, according to literature, exert the strongest solute drag effect.

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