

MODELLING GRAIN GROWTH KINETICS IN STEELS

Steel is a versatile material that has found widespread use because of its mechanical properties, its relatively low cost, and the ease with which it can be used in manufacturing process such as forming, welding and machining. Regarding to mechanical properties are strongly affected by grain size and chemical composition variations. Many industrial developments have been carried out both from the point of view of composition variation and grain size in order to exploit the effect of these variables to improve the mechanical properties of steels. It is also evident that grain growth are relevant to the mechanical properties of steels, thus suggesting the necessity of mathematical models able to predict the microstructural evolution after thermo cycles. It is therefore of primary importance to study microstructural changes, such as grain size variations of steels during isothermal treatments through the application of a mathematical model, able in general to describe the grain growth in metals. This paper deals with the grain growth modelling of steels based on the statistical theory of grain growth originally developed by Lücke [1] and here integrated to take into account the Zener drag effect and is therefore focused on the process description for the determination of the kinetics of grain growth curves temperature dependence.

Keyword: Grain growth, steel, computer simulation, kinetics.

1. Introduction

Controlling the evolution of microstructure is crucial to the optimization of steels properties through processing. Consequently, it has been a primary goal of computational efforts in metallurgy to develop the capability to model microstructural transformations under realistic conditions with predictive accuracy [2]. For this purpose, a great deal of attention has been devoted to the technologically important phenomenon of grain growth in polycrystalline materials, which has been studied experimentally and theoretically for decades, yet has proven surprisingly difficult to model analytically [3-4].

Grain growth after primary recrystallization is described as taking place in two different forms, either in form of continuous grain growth ("normal") or in form of discontinuous grain growth ("secondary recrystallization"). In the development of this area, two stages can be distinguished. In the first stage simple and mostly qualitative interpretations of grain growth have been given. For continuous grain growth, Beck [5] predicted an increase of the average grain diameter with time as $t^{1/2}$ which was practically never found. The second stage of the development of this area is characterized by more sophisticated approaches. Hillert [6] transferred the statistical treatment of Ostwald ripening of precipitates to grain growth according to the Lifshitz-Slyozov-Wagner theory [7-8]. Moreover Hunderi

and Ryum [9] introduced a deterministic model considering individual boundaries and describing the change of size of the individual grains by an extremely large set of differential equation (one for each grains) which they solved numerically. Finally Abbruzzese [1] developed further the Hillert model by calculating a critical radius that was only postulated by Hillert [6]. The main novelty of Abbruzzese study was to use discrete grain size classes which reduce significantly the numbers of differential equations (one for each class) and thus to the possibility to calculate numerically the evolution of the grain size distribution. In recent years the Monte-Carlo simulation was widely used to simulate grain growth including also the case of Zener drag [10]. Hesselbarth and Gobel used, with success, a method named cellular automata in simulation of the theory of Johnson-Mehl-Avrami-Kolmogorov and other successful mesoscale simulations for microstructural evolution including front tracking model [11], vertex model [12] and phase field model [13] have been developed. While analytical models, such as Abbruzzese and Lücke, predict all the characteristics of microstructural evolution (i.e., grain size and grain size distribution), the goal of mesoscale computational simulations is rather different: to generate snapshots of the evolving microstructure with time. Using the computational version of metallography, both local and ensemble properties of the microstructure may be determined from these snapshots.

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2. Description of the grain growth statistical model

As far as concerns grain growth, the statistical model, originally developed by Abbruzzese and Lucke [1], is based on the assumptions of:

- Super-position of average grain curvatures in individual grain boundaries. A grain v is characterized by volume V_v , is assumed to grow at the expense of a neighbouring grain μ with a rate:

$$\left(\frac{dV_v}{dt}\right)_\mu = Ms_{v\mu} \left(\frac{1}{R_\mu} - \frac{1}{R_v}\right) \quad (1)$$

Here the ‘‘radius’’ R_v of a grain v is defined according $V_v = 4\pi R_v^3/3$. $S_{v\mu}$ is the area of contact between the two grains v and μ , and m , γ and $M = 2m\gamma$ represent respectively, the mobility, the tension and the diffusivity of the grain boundary $v\mu$. By taking into account all N_v neighbours of this grain one obtain for all its total growth rate:

$$\frac{dR_v}{dt} = \frac{1}{4\pi R_v^2} \frac{dV_v}{dt} = \frac{M}{4\pi R_v^2} \sum_{\mu=1}^{N_v} S_{v\mu} \left(\frac{1}{R_\mu} - \frac{1}{R_v}\right) \quad (2)$$

With $v = 1, 2, \dots, N_G$ and N_G being the total number of grains, this expression represent a system on N_G differential equation for the unknown $R_v(t)$. However, because of the large numbers of grains N_G and thus a large number of equation is necessary to obtain a significant simulation, this leads to a great computational difficulties. Therefore, with the second and third simplifying assumptions will be easy to overcome these difficulties:

- Homogeneous surroundings of the grains. As a first approximation is assumed that for each grain v the individual neighbourhood of N_v individual grain can be replaced by a surrounding obtained by averaging over a neighbourhood of all grains of the same radius R_v . Since then all grains of the same radius would have the same surrounding, also their growth rate would be equal. This means that then all grains could be collected in classes characterized by their radius and that the behaviour of only different classes has to be considered, instead of single grains. In the following these classes will be denoted by the indices i, j, \dots, N_s being the total number of classes. From the mathematical point of view the simplification consists in replacing in Equation 3 the individual contact area $s_{v\mu}$ by averaged area $a_{ij} = A_{ij}/n_j$, n_j is the total number of grains in class j and A_{ij} is the total area of contact between the two classes i and j . Then, it follow that:

$$\frac{dR_i}{dt} = \frac{M}{4\pi R_i^2} \sum_{j=1}^{N_s} a_{ij} \left(\frac{1}{R_j} - \frac{1}{R_i}\right) \quad (3)$$

- A random array of the grains namely the probability of contact among the grains is only depending on their relative surface in the system. In this case the area of a grain of the class i is divided between the neighbouring grains of the class j in proportion to the individual surface area:

$$a_{ij} = 4\pi R_i^2 \frac{n_j 4\pi R_j^2}{\sum_j n_j 4\pi R_j^2} \quad (4)$$

The integration of all the above assumptions in the model leads to the following final form of the grain growth rate equation:

$$\frac{dR_i}{dt} = M \sum_j \left(\frac{1}{R_j} - \frac{1}{R_i}\right) \frac{n_j 4\pi R_j^2}{\sum_j n_j 4\pi R_j^2} \quad (5)$$

Where $M = 2m\gamma$ again the boundary diffusivity and in our case study m was evaluated according to the Stokes-Einstein relationship [14]:

$$m = \frac{D}{k_B T} = \frac{D_0}{k_B T} e^{-\frac{\Delta E}{k_B T}} \quad (6)$$

Where D is the diffusion coefficient, K_B is the Boltzmann constant, ΔE is the activation energy of the process and T is the annealing temperature. D was chosen proportional to the diffusion coefficient of Fe in Fe- γ .

According to Zener [4], particles cause inhibition of grain boundary motion which can be considered as a retarding forces acting homogeneously along the moving boundary and has a similar behaviour of a frictional forces. In order to apply this description to grain growth phenomena has to be taken into account the magnitude of this force I_Z . In fact, for a small external driving force P no motion of the boundary takes place. Only if external force surpasses a maximum force I_{Z0} the particles can exert, the boundary moves, but with a net driving force $\Delta P = P - I_{Z0}$. This lead to three ranges for this net force acting on boundary [15]:

$$\begin{aligned} \Delta P &= P - I_{Z0} && \text{for } P > I_{Z0} \\ \Delta P &= 0 && \text{for } -I_{Z0} < P < I_{Z0} \\ \Delta P &= P + I_{Z0} && \text{for } -I_{Z0} > P \end{aligned} \quad (7)$$

For the maximum Zener force I_{Z0} the will be used the usual expression where f_p is the volume fraction and r_p is the mean radius of the particles; β is a proportional constant in the range of 0.75-1.

$$I_{Z0} = \beta\gamma \frac{f_p}{r_p} \quad (8)$$

With Equation 7 and 8, one obtains the net force ΔP_{ij} in the three ranges:

$$\begin{aligned} \Delta P_{ij}(I) &= \gamma \left(\frac{1}{R_j} - \frac{1}{R_i} - I_{Z0}\right) && \frac{1}{R_j} > \frac{1}{R_i} + I_{Z0} \\ \Delta P_{ij}(II) &= 0 && \frac{1}{R_i} + I_{Z0} > \frac{1}{R_j} > \frac{1}{R_i} - I_{Z0} \\ \Delta P_{ij}(III) &= \gamma \left(\frac{1}{R_j} - \frac{1}{R_i} + I_{Z0}\right) && \frac{1}{R_j} < \frac{1}{R_i} - I_{Z0} \end{aligned} \quad (9)$$

Therefor the growth rate for each class i can be derived as:

$$\frac{dR_i}{dt} = M \left[\begin{aligned} &\psi_I \cdot \left(\frac{1}{R_C^I} - \frac{1}{R_i} \right) + \\ &+ \psi_{III} \cdot \left(\frac{1}{R_C^{III}} - \frac{1}{R_i} \right) - (\psi_I - \psi_{III}) \cdot I_{Z0} \end{aligned} \right] \quad (10)$$

Where:

$$\psi_\lambda = \frac{\sum \varphi_j R_j^2}{\varphi_j R_j^2} \quad (11)$$

$$\frac{1}{R_c^\lambda} = \frac{\sum \varphi_j R_j}{\sum \varphi_j R_j^2} \quad (12)$$

2.1. The power law for grain growth

Burke and Turnbull were among the first to model kinetics of grain growth and they assumed that the driving force for growth is the reduction in grain boundary area and subsequent release of boundary surface energy, γ_{gb} [16]. The model was proposed for the case of normal grain growth in a pure homogeneous material and the assumption was made that only forces acting on boundaries are those due to surface curvature. It was also assumed that the average radius of curvature of a boundary is proportional to the grain diameter, d , and the rate of change of the diameter is proportional to the driving force for growth F , as described by:

$$\frac{d(d)}{dt} = k * F \quad (13)$$

Where k is a constant of proportionality.

Integrating Equation 13 Burke and Turnbull were able to derive the parabolic equation for the kinetics of isothermal grain growth [16]:

$$d^2 - d_o^2 = K * t \quad (14)$$

Where d_o the grain size is at time equal to zero, t is time, K is the grain growth constant. Equation 15 defines the relation between the proportionality constant K and temperature T at which the grain grow.

$$K = K_0 e^{\frac{-Q}{RT}} \quad (15)$$

Where Q is the activation energy for grain growth, R is the gas constant and T is the absolute temperature.

Unfortunately, experimental result often do not obey to the parabolic equation. For this reason, the original equation is often modified and used in this form [18-19]:

$$d^n - d_o^n = K t \quad (16)$$

Where n is the grain growth exponent. The equation 16 is referred to as the power law for grain growth. In an ideal state, grain

growth is controlled only by diffusion. In this case the exponent $n = 2$ and the Equation 2 for ideal grain growth for simulation calculation is used. Furthermore, grain growth can be controlled, for example, by diffusion together with the precipitation phase in growing grains, the exponent $n = 3$ is used. If the common effect of precipitation and diffusion along grain boundaries is observed, the exponent $n = 4$ is used. In the case when the grain growth is mainly influenced by precipitation, the exponent is close to the value $n = 5$ [18].

3. Results

3.1. Determination of the grain growth kinetics

The model developed by Abbruzzese and Lucke allow to simulate the annealing process of the steel and gives as output the mean radius of the austenitic grains over time. Three temperatures, ranging from 1000°C to 1200°C, have been investigated to evaluate the grain growth constant value K . During the first phase the initial grain size value D_0 and partial pre-exponential constant are determined for every holding temperature. Each of the tested temperature are expressed by the trend line (Fig. 1) and its basic equation by the fitting method, with n parameter of the Equation 16 equal to 2 (for ideal grain growth). From the trend line slope it is possible to use the value of the partial pre-exponential constant K_T and the initial grain size D_0^2 for the particular temperature, which is obtained from the fitting line equation at the starting time. All measured and computed values are shown in Table 1.

TABLE 1

Partial pre-exponential values K_T and grain size D_0^2 .

	Simulated temperature		
	1000°C	1100°C	1200°C
K_T [$\mu\text{m/s}$]	0,23	0,86	5,82
D_0^2 [μm]	5,70	5,82	2,68

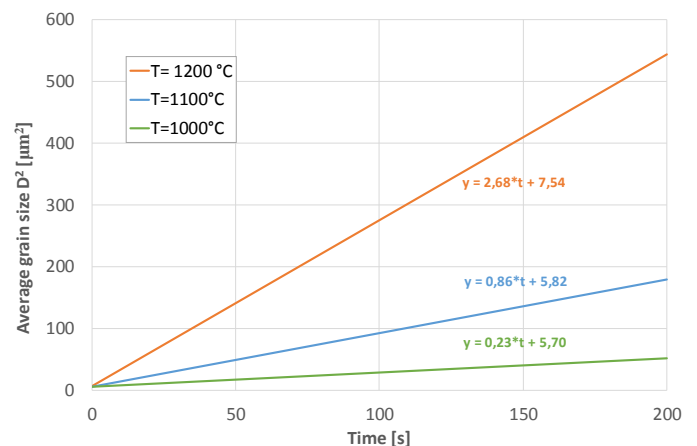


Fig. 1. Square dependence of average grain size on time

Result shows, according to Equation 15, that the values of the acquired partial pre-exponential constant K_T , which are dependant on reversed values of the absolute holding temperature (Fig. 2 and 3).

$$K = 6,80 * 10^{-5} e^{\frac{-1,49 * 10^{-4}}{T}} \quad (17)$$

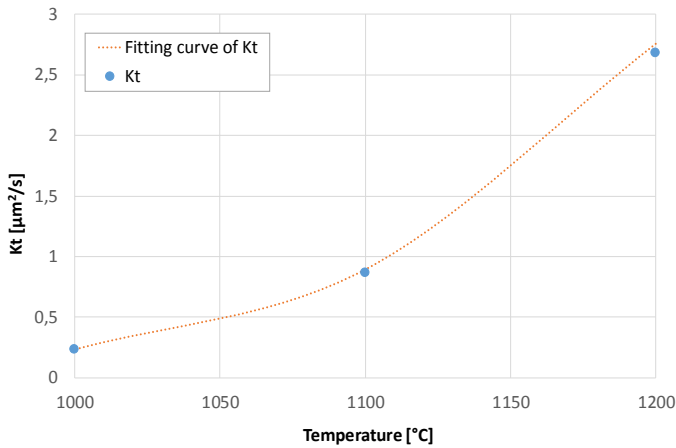


Fig. 2. Dependence of proportionality constant K_T (blue dot), obtained from the model, on absolute temperature values and exponential fitting curve of K_T (dashed orange line)

The activation energy Q and the total pre-exponential constants K_0 it is possible to obtain by the way shown in Fig. 3. The logarithm values of the acquired partial pre-exponential constant K_T , which are dependent on reversed values of the absolute holding temperature, are plotted in the graph. Thanks to that, the trend line can be created. Its equation can be expressed in the following format:

$$y = A * x + C \quad (18)$$

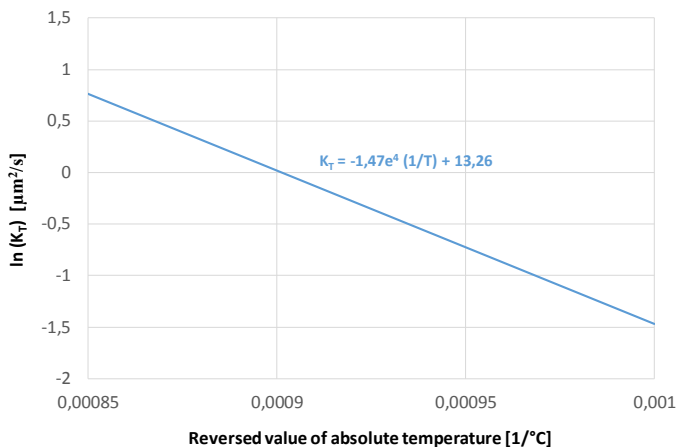


Fig. 3. Dependence of proportionality constant K on absolute temperature reversed value.

The constant C in Equation 18 defines the slope of the trend line, and will be, together with the gas constant R , further used to calculate the activation energy Q in agreement with Equation 19.

The constant B is used to calculate the total value of the total pre-exponential constants K_0 according to Equation 20.

$$Q = -(R * A) \quad (19)$$

$$K_0 = e^C \quad (20)$$

According to the trend line in Figure 3 and by means of Equation 19 and 20, activation energy values $Q = 123.88$ KJ/mol of the grain growth and the total pre-exponential constant $K_0 = 68.03 \times 10^4 \mu\text{m}^2/\text{s}$ were computed and this values is in agreement with the literatures data [20]. Inserting in Equation 15 the values of absolute holding temperature was possible to plot the curve for the grain growth kinetic. From the Figure 4 is evident that more intensive grain growth occurs under temperatures after 1200°C .

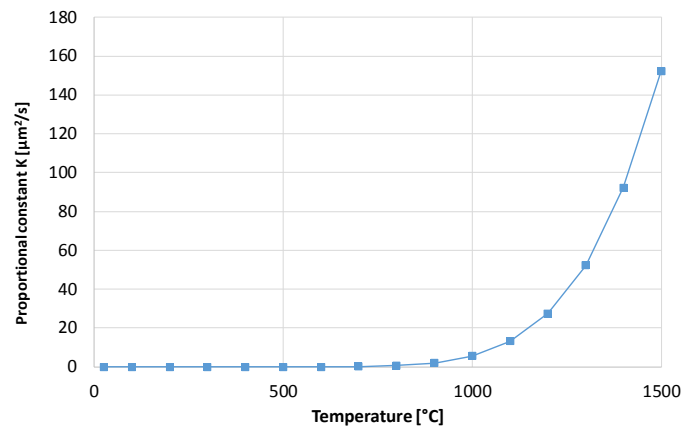


Fig. 4. Grain growth kinetic curves K

2.2. Grain growth kinetic in presence of Zener drag

One of the free input of the model parameter is the Zener drag parameter. One value of Zener parameter has been tested ($I_z = 500$) and the same analysis of the previous case (without precipitation) has been performed. The same three temperature, from 1000°C to 1200°C has been analyzed and in Fig. 5 the trend

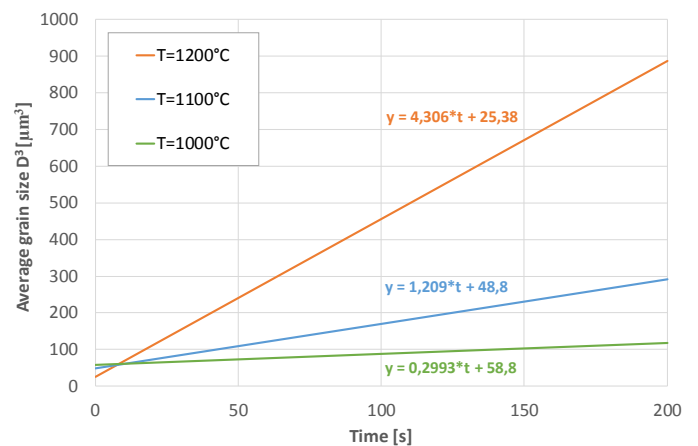


Fig. 5. Cubic dependence of average grain size on time

line for the three temperatures are plotted against holding time. In this case, according to [18], in Equation 16 for the n exponent a value of 3 has been tested due the presence of precipitates particles during the grain growth phase. In Table 2, values of K_T and D_0^3 (obtained from the trend line) are reported (Fig. 6).

TABLE 2
Partial pre-exponential values K_T and grain size D_0^3

	Simulated temperature		
	1000°C	1100°C	1200°C
K_T [$\mu\text{m}^3/\text{s}$]	0,2993	1,209	4,306
D_0^3 [μm]	58,03	48,8	25,39

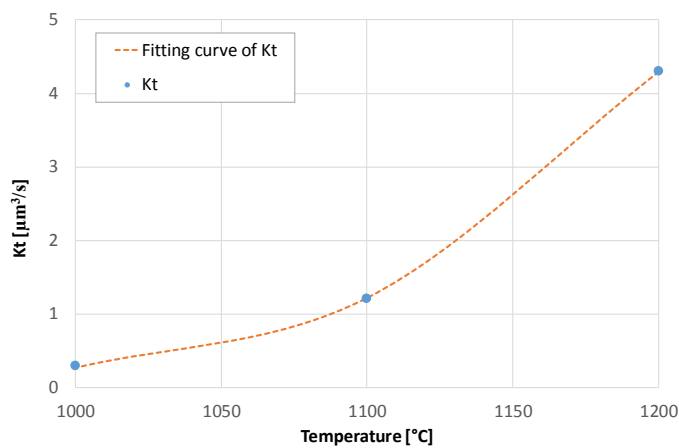


Fig. 6a. Dependence of proportionality constant K_T (blue dot), calculated from the model, on absolute temperature values and exponential fitting curve of K_T (dashed orange line)

As in the previous case, without precipitation, K_T follows an exponential trend as can be seen in Equation 21 that was obtained from fitting the model data.

$$K = 4,40 * 10^{-5} e^{\frac{-1,681 * 10^{-4}}{T}} \quad (21)$$

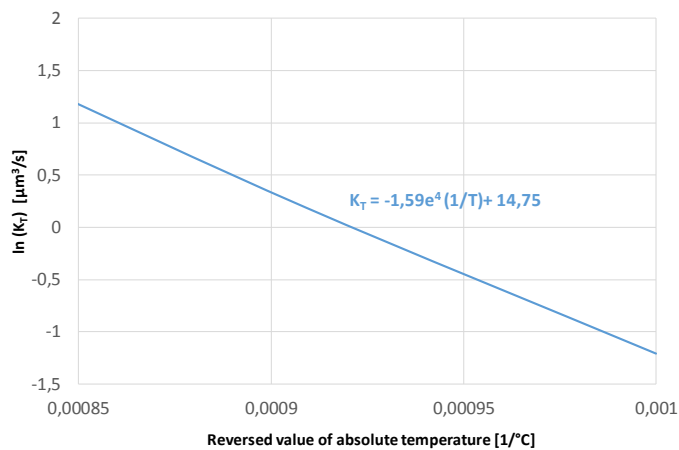


Fig. 6b. Dependence of proportionality constant K on absolute temperature reversed value

In the same manner of the first case, there were determined values $Q = -183$ KJ/mol and $K_0 = 25,5 \times 10^5 \mu\text{m}^3/\text{s}$ for the case with Zener drag effect. This data can be used to define the temperature dependence of proportionality K (Fig. 7).

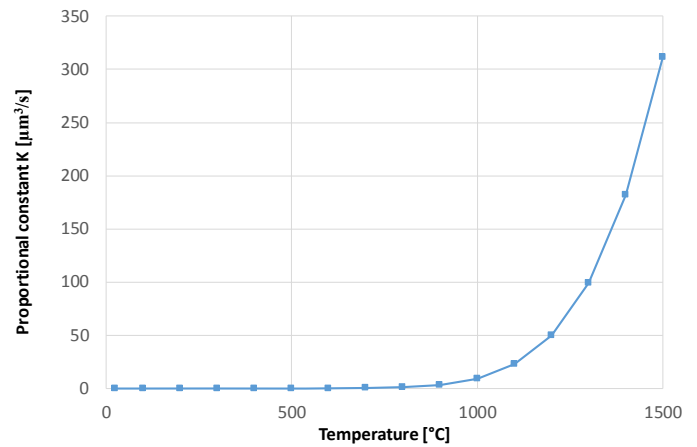


Fig. 7. Grain growth kinetics curve in case of Zener drag effect

4. Conclusions

The main conclusion are:

- The data of the mean radius of austenitic grains, obtained from the statistical model of grain growth, can be fitted according to Equation 16, with an exponent of n equal to 2 in the case without particles precipitations.
- In presence of Zener drag effect, the data can be fitted with n equal to 3, according to [18].
- In both cases, the proportionality constant K follows an exponential trend, similar to Equation 15, as suggested from [18] and more intensive grain growth occur under the temperature of 1200°C.

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