A New Approach on the Modelling of Isothermal Recrystallisation in Cold Rolled Ferritic Steels: An Application to Back-Annealing of Low Carbon Sheet Steels

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A new approach on the modelling of isothermal recrystallisation in cold rolled ferritic steels based on Avrami equation is presented here. The new model corrects some of the fundamental shortcomings of the classical JMAK modelling, such as non-random (clustered) nucleation and decreasing grain growth velocity. It is shown that the new model successfully predicts the recrystallisation degree of the deformed material during an isothermal annealing, depending on the cold rolling grade and annealing conditions. Finally, an application for back annealed cold rolled steels is illustrated. [doi:10.2320/matertrans.MRA2008149]

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1. Introduction

During deformation processes such as cold rolling, which is a necessary stage in production, a large amount of dislocations is introduced into the material, which can then be re-released during a subsequent annealing treatment. The microstructural observation of this recrystallisation process is characterized by a progressive substitution of deformed grains by new, relatively defect-free grains, usually described by a classic nucleation and growth mechanism.1) Due to its direct industrial relevance the influence of parameters such as cold rolling degree,2) chemical composition3) and annealing conditions4) of this phenomenon has been studied in detail. In some cases, the theoretical considerations and experimental verifications were then used to design a kinetic model which predicted the evolution of the fraction of the recrystallized material in function of the input parameters.5,6) However, the simplifications applied in some of these models were not always convincingly motivated, whereas in other cases the introduction of fitting parameters hindered a physical interpretation of the created model.

It is the objective of this work to present a model which uses physically relevant parameters, exclusively, in order to enable a precise prediction of the evolution of the recrystallization in cold rolled ferritic steels. This includes a careful selection of the relevant input parameters, theoretical considerations on the nucleation and growth process, set-up of the model and, finally, experimental validation. The proposed isothermal recrystallization model takes into account the deformation degree and the annealing conditions considering a single-type nucleation process, characterized by grains of the {111} type. The predicted values are compared with experimental results, showing good concordance between both series.

On the other hand, during cold rolling the steel becomes intensely hardened but loses almost all its ductility. In conventional processing the cold rolled steel is fully recrystallised by annealing for the purpose of restoring ductility, but at the expense of strength. Then, if higher strength is demanded it is achieved by either alloying the steel or by special heat treatments at higher temperatures. The above procedure entails wastage of energy and increased costs. On the other hand, by controlled low temperature annealing, it is possible to retain as much of the strength from cold rolling as possible while at the same time restoring adequate ductility. This allows to yield a material with properties that are a compromise between the high strength–low ductility of fully cold rolled sheet and the low strength–high ductility of fully recrystallised sheet. This process is variously termed as ‘back annealing’, ‘partial annealing’, ‘recovery annealing’, ‘stress relief annealing’, ‘temper annealing’ or ‘controlled incomplete annealing’. Basically, the process consists of two stages, deformation and annealing. It is in this context where the proposed model presented in this paper could be very useful to determine the annealing conditions required to reach a certain recrystallised volume fraction.

2. Materials and Experimental Techniques

Three cold-rolled laboratory casts of a low carbon (LC) steel (0.014C-0.19Mn-0.016S-0.007Si-0.055Al-0.0025N) with different cold rolling reductions (CR) were studied in this work (see Table 1).

Small cylindrical samples were cut out from the cold rolled strip and subsequently an isothermal heat treatment was simulated in laboratory conditions: the samples were rapidly heated (20°C/s) up to an annealing stage at 600, 620 and 640°C, which are typical annealing temperatures for the study of the recrystallisation in low carbon steels. Finally, the samples were quenched to room temperature.

<table>
<thead>
<tr>
<th>Sample</th>
<th>CR (%)</th>
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<tbody>
<tr>
<td>L-Def</td>
<td>59</td>
</tr>
<tr>
<td>M-Def</td>
<td>70</td>
</tr>
<tr>
<td>H-Def</td>
<td>80</td>
</tr>
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The samples were then mounted in bakelite for metallographic examination and subsequently ground and polished according to standardized techniques. Nital etching was used to reveal the microstructure and a point count method was applied to quantify the volume fraction of recrystallised material.7) Texture measurements were carried out by means of the Schulz reflection method, using a D-5000 X-ray diffractometer furnished with an opened Eulerian cradle. Details of the diffractometer used and the analysis method are given elsewhere.8) The diffraction studies were performed employing Cu-Kα radiation and a diffracted beam monochromator. After grinding and final polishing using 0.25 μm diamond paste, the samples were etched to obtain an undeformed surface. The pole figures (110), (200) and (211) were measured and a series expansion technique employed to calculate the ODFs, along with ghost correction.9) As both ferrite phases present a cubic crystal symmetry and the sample shape is orthorhombic, defined by rolling, normal and transverse direction (RD, ND, TD), the orientation density, f(g), was represented in the reduced Bunge-Euler space (0 ≤ ϕ₁, ϕ, ϕ₂ ≤ π/2). The typical cold rolling and recrystallisation textures of LC steels may be described by two texture fibers, i.e., first by the incomplete α-fiber which is characterized by (110) parallel to RD, and second by the complete γ-fiber which comprises all crystals with {111} plane parallel to ND, as reported elsewhere.10)

3. Results and Discussion

3.1 Evolution of the recrystallised volume fraction

Typical micrographs of as-cold rolled and partially recrystallised samples after annealing are shown in Fig. 1. It can be seen that the deformed grains are being gradually replaced by recrystallised grains and that both types of grains can be clearly distinguished. From these micrographs the evolution of the recrystallised volume fraction can be measured, for each holding temperature, annealing time and cold reduction degree. Results on recrystallised ferrite volume fraction are shown in Fig. 2.

As expected, the recrystallisation process is faster at higher annealing temperatures and at higher degree of deformation. It is now the objective of the present work to develop a model which is able to accurately predict the recrystallisation behaviour in these steels.
3.2 Relevant parameters that directly influence the recrystallisation process of cold rolled ferritic steels

Any recrystallisation model should include the details of the precise nucleation and grain growth processes taking place during annealing. Therefore, a study of the texture evolution in the steels has been carried out, since the preferred orientation of the grains reveals fundamental information on the recrystallisation mechanisms.\(^\text{11}\)

The evolution of the macrotexture obtained from the X-ray measurements is presented in Fig. 3 for the low (L-Def) and high (H-Def) deformed samples, indicating also the highest intensity obtained in each measurement. As seen from the figures, the cold-rolled samples exhibit a similar texture. One of the most important observations is that only the formation of a-fiber nuclei is detected for the cold-rolled samples, with the orientation of the a-fiber, \(a_1 || [001]\) and \(a_2 || (110)\), being approximately the same in both steels.

The a-fiber orientation is generally attributed to the initial cold rolling texture and the nucleation of the recrystallisation process. The intensity of the a-fiber orientation is higher in the steel with a higher cold rolling degree, as seen in the figures. This is expected, as the cold rolling process causes a texture development that influences the nucleation and growth processes during annealing.

3.3 Kinetic model of the isothermal annealing of cold rolled ferritic steels: Nucleation

In the classical Avrami approach\(^\text{14}\) the nucleation occurs randomly across the materials and the strain-free grains grow isotropically at a constant rate. Both fundamental hypotheses (random nucleation and constant growth rate) have been proved to be too simplistic descriptions of the recrystallisation process.\(^\text{15,16}\)

Firstly, it is known that new grains preferentially nucleate at grain boundaries and triple-points. Instead of the homogeneous nucleation mechanism proposed by Avrami, clustered nucleation has to be considered along edges and grain boundaries.

In the present work a single-type (i.e. corresponding to \(\gamma\)-fiber nuclei) and constant nucleation rate \(N\) is considered according to: \(^\text{17}\)

\[
N = \frac{4}{V_{\text{grain}}} f \exp\left(-\frac{Q_n}{RT}\right)
\]

where \(V_{\text{grain}}\) is the volume per grain in the unrecrystallised state, \(f\) is a nucleation attempt frequency, \(Q_n\) is the activation energy for nucleation, \(R\) is the gas constant and \(T\) is the absolute temperature. The attempt frequency, \(f\), can be taken as the boundary vibration frequency and is estimated by McLean\(^\text{18}\) to be \(1.3 \times 10^{9} \text{s}^{-1}\). The number density of nucleation sites per unit volume, \(4/V_{\text{grain}}\) is approximately equal to \(N_0 S_0^2\), where \(N_0\) is the number of nucleation sites per unit grain boundary area and \(S_0^2\) is the surface area per unit volume of the deformed grain, which can be easily measured.

According to Cahn\(^\text{19}\) \(N_0\) can be approximated by \(K_s/\delta^2\) where \(K_s\) is a constant (\(K_s = 1.0 \times 10^{-15}\)) and \(\delta\) is the grain boundary thickness which is approximately \(2.5 \times 10^{-10}\) m. According to McLean\(^\text{20}\) the nucleation attempt frequency, \(f\), is related with the strain rate \(\epsilon\) through the following relation: \(f \approx \epsilon^4 f_0\), where \(f_0\) is the vibration frequency, assumed constant. Combining the above equations, the following expression for the nucleation rate can be found:

\[
N \approx \frac{K_s}{\delta^2} S_0^2 f_0 \epsilon^4 \exp\left(-\frac{Q_n}{RT}\right).
\]

3.4 Kinetic model of the isothermal annealing of cold rolled ferritic steels: Grain growth

A well known equation for the velocity of a grain boundary is given by Christian: \(^\text{21}\)

\[
G = \delta \cdot v \cdot \exp\left(-\frac{Q_{\text{diff}}}{RT}\right) \left[1 - \exp\left(-\frac{\Delta G}{RT}\right)\right]
\]

In this equation, \(v\) is the characteristic frequency for the atomic jumps across the boundary (kT/h in Eyring’s theory), \(Q_{\text{diff}}\) is the activation energy for atom transfer across the boundary during growth (set equal to the activation energy for self-diffusion in pure \(\alpha\) iron which is quoted as \(286 \text{kJ mol}^{-1}\) by Hopkin); \(\Delta G\) is the driving force for grain boundary motion.

Some researchers\(^\text{23}\) suggested that the driving force, and thus the growth rate is not a constant but decreases with time. This has been experimentally verified in different materials and is believed to be one of the main shortcomings of classical modelling. Thus, \(\Delta G\) may be written as
\[ \Delta G(t) = \Delta G^0 \cdot f(t) \]  
(4)

The time dependence, \( f(t) \), is assumed to be of an potential type \( (t^{-r}) \), whereas the time-independent driving force \( \Delta G^0 \) will depend on the dislocation density, \( \frac{Q}{C^0} \), which is proportional to the true strain \( \varepsilon \). An Arrhenius relation is used to relate the driving force \( \Delta G^0 \) with the activation energy for grain growth \( Q_0 \). Equation (4) can thus be rewritten as:

\[ \Delta G^0 = C \cdot \varepsilon \cdot \exp \left( -\frac{Q}{RT} \right) \]
(5)

Taking into account that for small driving forces \( 1 - \exp(-\Delta G/RT) \approx \Delta G/RT \) and adding the activation energies, the following relationship for the rate of growth is suggested:

\[ G_t = C \cdot \varepsilon \cdot t^{-r} \cdot \exp \left( -\frac{Q_G}{RT} \right) \]
(6)

In this expression \( C \) is a fitting constant.

### 3.5 Recrystallised volume fraction determination

The extended recrystallised volume fraction formed during isothermal annealing after a holding time \( t \) can now be calculated as follows:

\[ X_{ex}(t) = \int_{\tau}^{t} V(t - \tau), N.d\tau \]
(7)

where \( V(t - \tau) \) is the total volume of all phantom grains nucleated at time \( \tau \) within a unit of volume and \( N \) is the constant nucleation rate at which nuclei are generated per unit volume. For isotropic growth rates \( V(t - \tau) \) can be rewritten as:

\[ V(t - \tau) = K_u \left( \int_{\tau}^{t} G_s(t)dt \right)^{3} \]
(8)

where \( K_u \) is a shape factor which describes the geometrical shape of the growing grain (for spheres the value of \( K_u \) will be \( 4\pi/3 \)).

Combining eqs. (2), (6) and (7) the following expression is obtained for the extended recrystallised volume fraction:

\[ X_{ex} = C \cdot \varepsilon \cdot S_0 \cdot \exp \left( -\frac{Q_{Re} x}{RT} \right) \cdot t^{4-3r} \]
(9)

To obtain the expression for the “real” recrystallised volume fraction, \( X_v \), a previous remark has to be made. Since the nucleation in the considered steels preferentially takes place at the grain boundaries, two nearest neighbour grains are generally closer than they would be in a sample where the nuclei are randomly orientated. Thus, the number of impingements in the “clustered” sample exceeds the random sample. Gokhale et al.\(^{25}\) have taken this effect into account and have come to the following general relationship:

\[ \frac{(1 - X_v)^{1-i - 1}}{i - 1} = X_{ex} \]
(10)

where \( i < 1 \) describes the case where the nuclei are ordered, i.e., when they are equi-distant, for randomly distributed nuclei \( i = 1 \) and \( i > 1 \) for clustered impingement. The best fit was found when \( i = 2 \) which leads to the following equation for the recrystallisation in LC steels:

\[ \frac{X_v}{1 - X_v} = C \cdot \varepsilon \cdot S_0 \cdot \exp \left( -\frac{Q_{Re} x}{RT} \right) \cdot t^{4-3r} \]
(11)

The experimental values for the exponential coefficient \( r \) can be determined using:

\[ \ln \left( \frac{X_v}{1 - X_v} \right) \propto (4 - 3r) \ln t \]
(12)

This fitting procedure has been carried out for all the samples and as an example the curves are fitted for L-Def and H-Def samples annealed at 620°C (Fig. 4). From the fitted results shown in Fig. 4 the characteristic value of 4-3r for \( t \) in eq. (11) can be obtained in each case, and it can be seen that higher values of 4-3r are obtained in samples with higher \( CR \), as was expected. These fitting values (\( r = 1.20 \) for H-Def, \( r = 1.22 \) for M-Def, and \( r = 1.24 \) for L-Def samples, respectively) are then introduced in the kinetic model and a prediction of the recrystallised volume fraction is now possible. A validation of this procedure is carried out in the next section.

### 3.6 Validation of the proposed kinetic model for recrystallisation in cold rolled ferritic steels

The result of the modelling of the annealing process in cold rolled steels is showed in Fig. 5. It can be seen that an excellent agreement is obtained between the experimental values and the theoretical predictions.

When comparing the model at different annealing temperatures (Fig. 5(a)) it can be seen that only at intermediate temperatures (620°C) some deviation can be observed, mostly at low recrystallised fractions. This is probably due to an initial overestimation of the grain growth velocity at these conditions. When comparing the recrystallisation behaviour at different deformation conditions (Fig. 5(b)) it can be seen that the model leads to excellent results in case of high deformed steels, whereas at lower rolling degrees some deviation is noticed. This can be explained by the fact that the model considers only a single type of (high-energy) nucleation, which corresponds to the typical \{111\} grains. However, at lower deformations this hypothesis can no longer be sustained, since lower energy nuclei may be formed.
3.7 Application of the proposed model for back-annealed steels

Back annealing can be a successful industrial practice provided that it produces consistent mechanical properties in the sheet steel in a routine manner. It is for this reason that, for production purpose, the operator must have maximum flexibility in annealing temperature and time. In this sense, the following paragraphs will illustrate the ability of the model to predict the recrystallised volume fraction in the LC steel studied in the present work and hence the feasibility of the back annealing process of this type of material.

In the following discussion, the time to obtain 50% of recrystallised volume fraction, i.e. \( t_{50} \), will be considered as an output parameter from the proposed model. The first important variable that has been investigated is the annealing temperature for a cold rolling degree of CR of 70% (Fig. 6).

Moreover, the relation between \( t_{50} \) and \( \varepsilon \) (which is related with CR through the following expression: \( \varepsilon = 2/\sqrt{3}[100/(100 - CR)] \)) is given in Fig. 7 for different annealing temperatures.

As can be seen in these figures, the model successfully predicts that higher annealing temperatures and high deformations lead to faster recrystallisation kinetics. These results reflect the high sensitivity of the steel to annealing temperature (and time) and cold rolling degree, leading to a small processing window, in which a minor variation of temperature or deformation grade may result in large considerable modifications of the mechanical properties. This emphasizes the fact that not all the desired combinations of strength and ductility will be achievable by back annealing unless the possibility is explored of using the partly recrystallised region of the annealing curve. This will, of course, be determined by practical processing conditions.

Industrial practice can be made more flexible by providing a wider recrystallisation annealing ‘window’. It is clear from Fig. 7 that this can be achieved by annealing at lower temperatures and/or applying lower cold reductions (\( CR < 50% \)). This practice will effectively lengthen the recrystallisation ‘plateau’ and provide a high operating flexibility. However, because of this limitation on the amount of cold reduction, and the present hot band thickness limitation (1.9 mm minimum), the LC recrystallised grades are currently produced only in gauges of 1.14 mm or thicker. This, unfortunately, represents a limited market potential for recrystallised steel sheets.

Although it is out of the scope of this paper, one possible way to circumvent this problem could be to use a higher level of microalloying elements in the steel which may allow a longer recrystallisation window during annealing after heavier cold rolling reductions. It is suggested from the
modelling work presented above that an increase of the \( r \) exponent in eq. (11) will increase the value of \( t_{50} \). Previous works\(^{26}\) demonstrated that the presence of microalloying elements such as Nb or Ti will increase the value of \( r \) by a factor of ten.

4. Conclusions

A new isothermal recrystallisation model is proposed to predict the evolution of the recrystallised volume fraction during annealing of cold rolled ferritic steels, taking into account the deformation degree. The model corrects some of the fundamental shortcomings of the classical Avrami modelling, such as non-random (clustered) nucleation and decreasing grain growth velocity. The new model assumes a single class of nucleation, of \( \gamma \)-fiber grains nucleating at grain boundaries, which was experimentally confirmed by X-ray macrotexture measurements. The variation of the cold rolling degree is reflected in the model through quantitative differences of the applied values, e.g. regarding the decrease of the driving force for nucleation with annealing time. It is observed that the model presents good fitting of the experimental values, especially at high deformations and low and high holding stages.

Finally, the model is applied to analyze the feasibility of back annealing in LC sheet steels, concluding that the processing parameters normally used in production lines will provide a higher degree of operational flexibility leading to back annealing to be successful processing technique for LC sheet steels.

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