In-Situ Studies of the Ferrite Nucleation and Growth in C-Mn Steels

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Abstract

In-Situ Neutron Depolarization (ND) measurements were performed during continuous cooling at moderate cooling rates for several C-Mn steels. During cooling, the phase transformations from austenite to ferrite and pearlite were monitored by the simultaneous determination of the ferrite fraction and the particle size by means of ND. The onset of the pearlite formation is very clearly distinguished. In isothermal experiments, a change in temperature does not lead to renewed nucleation. In order to obtain a more detailed insight in the transformation mechanism, a three-dimensional model of the austenitic microstructure decomposing into ferrite was developed and compared with experimental results. This comparison yields an estimate of the nucleation behaviour in the first stages of the ferrite formation.

THE FINAL MICROSTRUCTURE OF construction and engineering steels as produced in a hot-rolling mill is controlled by the phase transformation of austenite (γ) into ferrite (α) and pearlite. Quantitative information on the kinetics of this transformation is therefore of importance for gaining control of the eventual microstructure of the steel.

Neutron Depolarization (ND) has been used to study the decomposition of austenite that is supersaturated in carbon, into ferrite and pearlite during isothermal and isochronal heat treatments (see also [1, 2, 3]). In an ND experiment, the change in the polarization vector of the polarized neutron beam after transmission through a sample is determined. The mean magnetization of the sample results in a rotation of the polarization vector, and fluctuations in the magnetization give rise to a shortening of this vector (depolarization) [4]. The rotation can therefore be related to the fraction magnetic material in the sample, and the depolarization to the average magnetic domain size [5, 6].

Since austenite is paramagnetic and ferrite is, below \( T_C \approx 1043 \text{ K} \), ferromagnetic, the magnetic fraction and the domain size equal the ferrite fraction and the ferrite particle size, respectively. The simultaneous and in-situ determination of these two parameters with ND gives insight in the nucleation and growth kinetics that govern the phase transformation.

Neutron Depolarization

In a Neutron Depolarization experiment the polarization vector \( \vec{P} \) of a polarized neutron beam is analyzed after transmission through a magnetic sample. The change in \( \vec{P} \) after passage through magnetic material can be written in matrix form [4, 5]:

\[
\vec{P}^f = \vec{D} \vec{P}^0,
\]

where \( \vec{P}^0 \) and \( \vec{P}^f \) are the initial and final polarization vectors, respectively, and \( \vec{D} \) is the \( (3 \times 3) \) depolarization matrix. The elements of \( \vec{D} \) are calculated from the measured intensities \( I_{ij} \), where \( i, j = \{x, y, z\} \) represent the directions in which the beam is analyzed and polarized, respectively, by

\[
D_{ij} = 1 - I_{ij} / I_0.
\]

\( I_0 \) is the intensity of the fully depolarized beam. The local magnetic induction in a domain at site \( \vec{r} = (x, y, z) \) in a sample can be written as the sum of the mean magnetic induction \( \langle \vec{B} \rangle \) and the local deviation from the mean induction, \( \Delta \vec{B}(\vec{r}) \). The mean magnetic induction results in a rotation of the polarization vector over an angle \( \varphi \), and can be represented by a rotation matrix \( R(L) \) with \( L \)
the transmission length of the sample. The relationship between $\langle B \rangle = \langle \bar{B} \rangle$ and $\varphi$ is given by [6]

$$\varphi = \langle B \rangle > L \sqrt{c} = m f B_s(T) L \sqrt{\xi},$$  \hspace{1cm} (3)

with the parameter $c = 2.18 \times 10^{20} \lambda^3 \text{T}^{-2} \text{m}^{-2}$, with $\lambda$ the wavelength of the neutrons, $f$ the volume fraction of magnetic material, $m$ the reduced magnetization, and $B_s(T)$ the temperature-dependent spontaneous magnetic induction.

The local fluctuations result in a depolarization (length reduction of the polarization vector) that is proportional to the correlation length of $|\Delta \bar{B}(\theta)|^2$ along the neutron propagation direction (the $z$-direction). In a thin layer of thickness $L_W$ and volume $W$ at position $x_0$, the depolarization can be represented by the matrix $\hat{D}$, with elements [6]:

$$D_{ij} = \delta_{ij} (1 - c L_W \xi) + c L_W \alpha_{ij},$$  \hspace{1cm} (4)

with

$$\alpha_{ij} = \frac{1}{W} \int_{-W}^{W} \int_{x_0}^{x_0} \int_{-W}^{W} \int_{x_0}^{x_0} dx' \Delta B_i(x, y, z) \Delta B_j(x', y, z),$$  \hspace{1cm} (5)

$$\xi = \sum_i \alpha_{ii}.\hspace{1cm} (6)$$

Here, $\delta_{ij}$ is the Kronecker delta, $\Delta B_i(x, y, z)$ is the $i$-component of $\Delta \bar{B}(x, y, z)$. $\alpha$ is the correlation matrix, and $\xi$ is the correlation function. If $\langle B \rangle >$ is directed along the $x$, $y$, or $z$ axis, then $\alpha_{ij} = 0$ for $i \neq j$.

If the sample is described by $N = L/L_W$ identical layers perpendicular to the neutron propagation direction, the total depolarization matrix $\hat{D}$ is given by

$$\hat{D} = \left[ \hat{R}_z \left( \frac{L_W}{2} \right) \right] \hat{D} \left[ \hat{R}_z \left( \frac{L_W}{2} \right) \right]^{\dagger},$$  \hspace{1cm} (7)

In order to determine $\varphi$ and $\alpha$ from the measured depolarization matrix $\hat{D}$, corrections have to be made to account for depolarization caused by the instrument. Sources of depolarization are the polarizer, analyzer, and the wavelength distribution of the neutron beam. The measured matrix $\hat{D}$ (Eq. (2)) is fitted to Eq. (7), with fit parameters $\langle B_x \rangle$, $\langle B_y \rangle$, $\langle B_z \rangle$, $\alpha_{xx}$, $\alpha_{yy}$, and $\alpha_{zz}$, after taking the necessary corrections into account.

If it is assumed that each ferrite particle contains only one magnetic domain, the correlation function $\xi$ can be related to the average ferrite particle size $\delta$. If an external field is applied to the sample in the $y$-direction, and it is assumed that the particles are homogeneously distributed spheres, without overlap between the demagnetization fields of the particles, this relationship is [6]:

$$\delta = \frac{3 c_2 \xi}{2 f B_s^2 (1 - c_3 c_5 m^2)}, \hspace{1cm} (8)$$

$$c_2 = \left( \frac{4 \pi f^2}{81} \right)^{1/3}, \quad c_5 = \frac{32}{9 (3 - < n_2^2 >)}. \hspace{1cm} (9)$$

$< n_2^2 >$ is the average of the square of the local magnetic induction direction cosine and can be derived from:

$$\gamma = \frac{\alpha_{yy}}{\xi} = \frac{\left( \frac{2.25+1.25 < n_2^2 >^2}{\frac{2.25+1.25}{1-2 < n_2^2 >}} - c_3 \right) m^2}{1 - c_3 m^2}.$$  \hspace{1cm} (10)

$\gamma = \alpha_{ii}/\xi$ are the mean direction cosines and are a measure for the magnetic texture in direction $i$.

**Experimental Set-up**

The experiments were performed at the Interfaculty Reactor Institute. A schematic view of the instrument is given in Fig. 1. A thermal neutron beam from a nuclear reactor is monochromatized and polarized before entering the sample chamber. Before and after the sample chamber rotators are installed that rotate the polarization vector into one of three perpendicular directions. After the second rotator, the beam passes through the analyzer and enters the BF$_3$-detector. The detector intensities are determined for the three polarization and three analyzer directions composing a $3 \times 3$ depolarization matrix that describes the changes of the polarization vector. In the experiments presented here, the intensity for each measured matrix element was determined with a statistical error less than 0.5%. The samples are ring shaped, with a thickness of 0.3 mm. A toroidal coil is wound around the sample in order to apply a magnetic field of 780 A/m. The furnace in which the sample is placed is operated at a pressure of $10^{-3}$Pa.

![Figure 1: Schematic drawing of the experimental set-up.](image)

**Simulation Model**

In order to attain a better interpretation of the experimental results, the transformation is simulated with a nucleation and growth model in three-dimensional space. The austenite grains are represented by Voronoi cells constructed from randomly placed points. The cell corners are assigned as possible nucleation sites for ferrite. A sigmoidal function is used to describe the number of ferrite...
nuclei as a function of time. The simulation starts from a completely austenitic structure, and time is increased in discrete steps. At each time step a random selection is made from the possible nucleation sites in order to determine at which site ferrite is to nucleate. The growth of each ferrite particle is modeled by applying diffusional growth theory for finite media [7] to spherical particles. This means that the growth rate of the particles depends parabolically on time, on temperature through the activation energy for carbon diffusion in austenite, and in a rather complex manner [7] on the carbon concentration in the remaining austenite. It is assumed that each ferrite particle contains only one magnetic domain and that the direction of the magnetization is determined by the sum of the applied field and the magnetic dipole interactions of neighboring particles [8]. The Neutron Depolarization properties for the simulated structure is calculated using equations derived from Eq. (4-6) [6, 9].

Results: Isochronal experiments

Isochronal cooling experiments were performed on commercial C60, C45, and C35 steel samples (for compositions, see Table 1). The samples were initially heated and annealed above the $A_s$ temperature, i.e. in the one-phase γ-region, at 1073, 1073, and 1148 K, respectively. ND experiments were conducted during cooling at a rate of 9.8 K/hour.

<table>
<thead>
<tr>
<th>Material</th>
<th>Element (in wt.%)</th>
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<tbody>
<tr>
<td></td>
<td>C</td>
</tr>
<tr>
<td>C60</td>
<td>0.68</td>
</tr>
<tr>
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<td>0.47</td>
</tr>
<tr>
<td>C35</td>
<td>0.36</td>
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Table 1: Compositions of the steel samples.

In the above equations for the ferrite fraction $f$ (Eq. 3) and mean ferrite particle radius $\delta$ (Eq. (8-10)) all parameters are either known from literature or measured (c.q. fitted), except the reduced magnetization $m$, which is a crucial parameter in all equations. The reduced magnetization $m$ was determined from the measured rotation after completion of the transformation to pearlite, by assuming that at this point the ferrite fraction was equal to that determined with quantitative image analysis (QIA) of the samples performed after the ND experiments. This value for $m$ was applied to the whole measurement. From the final ferrite fractions of 90%, 91%, and 93% for C60, C45, and C35, resp., values of 0.66, 0.75, and 0.51 were determined for $m$.

Figures 2 and 3 show the ferrite fraction $f$ calculated from the measured rotation of the polarization vector. The difference in the carbon concentration of the samples can be seen in the temperature at which the transformations start. For the higher carbon steels (C60 and C45) the transformation to ferrite starts below the Curie temperature at 1015 and 1030 K, respectively. In the case of C35, the transformation to ferrite has started above $T_C$, resulting in a rapid increase in the rotation of the polarization vector and in $f$ just below $T_C=1043$ K. The transformation to pearlite starts at approximately the same temperature (968 K) for C45 and C35, while for C60 the transformations to pearlite starts at 980 K.

Furthermore, at the onset of the transformation to pearlite there is a large difference in the ferrite fraction, 2.5%, 14%, and 22% for C60, C45, and C35, respectively. For all samples this pro-eutectoid ferrite fraction, $f_{pro}$, measured with ND, is less than the equilibrium fraction calculated with the thermodynamical programs MTDATA, although the cooling rate was low (see Table 2). However, in the case of C45, $f_{pro}$ is consistent with
the fraction determined with QIA. In the case of C35, \( f_{pro} \) determined with ND is much lower than that determined with QIA. The final ferrite fractions determined with QIA, assuming a fixed ferrite fraction in pearlite, were in agreement with values calculated with MTDATA.

<table>
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<th>ND</th>
<th>QIA</th>
<th>MTDATA</th>
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<tr>
<td>C60</td>
<td>2.5</td>
<td>-</td>
<td>7</td>
</tr>
<tr>
<td>( \delta (\mu m) )</td>
<td>5</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>C45</td>
<td>14</td>
<td>16</td>
<td>31</td>
</tr>
<tr>
<td>( \delta (\mu m) )</td>
<td>10</td>
<td>6</td>
<td>-</td>
</tr>
<tr>
<td>C35</td>
<td>22</td>
<td>46</td>
<td>47</td>
</tr>
<tr>
<td>( \delta (\mu m) )</td>
<td>13</td>
<td>13</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 2: The pro-eutectoid ferrite fractions \( f_{pro} \) and the pro-eutectoid particle size \( \delta \) measured with neutron depolarization (ND), quantitative image analysis (QIA), and calculated with MTDATA.

The mean ferrite particle radius \( \delta \) determined with ND is given in Fig. 4. For all samples, \( \delta \) increases until the onset of the pearlite transformation, where a very sharp peak occurs. After completion of the pearlite transformation, \( \delta \) drops to 4 \( \mu m \) for C60 and 5 \( \mu m \) for C45 and C35. The origin of the peak at the start of the pearlite transformation is not yet understood, but it is not believed to be an artefact, since it has been observed for all steel samples that we have measured [2, 3]. The size of the ferrite particles before the pearlite transformation increases with decreasing carbon content. However, the growth rate at the beginning of the transformation for each composition seems similar. Since \( \delta \) is still small at \( T_C \) for C35, it can be assumed that the transformation in this steel started at approximately \( T_C \) and not much above it.

QIA showed that for C45 and C35 the mean particle radii of the pro-eutectoid ferrite were 6 and 13 \( \mu m \), respectively. The value for C45 is less than the value determined with ND. Systematic errors in the determination of \( f \) and \( \delta \) could be caused by two effects. First, when the rotation of the polarization vector in a single particle becomes much greater than \( \pi/2 \), the assumptions made in determining the mean particle radius from the correlation function are no longer valid, and therefore producing incorrect values of \( \delta \). Another uncertainty in the present data interpretation stems from the value of \( m \). The assumption of \( m \) being constant over the entire transformation range possibly introduces a gradually changing error, specifically in \( f \).

Results: Isothermal experiment

After reheating and annealing the C60 sample at 1073 K, a two-step isothermal experiment was performed. The temperature-time sequence imposed on the sample is given in Fig. 5, along with the ferrite fraction and particle size resulting from the ND experiments. Initially, \( f \) increases rapidly when cooled below \( A_3=1013 \) K (i = 48 min.). At this stage of the transformation, the rotation is significant while the depolarization remains small. This effect can only be caused by a homogeneous magnetic layer. Such a layer is likely to be formed on the surface due to surface-enhanced nucleation. Metallographic analysis of the sample after the experiments indeed shows a surface layer mainly consisting of ferrite.

![Figure 5: The two-step isothermal experiment on C60 steel. The ferrite fraction \( f \) and average particle radius \( \delta \) measured (symbols) and simulated (lines), and the sample temperature as a function of time.](image)

During the period that the temperature is held constant in the two-phase \( \gamma + a \) region at 1003 K, \( df/dt \) decreases, yet \( f \) does not reach its equilibrium value of approximately 4%, calculated with MTDATA. \( \delta \) increases approximately proportionally to \( \sqrt{t} \), consistent with a
diffusion controlled growth [7]. When the temperature is reduced to 998 K, both $f$ and $\delta$ increase further with initially increased rates, yet $df/dt$ is much smaller than at the beginning of the transformation. Again, the MT-DATA equilibrium ferrite fraction of approximately 7% is not reached.

Fig. 6 shows that there is a linear relationship between the average particle volume $\frac{4}{3}\pi f^3$ and $f$ over a large range of the experiment. No change in slope occurs at the transition from 1003 to 998 K. This leads to the conclusion that the ferrite particle density remains unchanged, and the increase in $f$ at the temperature change can be concluded to be due to growth only.

![Figure 6: The two-step isothermal experiment on C60 steel. The average ferrite particle volume $\frac{4}{3}\pi f^3$ as a function of $f$, measured (symbols) and simulated (lines).](image)

Simulated values for $f$ and $\delta$ resulting from the three-dimensional model are also given in Fig. 5. In addition to bulk nucleation and growth, relatively rapidly nucleating (in 7 minutes) and growing (1.8 % in 50 min.) surface layers of ferrite are introduced, in agreement with the experimental observations. Bulk nucleation in the simulations takes place during the first 50 minutes of the transformation only, during which a total fraction $f = 1.8\%$ is reached, which is mainly due to the growth of the surface layers.

The agreement between model and experiment shows that the initial sigmoidal nucleation function and the absence of renewed nucleation after the temperature change correctly describe the nucleation characteristics. The assumption of diffusion-controlled growth appears to be valid for these steels and cooling conditions.

**Conclusions**

The present Neutron Depolarization study of the transformation of austenite into ferrite and pearlite shows the potential of this technique. For several steels, simultaneous in-situ determinations of the ferrite fraction and the ferrite particle size have been performed. Three-dimensional simulations based on the experimental results from neutron depolarization give a detailed insight into the transformation kinetics. Distinction between nucleation and growth becomes possible and a spatial view of the transforming sample is obtained. Nucleation is found to occur only at the beginning of the transformation, not at a further decrease in temperature after the formation of a few percent ferrite. The growth of ferrite particles is essentially diffusion controlled.

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**References**


