NUCLEATION AND GROWTH OF FERRITE AND PERLITE
IN A MEDIUM CARBON STEEL
STUDIED USING NEUTRON DEPOLARIZATION

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ABSTRACT
Neutron Depolarization (ND) experiments on ferrite phase transformations in 57Cr3 steel (DIN 1.7176, UNS H51550) during continuous cooling and heating are presented. In the experiments the mean magnetization and the magnetic correlation length are determined simultaneously as functions of time and temperature. From these parameters, both the ferrite fraction and the average ferrite particle size, during the course of the transformation, are calculated. In this way, the contributions of both nucleation and growth processes to the total transformation are determined. The results show that the previous thermal history of the sample primarily influences the nucleation process. This applies to both the pro-eutectoid ferrite and perlitic formation. The results are an indication of the potential of the ND technique.

INTRODUCTION
The ferrite phase transformation plays an important role in the evolution of the microstructure of steels, and is therefore an important step in the steel production process. The kinetics of the transformation are usually studied by techniques such as dilatometry, differential scanning calorimetry (DSC), optical or (in situ) electron microscopy. These methods, however, require the interruption of the transformation process, or give only limited information on the transformation. In contrast, with neutron depolarization, it is possible to measure, simultaneously and in-situ, the ferrite fraction and the average ferrite particle size, over a relatively large amount of material. These parameters are derived from the rotation and the shortening of the polarization vector after transmission through a specimen. Simultaneous determination of the ferrite fraction and the particle size allows the determination of the ratio between nucleation and growth, which are important parameters in studying transformation kinetics.

A major limitation of the use of neutron depolarization in steel transformation studies, is that the transformation product has to be ferromagnetic. As the Curie temperature Tc of ferrite is 1045 K, only transformations that take place below this temperature can be studied. However, this imposes no restrictions when studying steels with carbon concentrations between 0.45% and 0.77%. Alloys of lower carbon mass percent can also be studied, provided the sample can be cooled to below 1045 K before the onset of the transformation (isothermal experiment).
This work deals with the pro-eutectoid ferrite and perlite formation in a 57Cr3 steel during slow cooling and heating experiments and presents a further evaluation of the method introduced by Krielaart et al. (1994).

**NEUTRON DEPOLARIZATION THEORY**

Neutron Depolarization makes use of the fact that a neutron beam can be polarized. The polarization direction is the direction in which the average spin component of the neutrons have an extreme which is not equal to zero. The polarization vector of the neutron beam will rotate around a magnetic field that is passed during transmission. This is described by the Larmor equation:

\[
\frac{d\hat{P}}{dt} = \gamma [\hat{P}(t) \times \hat{B}(t)]
\]  

(1)

Here, \( \hat{P} \) is the polarization vector, \( \gamma \) is the gyromagnetic ratio \( (1.83 \times 10^8 \text{ s}^{-1} \text{T}^{-1}) \) and \( \hat{B} \) is the local magnetic induction. The change in the polarization direction is determined by the transmission time of the neutron beam in a magnetic domain and therefore, depends on the velocity of the neutrons and the size of the magnetic domain. The solution of equation (1) for the full beam cross-section can be written in matrix form (Halpern and Holstein, 1940; Rekveldt, 1973):

\[
\hat{P}' = \hat{D} \hat{P}_0
\]  

(2)

Where \( \hat{P}_0 \) is the initial polarization vector, \( \hat{P}' \) is the polarization vector after transmission, and \( \hat{D} \) is the \((3 \times 3)\) depolarization matrix. The local magnetic induction in a domain can be described by the sum of the mean magnetic induction \( \langle \hat{B} \rangle \) and a local fluctuation of the magnetic induction \( \Delta \hat{B}(\vec{r}) \):

\[
\hat{B}(\vec{r}) = \langle \hat{B} \rangle + \Delta \hat{B}(\vec{r})
\]  

(3)

The polarization vector is influenced by each of these two components in a different way. The mean magnetic induction will result in a rotation of the polarization vector by an angle \( \varphi \). The relationship between the mean magnetic induction and \( \varphi \) can be written as (Rosman and Rekveldt, 1991):

\[
\varphi = \langle B \rangle L \sqrt{c} = mb_4(T) L \sqrt{c}
\]  

(4)

Here \( c = \gamma^2 / v^2 = 2.18 \times 10^{26} \lambda^2 \), where \( v \) is the velocity and \( \lambda \) is the wavelength of the neutrons. \( L \) is the total transmission length of the sample. The mean magnetic induction is the product of the reduced magnetic induction \( \omega \) the volume fraction of ferrimagnetic material, thus the fraction of ferrite \( f \), and the temperature dependent spontaneous magnetic induction \( B_4(T) \).

The local fluctuations \( \Delta \hat{B}(\vec{r}) \) will result in the shortening of the polarization vector by an amount proportional to the correlation length of \( \langle \Delta B \rangle^2 \) along the neutron path. This is called depolarization. If it is assumed that each ferrite particle consists of only one magnetic domain, this correlation length is the average ferrite particle size. The rotation and depolarization are determined (partially using fit procedures) from the elements of the depolarization matrix \( \hat{D} \), which are calculated from the measured intensities \( I_y \), where \( i,j = x,y \), and \( x \) represent the direction in which the beam is analyzed and polarized, respectively:

\[
D_y = \frac{1 - I_y}{I_x} \frac{I_y}{I_x}
\]  

(5)
\( I_s \) is the shim intensity which is the average of the spin up and spin down intensities measured in any direction. The quantity \( Q_0 \) is the polarizing power of the polarizer/analyser combination and is approximately 0.95 for the set up used.

If an external field is applied to the sample in the \( y \)-direction, and it is assumed that the particles are identical spheres, homogeneously distributed, and there is no overlap between the demagnetization fields of the particles, the relationships between the depolarization matrix elements and the fraction \( f \) and the average radius of the ferrite particles \( \delta \) are (Rosman and Rekveldt, 1991; Rekveldt, 1976):

\[
f = \frac{\varphi}{m_B L \sqrt{c}} = \arctan \left( \frac{D_{zz} - D_{xy}}{D_{xy} + D_{zz}} \right)
\]

(6)

\[
\delta = -\frac{\ln[\det(\hat{D})]}{\mu_B^2 B^2 (1 - g m^2)}
\]

(7)

\[
\gamma_0^0 - g m^2 = \frac{\ln[\det(\hat{D})]}{2 \ln D_{yy}}
\]

\[
\gamma_0^0 = \frac{\ln[\det(\hat{D})]}{2 \ln D_{yy}}
\]

(8)

with \( \langle \gamma_0^0 \rangle \) is the average of the square of the local magnetic induction direction.

As equations (6) through (8) show, \( f \) and \( \delta \) also depend on \( m \) and \( \langle \gamma_0^0 \rangle \). This means that the fraction and average particle radius can only be derived directly from the measurements if one of these last mentioned quantities is known. This is the case when the applied field is strong enough to saturate the magnetization in the sample, then \( m=1 \).

**EXPERIMENTAL SET UP**

The experiments were performed at the Interfaculty Reactor Institute (IRI) at the Delft University of Technology. The experimental set-up used is referred to as the KP (crystal polarimeter). From the nuclear reactor a white neutron beam is obtained and monochromized to a wavelength of 0.16 nm by Bragg reflection from a pyrolytic graphite crystal. The neutron beam is polarized by a multichannel neutron polarizer (MCNP) composed of stacked glass mirror substrates coated with CoFeVTiZr multilayers. The neutron beam is analyzed after passage through the sample chamber, by a MCNP with Co/Ti multilayers as the coating. At the end of the set-up the neutrons are counted by a BF\(_3\) detector. Between the polarizers and the sample chamber polarization rotators are placed so that the incoming and emerging beam can be polarized and analyzed in all three directions in order to measure the nine elements of the depolarization matrix \( \hat{D} \), successively (figure 1).

![Figure 1: Schematic layout of the neutron depolarization set-up.](image-url)

**FIGURE 1: SCHEMATIC LAYOUT OF THE NEUTRON DEPOLARIZATION SET-UP.** P: polarizer, R: rotator, S: sampleholder (BN), A: analyzer, D: detector, D: diaphragm
The sample used is a ring shaped sample with an inner and outer diameter of 20 and 26 mm, respectively. The thickness of the sample is 0.305 mm. The sample is placed in a circular sample holder of boron nitride (BN), which contains a diaphragm of 2 × 10 mm. Around the sample a toroidal coil of molybdenum wire, with 550 turns/m, is wrapped, in order to induce an homogeneous external field. The sample holder is placed in a stainless steel cylinder which has a thermocouple heating element wrapped around it. The temperature of the sample is monitored using an Alumel-Chromel thermocouple which is situated at the center of the sample holder. Each matrix element intensity was measured in 10 seconds with a statistical error of 0.5%.

Experiments were carried out on a 57Cr3 steel (DIN 1.7176, UNS H51550) (in wt.%: 0.56% C, 0.82% Cr, 0.92% Mn, 0.24% Si, 0.07% Ni, 0.02% Mo, 0.17% Cu, 0.013% P, 0.009% S, 0.05% V, 0.035% Al, 0.01% Co, 0.01% Ta). The sample was mechanically polished as to be plan parallel within 3 µm. The oven was evacuated with a turbo molecular vacuum pump to 10⁻⁴ Pa, during the experiments. Experiments were done with a constant external fields of 1705 A/m.

RESULTS
The transformation behavior of the 57Cr3 steel sample was studied during isochronal heating and cooling experiments and during an isothermal transformation.

Isochronal Experiments
The isochronal transformation experiments consisted of a set of 4 consecutive transformation experiments. The sample was initially heated for 3 hours at the austenitization temperature of 1123 K. In run 1 the sample was cooled from 1123 K to 828 K at a cooling rate of 21 K/h. The sample was then reheated to 1123 K also using a heating rate of 21 K/h. In run 2 the same cooling and heating cycle was followed but at 23 K/h. After run 2 the sample was heated for 24 hours at 1123 K to erase the effects of the previous thermal cycles as much as possible. In run 3 the same cooling and heating cycle was imposed as in run 2. Finally, in run 4 the sample was just cooled to 828 K with a rate of 23 K/h and then cooled more rapidly to room temperature.

The rotation of the polarization vector as a function of the temperature is shown in figure 2 for these four experiments. The figure shows that the rotation of the polarization vector has a characteristic temperature dependence. The polarization vector starts to rotate at a temperature of about 1018 K for all four experiments on cooling. Upon further cooling the rotation only increases moderately, until a second transition temperature is reached at which the rotation of the polarization vector increases far more rapidly with decreasing temperature. At a temperature of about 964 K the rotation seems to have reached a saturation value, and increases only little upon further cooling. Upon heating, the angle of rotation decreases slowly with temperature until a temperature of 1010 K is reached. Above this temperature, which does not vary much between experiments, the angle of rotation decreases rapidly. Once an angle of rotation of approximately 1.5 rad is obtained the polarization angle decreases less rapidly with further increase of the temperature. At a temperature of 1044 K the angle of rotation has become zero.

The change of the rotation angle with temperature during cooling and heating can be explained qualitatively rather easily. The initial rotation of the polarization vector during cooling is attributable to the pro-eutectoid ferrite formation, while the rapid increase in rotation in the temperature interval of approximately 984 to 964 K is due to the perlitic formation. A similar explanation can be given for the behavior during heating, indicating that perlitic dissolution proceeds that of the pro-eutectoid ferrite. The transition temperatures agree well with the reported values for the A₃ and the A₅ temperatures (1013 K and 1032 K respectively).

To determine the ferrite fraction (either present as pro-eutectoid ferrite or as ferrite in perlitic) more quantitatively the temperature dependence of B₅ should be taken into account. It is assumed that the same temperature dependence applies as in pure iron (Stiesser et al., 1985). The reduced magnetic induction m is taken equal to 0.7 which yields the expected ferrite fraction of 0.92 at temperatures below the A₅ temperature.

30
The ferrite fractions calculated from experiments are shown in figure 3a. The pro-eutectoid ferrite fraction is also shown more clearly in figure 3b. The figures show quite clearly that the amount of pro-eutectoid ferrite formed, \( f_{\text{p}} \), increases with the order of the experiment (see also table 1). However, the onset temperature for the pro-eutectoid formation hardly changes, and remains at approximately 1018 K. These results suggest that the ferrite nucleation is not strongly affected by the prior thermal history.

The balance between nucleation and growth can be determined more quantitatively by examining the average particle radius \( \delta \) as a function of temperature and order of the experiment. As indicated in equation 7 the particle radius can be derived from the shortening of the polarization vector.

Figure 4 shows the average particle radius for all four runs during cooling. Due to numerical reasons the particle radius at temperatures close to the onset temperature of pro-eutectoid formation was difficult to determine. However, the average particle radius can be determined with sufficient accuracy at lower temperatures. The figure shows that in each run the pro-eutectoid particle size increases upon cooling to the most 3 \( \mu m \) before the onset of the perlite transformation at the temperature \( T_p \). At the onset of the perlite transformation a large increase in the average particle radius can be seen in each run. This rapid increase is probably due to the fact that the microstructure assumed in the derivation of the ND equations is an incorrect representation of the actual microstructure.

A summary of the characteristics of the pro-eutectoid ferrite particles determined at 990 K, which is above the onset temperature of the perlite transformation for all four runs, is given in table 1. The ratio between nucleation and growth in the different runs can be determined from \( N \), which is the number of particles per \( m^3 \). This data shows that the increase in the pro-eutectoid fraction with the order of the experiment is largely due to an increase in the number of particles for the first three runs. However, in the last run the increased fraction is the result of larger particles. As a result of the austenitization between runs 1 and 2, it can be expected that the sample will homogenize and that, as a result, the number of pro-eutectoid ferrite nuclei will decrease. Therefore it was expected that the transformation of run 3 would be similar to that of run 1. This is, however, not the case for the pro-eutectoid ferrite formation. In run 3 there is an increase in the number of particles, with respect to the previous run.

The results of figure 4 suggest that the increase in the pro-eutectoid ferrite fraction, during one run, is for the most part governed by grain growth and not by additional nucleation. However, the increase of the
fraction between consecutive transformation cycles is mostly the result of an increase in the number of particles (table 1).
Similarly, the effect of multiple transformations on the perlite formation during cooling can be examined. Figure 3 shows that the onset temperature for perlite formation increases with the order of the experiment from 973 K to 984 K. Clearly, the nucleation of perlite is facilitated. Again the effect of the intermediate annealing at 1123 K between run 2 and run 3 on the perlite nucleation is negligible, as the transformation behavior during run 3 resembles that of run 2 closer than that of run 1. Most of the transformation to perlite takes place within 10 to 15 K for all the runs. Below 960 K the ferrite fraction only increases very slowly by about 0.05.

**Figure 3A: Ferrite Fraction During Four Successive Transformation Cycles**

**Figure 3B: Pro-eutectoid Ferrite Fraction Determined Upon Cooling**
TABLE 1: CHARACTERISTICS OF THE PRO-EUTECTOID FERRITE PARTICLES

<table>
<thead>
<tr>
<th>Run</th>
<th>$T_p$ (K)</th>
<th>$f_{pro}$</th>
<th>$f_{pro}$</th>
<th>$\delta$ (µm)</th>
<th>$N\left(10^{15} \text{m}^{-3}\right)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>973</td>
<td>0.038</td>
<td>0.026</td>
<td>1.6</td>
<td>1.5</td>
</tr>
<tr>
<td>2</td>
<td>980</td>
<td>0.085</td>
<td>0.059</td>
<td>1.4</td>
<td>5.1</td>
</tr>
<tr>
<td>3</td>
<td>978</td>
<td>0.117</td>
<td>0.101</td>
<td>1.6</td>
<td>5.9</td>
</tr>
<tr>
<td>4</td>
<td>984</td>
<td>0.138</td>
<td>0.123</td>
<td>2.5</td>
<td>1.9</td>
</tr>
</tbody>
</table>

Additional information on the perlite formation can again be obtained from the average ferrite particle radius. It should be realized that the equations relating the shortening of the polarization vector to the ferrite particle radius were derived assuming a spherical morphology for the ferrite particles. This is clearly not the case when the ferrite is present in the form of perlite. However, the particle size derived in this fashion is a measure for the average dimensions of ferrite lamella in perlite. As the perlite morphology is unlikely to change with the order of the experiment, the relative change in the ferrite particle size in the four transformation runs is considered to be correct. It can also be expected that the lamellar spacing in the perlite is the same in all the runs because the temperature-time profile was the same. This means that differences in the determined average ferrite particle radius in perlite are related to differences in the dimensions of the perlite colonies. Microscopic examination of the sample indicate that the average lamella spacing is 0.3 µm.

Figure 4 shows that below 960 K, where the transformation to perlite seems to be completed, $\delta$ changes very little, as is to be expected. There is, however, a large difference in the average particle size between the different runs. The ferrite particle characteristics at a temperature of 900 K, where the transformation is fully completed, are listed in table 2.

![Figure 4: Average ferrite particle radius during four successive transformation runs](image-url)

<table>
<thead>
<tr>
<th>Run</th>
<th>$f_{\text{max}}$</th>
<th>$\delta$ (µm)</th>
<th>$N$ (10$^{15}$ m$^{-2}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.93</td>
<td>4.4</td>
<td>2.5</td>
</tr>
<tr>
<td>2</td>
<td>0.92</td>
<td>2.9</td>
<td>8.5</td>
</tr>
<tr>
<td>3</td>
<td>0.92</td>
<td>4.1</td>
<td>3.0</td>
</tr>
<tr>
<td>4</td>
<td>0.92</td>
<td>3.5</td>
<td>5.0</td>
</tr>
</tbody>
</table>

The data indicate that the perlite particle size decreases after each transformation cycle. In contrast, the intermediate annealing between run 2 and run 3 had an opposite effect on the ferrite particle size, as approximately the same particle radius in run 1 and run 3 was observed. This is most likely the result of the dissolution of perlite nuclei and a coarsening of the austenite grains during the annealing, which results in larger perlite colonies.

**Isothermal Experiment**

The ferrite and perlite formation was also studied during (quasi-) isothermal transformation. To this aim a sample was preannealed at 1123 K for 25 minutes and cooled to 973 K at a rate of 200 K/h. The sample was then held at 973 K for 260 minutes. The temperature undershoot was less than 2 K. Finally, the sample was cooled to 640 K.

The rotation of the polarization vector during this experiment is shown in figure 5. The change of the temperature with time is also plotted, to facilitate its interpretation. The figure shows a small discontinuity in the time dependence of the rotation of the polarization vector after 39 minutes. This marks the transition from pro-eutectoid ferrite formation to perlite formation. The perlite formation continues until the end of the isothermal transformation. The increase in rotation of the polarization vector after further cooling (t > 300s) is only due to the temperature dependence of $B_0$ (see equation 6).

![Figure 5: Rotation of the polarization vector during the temperature-time profile imposed](image)

**FIGURE 5: ROTATION OF THE POLARIZATION VECTOR DURING THE TEMPERATURE-TIME PROFILE IMPOSED**
Figure 6 presents both the ferrite fraction and the average ferrite particle radius as a function of time and shows that the ferrite fraction after the perlite transformation remains constant. As in the previous results, the reduced magnetization was taken equal to 0.7, which resulted in a maximum ferrite fraction of 0.96. The slight increase in the determined fraction between $t = 305$ and 340 minutes is purely the result of the use of an incorrect value of $B_n$, which is the result of a possible discrepancy in the actual temperature of the sample and the temperature measured, during the rapid change in temperature that was initiated at $t = 305$ min.

The average ferrite particle radius before the onset of perlite formation is approximately 2 $\mu$m, but difficult to determine due to the rapid increase in particle size at the onset of perlite formation. This increase has the same origin as the rapid increase in the particle size measured in the isochronal transformation measurements just below $T_p$. However, the transition from pro-eutectoid ferrite formation to perlite formation is clearly distinguishable. Surprisingly, the average ferrite particle size decreases continuously during the perlite formation. This suggests, within the parameters of the present model, that the transformation proceeds by continuous nucleation rather than growth of a limited number of perlite nuclei.

FINAL REMARKS
While the analysis of the neutron depolarization during isochronal and isothermal transformations in a medium carbon steel like 57Cr3 clearly needs further theoretical work to take all microstructural effects related to the ferrite and the perlite formation into account properly, it has been shown that the ND technique offers many advantages in studying phase transformation kinetics and in resolving the transitions between the various types of transformations.

Additional metallographic and local microstructural analysis is in progress to validate the results obtained and to guide future refinements in the theoretical analysis.

REFERENCES