Neutron depolarisation: a novel technique for studying phase transformation kinetics in steels

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Neutron depolarisation experiments on ferrite phase transformations in an unalloyed steel during continuous cooling and heating are presented. In the experiments the mean magnetisation and the magnetic correlation length are determined simultaneously as functions of time and temperature. From these parameters the ferrite fraction and the mean ferrite particle size, respectively, are determined. The potential of the technique is illustrated on a C60 steel.   IS/1188

INTRODUCTION

The austenite–ferrite phase transformation plays an important role in the evolution of the microstructure of steels and, therefore, is 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), and optical or *in situ* electron microscopy. From dilatometry and DSC, information about the total fraction transformed only can be acquired. From optical and electron microscopy, information about the particle size and morphology can be obtained. These latter methods, however, require the interruption of the transformation process, or give information only about a small amount of the material. In contrast, with neutron depolarisation, it is possible to measure, simultaneously and on line, the fraction transformed and the mean ferrite particle size, and to obtain information on the average morphology of the particles. These parameters are derived from the rotation and the shortening of the polarisation vector after transmission through a specimen.

A major limitation of the use of neutron depolarisation in steel transformation studies is that the transformation product must be ferromagnetic. As the Curie temperature T_C of ferrite is 1045 K, only transformations that take place below this temperature can be studied. In the case of an experiment with slow cooling, the weight per cent of carbon in the steel should be between 0·45 and 0·77% to study the transformation from its beginning. Alloys of lower carbon content can also be studied, provided the sample can be cooled to below 1045 K before the onset of the transformation. This work deals with the ferrite and pearlite formation in a C60 steel during slow cooling and presents a further evaluation of the method introduced in Ref. 1.

THEORY

Neutron depolarisation makes use of the fact that neutrons possess a spin, represented by the spin vector $\vec{S}$. A neutron

beam is polarised if the average spin component in a certain direction is not equal to zero. The direction in which the average has an extreme is the polarisation direction. The polarisation vector of the neutron beam will rotate around any magnetic field which is passed during transmission. This is described by the Larmor equation

$$\frac{\mathrm{d}\vec{P}}{\mathrm{d}t} = \gamma[\vec{P}(t) \times \vec{B}(t)] \qquad \qquad (1)$$

where $\vec{P}$ is the polarisation vector, γ is the gyromagnetic ratio ($= 1·83 \times 10^8 \mathrm{~s}^{-1}\mathrm{T}^{-1}$), and $\vec{B}$ is the local magnetic induction. The change in polarisation direction is determined by the time that the neutron beam is 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[2,3]

$$\vec{P}' = \hat{D}\vec{P}^\circ \qquad \qquad (2)$$

where $\vec{P}^\circ$ is the initial polarisation vector, $\vec{P}'$ is the polarisation vector after transmission, and $\hat{D}$ is the 3×3 depolarisation matrix.

The local magnetic induction in the sample can be described by the sum of the mean magnetic induction $\langle\vec{B}\rangle$ and the local fluctuation of the magnetic induction $\Delta\vec{B}(\vec{r})$

$$\vec{B}(\vec{r}) = \langle\vec{B}\rangle + \Delta\vec{B}(\vec{r}) \qquad \qquad (3)$$

The polarisation vector is influenced by each of these two components in a different way. The mean magnetic induction will result in a rotation of the polarisation vector by an angle ϕ. The relationship between the mean magnetic induction and ϕ can be written as[4]

$$\phi = \langle B\rangle f L \sqrt{c}$$
$$= mB_s f L\sqrt{c} \qquad \qquad (4)$$

where $c = \gamma^2/v^2 = 5·6 \times 10^9 \mathrm{~m}^{-2}\mathrm{T}^{-2}$ for wavelength $\lambda = 0·16$ nm (with v the velocity of the neutrons), L is the total transmission length of the sample, and f is the fraction of ferromagnetic material (hence the fraction of ferrite). For a ferromagnet, the mean magnetic induction is the product of the reduced magnetic induction m and the temperature dependent spontaneous magnetic induction B_s. In the case of Fe, the temperature dependence of the spontaneous magnetic induction is[5]

$$B_s(T) = 3·45\left(\frac{T_C - T}{T_C}\right)^{0·373}\left[1 - 0·5\left(\frac{T_C - T}{T_C}\right)^{0·55}\right] \qquad (5)$$

The local fluctuations $\Delta\vec{B}$ will result in the shortening of the polarisation vector by an amount proportional to the correlation length of $(\Delta B)^2$ along the neutron path. This is termed depolarisation. This correlation length is the mean ferrite particle size. The rotation and depolarisation are determined (partially using fit procedures) from the elements of the depolarisation matrix $\hat{D}$, which are calculated from the measured intensities I_{ij}, where $i, j = x, y,$ and z represent the directions in which the beam is,

respectively, analysed and polarised

$$D_{ij} = \frac{1 - I_{ij}/I_s}{Q_0} \qquad \ldots \ldots \ldots \ldots \ldots \quad (6)$$

The parameter I_s is the shim intensity, which is the average of the spin up and spin down intensities measured in any direction: $I_s = \frac{1}{2}(I_+ + I_-)$. The quantity Q_0 is the polarising power of the polariser–analyser combination and is approximately 0·95 for the setup used.

If an external field is applied to the sample in the y direction and if it is assumed that the particles are identical spheres, homogeneously distributed, and that there is no overlap between the demagnetisation fields of the particles, the relationships between the depolarisation matrix elements and the fraction f and mean radius of the ferrite particles δ are[4,6]

$$f = \frac{\phi}{mB_s L\sqrt{c}}$$

$$= \frac{\arctan[(D_{xz} - D_{zx})/(D_{xx} + D_{zz})]}{mB_s L\sqrt{c}} \qquad . \quad . \quad (7)$$

$$\delta = \frac{-\ln[\det(\hat{D})]}{f L B_s^2 c(1 - gm^2)} \qquad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

$$\frac{\langle n_y^2 \rangle - gm^2}{1 - gm^2} = \frac{\{\ln[\det(\hat{D})]\} - 2\ln D_{yy}}{\ln[\det(\hat{D})]} \qquad . \quad . \quad . \quad . \quad (9)$$

$$g = 1\cdot 91 \frac{f^{2/3}}{3 - \langle n_y^2 \rangle} \qquad . \quad . \quad . \quad . \quad . \quad . \quad (10)$$

Here, $\langle n_y^2 \rangle$ is the average of the square of the local magnetic induction direction and is a measure for the mean shape factor of the magnetic particles.

As equations (7)–(10) show, f and δ also depend on m and $\langle n_y^2 \rangle$. This means that the fraction and mean particle size 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 magnetisation in the sample; then $m = 1$. Equations (7)–(9) can also be solved by comparing two measurements made with different external fields and, thus, having different values for m and $\langle n_y^2 \rangle$, but the same values for f and δ.

EXPERIMENTAL SETUP

The experiments were performed at the Interfaculty Reactor Institute (IRI) at the Delft University of Technology. The experimental setup used is referred to as the crystal polarimeter. From the nuclear reactor a white neutron beam is obtained and monochromatised to a wavelength of 0·16 nm by Bragg reflection from a pyrolytic graphite crystal. The neutron beam is polarised by a multichannel neutron polariser (MCNP) composed of stacked glass mirror substrates coated with CoFeV/TiZr multilayers. The neutron beam is analysed after passage through the sample chamber, by a MCNP with Co/Ti multilayers as the coating. At the end of the setup the neutrons are counted by a BF₃ detector. Between the polarisers and the sample chamber polarisation, rotators are placed so that the incoming and emerging beam can be polarised and analysed in the x, y, or z direction to measure the nine elements of the depolarisation matrix $\hat{D}$ successively (Fig. 1).

The sample used is ring shaped with inner and outer diameters of 20 and 26 mm, respectively. The thickness of the sample is approximately 0·3 mm. The sample is placed in a circular sample holder of boron nitride, which contains a diaphragm of 2×10 mm. Around the sample a toroidal coil of molybdenum wire, with 550 turns m⁻¹, is wrapped, in order to induce an homogeneous external field. The sample holder is placed in a stainless steel cylinder which

P	polariser
R	polarisation rotator
S	sample holder (BN)
A	analyser
De	BF₃ detector
D	diaphragm
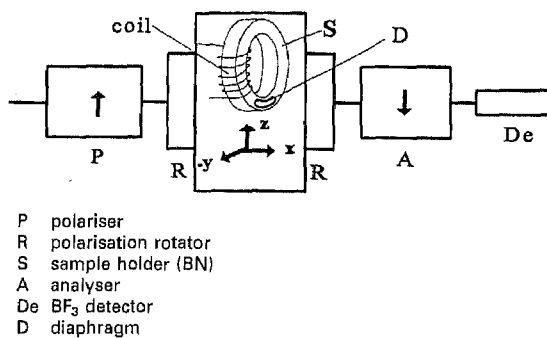
1 Schematic drawing of experimental setup

has a thermocoax heating element wrapped around it. The temperature of the sample is monitored using an alumel–chromel thermocouple which is situated at the centre of the sample holder. Each matrix element intensity was measured in 15 s with a statistical error of 0·5%.

RESULTS

Experiments were carried out on an unalloyed steel containing 0·6 wt-%C (C60, DIN 1.0601), with a thickness of 0·285 mm. The sample was mechanically polished so as to be plan parallel within 3 μm. The oven was sealed off during the experiments. Experiments were done with constant external fields between 275 and 1650 A m⁻¹. Temperature changes were applied linearly with rates between 5 and 30 K h⁻¹. Cooling and heating experiments were done consecutively, by cooling from temperatures of 1075 to 850 K or lower and then increasing the temperatures back to 1075 K.

Figure 2a shows the rotation angle ϕ measured in two successive heating and cooling cycles, with applied fields of 275 and 550 A m⁻¹ in the y direction. Both cooling and heating rates were 22 K h⁻¹. On cooling below T_C (1045 K), there is a rapid increase in the rotation by at least 1·4 rad within a few degrees. The rapid increase indicates that there is already ferrite present. This means that it is very likely that the actual carbon concentration in the sample has become less than 0·6 wt-%*. The rotation angle then gradually increases by approximately 2·5 rad over 70°. This increase is mainly the result of the increasing ferrite fraction and the increasing $B_s(T)$. Below 972 K, within 15°, there is a large increase in ϕ. This is the result of the transformation of enriched austenite to pearlite. The further increase in ϕ is mainly the result of the increasing $B_s(T)$. On heating, the dissolution of the pearlite occurs at 1010 K, indicated by the large decrease in the rotation. The rotation then further decreases slowly until T_C is exceeded, at which ϕ drops to zero. The austenite–pearlite transformation shows the usual hysteresis. Also, the rate of formation is lower than the rate of dissolution.

From the depolarisation (Fig. 2b) the transition from enriched austenite to pearlite and vice versa can also clearly be seen by the relatively large change in the depolarisation when cooling below 960 K and heating above 1005 K. In the ferritic–pearlitic state, the depolarisation is smaller. This means that the particles are smaller than in the austenitic–ferritic state. The constant increase at lower temperatures is again the result of the increasing $B_s(T)$.

From a comparison of the results of the measurements made with different applied fields, it is apparent that the saturation magnetisation was not yet reached. This is clear from the fact that there is a difference in the rotation angle

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* The decarbonisation of the sample was due to malfunction of the vacuum installation during the measurements. Therefore, decarbonisation can be prevented if vacuum conditions are imposed during the measurements.
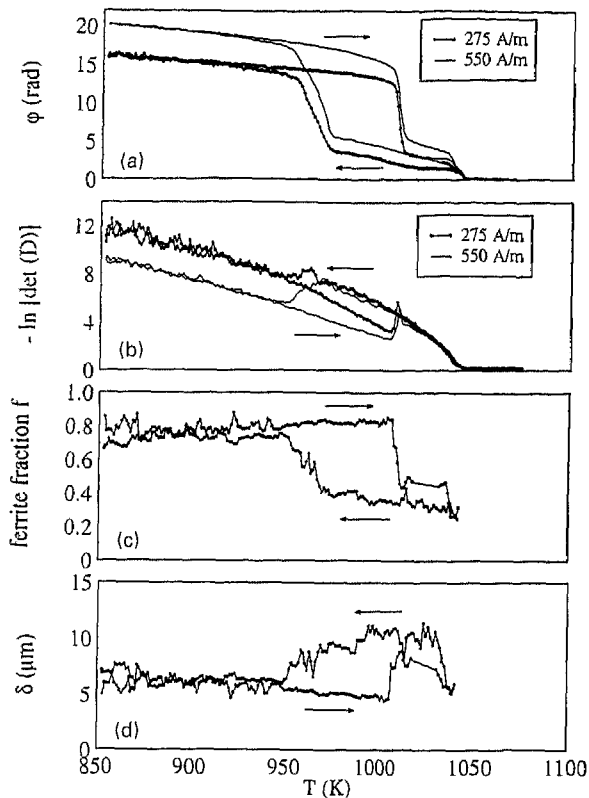
2 *a* Rotation angle, *b* depolarisation, *c* ferrite fraction, and *d* mean radius of ferrite particles as function of temperature for both cooling and heating experiments; arrows indicate direction of temperature change

over the whole temperature range. Because $m \neq 1$ the measurements were analysed together to estimate the fraction and the mean particle size, using the equations discussed. The analysis is based on the assumption that the transformation behaviour is fully reproducible, i.e. the ferrite fraction is only a function of the temperature and the direction of the temperature change. Figure 2c shows the ferrite fraction determined. The ferrite fraction in the austenitic–ferritic state is approximately 30% and increases to 75% in the ferritic–pearlitic state. The fraction obtained for the ferritic–pearlitic state is higher than would be expected for a steel with 0·6 wt-%C (~20% is expected). From this result it is concluded that decarbonisation had occurred in previous runs. In contrast, the fraction at low temperatures is lower than the expected value of 92%. This implies that the actual thickness of the sample has decreased by ~10%. A decrease in sample thickness was confirmed by analysis of the sample after the measurements.

Figure 2d shows the determined mean radius of the ferrite particles. In the cooling curve, a mean radius of 10 μm is observed above 970 K. Below 970 K δ decreases to 6 μm, yet increases with further decrease of the temperature. It is clear that in the ferrite–pearlite phase, δ is smaller than in the austenite–ferrite phase. This smaller value of the mean radius is due to the formation of lamellar ferrite as part of the pearlite structure. If the sample thickness is actually 90% of the original thickness (0·285 mm), these values for δ will increase by 10%.

DISCUSSION

It has been shown that neutron depolarisation is a powerful technique with which the change in magnetisation due to the formation or dissolution of ferrite can be followed closely without disrupting the transformation process. From the depolarisation matrix is was possible to determine the rotation angle ϕ and the depolarisation. Both parameters yielded information on transformations occurring. The smallest rotation angle that can be detected, within an error margin of 1%, is approximately 0·02 rad. Using equation (7) this means that, if $m = 1$, $T = 1040$ K, and $L = 0·3$ mm, the minimal detectable fraction is 0·002. This value depends on both the temperature and the sample thickness. By choosing larger sample thicknesses even smaller fractions can be detected. Exact determination of the mean particle size from the depolarisation becomes difficult when the rotation per particle is much greater than $\pi/2$ rad. This means that the maximum average magnetic transmission length per particle that can accurately be determined is approximately 20 μm, at 1000 K.

Because of the relatively small measuring times, it was possible to follow the rapid transformations. With a larger diaphragm of 100 mm² and the present neutron beam intensity on the CP setup, it is even possible to measure the whole depolarisation matrix within 2 s, with a statistical error of 1%, making the accurate measurement of even faster transformations possible.

In the analysis it was assumed that the temperature dependence of f was the same for both measurements. Because a decrease in the carbon concentration was also observed, it is possible that the carbon concentration was not identical while performing the two measurements. This would mean that f was not the same in both cases. This problem would not occur if a field was applied such that $m = 1$ throughout the transformation.

Whereas the full austenite–ferrite/ferrite–pearlite transformation could clearly be followed, the onset of the austenite/austenite–ferrite transformation was not observed. This is due to the fact that the transformation had started above the Curie temperature due to the decrease in the carbon concentration. Close to T_C the interpretation is also more difficult due to the rapid increase in B_s and the importance of the exact value of T_C. For the measurement of the full transformation behaviour in steels with 0·45 wt-%C (i.e. with an A_3 temperature above T_C) a higher cooling rate is required to achieve sufficient undercooling. If the transformation occurs reproducibly, the data of consecutive cycles can be superimposed and the transformation can be followed with a much higher time resolution but with the same accuracy.

In the future it is intended to perform this last mentioned type of measurement. Furthermore, additional refinement of the theory and the fit procedures will have to be made to make full use of the technique.

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