Use of Magnetic Techniques for Characterisation of the Microstructure Evolution during the Annealing of Low Carbon Steels

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Abstract. In this paper, how magnetic hysteresis loop and magnetic Barkhausen noise (MBN) measurements can provide non destructive information about the evolution of the microstructure during the annealing of cold rolled low carbon steel sheets is shown.

The effect of recovery and recrystallization processes on hysteresis loop and MBN signal measurements is analysed and related to the microstructural changes produced due to these softening processes, which have been previously characterised by optical and transmission electron microscopy.

The results obtained prove that the coercive field and the remanent induction from hysteresis loop measurements, as well as the total number and average amplitude of pulses of the MBN signal could successfully be employed to non-destructively monitor the kinetics of recovery and recrystallization processes in cold rolled low carbon steels. These parameters are sensitive to the reduction in the dislocation density taking place during recovery and to the combined effect of the decrease in the dislocation density and the average grain refinement during recrystallization.

Additionally, some correlations are considered between parameters derived from hysteresis loops, and from MBN measurements that are satisfied during recovery, and can also be used to distinguish recovery from recrystallization.

1. Introduction

Among the existing numerous non-destructive testing techniques, micromagnetic methods such as hysteresis (B-H) loop and magnetic Barkhausen emission measurements are known to be very effective for the inspection of ferritic steel structures. The microstructure and composition of steels define their mechanical and ferromagnetic properties. Consequently, the above mentioned techniques can be used for non-destructive evaluation of several material properties and for characterising numerous microstructural features of steels.

When a varying magnetic field is applied to a polycrystalline ferromagnetic material its magnetic response is strongly influenced by its microstructure. Magnetic Barkhausen noise (MBN) occurs due to sudden irreversible motions of magnetic domain walls as they overcome local pinning sites. It is well known that microstructural features, such as dislocation density and their arrangement, distribution and size of grains, grain boundaries, second phase precipitates and applied or residual stresses (either tensile or compressive) act as local pinning sites that strongly influence the magnetic domain wall movements and the overall magnetisation processes. These microstructural features affect both the pinning strength and the mean free path of the displacement of domain walls during magnetisation, in such a way that the magnetic hysteresis loop and MBN are sensitive to the microstructure.

Low carbon steel sheets for packaging or other applications are usually cold rolled and subsequently annealed, in most cases, to soften the material, enhance its formability and develop the appropriate texture for drawing. During the annealing treatment, the cold rolled steel substructure experiences recovery and recrystallization processes. Recovery involves both the annihilation of dislocations and their rearrangement into low energy configurations. Recrystallization leads to the suppression of dislocations by the nucleation of defect free volumes and the migration through the material of the recrystallization front, resulting in a new grain structure with a low dislocation density. The loss via both processes of the dislocations introduced by work hardening produces the softening of the steel [1].

As mentioned, both dislocations [2-5] and grain boundaries [6-10] act as effective barriers to domain wall motion, and influence the mean free path of the movement of the domain walls. Consequently, both magnetic hysteresis loop and MBN parameters are expected to be affected by both recovery and recrystallization due to the changes these processes produce on the microstructure.

Thus, the aim of the present contribution is to show how parameters derived from these techniques can provide information about the microstructural changes produced due to recovery and recrystallization during the annealing of cold rolled extra low carbon (ELC) steels. With that purpose, cold rolled samples of an ELC steel were isothermally annealed in the laboratory under different conditions in order to impart various degrees of recovery or recrystallization. In a previous paper by authors and co-workers [11] the evolution of the substructure during recovery was studied by transmission electron microscopy and electron backscattering diffraction observations, and the changes in the microstructure as a consequence of recrystallization were analyzed by optical microscopy. In the present paper the effect of these softening processes on hysteresis loop and Barkhausen noise parameters is analysed and related to the microstructure, previously characterised by conventional techniques.

2. Experimental procedure

2.1 Sample preparation

An industrially produced extra low carbon steel (ELC1), cold rolled to a final thickness of 0,3mm through a reduction of 84%, was used in this study. The composition of the steel was the following: 0.03C – 0.19Mn – 0.13Al – 0.0035N – 0.012P – 0.01Si (wt-%). In order to obtain samples with various degrees of recovery or recrystallization, a series of interrupted isothermal annealing cycles were performed on a number of cold rolled samples. Details on the annealing treatments carried out can be found in [11].

2.2 Experimental device for magnetic hysteresis loop and Barkhausen Noise measurements

The magnetic measurements were made at room temperature once the samples had been removed from the annealing device. The measuring system was designed and constructed in the authors' laboratory. This is basically composed of an excitation unit for magnetising the samples and an acquisition and processing unit, shown schematically in Figure 1. As shown, both types of tests share the excitation unit, the sensors and part of the signal conditioning circuit.

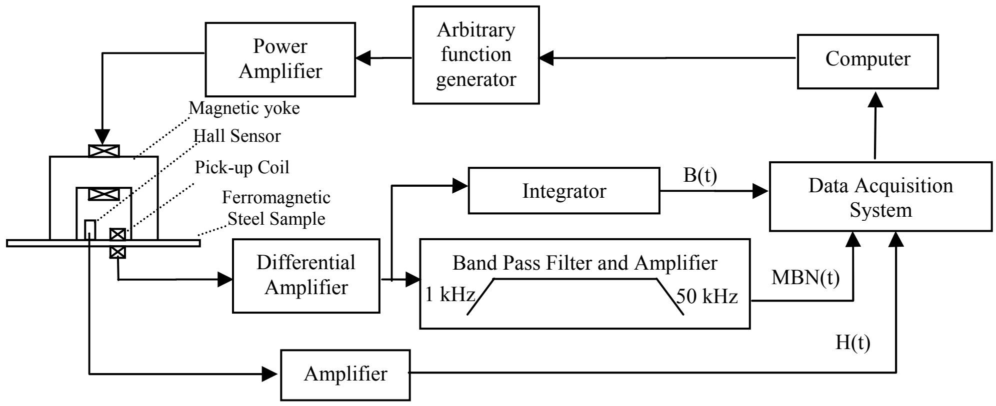


Figure 1 Schematic diagram of experimental setup for magnetic hysteresis loops and MBN measurements.

The excitation part consists of a programmable function generator, a power amplifier and a magnetic yoke consisting of a 200 turn coil wound around a U-shaped magnetic laminated core. For the magnetic measurements the steel sample under test is placed below the magnetic core so that the magnetic circuit is closed through the sample.

A small 50 turn encircling search coil wound around the samples is used to detect the induced voltage. A signal proportional to the magnetic induction signal, B(t), is obtained after integrating the induced voltage. The signal at the output of the differential pre-amplifier is further amplified and band pass filtered in the range of 1kHz to 50kHz to obtain a signal proportional to the MBN (the results will be shown in arbitrary units (a.u.)). Furthermore, the tangential magnetic field strength, $H_t(t)$, is measured using a small Hall probe placed at the surface of the steel samples. The signals picked up from both sensors are acquired by a data acquisition system, stored in a personal computer and post-processed using MATLAB software.

Sinusoidal magnetizing currents, producing maximum magnetic field strengths of about 4.1kA/m, sufficiently high to saturate the measured samples, were used at 1Hz for hysteresis loop and at 0.1Hz for MBN measurements, in order to cause a relatively slow domain wall movement during the magnetisation and to obtain sensitive parameters dependent on the changes in the microstructure. The digitising rate for hysteresis loop is 5kHz and 50kHz for MBN measurements, which limits the effective bandwidth of the MBN signal to 25kHz. Before each measurement, the samples are demagnetised at the test frequency, by applying a sinusoidal signal whose amplitude diminishes gradually, in several cycles, to values close to zero.

2.3 Magnetic parameters derived

Coercive field (H_c), remanent induction (B_r), relative differential permeability (μ), maximum relative differential permeability (μ_{max}), maximum induction (B_{max}) and hysteresis loss (W_h) values were derived as parameters from each hysteresis loop. The parameters obtained from the four hysteresis loops applied in each test were averaged and the measurements shown were calculated as the average of two complete tests.

An example of the MBN signal and the intensity of the tangential magnetic field, $H_t(t)$, acquired during two complete magnetising cycles applied to a low carbon steel sample is shown in Figure 2. In each measurement, only the signals corresponding to half of an excitation cycle, shown inside the dotted rectangle in Figure 2, are stored. The MBN parameters are derived from this part of the MBN signal, averaging the results obtained from five independent measurements. Part of the MBN signal with an expanded time scale is shown in Figure 3.

The digitised data set of the MBN signal is transformed by an algorithm analogous to a pulse height analyser, which is based on the one proposed in [12] and slightly modified. Further details on the algorithm used to detect the MBN pulses and for calculating their amplitude can be found in [13,14]. The total count of pulses and their average amplitude are considered as MBN parameters in this paper.

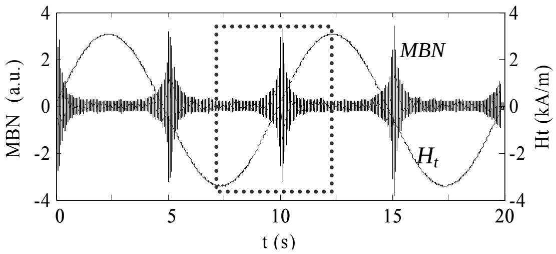


Figure 2 The MBN and H_t signals acquired during two complete hysteresis cycles applied to a low carbon steel sample.

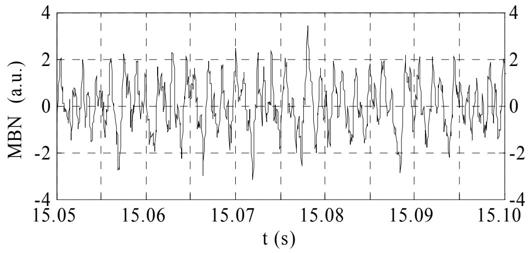


Figure 3 The MBN signal with an expanded time scale.

3. Experimental results and discussion

3.1 Microstructural evolution during the annealing treatment

A previous analysis on the microstructure and hardness evolution during the annealing of the cold rolled steel [11] revealed on the one hand that hardness measurements in this type of steels do not give any information about the recovery processes (300-500ºC, and up to 11s at 600ºC, see Figure 4-a), that they only experience an important drop at the onset of recrystallization (after 11s at 600ºC, see Figure 4-b) and that they can be used to monitor the progress of recrystallization (between 11s and 5400s at 600ºC, see Figures 4-b to 4-d). The full softening of the steel finishes after a soaking time of about 400s at this temperature, when the microstructure showed a fraction of more than 90% recrystallization.

The recrystallization produces a large decrease in the dislocation density via the nucleation of dislocation free grains within the deformed or recovered microstructure (Figures 4-b,4-c). Then, these grains grow consuming the old grains, resulting in a new grain structure with a low dislocation density (Figure 4-d). In summary, recrystallization removes dislocations, but at the same time creates a microstructure full of new grain boundaries. As it can be seen in Figures 4-b to 4-d the recrystallization process produces an effective grain refinement of the microstructure. The initial grain size of the cold rolled material is quite large, 60µm, compared to that of the nearly fully recrystallized one, 8 µm.

On the other hand, it was observed that during recovery the initial grain structure of the cold rolled steel remains constant and microstructural changes only occur in the cold rolling dislocation substructure inside the grains (see transmission electron microscopy images in [11]). The dislocations arrange into more perfect walls and the cell boundaries evolve into a well defined subgrain substructure.

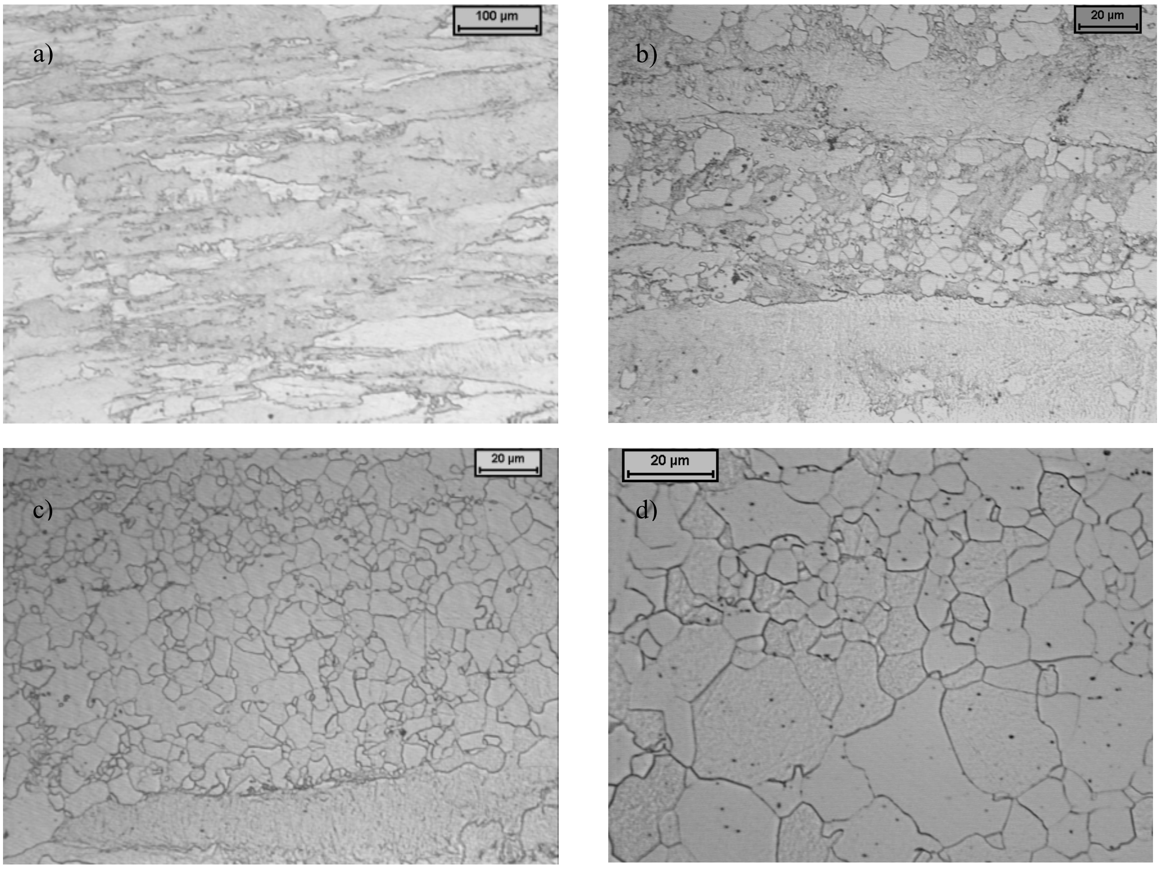
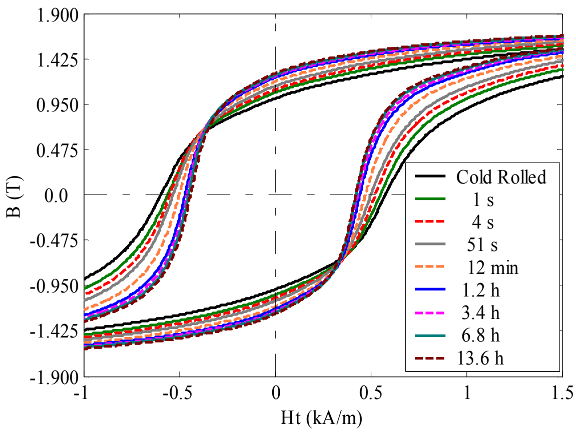


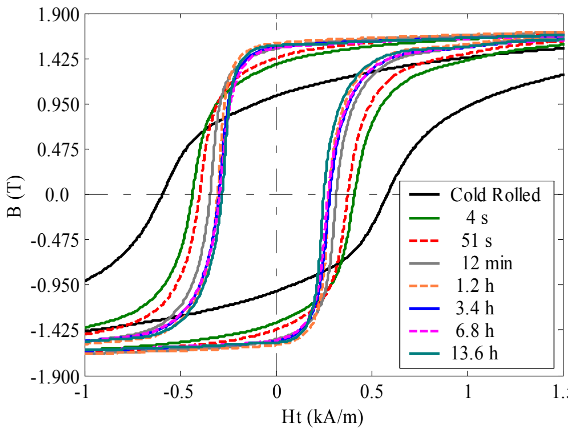
Figure 4 Optical micrographs showing the steel microstructure after different annealing treatments: a) 500 ºC-4,8h; b) 600 ºC-11s; c) 600 ºC-240s; d) 600 ºC-5400s.

3.2 Evolution of hysteresis loop parameters with annealing

The graphs in Figure 5 show the effect of increasing the annealing time on the hysteresis loops of the ELC1 steel. It is observed that the B-H curves become steeper with annealing, presenting lower coercive field and hysteresis losses, and higher remanent induction and permeability values.



a) 300ºC-ELC1



b) 450ºC – ELC1

Figure 5 Evolution of the cold rolled ELC1 steel samples isothermally annealed and quenched at various stages.

In Figure 6 the coercive field values are represented as a function of annealing time in logarithmic scale. It is observed that H_c progressively decreases with the soaking time at low temperatures due to the recovery processes (300-500ºC, and up to 11s at 600ºC). As theoretical considerations specify that H_c is proportional to the square root of the dislocation density [5], the evolution of H_c can directly be related to the evolution of the average dislocation density during the recovery at low temperature annealing. Then, in the 30-2460s range at 600ºC H_c stagnates when a decrease in H_c would be expected as a consequence of the decrease in the dislocation density caused by the movement of the recrystallization front through the material. However, it is also well known that H_c also depends on the inverse of the grain size [10]. So, these results show that the effective grain refinement occurred in this steel during recrystallization produces an increase in H_c, which compensates the effect of its decrease due to the reduction in the dislocation density.

Figure 7 shows the evolution of the remanent induction as a function of the annealing time. It is seen that B_r presents a progressive increasing tendency with the annealing time during recovery, in the whole 300-500ºC temperature range, and up to 11s at 600ºC. Then, it shows a slower slope increase up to 240s during recrystallization, up to a recrystallized fraction of 85%, to eventually decrease at the final stages of recrystallization.

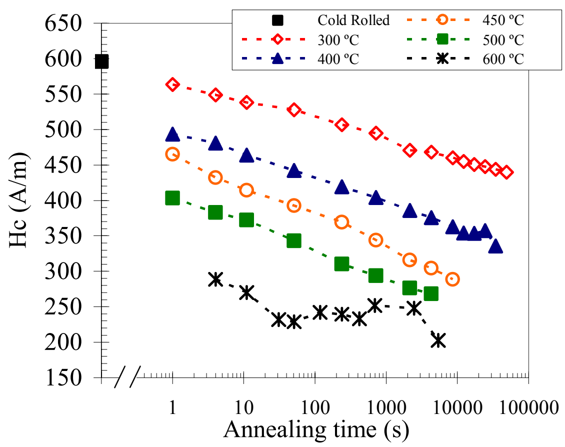


Figure 6 Evolution of the coercive field with the isothermal annealing time.

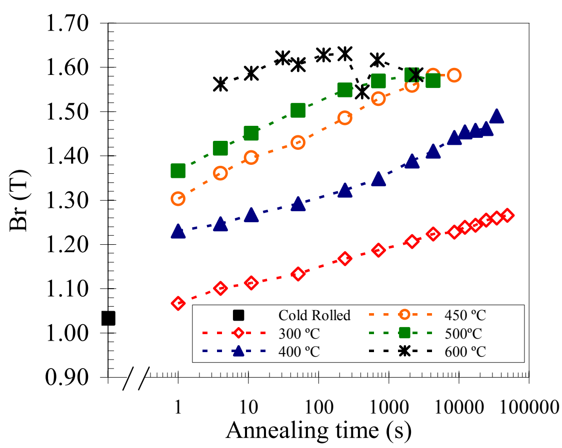


Figure 7 Evolution of the remanent induction with the annealing time.

These results show that both H_c and B_r parameters derived from hysteresis loops show a higher degree of resolution than hardness measurements to monitor the recovery processes, but that they can not be used to quantitatively follow the progress of recrystallization in this steel. The evolution of some other parameters derived from the hysteresis loops, the maximum differential relative permeability and the maximum induction have been reported in [15].

3.3 Evolution of MBN parameters with annealing

Figures 8 and 9 show the total number of pulses identified after post-processing the MBN signal and their average amplitude at each annealing stage as a function of the annealing time, respectively. It can be seen that at temperatures in the range from 300ºC to 500ºC, the total count of pulses decreases progressively, while their average amplitude increases gradually, both with a higher slope as the temperature increases, as a consequence of the recovery processes taking place at these temperatures. As mentioned above, the dislocations act as effective barriers to the domain wall movement. The number of pulses detected in the MBN signal depends on the number of unpinning events. Therefore, the reduction in the dislocation density produced during the recovery occurring in this range of temperatures can explain the reduction in the number of pulses detected. Furthermore, the dislocations also have an effect on the mean free path of the displacement of domain walls during magnetisation. The reduction in the dislocation density and the sharpening of the cell walls enhance the domain wall movement allowing the domain walls to move longer distances or give larger jumps, which is reflected as a gradual increase in the average amplitude of the pulses.

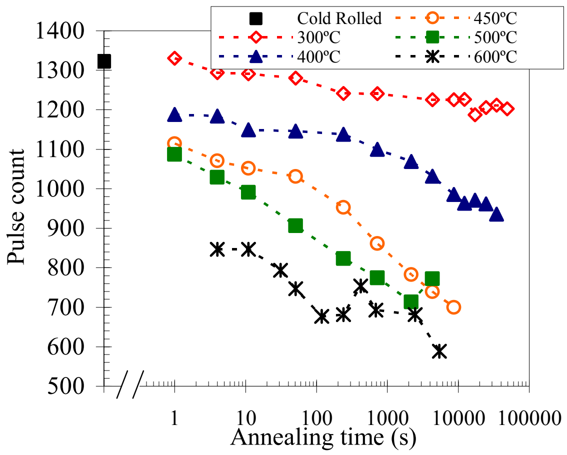


Figure 8 Total number of MBN pulses as a function of the annealing time.

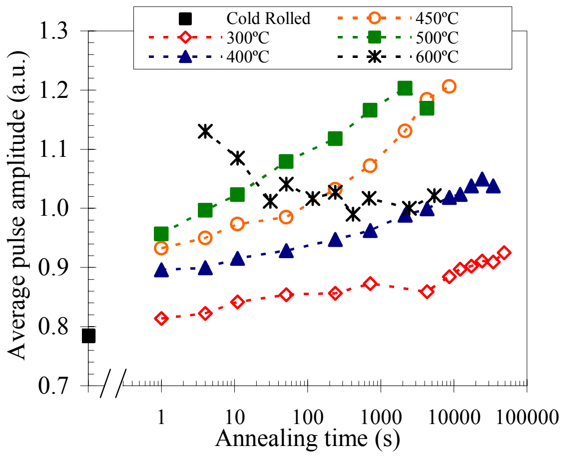


Figure 9 Average amplitude of MBN pulses as a function of the annealing time.

However, at 600ºC, on the one hand the total count of pulses shows an initial decrease, up to about 120s, afterwards the count almost stagnates at a constant value up to about 2500s, and finally decreases for an annealing time of 5400s. The stagnation in the number of pulses at intermediate recrystallization states can be explained by taking into account, as mentioned above, that the new grain boundaries generated during recrystallization also act as effective barriers to domain wall movement, compensating the reduction produced by the decrease in the dislocation density in the number of pinning sites and therefore in the unpinning events detected. The effect of increasing the total number of pulses with the decrease in the average grain size, is consistent with the results reported in [6] for nickel and in [8] for silicon steel. Moreover, Ranjan *et al*. [2] also observed a valley in the number of pulses at the recrystallization region during isochronal annealing treatments of nickel samples, due to the combined effect of change in grain size and dislocation density during recrystallization.

On the other hand, at 600ºC, the average pulse amplitude increases only during the first annealing stage compared to the initial deformed state, to decrease more or less gradually during the whole recrystallization process. This effect can be attributed to the fact that as soon as the recrystallization activates, the formation of new grains and the continuous grain refinement during recrystallization considerably reduce the mean free path of the displacement of domain walls during magnetisation, decreasing the mean amplitude of the pulses detected. The decreasing average amplitude of the pulses with the reduction in the average grain size agrees with the results reported in [7] for low-alloy low carbon steel and in [8] for silicon steel. Furthermore, the opposite evolution with annealing time of the average amplitude during recovery and recrystallization is an interesting feature because it allows the recovery and the recrystallization processes to be distinguished and the recrystallization process to be monitored.

3.4 Correlations between magnetic parameters and their relationship with microstructure

The correlation between H_c and B_r values measured on the analysed annealing states is represented in Figure 10. It has been found that the linear correlation between these parameters is only satisfied during recovery, while the changes in the microstructure only occur in the cold rolling dislocation substructure inside the grains and the varying dislocation density is the only microstructural feature affecting the magnetisation processes.

Figure 11 shows the correlation between the amplitude of the MBN pulses and H_c, which is also verified only during recovery and demonstrates that the parameters derived from the MBN can also be used to monitor the recovery kinetics.

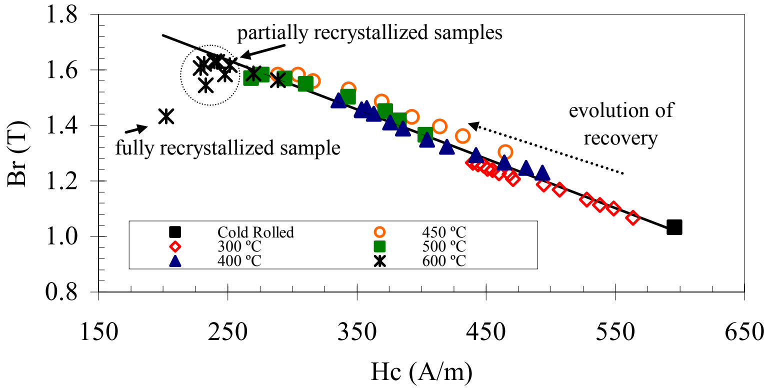


Figure 10 Correlation between remanent induction and coercive field values for the analysed temperatures during recovery and recrystallization.

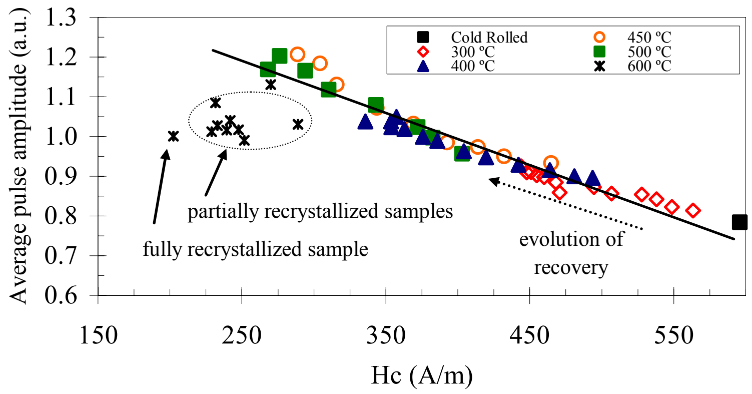


Figure 11 Correlation between the average amplitude of MBN pulses and the coercive field values during recovery and recrystallization.

The correlation between the number and the average amplitude of the pulses in the MBN profile is shown in Figure 12. Again, the linear correlation is only verified during recovery processes, where the number of pulses decrease and their average amplitude increases. However, as soon as recrystallization is activated the same relation is no longer satisfied, because of the additional effect of the varying grain size and the generation of new grain boundaries, which produces the decrease of the amplitude during recrystallization and the compensation effect on the total count of pulses.

Therefore, it can be concluded that all these correlations, both obtained between the same magnetic type of test (either hysteresis loop or MBN), and when combining parameters derived from different tests can be used to distinguish between recovery and recrystallization and to recognise the onset of the recrystallization process during the annealing.

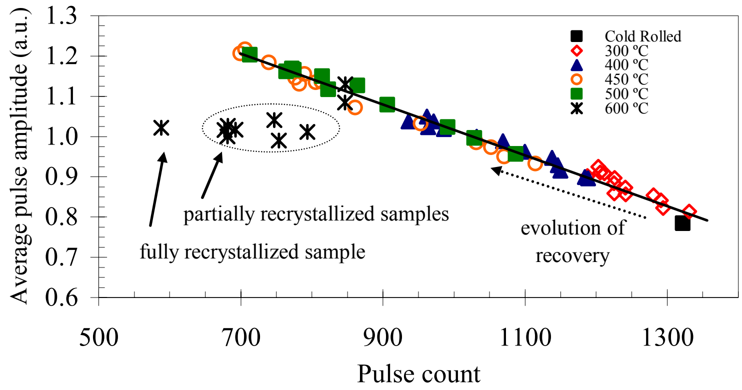


Figure 12 Correlation between the average amplitude and the total number of MBN pulses during recovery and recrystallization.

3.5 Relative significance of grain refinement during recrystallization

In Sections 3.2 and 3.3 that some parameters derived from both hysteresis loop and MBN are influenced by two competing effects during recrystallization has been analysed. That is, on the one hand, the decrease in the dislocation density due to the movement of the recrystallization front through the material causes the coercive field and the number of pulses to decrease; and on the other hand, the decrease in the average grain refinement taking place with the progress of recrystallization causes H_c and the number of pulses to increase. The opposite effect of these phenomena on the magnetic parameters indicated would not allow the use of them to monitor the progress of recrystallization.

In order to further research on this fact, magnetic measurements were taken of another ELC steel (ELC2: composition: 0,03C - 0,38Mn - 0,037P - 0,11Si (wt%), cold reduction 76%), with a initial cold rolling grain size of 12µm. Several samples were annealed at 575ºC in order to promote various degrees of recrystallization. Magnetic measurements were made and then the fraction of recrystallization was measured by conventional techniques [16]. The complete recrystallization grain size resulted to be of 6µm.

The correlations of the recrystallized fraction with H_c and with the total number of pulses obtained for ELC2 steel are shown in Figures 13 and 14, respectively. Figure 14 shows the comparison of the correlations between the number of pulses and the recrystallized fraction obtained in ELC1 and ELC2. It is observed how better correlations are obtained when the effective grain refinement due to recrystallization is not so significant in magnitude as in the case of ELC2 in comparison with ELC1.

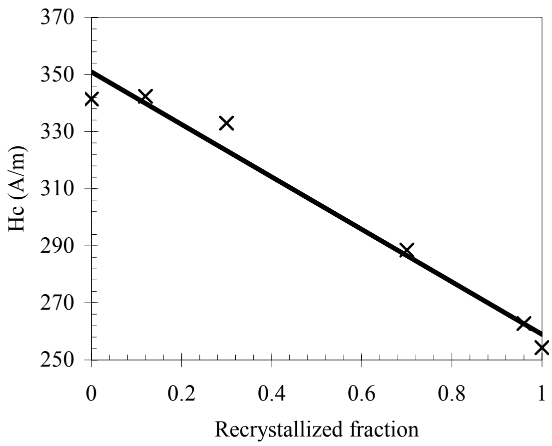


Figure 13 Correlation between the coercive field and the recrystallized fraction in ELC2.

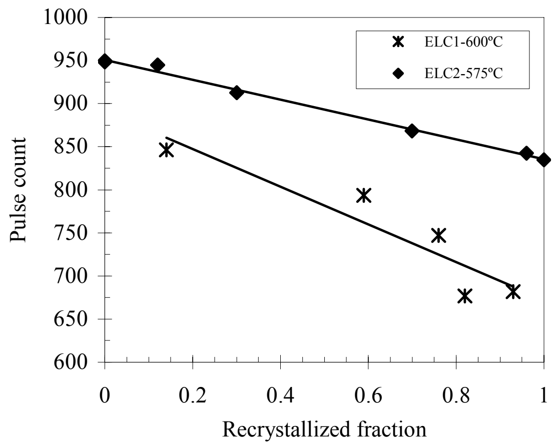


Figure 14 Correlation between the MBN number of pulses and the recrystallized fraction in ELC1 and ELC2.

Therefore, in those cases where the grain refinement caused by recrystallization is not so relevant as to compensate the opposite effect of the decrease in the dislocation density, these parameters could also be used to monitor the recrystallization in extra low carbon low alloy steels. The result has been also confirmed by authors in a Ti-Nb stabilised interstitial free (IF) ultra low carbon steel [17] with an initial cold rolled grain size of ~30µm and a final recrystallized grain size of 11µm. Additionally, in this type of steel, H_c and also other parameters derived from hysteresis loop measurements have also shown a higher degree of resolution to monitor recovery than conventional hardness measurements [17-18].

4. Conclusions

The coercive field and the remanent induction from hysteresis loop measurements as well as the total number and the average amplitude of MBN pulses have revealed to adequately monitor the reduction in the dislocation density occurring due to the recovery processes during the annealing of cold rolled low carbon steels.

These parameters can also be used to monitor the fraction of recrystallization when the effective grain refinement is not so relevant as to compensate the opposite effect of the decrease in the dislocation density occurring during recrystallization.

Linear correlations between the coercive field and the remanent induction, between the coercive field and the average pulse amplitude and between the total number of pulses and their average amplitude are satisfied only while the recovery processes take place in the substructure and the global grain size structure remains constant.

Even in the cases when the grain refinement effect takes place during recrystallization, this process can be distinguished by the recovery processes by the stagnation of the magnetic parameters during the annealing and by the loss of linearity of the correlations found between them.

Acknowledgments

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