Microstructure, Texture, Grain Boundary Characteristics and Mechanical Properties of a Cold Rolled and Annealed Martensitic Steel

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The microstructural and textural evolution and changes in Grain Boundary Character Distribution (GBCD) during annealing of a prior cold worked (30%, 50% and 80%) Fe–C martensitic steel have been studied, and correlated with mechanical properties. It has been demonstrated that ultrafine grains in the range 50–250 nm can be obtained by choosing appropriate amounts of cold rolling and annealing. Increasing the annealing temperature in all the three materials produces the expected results, namely decrement of strength with a concomitant increase in ductility. Although reasonably sharp γ-fibres were obtained in most of the cases, the very low r-bar values (<1.0) make the steels unsuitable for deep drawing purposes. It has been suggested that grain boundary engineering may lead to better strength–ductility combinations in this steel for enhanced range of applications.

KEY WORDS: texture; cold rolling; annealing; mechanical properties; Grain Boundary Character Distribution; martensite.

1. Introduction

In the recent years there has been an increasing interest to produce ultrafine grained (UFG) steels which are expected to possess high strength coupled with high toughness. The challenge of obtaining UFG steels with minimum alloy costs is being pursued with great endeavor in quite a few research laboratories. Currently, there are two main groups of laboratory methods for refining the grain structure down to the ultrafine grain regime in bulk steels. The first group comprises the various severe plastic deformation (SPD) processes, such as torsion straining under high pressure, equal channel angular pressing (ECAP), mechanical milling for metallic powder, accumulative roll bonding (ARB) etc. However, all these processes need special equipments and techniques, and they also require to apply high enough strain (above ~4) to materials in order to form the UFG structure. Most of these routes cannot be applied in continuous manufacturing processes used in industries. The second group of methods consists of advanced thermomechanical processing. These processes pursue alternative strategies to produce ultrafine grain microstructure. For instance, deformation induced grain subdivision is known to lead to the formation of ultrafine grains. In fact, in recent years, several studies have been reported on the grain refinement and strengthening aspects of cold rolled and annealed Fe–C based martensitic steels at normal temperature as well as at warm rolling conditions.

In the present investigation, an attempt has been made to produce ultrafine grained low carbon martensitic steels by conventional cold rolling and annealing route. It is generally known that the recrystallized grain size decreases with increasing cold reduction because of an increasing density of recrystallization nuclei with large rolling strains. However, in the present investigation the main aim is not only to produce ultrafine grained structure but also to study the microstructural, textural and grain boundary evolution in such steels and to evaluate the relevant mechanical properties with an eye on the possible applications of such materials.

2. Experimental Details

The experiments were conducted on a commercial low carbon steel in the form of hot rolled plate, with chemical composition as tabulated in Table 1. Pieces were cut from the initial plate to the dimensions of 5 mm×25 mm×200 mm (thickness×width×length) and austenitized at 950°C (above Ae3) for 30 min in a laboratory furnace followed by iced brine quenching to produce martensitic structure. Before rolling, the pieces were aged at 300°C for 30 min to reduce their brittleness. Three different cold rolling reductions were given, namely, 30%, 50% and 80%. The rolling was carried out using a two high laboratory

| Table 1. Chemical composition in wt%.
<table>
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<tr>
<th>C</th>
<th>Mn</th>
<th>Si</th>
<th>Cr</th>
<th>V</th>
<th>S</th>
<th>P</th>
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<td>1.6</td>
<td>0.52</td>
<td>0.083</td>
<td>0.13</td>
<td>0.014</td>
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rolling mill with a roll diameter of 250 mm. The rolling was performed at a roll peripheral speed of 27.5 m·min⁻¹ with machine oil as lubricant. The cold rolling could be successfully done without any edge cracking. Specimens of 15 mm in width and 100 mm in length were cut from the cold rolled sheets along the rolling direction (RD) for subsequent annealing and mechanical testing. The annealing of the cold rolled specimens was carried out at different temperatures ranging from 200–700°C, for 30 min with an interval of 100°C. From the annealed samples tensile specimens were prepared in accordance with ASTM E8M-04 standard and were subsequently polished. Tensile tests of polished samples were carried out at room temperature in Instron 4210 machine with a constant cross-head speed of 0.5 mm·min⁻¹. The bulk hardness values (Hv) were calculated from the average of seven separate measurements taken at randomly selected points by using a load of 5 kg for 15 s. A detailed microstructural characterization was performed using Field Emission Gun Scanning Electron Microscope (FEG-SEM) and Transmission Electron Microscope (TEM). Specimens after etching with 2% Nital, were observed along the cross section in a FEG-SEM operated at 15 kV. Thin foil TEM study was conducted in a PHILIPS CM200 TEM operated at 100 kV. Thin foils were prepared by twin jet electro polishing using a solution of 10% HClO₄+90% CH₃COOH. Global texture measurements were carried out on the half thicknesses of the 30%, 50% and 80% cold rolled as well as annealed (at 500°C and 700°C) samples using X-ray Diffraction (XRD) method in a PANalytical MRD machine. The (110), (200), (112) and (222) pole figures were determined, from which the Orientation Distribution Functions (ODFs) were calculated using the method of Bunge.¹¹)

The grain boundary characterization was carried out in FEG-SEM operated at 20 kV equipped with an OIM/EBSD attachment. Samples after polishing with colloidal silica, were put in the sample holder of the SEM/EBSD equipment. Using a step size of 1.0 μm a number of different areas were selected for scanning. The final scanned data was processed by the HKL software to calculate the Grain Boundary Character Distribution (GBCD) chart.

3. Results

3.1. Microstructural Changes

The FEG-SEM micrographs of the virgin martensite, and the 30%, 50% and 80% cold rolled steels are presented in Figs. 1(a)–1(d). A distinct fragmentation of the martensitic structure and its gradual alignment along the rolling direction, with increasing rolling deformation, can be seen in these micrographs. Obviously the martensite becomes finer as the amount of rolling reduction increases.

FEG-SEM studies of the microstructural features for the cold rolled steel samples show significant changes only after annealing at 500°C and above. Figures 2 and 3 depict the microstructures of the steel samples rolled at different levels and annealed at 500°C and 700°C respectively. It is clear from Fig. 2 that although the martensitic structure is very much retained in the 30% cold rolled material after annealing, the microstructures of the 50% and 80% cold rolled steel samples exhibit partial recrystallization along with some precipitation of carbide particles. In both Figs. 2(b) and 2(c) at many places large and clear ferrite grains with sharp boundaries have been observed, indicating partial recrystallisation of deformed martensite. Figure 3(a) shows a typical FEG-SEM microstructure of the 30% cold rolled steel annealed at 700°C. The blocky and fibrous nature of the martensite is clearly visible in this micrograph, although a few ferritic areas can also be seen. The FEG-SEM microstructures of the 50% and 80% cold rolled steel samples exhibit partial recrystallization along with some precipitation of carbide particles. Both these micrographs show three phases, ferrite, martensite and carbide. In the former the volume fraction of martensite is much larger and these appear as a string of islands along the ferrite grain boundaries. The interiors of the recrystallized ferrite grains are delineated with profusion of carbide particles. The recrystallized ferrite grains are much finer in the 80% cold rolled and 700°C annealed steel and
ferrite grain interior are decorated with carbide particles which are finer than in Fig. 3(b). The volume fraction of martensite at this stage is also drastically reduced, only a few small martensitic islands appear at ferrite grain boundaries.

In order to examine the microstructures in greater detail, transmission electron microscopy of thin foils from selected number of samples was undertaken. Although there are minor variations in the microstructure over extended areas in any one sample, the images reported here represent the predominant features only. **Figures 4(a)–4(c) show the TEM micrographs of 30%, 50% and 80% cold deformed steel. Fragmented martensite laths can be seen in each of these micrographs. The lath boundaries appear to become more prominent and the dislocation density within the laths increases with increase in the level of cold deformation. Simultaneously, there is a general decrease in the lath width and a definite alignment of the laths as the amount of cold rolling increases.**

The TEM microstructures of the 30% cold rolled steel (Figs. 5(a)–5(c)) do not show significant change even after annealing at 700°C. The general process which occurs in this steel on annealing is extensive recovery. In fact, highly recovered martensitic laths are obtained after annealing at 700°C. **Figures 6(a)–6(c) represent the TEM micrographs of the 50% cold rolled steel, annealed at different temperatures. Extensive amount of recovery appears to take place in this material right from the annealing temperature of 400°C [Fig. 6(a)]. Further recovery and subgrain growth takes place after annealing at 500°C [Fig. 6(b)]. Finally, annealing at the higher temperature of 700°C leads to the formation of large recrystallized grains with profusion of mostly globular carbide precipitation inside.**

**Figures 7(a)–7(c) represent the TEM micrographs of the 80% cold rolled steel annealed at 400°C and 500°C. Figure 7(a) shows extensive recovery of the initial martensitic structure and formation of subgrains. There are a few nano-sized regions which are practically free of any dislocation structure inside. These could be the nuclei of nano-sized grains. A higher magnification micrograph [Fig. 7(b)] shows that many of the martensitic laths are recovered to a very great extent. Annealing at 500°C leads to almost complete recrystallization and formation of a high density of ul-
3.2. Textural Changes

The textural data were collected from all the samples annealed at 500°C and 700°C. Extensive recovery and recrystallization of the martensitic structure were observed after annealing within this temperature range.

The $\phi_2=45^\circ$ sections of the ODFs of 30%, 50% and 80% cold rolled materials after annealing at 500°C and 700°C are presented in Figs. 8(a)–8(f). The 30% rolled and annealed material shows the formation of a $\gamma$-fibre after annealing at 500°C; however this fibre is totally absent after the material is annealed at 700°C [Figs. 8(a) and 8(d)]. Formation of a distinct $\gamma$-fibre can be seen in both the 50% and 80% cold rolled steels after annealing at 500°C and 700°C [Figs 8(b), 8(c), 8(e) and 8(f)]. A plot of the $\gamma$-fibres for the
different cold rolled and annealed steels is shown in Fig. 9. Excepting the 30% cold rolled and 700°C annealed material, all the others exhibit a reasonably uniform γ-fibre, the intensities of which lie within a band of 2.0 and 3.5 times random. The 80% cold rolled and 500°C annealed material shows the highest intensities of the γ-fibre. The calculated r-bar values for all the cold rolled and annealed samples have been found to be less than 1.0. The procedure for calculating the r-bar value from texture is given in reference. 12)

3.3. Changes in Grain Boundary Character Distribution (GBCD)

Figures 10–12 represent the total grain boundary characteristics of the steel rolled to different levels and annealed at 500°C and 700°C. Figures 10(a)–10(c) show the distribution of the Low Angle Grain Boundaries (LAGB), High Angle Grain Boundaries (HAGB) and Coincidence Site Lattice (CSL) boundaries at different cold deformation levels. It is clear that with the starting martensitic microstructure, increasing the cold deformation level from 30 to 80% leads to an increase in the HAGB and decrease in the LAGB fraction respectively. The CSL fraction initially decreases from virgin martensite to 30% deformation level and then again increases from there to 50% and 80% deformation levels. Figures. 11(a)–11(c) compare the changes in LAGB, HAGB and CSL boundary fractions between the cold rolled and the cold rolled and annealed materials (at 500°C and 700°C). In general, the LAGB fraction has been found to increase after 500°C annealing from the corresponding cold rolled level, whereas it decreases after 700°C annealing. Reverse is the trend for the HAGB fraction. No significant change in the overall CSL fraction has been observed as a function of annealing. Figures 12(a)–12(d) show the % frequency variation of different CSL boundaries in the virgin martensite, cold rolled as well as the annealed materials. In all the cases the most prominent CSL boundary has been the Σ3 type, followed by Σ25b, Σ11, Σ17b, Σ7 and Σ9 in decreasing order of importance. The Σ3 boundary fraction in all the cold rolled materials is significantly lower than its value for the virgin martensite, although the 80% cold rolled steel shows a decidedly higher Σ3 fraction as compared to the 30% and 50% rolled materials. Not much change has been observed in the Σ3 fraction on annealing, either in the 50% or in the 80% cold rolled steels, although for the 30% cold rolled material Σ3 fraction shows a significant decrease after 500°C anneal and a very prominent increase after 700°C anneal. The variation in the fraction of other CSL boundaries with annealing is not much and hence will not be discussed any further here.

Fig. 9. Variation in γ-fibre for the different cold rolled and annealed steels.

![Graph](image1.png)

Fig. 10. Grain Boundary Character Distribution (GBCD) of the steel: (a) LAGB, (b) HAGB and (c) CSL distribution in the martensite after 30%, 50% and 80% cold rolling.

![Graph](image2.png)

Fig. 11. The number fraction distribution of different types of boundaries in the (a) 30%, (b) 50% and (c) 80% cold rolled, 500°C and 700°C annealed steels.
3.4. Mechanical Properties

Figures 13(a) and 13(b) show the percentage elongation and UTS values of the cold rolled steels after annealing at different temperatures. The % elongation achieved in the 50% cold rolled material is the highest whereas the 80% cold rolled material shows the lowest % elongation values. The 30% cold rolled material exhibits % elongation values which lie within the two extremes. Obviously, the % elongation values, for any of the cold rolled material, increases with the increase in annealing temperature. The UTS value at any particular annealing temperature is the highest for
the 80% cold rolled steel and lowest for the 30% cold rolled steel. The UTS of the 50% cold rolled steel lies within these two extremes. Understandably, the UTS values show a progressive decline with annealing temperature for any particular cold rolling level. Figure 13(c) shows a plot of failure–ductility values (UTS×%Elongation) for the steel, at three different cold rolling levels, as a function of annealing temperature. It is clear from this figure that the highest values for this parameter at any annealing temperature are achieved for the 50% cold rolled material. The bulk hardness values of the steel rolled at different levels are shown in Fig. 13(d), as a function of annealing temperature. The hardness plot shows a trend similar to the UTS plot shown in Fig. 13(b).

The plot of the strain hardening exponent (n) values calculated from the tensile stress–strain data of the three cold rolled materials after annealing at 500°C and 700°C are shown in Figs. 14(a) and 14(b). This diagram clearly shows that the 80% cold rolled and annealed steel shows the highest n values amongst the three different cold rolled steels, after annealing at 500°C as well as 700°C.

4. Discussion

The martensitic steels are traditionally high strength steels with low ductility. Ductility can be increased by subjecting the steel to recrystallization anneal, but this always takes place at the cost of strength. Annealing of martensite may lead to two types metallurgical processes, namely, precipitation of carbides from martensite leading eventually to the formation of ferrite and secondly, relaxation of martensite/ferrite by the process of recovery and recrystallization. In the 30% cold rolled material recovery continues to occur till 700°C, the highest annealing temperature used in the current study, without any visible recrystallization setting in. On the other hand, both the 50% and the 80% cold rolled steels have been found to undergo recrystallization, the kinetics being faster in the latter as compared to the former. The above observations can be clearly explained by the facts that increase in the deformation level increases the amount of stored internal energy, which triggers the process of recrystallization.

That cold rolling and annealing of a martensitic structure can lead to the formation of ultrafine grains has been shown by earlier researchers. The present study also clearly indicates that it is possible to produce ultrafine ferrite grains after annealing the 80% cold rolled material at 500°C. In fact, several nano-sized grains were found to nucleate when the same material was annealed at a lower temperature. These could be the precursors to the final ultrafine grains.

Saha and Ray claim to have obtained submicron to nano-sized grains by subjecting a hot rolled IF steel to 98% and 99.5% of cold rolling. Severe cold rolling of the ferritic steel from 90 to 98% led an increment of the HAGB fraction from 0.39 to 0.50 in their case. In the present case the HAGB fraction of the 80% cold rolled martensitic steel has been found to have a value of ~0.6. Literature also shows that severe plastic deformation caused by techniques such as ECAP or ARB significantly increases the fraction of the HAGBs. As in the case of Saha and Ray’s work in the present case also there has been a continuous increase in the CSL boundaries with increasing amount of cold rolling.

Textural results clearly show the formation of γ-fibre in the annealed materials together with some α-fibre components. The α-fibre components are more prevalent in the 30% cold rolled material. Excepting the 30% cold rolled and 700°C annealed steel, all the other annealed materials show a reasonably uniform γ-fibre (within a band of intensity between 2.0 to 3.5 times random). The presence of such γ-fibres is indicative of reasonable deep drawability for such materials. However, their very low r-bar (<1.0) clearly indicates that this is not the case. The microstructures of the 50% and 80% deformed martensitic steels after high temperature annealing, consist of three phases, namely, ferrite, tempered martensite and carbide. Thus, basically in the annealed condition these steels very much resemble the traditional ferrite–martensite dual phase steels with low r-bar values. Therefore, such materials can not be considered in applications where high deep drawability is required.

The mechanical property values for the different materials clearly indicate the expected trends, namely that an increase in strength is always associated with a decrease in elongation. For example, when the 80% cold rolled material is annealed at 500°C, a strength–elongation combination of 900 MPa–11% is obtained. Thus it is clear that such materials can be used in applications which require high
strength without very high amount of ductility. The strain hardening exponent values (Fig. 14) show a general increase with an increase in the level of cold deformation both in the 500°C and 700°C annealed materials. This can be explained on the basis of the carbide precipitation in the ferrite matrix. The fine second phase particles may increase the strain hardening values by the creation of strain gradients near the particles owing to the generation of Orowan loops. The 30% cold rolled steel does not show much of precipitation. The 50% cold rolled steel shows the presence of carbide precipitates of slightly larger size as opposed to the 80% cold rolled one which exhibits highly dense precipitates of very small size. This could be the reason why the strain hardening exponent increases with the increase in cold deformation level.

The microstructural evolution in these martensitic steels, after cold rolling and annealing, appear very similar to that observed by Ueji et al. Ultrafine grains of comparable sizes have been observed in both the cases after similar levels of cold rolling and annealing. It has been observed that the fine grains are obtained essentially by discontinuous recrystallization of the cold rolled martensitic matrix. Recently, Song et al. has introduced a new concept for the production of ultrafine grains in steels with lean compositions. They had adopted the route of large strain warm deformation and subsequent annealing. The grain sizes achieved by these workers are comparable to those obtained by the present authors and by Ueji et al. although the texture developed in their material is primarily the α-fibre ((110)||RD), in contrast to the γ-fibre ((111)||ND) developed in the steels studied in the present investigation. Song et al. further suggested that in their steels, ultrafine grains are produced by a pronounced recovery process during which new grains are created without preceding nucleation, a process very much similar to “continuous recrystallization”.

Although Song et al. have not provided any result on the mechanical properties of their ultra fine grained steels, Ueji et al. have published some results on their steels. The latter group of researchers achieved the best strength–elongation combination of 870 MPa–9% in their 0.13%C–0.37%Mn martensitic steel after 50% cold deformation followed by annealing at 550°C. This compares favorably with the 900 MPa–11% strength–elongation values obtained in the present 0.17%C–1.6%Mn martensitic steel after 80% cold deformation followed by annealing at 500°C. The present results clearly indicate that the major reason for which the application of such high strength martensitic steels may be limited, is due to their low ductility. Obviously the next step will be to improve the ductility of such steels without losing much of the strength. In this connection application of grain boundary engineering (GBE) may be worth investigating. Randle and Davies have given some clue in this regard. They applied strain-recrystallization cycles to alpha-brass in order to enhance ductility without sacrificing tensile strength. Their results have clearly demonstrated that increase in the proportion of Σ3 boundaries leads to a general increase in the strain-to-failure values. It is therefore envisaged that by subjecting the martensitic steels to iterative processing may lead to a beneficial engineering of the grain boundaries, thereby increasing the overall ductility of the material. Figure 15 shows the traditional “banana” diagram depicting the entire range of steels currently available. It may be conjectured that Grain Boundary Engineering (GBE) of the present steel may lead to the formation of a new grade with properties ranging between those of TRIP and TWIP steels.

5. Conclusions

The following conclusions can be drawn from the present study:

1. Appropriate amounts of cold rolling and annealing of Fe–C martensitic steel can lead to the formation of ultrafine grains in the range of 50–250 nm.
2. The nucleation of nano sized grains in the matrix, when the 80% cold rolled steel is annealed at 400°C, is associated with a high angle boundary fraction of around 0.6.
3. With the increase in annealing temperature decrease in strength and simultaneous increase in ductility occur.
4. In most of the cases reasonably sharp γ-fibres are obtained, but the r-bar values are pretty low (<1.0) in all the steels.
5. Grain boundary engineering may be a good option to produce a better strength–toughness combination.

REFERENCES