Magnetic characterization of cold rolled and aged AISI 304 stainless steel

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Abstract

Magnetic easy axis predicted by the orientation distribution of the maximum amplitude of magnetic Barkhausen emission (MBE), which is obtained by magnetization in radial directions from the center of the specimens has been applied to determine the magnetic anisotropy on cold rolled and aged 304 SS in two sets of specimen. The maximum of the MBE has been found to orient along the rolling direction (RD) compared to the transverse direction (TD), indicating the presence of magnetic easy axis along the rolling direction for both sets. The strain induced martensite phase transformation has been determined using X-ray diffraction technique. The orientation distribution function (ODF) analysis has been carried out to obtain the crystallographic texture with cold rolling. ODF analysis revealed the $h110i$ texture as the major. The magnetic anisotropy factor has also been determined with cold deformation and noticed that the strength of magnetic anisotropy decreases above 50% deformation for both the sets. Results have been explained considering two competitive effects, formation of crystallographic texture in the martensite phase and presence of compressive residual stresses along RD during cold rolling.

Keywords: Stainless steel; Cold rolling; Martensite; X-ray diffraction; Texture; Residual stress; Barkhausen emission

1. Introduction

Texture or crystallographic preferred orientation is found in a polycrystalline metallic alloy, unless the constituent crystals are randomly oriented in space (which is less common). Thermo-mechanical treatments given to a metallic alloy always introduces texture. The importance of texture stems from the fact that it affects the physical, mechanical and corrosion properties. The standard techniques for characterizing texture is by diffraction—using mainly X-rays as the interrogating medium [1,2] and sometimes by neutron [3]. These being laboratory techniques, there is a need to avail techniques which can be used in the shop floor. Ultrasonic velocity measurement is a possibility since the velocity depends on the crystallographic direction along which the ultrasonic wave propagates [4]. Another possibility is by the use of magnetic properties since the magnetic behaviour is dependent on the crystallographic direction along which the properties are measured [5]. It should be noted that neither the ultrasonic nor the magnetic techniques gives detailed information on texture as much as given by the diffraction techniques. They are nonetheless important because they are quick and can be used in a shop floor condition. These techniques, however, require to be qualified by the results of a diffraction technique [4].

This paper discusses the application of magnetic Barkhausen emission (MBE) technique in characterizing texture in cold worked (and cold worked annealed) AISI 304 stainless steel (SS). MBE signals are generated essentially by the motion of 180° magnetic domain walls occurring around the coercivity point of a magnetic hysteresis loop [6]. The strength of the signal depends, among others, on the testing parameters such as maximum of the applied field strength, and micro-structural features which act as pinning points for the moving domain walls or domain generation sources—examples being, grain boundaries, dislocations, twin boundaries, precipitates etc. There has been a great amount of interest in studies on MBE in regard of its application in characterizing microstructures...
and residual stress [6]. The MBE data have been qualified by optical microscopy and XRD data. The work is part of a project on studying the evolution of texture and their effects on various properties of the steel. A solution annealed 304 SS is constituted by a non-magnetic austenite phase having face centered cubic crystallographic structure. When it is cold deformed, ferromagnetic martensite (distorted body centered cubic structure) α′ phase nucleates and grows having specific crystallographic relationship with the austenite phase [7]. Among others, the following aspects are important to be known: (i) relative amounts of the ferromagnetic and the non-ferromagnetic phases in the microstructure, (ii) development of texture and its measurement in the two phases, and (iii) the type and the magnitude of the residual stress developed. If the cold worked samples are annealed (which is a common industrial practice), there will not be any change in the relative amounts of the phases if the annealing temperature is carried out below the curie temperature of the ferromagnetic phase. There will, however, a change in the texture as well as the residual stress. In the literature, the use of magnetic techniques including MBE, magnetic hysteresis loop parameters and magnetic fluxgate magnetometer have been reported for the measurement of the amount of the magnetic phase [8–12]. Texture or preferred crystallographic orientations have been characterized by torque magnetometer [13] and MBE technique [14–16]. However, we have found that although attempts have been made to understand the nature of domain movements (leading to the preferred orientations), and reference to the data pertaining to texture by XRD, no systematic study comparing the data obtained by a diffraction technique and a magnetic technique like MBE are reported. This work is an attempt towards this direction.

2. Experimental

2.1. Material and thermo-mechanical treatment

AISI 304 SS grade steel plate of thickness 8 mm having chemical composition as shown in the Table 1 have been solution annealed at 1080 °C followed by quenching in water at room temperature. Annealed plates were cold rolled to various levels (in terms of % reduction in thickness) as 10, 30, 50, 70 and 90% under two different rolling conditions: (a) Set I, without inter pass cooling and (b) Set II, with inter pass cooling.

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Chemical composition of the AISI 304 stainless steel sheets</th>
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<td><strong>C</strong></td>
<td>0.03</td>
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</table>

2.2. Optical microscopy

The optical microscopy has been carried out after standard metallographic preparation.

For etching a solution has been prepared using 48% of Ammoniumdifloride, 800 ml distilled water and 400 ml conc. HCl. One gram of Potassium bi-sulphite (K2S2O5) has been added to 100 ml of the above solution and the specimen kept in this reagent for 10 s at 20 °C.

2.3. XRD measurements

The volume percentage of strain induced α′ martensite and residual stress measurement has been carried out using X-ray residual stress analyser, AST X2001. This equipment has a facility for the quantitative measurement of the amount of a ferromagnetic phase in the presence of a non-ferromagnetic phase in the microstructure. Seifert XRD 3003PTS was used to determine the Pole figures for {110}, {200}, and {220} planes of α′ martensite, using Cr Kα radiation.

2.4. Magnetic measurements

μ-Scan 500-1 equipment from American stress technologies, USA has been used to acquire and analyse the magnetic Barkhausen emission (MBE) signal. MBE signals are generated during the motion of the 180° magnetic domain around the region of the coercivity point when a ferromagnetic or a ferrimagnetic material is swept through a magnetic hysteresis cycle. The origin of the MBE signals is the local change in magnetization when a domain wall moves from one pinning point to another. A surface probe with a test frequency of 120 Hz was employed. The probe is made up of a ‘U’ shaped yoke to induce the cyclic magnetic field in the material. Within the space between the two ends of the yoke (e.g. the magnetic poles) is situated a pick-up coil which detects and measures the MBE signals. The output signals in the frequency range of 1–500 kHz has been acquired. The peak values of the envelope of the MBE signals generated (VP) has been used as the magnetic parameter. To characterize oriented nature of the magnetic properties, the values of VP has been measured radially around the centre point of the specimen plates along the line joining the centers of the pole surfaces of the yoke, and the centre point of the specimen. The measurements has been made at 15° intervals.

3. Results

3.1. Optical microstructures

The optical micrographs of 0, 30, 50 and 90% cold rolled specimens of set II are shown in Fig. 1. In a bright field (optical) microstructure, the etchant used brings out
the martensite phase as a ‘black’ constituent, the austenite phase remaining white. Fig. 1(a) shows essentially a single phase. The grain boundaries are faintly visible. Because of this, no attempt has been made to measure the average grain size by the standard intercept method. However, from Fig. 1(a), one can conclude that it was a coarse grain microstructure with many of the grains exceeding 50 μm.

The wavy features in the microstructure going in diagonal directions signify the manifestations of the deformation layer on the specimen surface. The small amount of black features seen in this micrograph is due to residual fine scratches and inclusions. It is pointed out that much difficulty has encountered in preparing the specimen surface in the solution annealed condition. The problems were less in the specimens in the cold worked conditions. From the Fig. 1(b)–(d) one can see the increase in the amounts of the black phases signifying the increase in the amounts of the martensite phase. It is apparent that the slip lines and the twins had a bearing to the nucleation and the growth of this phase. It is known that the phase nucleates at the intersections of shear bands. The details of these features have been investigated by several investigators and are beyond the scope of this paper.

### 3.2. Amount of martensite phase determined by XRD

The volume percentage of strain induced α’ martensite of cold rolled samples is determined by XRD quantitative phase analysis in both sets and is given in Table 2.

Table 2 reveals that the level of α’ content is same in both the sets of specimens till 50% reduction and above this, α’ content is more in set-II specimen where inter-pass cooling has been used. The reason is the adiabatic heating of

<table>
<thead>
<tr>
<th>Cold work level</th>
<th>0%</th>
<th>10%</th>
<th>30%</th>
<th>50%</th>
<th>70%</th>
<th>90%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Vol% of set I</td>
<td>0%</td>
<td>2.5</td>
<td>10</td>
<td>25</td>
<td>28</td>
<td>58</td>
</tr>
<tr>
<td>Vol% of set II</td>
<td>0%</td>
<td>2.5</td>
<td>10</td>
<td>25</td>
<td>50</td>
<td>82</td>
</tr>
</tbody>
</table>

Table 2
Volume percentage of martensite with % of Cold rolling as determined by XRD
the specimen during cold rolling which in turn reduces the driving force for \( \gamma \) to \( \alpha \)\(^\prime\) transformation. By changing the rolling conditions it has been possible to vary the \( \alpha \)\(^\prime\) martensite content from 10 to 82\% in volume fraction in the samples.

3.3. Texture data obtained by XRD

\{110\} pole figures obtained by XRD in the set I and set II of the specimens are shown in Fig. 2(a) and (b), respectively. Progressive development of texture during cold rolling can be very clearly seen from the pole figures. The main texture component forms at around 50\% cold deformation and its intensity continuously increases with increasing cold reduction. Cold rolling also decreases the spread of the main texture component. The \( \alpha \)\(^\prime\)-martensite phase exhibits relatively weak texture till 50\% reduction in set-I specimen as compared to set-II specimen though the volume fraction of the martensite is nearly same in both the samples. The low volume fraction of martensite phase in 30\% CR specimens exhibits very weak peak intensity. This has restricted the measurement of experimental pole figures at lower cold reductions.

The orientation distribution function (ODF) analysis has been carried out for all the samples. The ODF analysis exhibits \{001\}\{110\} and \{112\}\{110\} texture component as main texture component at all cold reduction levels. At 90\% CR, in addition to major components \{332\}\{113\} texture component also develops in both the set of samples.

3.4. Magnetic Barkhausen emission data

The polar plots of the \( V_p \) (meaning explained earlier under section Section 2) for 30, 50, 70 and 90\% cold rolled specimens for set-I and set-II are shown in Fig. 3(a) and (b), respectively. The vertical axis (0–180\°) pertains to the rolling direction (RD). The axis perpendicular to this (the 90–270\° axis) is referred to as the transverse direction (TD) axis. It can be seen that for all the plots, the maximum values of \( V_p \) lie on the RD. The plots are dumbbell shaped with a constriction at the TD axis. The amounts of the constrictions are not the same in the various plots.

4. Discussion

4.1. Magnetic medium and easy axis.

In a polycrystalline alloy the dynamics of the magnetic domains during a hysteresis sweep is complex because of the following reasons. Domains are nucleated at various microstructural features like grain surfaces, twin
boundaries, slip lines and secondary precipitates etc. The morphology of the nucleated domains depends on the physical nature of the surfaces of these features. The motion of the domain walls depend on the free space available for the motion between the various microstructural features acting as pinning points for the domain walls, the pinning strengths of the features, the nature of the crystallographic directions lying parallel to the directions of magnetization (as to whether they are easy, medium or hard axes). The polar plots of $V_p$ will be affected by these features acting sometimes in a synergistic manner and sometimes in an opposing manner.

As shown in Fig. 3, the peak amplitude of Barkhausen emission ($V_p$) is maximum along RD and minimum along TD in both the rolling conditions. This indicates that magnetic easy axis predominantly lies along the RD. It is also important to note that unlike in ferritic alloy steels where a shift in the orientation in the magnetic easy axis has been reported [15] under different levels of deformation, in the present case, there is no shift and it lied along the RD. This is attributed to be due to the presence of austenite phase in the microstructure and the requirement of maintenance of an orientation relationship between the austenite and the ferrite phase [6]. The dumbbell nature of the polar plots shown in Fig. 3 is attributed to the complex nature of the domain nucleation and dynamics explained earlier. If there is a texture like $\{100\}\{100\}$ in which the $\{100\}$ planes of all the grains of the sheet lie parallel to the major surfaces of the sheet and the one of the $\{100\}$ directions $\langle100\rangle$ lie along the RD, then as explained on page 225 of Ref. [1], the magnetic domain dynamics from the demagnetized state to saturation and while completing the loop, will only consist of domain wall motion at a very small magnetic field. In such a case, the polar plot will show only a line along the $0$–$180^\circ$ axis with a high magnitude, because it will have a probability to find a direction of magnetic easy axis only along this direction. Along other directions this probability will ideally be zero. On the other hand, if there is an ideally random texture, the polar plot should show a circle, because there will be equal probability of one of the easy magnetic axes to lie along any of the poles around the polar origin. Between these two cases, it is expected to get a dumbbell shaped plot with various extents of constriction as seen in Fig. 4 of Ref. [16].

Comparing the XRD texture data and the MBE polar plots, it can be concluded that RD coincided with the $\langle110\rangle$ crystallographic direction which is a magnetic medium axis—the easy axis being $\langle100\rangle$ and the hard axis being the $\langle111\rangle$ direction [5]. XRD analysis also showed the presence of $\langle110\rangle$ crystallographic direction along TD. As a result, with increasingly cold work, the constriction along the $90$–$270^\circ$ has been reduced. This means a direction perpendicular to the magnetic medium axis started occurring as a texture direction. The $\langle100\rangle$ direction in the MBE polar plot (Fig. 3) would lie at an angles of $45$–$270^\circ$ and the $135$–$315^\circ$ direction. As can be seen for both the sets I and II, the probability of the easy magnetic axis lying parallel to the direction of magnetization increased with increasing levels of deformation.

![Fig. 3. Magnetic pole figures of (a) Set I (without inter pass colling) & Set II (with inter pass colling).](image1)

![Fig. 4. Variation of peak voltage of Barkhausen emission along the rolling direction with volume % of martensite of Set I (without inter pass colling) and Set II (with inter pass colling).](image2)
4.2. Comparative sensitivity of MBE plot and XRD pole figure

It has been found that at as low as 30% cold reduction (when the amount of martensite is about 10%), the MBE plot clearly shows the presence of texture. On the other hand, XRD pole figure shows the presence of texture only at or after 50% of cold reduction. Below this level, the MBE signal strength was not significant. This is in line with reports made by other investigators.

4.3. Empirical texture parameter from MBE plots

From the viewpoint of fabrication process, a simple texture parameter would be useful. The maximum value of $V_p$ in the RD could be one possibility. The variation of $V_p$ along RD with vol% of martensite for set I and set II are shown in Fig. 4. It is seen that for both set I and set II, the plot continually increases with a tendency to saturate at higher levels of cold reduction. Table 2 also shows that the continuous increase in the amount of martensite with an increase in cold reduction in both the sets. Fig. 5 shows the plot using the ratio of $V_p$ along RD and TD with vol% of martensite. It can be seen that the trend for set I and set II are same; there is a decrease after attaining a maximum at 52% of martensite (corresponding to 70% cold reduction).

In the literature, a parameter defined as below has been used as an empirical texture parameter [14]

$$k = \frac{(V_p)_{RD} - (V_p)_{TD}}{(V_p)_{RD} + (V_p)_{TD}} \tag{1}$$

Fig. 6 shows the plots on the variation of $k$ as a function of the amount of martensite. This shows similar variations as in Fig. 5. For both sets, a maximum is attained which is followed by a decrease. This reduction is attributed to the increase in the probability of finding (110) direction along the TD. We can thus see that all the three parameters invoked to describe the texture give consistent information.

It is well known that one of the strong influencing factors on MBE signals is residual stress. To understand the possible role of residual stress, its value along the RD has been measured by XRD and plotted as a function of cold work level in Fig. 7. It is seen that at higher levels of cold work, the residual stress is highly compressive. The stress anisotropy factor expressed by the following equation then becomes important [17]

$$K_s = \frac{3}{2} \lambda \sigma \tag{2}$$

Where $\lambda$ is the magnetostriction constant and $\sigma$ is the stress. For compressive stress, $K_s$ becomes negative, which causes the stress axis to be the hard axis whereas the plane normal to the stress axis is the easy plane of magnetisation. Hence in cold rolled 304 SS presence of compressive residual stress along RD, for higher percentage of deformation, tries to orient the magnetic domains in the transverse to the rolling direction. As a result $V_p$ along TD (($V_p)_{TD}$) increases, which in turn reduces the $(V_p)_{RD}/(V_p)_{TD}$ value as well as the magnetic anisotropy factor $k$ (as defined in Eq. 1) beyond 50% deformation for both sets. To have still a better understanding on the role of residual stress, 70 and 90% cold rolled specimens of both sets are annealed at 400 °C to relieve the residual stress and MBE polar plots have been made for 90% cold rolled and cold rolled annealed
specimens for both sets (Fig. 8(a) and (b)). These figures show a more prominent texture in the annealed specimen with an increase in the value of \((V_p)^{RD}\) and decrease in \((V_p)^{TD}\) compared to the rolled specimen. Which is more clear in Figs. 5 and 6 where continuous increase in \((V_p)^{RD}/(V_p)^{TD}\) and the magnetic anisotropy factor \((k)\) with vol% of martensite are observed for rolled and then annealed specimen (as shown by dotted lines in Figs. 5 and 6).

5. Conclusion

The magnetic Barkhausen emission technique has been used to qualitatively study the texture (preferred crystallographic orientation) in two sets of cold worked AISI 304 SS specimens: set I, without inter pass cooling and set II, with inter pass cooling. The nature of the plots of MBE signals in polar co-ordinates as obtained by recording the maximum amplitude of MBE \((V_p)\) in different magnetization directions around the centre point of plate specimens have been compared with X-ray pole figures. During cold working magnetic martensite phase nucleates and grows within the non-magnetic austenitic phase. The texture within the martensite phase is influenced by the cold work as well as the texture developed within the austenite phase. The martensite phase transformation has been observed to be more in set II for higher percentage of deformation compared to set I because of the adiabatic heating of the specimen during cold rolling, which in turn reduces the driving force for \(\gamma\) to \(\alpha'\) transformation.

Micro-magnetic Barkhausen emissions technique indicated the occurrence of rolling texture along RD from cold work level of 30% onwards (12 vol% martensite) whereas by XRD below 25 vol% of martensite no distinct crystallographic texture has been observed. Three different empirical texture parameters have been considered. Variation of all these parameters with vol% of martensite follows the texture result as determined by ODF analysis.

The compressive residual stress introduced on the surface by increasing levels of cold work have tended to saturate the peak value, \(V_p\) along the rolling direction (RD), and even to reduce it. This factor seems to have contributed to increase the magnetic medium axis along the transverse direction (perpendicular to RD).

The present work has shown that the simple low cost MBE technique can be used to evaluate qualitatively the rolling texture in a material even at relatively lower levels of cold work when the volume percentage of the magnetic phase is low. However, more work is needed using transmission electron microscopy and magnetic force microscopy to understand the correlation of the crystallographic anisotropy and the magnetic domain configurations.

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References


