Synergistic effects of holding time at intercritical annealing temperature and initial microstructure on the mechanical properties of dual phase steel

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ABSTRACT

The synergistic effects of the initial microstructure and the intercritical annealing time on the microstructure and tensile properties of low carbon steel were studied. It was found that by refining the pre-intercritical annealing microstructure, the resulting duplex DP microstructure becomes finer, which results in the enhancement of the tensile properties and work-hardening behavior. Based on the hardness measurements, three stages were identified for intercritical annealing: initial rise or fall, reaching a plateau, and final fall to a low value. The pre-intercritical annealing microstructure did not show any pronounced effects on these stages, with the exception of the short-lived initial stage. It was shown that the DP steel obtained right after reaching the abovementioned plateau can exhibit improved mechanical properties and work-hardening behavior. During the third stage, the enrichment of manganese in austenite and the concurrent grain growth were found to be the main factors taking part in the change of hardness and tensile strength and reappearance of the yield-point elongation. The impact of the microstructural features (austenite and ferrite) on the grain growth behavior during intercritical annealing and the effect of the grain size on the Lüders strain were also discussed.

1. Introduction

Weight reduction and the resulting fuel efficiency is one of the main aims of the vehicle manufacturers. This can be achieved via thinner gauges of the parts, where, in return, increasing the strength is a vital requirement. However, the resulting deterioration of formability effectively retards the applicability of the used material. The response of the steel industry was the introduction of advanced high strength steels (AHSS) [1–4]. Among them, the ferritic-martensitic dual phase (DP) steels have been developed, where the co-presence of martensite governs the noticeable work hardening rate and remarkable ductility-strength balance. Their tensile properties can be adjusted by the grain size of ferrite and also the volume fraction, size, and morphology of the martensite islands [5–8]. These microstructural features are dependent on the initial microstructure and the parameters of the intercritical annealing as summarized in the following:

(I) Heat treatment [9–12] and thermomechanical processing routes [13–21] can be used to control the initial pre-intercritical annealing microstructure. It has been shown that the cold rolled martensite [16–19] and the cold rolled DP steel [20] are appropriate ones for the subsequent intercritical annealing to obtain fine-grained DP steel. In these cases, the heating rate to the intercritical annealing temperature is quite important [16–18]. Moreover, for enhancement of mechanical properties of the resulting DP steel, the martensitic microstructure is generally a better initial microstructure compared with the ferritic-pearlitic ones for intercritical annealing [9,10].

(II) Beside the heating and cooling rates, the temperature and time of the intercritical annealing are the main parameters for the control of DP microstructure. The effect of intercritical annealing temperature on martensite content has been widely studied [22–24], where the decrease in the carbon content of martensite with increasing its volume fraction has been considered as an important factor [24]. There are few reports on the effect of the intercritical annealing duration [16,25–30], where elucidating its importance on the adjustment of martensite fraction has been the main aim so far. This has been investigated in the case of the cold rolled steel sheets [16,17] and also for the medium-carbon steels [28]. Moreover, the partitioning of manganese has been a subject of several investigations [29–31]. However, despite the importance of the grain size in DP steels [13–20], grain growth during intercritical annealing has received less attention [32–34].

Based on the importance of the initial microstructure and the intercritical annealing time, the present work has been dedicated to study...
their synergistic effect for a typical low carbon steel (0.035 wt% C - 0.268 wt% Mn - 0.035 wt% Si). A wide range of intercritical annealing durations and three initial microstructures (normalized, full annealed, and martensitic) were considered. The resulting DP microstructures, mechanical properties, and work-hardening behaviors were characterized to elucidate these synergistic effects.

2. Experimental details

2.1. Processing

A 0.035C-0.268Mn-0.035Si (wt%) steel sheet was received in the normalized condition. This sheet was austenitized at 1323 K (1050 °C) followed by furnace cooling/water quenching to develop full annealed ferritic-pearlitic/dominantly martensitic microstructures, respectively. The as-received, full annealed, and quenched sheets were considered as the pre-intercritical annealing microstructures. The temperature of 1073 K (800 °C) in the intercritical annealing range with ~ 21 vol% martensite was considered (the amount of martensite was measured based on the area fraction of the 2D micrographs). Various intercritical annealing durations from 0.5 min up to 120 min were considered and some of them are shown in Fig. 1. In this figure, a code has been assigned for each sample to make the presentation of the results more straightforward.

2.2. Characterisation

The obtained microstructures were etched by the LePera’s reagent and Nital solution. An optical microscope and a Vega Tescan SEM equipped with a Bruker detector for energy-dispersive spectroscopy (EDS) were used for microstructural investigations. The JIS Z 2201 standard for tensile test specimens was used. A universal testing machine was used for tensile tests at room temperature with the initial strain rate of 0.001 s\(^{-1}\). The reproducibility of results was evaluated by the repetition of the tests. The hardness measurements were based on the Vickers hardness under a load of 30 kg. The values of work-hardening rate (\(\varepsilon = \varepsilon_0 + \alpha \varepsilon\)) were calculated based on the equation of \(\varepsilon = \varepsilon_0 + \alpha \varepsilon\) (35-37) to apply the modified method of the Crussard-Jaoul analysis [38-40]. This technique is based on the relation of \(\varepsilon = \varepsilon_0 + \alpha \varepsilon\), where \(\varepsilon_0, k, n\) are constants. So, \(\varepsilon = \varepsilon_0 + \alpha \varepsilon\), then \(\varepsilon = \varepsilon_0 + \alpha \varepsilon\), and eventually \(\varepsilon = \varepsilon_0 + \alpha \varepsilon\). Therefore, the plots of \(\varepsilon = \varepsilon_0 + \alpha \varepsilon\) will be taken into account.

3. Results and discussion

3.1. The effects of the initial microstructure

The microstructure of the as-received sheet (normalized) is shown in Fig. 2a, which reveals a typical ferritic-pearlitic microstructure in low-carbon steels. After austenitization at 1323 K (1050 °C) followed by furnace cooling, a much coarser ferritic-pearlitic microstructure was achieved (full annealed microstructure in Fig. 2a), where the average ferrite grain size increased from 17 µm to 36 µm. For the austenitized and water quenched sample, the presence of martensite with lath morphology is evident, where some amount of ferrite ~ 10% has formed as a result of very low hardenability of this steel with low carbon and manganese contents (0.035 wt% C and 0.268 wt% Mn) [41-43].

Fig. 2b shows the tensile properties of these sheets. The low strength of the full annealed sheet can be related to the presence of coarse ferrite grains. The as-received sheet with finer microstructure has higher tensile strength compared with the full annealed sheet (326 MPa versus 257 MPa). Both sheets show the yield-point elongation at the onset of plastic flow, which is common in low-carbon steels and it is related to the formation of Cottrell atmospheres produced by interstitial atoms around dislocations [44,45]. The quenched sheet shows higher tensile strength (465 MPa) due to the presence of martensite. However, the strength of the quenched sheet is not comparable with those of martensitic steels [42], which can be related to the co-presence of ferrite in the microstructure of this sheet. In fact, it has a DP microstructure, and hence, shows relatively good total elongation of ~ 20%. Beside these findings, due to the attributes of the lath martensitic structure (presence of prior austenite grain boundaries, pockets, blocks, and laths [46]), this microstructure can be considered to be much finer compared with the as-received sheet [47].

Typical microstructures after intercritical annealing (for 5 min) are also shown in Fig. 2a. The absence of yield-point elongation in DP steels is evident, which is related to the transformation of austenite to martensite during quenching from the intercritical annealing temperature and generation of free dislocations in the surrounding ferrite [16,18,42].

Fig. 2a also shows that with refining the pre-intercritical annealing microstructures (the quenched and full annealed sheets respectively
show the finest and the coarsest microstructures), the resulting DP microstructures become finer and both ferrite grain size and the size of martensite islands decrease as summarized in Table 1. The refinement of the DP microstructure [13–20] has a direct effect on the enhancement of the tensile properties as shown in Fig. 2b and Table 1. These observations can be related to the improved work-hardening capacity as shown in Fig. 2c. It can be seen that, at each flow stress, the DPQ5 steel exhibits a higher work-hardening rate compared with the DPN5 and DPF5 steels. Due to the yield-point phenomenon at the beginning stages of the plastic flow, the as-received sheet shows low work-hardening rate. Distinct stages can be identified for the work-hardening of the DP steels: The initial transient stage of glide in ferrite resulted from the mobile dislocations (Stage 1), the normal work-hardening of ferrite (Stage 2), and the deformation of already hardened ferrite and martensite (Stage 3) [9,16,39].

3.2. The effects of the intercritical annealing time on the full annealed sheet

As shown in Fig. 1, the full annealed and quenched sheets were intercritically annealed for different durations. The hardness values of

Table 1
Summary of microstructural observations and tensile properties.

<table>
<thead>
<tr>
<th>DP Steel</th>
<th>Average Ferrite Grain Size (µm)</th>
<th>Average Size of Martensite Islands (µm)</th>
<th>Yield Stress (MPa)</th>
<th>Ultimate Tensile Strength (MPa)</th>
<th>Total Elongation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>DPF5</td>
<td>35.1</td>
<td>30.0</td>
<td>160</td>
<td>373</td>
<td>44</td>
</tr>
<tr>
<td>DPN5</td>
<td>15.1</td>
<td>8.8</td>
<td>162</td>
<td>387</td>
<td>42</td>
</tr>
<tr>
<td>DPQ5</td>
<td>8.1</td>
<td>4.5</td>
<td>230</td>
<td>405</td>
<td>36</td>
</tr>
</tbody>
</table>

Fig. 2. The effects of the initial microstructure: (a) Micrographs of the pre-intercritical annealing sheets (full annealed, normalized, and quenched) along with the corresponding DP sheets (DPF5, DPN5, and DPQ5), (b) Tensile stress-strain curves, and (c) Work-hardening rate plots.

Fig. 3. Hardness versus intercritical annealing time at 1073 K (800 °C) for different initial microstructures.
the obtained sheets are reported in Fig. 3. It can be seen that the hardness of the full annealed sheet initially increases (Stage I), and then, the hardness experiences a plateau from ~ 5 min to ~ 20 min (Stage II), and eventually falls to a low value (Stage III). The corresponding tensile stress-strain curves and microstructures are shown in Fig. 4. It can be seen that the trend of changes in tensile strength (Fig. 4b and Fig. 4c) are generally consistent with that observed for hardness in Fig. 3.

The initial increase in hardness and tensile strength is related to the substitution of pearlite and some amount of ferrite by austenite (martensite after quenching). The tensile curve of DPF0.5 in Fig. 4b exhibits the shortening of the yield-point elongation, which is consistent with the formation of some amount of martensite that is not sufficient to remove this phenomenon completely. By increasing the intercritical annealing time, the appearance of the hardness plateau at ~ 5 min is related to the formation of the nearly equilibrium ferrite-austenite microstructure, which remains nearly unchanged up to ~ 20 min. By further intercritical annealing, the grain size remains nearly unchanged (See DPF120 in Fig. 4a) but the hardness and tensile strength drop considerably in Stage III. Therefore, the latter observation should be related to the partitioning of alloying elements as discussed in the following.

It has been shown by many researchers that the intercritical annealing has three stages [25–27,30]: (1) Very rapid pearlite dissolution (controlled by the diffusion of carbon), (2) Increasing the fraction of the austenite by growing into the ferrite grains and along grain boundaries (controlled by the diffusion of manganese and carbon), and (3) Final slow equilibration of the Mn contents of the austenite and ferrite phases (controlled by the diffusion of manganese in austenite). Therefore, applying long intercritical annealing times is identical to increasing the Mn content of austenite based on the stage (3). To support this claim, EDS point analysis were taken from the martensite islands of DPF5 and
DPF120 samples as shown in Fig. 5. It can be seen that the Mn content of martensite after 5 and 120 min respectively is 0.11 and 0.43 wt%, which supports the enrichment of austenite by Mn during intercritical annealing. It has been known that all alloying elements, except cobalt, lower the martensite start temperature \((M_s)\) \([42,48]\): 
\[ M_s = 812 - 423C - 30.4Mn - 12.1Cr - 17.7Ni - 7.5Mo. \]
Therefore, by increasing the Mn content of austenite for long holding time at intercritical annealing temperature, \(M_s\) decreases. It has been also reported that the volume change of austenite to martensite transformation decreases as the \(M_s\) temperature decreases \([49]\). By consideration of the tensile curve of DPF120 in Fig. 4b, it can be clearly seen that the yield-point elongation has reappeared, which implies that enough free dislocations have not been generated in ferrite. This supports the above discussion. Therefore, the DPF120 steel should have inferior mechanical properties compared with DP steels obtained at lower intercritical annealing times. This can be better evaluated based on the work-hardening analysis, which is shown in Fig. 6d. It can be seen that DPF120 steel shows an initial poor work-hardening behavior (notice the presence of the yield-point elongation regime). Moreover, in the normal work-hardening regime, its work-hardening curve falls below that of DPF5 steel, which is responsible for its inferior mechanical properties. However, its work-hardening curve locates above that of the as-received sheet due to the presence of martensite.

3.3. The effects of the intercritical annealing time on the quenched sheet

Fig. 3 also shows the change in the hardness of the quenched sheet versus intercritical annealing time. For this initial microstructure, the hardness initially decreases (Stage I), and then, the hardness experiences a plateau from ~ 5 min to ~ 20 min (Stage II), and eventually, falls to a low value (Stage III). Except the initial stage (Stage I), the change in the hardness versus intercritical annealing time curve for the full annealed and quenched sheets coincide with each other. The hardness drop in Stage I of intercritical annealing can be related to the tempering of martensite and substitution of some amount of martensite with ferrite, which is well-known and will not be considered further in this work.

The equilibrium DP microstructures (Stage II) have hardness values identical to those obtained from the full annealed sheet. The corresponding tensile stress-strain curves and microstructures are shown in Fig. 6. It can be seen that the change in tensile strength in Fig. 6a and Fig. 4b is consistent with that observed for hardness in Fig. 3. The decline of hardness and tensile strength in Stage III for the quenched sheet can be related to the partitioning of alloying elements (as discussed for Fig. 5) and also to the obvious abnormal grain growth (AGG) as can be seen in Fig. 6c for the intercritical annealing time of 120 min. These microstructural changes have adverse effects on the work-hardening response as shown in Fig. 6d, where DPF_{Q120} steel shows inferior...
behavior when compared with the DPQ5 steel. An important question remains to be answered: Why long-term intercritical annealing of the quenched sheet results in AGG of ferrite; while this is not the case for the full annealed sample? This will be elaborated in the following.

It has been shown by Humphreys and Hatherly [50] that a dispersion of second-phase particles in the microstructure can prevent growth above the limiting grain size. However, under certain circumstances, abnormal grain growth may still be possible. By consideration of the volume fraction \( f \) and diameter \( d \) of the second phase in a microstructure containing grains of mean radius of \( R \), Fig. 7 has been developed to distinguish the regimes of grain growth based on the plot of \( R \) versus dispersion level \( f/d \).

Based on the \( d \) and \( 2\bar{R} \) values reported in Table 1 and \( f = 0.21 \), the points corresponding to DPF5 and DPQ5 are also shown in Fig. 7. The intercritical annealing time of 5 min was used because both sheets at this stage show DP microstructures with equilibrium martensite fraction. Continued annealing at the intercritical annealing temperature can be considered as grain growth annealing. It can be seen in Fig. 7 that the point corresponding to DPQ5 is located at the center of the abnormal growth zone; whereas the point corresponding to DPF5 is located near the boundary of the normal growth zone. The observed trend in the present work is consistent with this analysis.

Fig. 6 also shows that DPQ120 does not show the yield-point phenomenon, while as shown in Fig. 4b, DPF120 exhibits this phenomenon. This needs more explanation. It has been shown by Hall [51] that the Lüders strain is a function of grain size, where this feature was particularly marked in the case of mild steel as shown in Fig. 8. When the grain size is fine, the bands appear sharp and deformation virtually proceeds grain by grain along the specimen. With coarse grain sizes, however, the sharp front to the bands may be lost, leading to the so-called diffuse Lüders bands. The ferrite grain size of DPF120 and DPQ120 are \( \sim 0.036 \text{ mm} \) and \( \sim 0.2 \text{ mm} \), respectively. Since at the onset of plastic flow, the large grains are normally being deformed, only large grains for DPQ120 are considered in the microstructure shown in Fig. 8. These points are also shown on Fig. 8, where the Lüders strain of DPF120 and DPQ120 can be estimated as 2.5% and 0.5%, respectively. It can be seen that the Lüders strain of DPF120 is large, which is consistent with Fig. 4b. However, the Lüders strain of DPQ120 is very small.

4. Conclusions

The synergistic effects of the initial microstructure and intercritical annealing time on the microstructure and tensile properties of low carbon steel were studied. The following conclusions can be drawn from this work:

(1) By refining the pre-intercritical annealing microstructure, the resulting DP microstructure became finer and both ferrite grain size and the size of martensite islands decreased. Normalizing, and particularly austenitization and quenching, were found to be effective heat treatments in this respect. The refinement of the DP microstructure had a direct effect on the enhancement of the tensile properties, which was related to the improved work-hardening...
behavior.

(2) Hardness measurements during intercritical annealing revealed the presence of three distinct stages: initial rise or fall (Stage I), reaching a plateau at ~ 5 min (Stage II), and finally fall to a low value (Stage III). The pre-intercritical annealing microstructure did not show any pronounced effects on these stages, with the exception of the Stage I. It was shown that the DP steel obtained at Stage II exhibits improved mechanical properties and work-hardening behavior. During Stage III, the enrichment of manganese in austenite and also grain growth were found to be responsible for the decrease in hardness and tensile strength.

(3) Long-term intercritical annealing resulted in the reappearance of the yield-point elongation in the tensile stress-strain curves, which was related to the enrichment of austenite with manganese. The latter resulted in the decrease of the martensite start temperature, which is known to be responsible for the decrease in the volume change of austenite to martensite transformation. However, this reappearance was also found to be sensitive to the initial microstructure, where the effect of the microstructural features (austenite and ferrite) on the grain growth behavior during intercritical annealing and the effect of the grain size on the Lüders strain were also discussed.

Acknowledgment

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Data availability

The raw data required to reproduce these findings are available to download from Mendeley Data [https://data.mendeley.com/datasets/9c3yxxvvh1/draft?a=9df9909a7-108d-4883-afaf-82012271b11c].

References