Phase Transformation Products in a Low-Mn High-Nb X65 Steel Produced by Accelerated Cooling

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ABSTRACT

The identification of acicular ferrite (AF) and bainite (B) in linepipe steels is important for the determination of toughness by means of drop-weight tear test (DWTT), as those microconstituents give different responses for crack propagation during pipe usage. For this assessment, the phase transformation behavior in a low Mn and high Nb microalloyed steel plate was investigated for the characterization of AF and B, through dilatometry and SEM examination. The local misorientation was determined through electron backscatter diffraction, EBSD, (kernel average misorientation - KAM) for further assessment of the microstructure. The findings from this study showed no evidence for the distinction between AF and B in terms of start and finishing temperatures, as well as cooling rates. Results suggest that AF and B cannot be separated by distinguishable process conditions in this particular steel and other toughening mechanisms should be activated to improve the response to crack propagation.

Keywords: phase transformation; bainite; acicular ferrite; dilatometry; EBSD

INTRODUCTION

For the development of steel plates applied in offshore sour service pipelines, phase transformation of austenite is a fundamental mechanism towards obtaining high levels of strength, toughness and resistance against hydrogen induced cracking (HIC).

For strength the main contribution comes from the presence of very fine AF grains and from low angle grain boundaries (LAGB) misorientations, typically found in bainite ¹². Toughness performance relies on the presence of high angle grain boundaries (HAGB), above 15° ³, capable of deflecting the crack propagation. This type of misorientation is prevalent in AF due to its inherent intragranular nucleation nature. The presence of intragranular sites, such as deformation bands and non-metallic inclusions, are beneficial to the maximization of AF ⁴,⁵. Since non-metallic inclusions are severely detrimental to the HIC performance in steels for sour service applications, these nucleation sites are more commonly pursued in welds. The type of AF focused on this study nucleates from deformation bands and dislocation substructures, displaying and displays fine irregular grains. For this purpose Nb plays an important role by delaying the recrystallization of austenite, allowing the proper conditioning of the latter.
In HSLA steels, AF was defined by Smith et al. in 1972 as a highly substructured, non-equiaxed ferrite that forms upon continuous cooling by a mixed diffusion and shear mode of transformation that begins at a temperature slightly higher than the upper bainite transformation range. Although AF in HSLA steels nucleates intragranularly and its transformation mechanism is bainitic, as AF is the preferred microconstituent regarding to combining strength and toughness, and the temperature conditions are like those of B, it is important to assess the phase transformation products of austenite to obtain the optimal processing parameters during the industrial hot rolling.

**RESULTS**

1. Materials and Methods

Samples were taken from low Mn, high Nb X65 steel plate hot rolled to 32 mm thickness and cylindrical test pieces were machined with 5 mm diameter and 10 mm length. The chemical composition consisted of 0.06%C, 0.97%Mn, 0.07%Nb, 0.60%(Cu+Cr+Ni). The non-recrystallization temperature (Tnr) was determined by multi-pass hot torsion tests applying the method developed by Boratto et al. and was 1010°C. The Ar3 temperature was determined during the same hot torsion test and was 770°C.

The thermomechanical cycles were performed in a DIL805 A/D/T dilatometer coupled with a compression module. Two different deformation schedules were specified to evaluate the influence of the austenite conditioning in the transformation products and to determine the transformation temperatures within the industrial cooling range. The start and finish transformation temperatures were determined by analyzing the test piece length variation versus temperature curves and applying the method described in 11). The derivative curves were also determined as an additional support for confirming the inflexions representing the phase transformations. All test pieces were heated at 1250°C and kept for 15 min. Cycle 1 consisted of a 25% roughing pass at 1230°C and a 25% finishing pass at 850°C. Cycle 2 consisted of a 15% roughing pass at 1230°C and a 40% finishing pass at 850°C. The deformation temperatures were selected based on actual industrial rolling schedules and the cooling rates (10, 20 and 30°C/s) represent the actual range possible for a 32 mm plate. The thermomechanical cycles are shown in Figure 1. Additionally, the austenite grains were characterized by quenching the microstructure at three different stages: before the first pass (AG 1), before the 40% finishing pass (AG 2) and before the cooling stage (AG 3).

The microstructure was characterized by optical microscopy (OM) using an Olympus BX51M microscope and by scanning electron microscopy (SEM) using a JEOL microscope model JSM-7100F with field emission gun (FEG). Samples were prepared by conventional grinding and polishing procedures and etched with 2% nital. EBSD analyses were carried out for the characterization of the level of strain in each sample by using the kernel average misorientation mapping and the preparation followed standard procedures. The step size for data acquisition was 0.5 µm and analyses was done by Oxford Instruments Channel 5 software application.

Vickers hardness measurements were carried out with a 5 kgf load (HV5) applying 4 indentations on random areas of the test pieces.

![Figure 1. Thermomechanical cycles applied for the dilatometry tests.](image-url)
2. Results

2.1 Prior Austenite Grains
For the purpose of verifying the prior microstructural condition to the cooling step, the austenite grain size was determined in 3 different stages of thermomechanical cycle 2: AG1, AG2 and AG3. AG1 is shown on Figure 2a and represents the austenite condition after the reheating process and immediately prior to the roughing deformation. The measured grain size was 57 μm. AG2 is displayed on Figure 2b and shows the austenite grains after recrystallization from the first deformation pass and after the holding process, when temperature drops to 850°C. The grain size is reduced to 40 μm. Figure 2c shows the austenite grains after the finishing pass and before the accelerated cooling process starts. The micrograph (Fig. 2c) shows pancaked grains, confirming the absence of austenite recrystallization during the finishing stage.

![Figure 2. Austenite grains after different stages of the dilatometry tests (a) AG1, (b) AG2 and (c) AG3.](image)

Typically, the initial austenite grain size of a reheated slab lies between 150 – 400 μm, which is significantly larger than the one found on the tested sample. However, since dilatometry is uncapable to deliver deformation levels like the actual industrial rolling process, the initial grain size of 57 μm was adequate to promote a final grain size compatible to the actual plate at 40 μm.

2.2 Transformation Temperatures
The start and finish temperatures of austenite transformation are displayed in Table 1. The length variation versus the temperature curves demonstrated only inflexions for the start and finish of a single phase transformation, as displayed on Figure 3. Various studies have showed CCT diagrams where B and AF were separated in different fields, as well as B and PF. The microstructures showed the presence of B, AF and in one particular condition polygonal ferrite (PF).
Figure 3. Length variation of the test pieces during cooling along with the differentiated curve and the start and the finish transformation temperatures. (a, c, e) Refer to thermocycle 1 at cooling rates of 10°C, 20°C and 30°C, respectively. (b, d, f) Refer to thermocycle 2 at cooling rates of 10°C, 20°C and 30°C, respectively.

Table 1. Start and finish temperatures of austenite transformations for each cooling rate and deformation cycle.

<table>
<thead>
<tr>
<th>Cooling Rate (°C/s)</th>
<th>Start Temp.(°C)</th>
<th>Finish Temp. (°C)</th>
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<tbody>
<tr>
<td></td>
<td>Deformation Cycle 1</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>720</td>
<td>498</td>
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<tr>
<td>20</td>
<td>701</td>
<td>481</td>
</tr>
<tr>
<td>30</td>
<td>681</td>
<td>468</td>
</tr>
<tr>
<td></td>
<td>Deformation Cycle 2</td>
<td></td>
</tr>
<tr>
<td>10</td>
<td>732</td>
<td>531</td>
</tr>
<tr>
<td>20</td>
<td>705</td>
<td>484</td>
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<tr>
<td>30</td>
<td>700</td>
<td>454</td>
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</table>
2.3 Microstructural Characterization

The microstructures from cycles 1 and 2 were evaluated by SEM and are depicted in Figure 4a-h. The main microconstituents observed were B (granular and upper bainite) and AF. The exception for this observation was found in the sample cooled at 10°C/s and deformed according to the cycle 2 schedule, where a small amount of PF was identified. Dashed lines were drawn to highlight an estimate of the prior austenite grain boundaries and the red arrows point out examples of the AF grains. The SEM images displayed on Figures 4a-f show the grain size difference between B and AF, with the latter being much finer and presenting ferrite plates with a non-parallel orientation. This feature originates from the intragranular nucleation behavior of AF, which is augmented by the presence of deformation bands. The nucleation of AF at the deformation bands in low carbon steels was evidenced by various studies [16-19], exhibiting similar microstructures as presented in Figure 5. Bai et al. analyzed the effect of deformation and cooling rate on the microstructures of low carbon Nb-B steels. Using TEM micrographs made from the rapidly cooled sample, the authors found that the lengths of the lath ferrite (AF) can attain 20 μm and the width can be as narrow as 0.3 μm. Several individual ferrite laths, also called subunits are parallel and of identical crystallographic orientation to form a packet. These packets grow into the austenite in a specific direction until they meet one another or are interrupted by austenite grain boundaries or other interfaces. There was no evident occurrence of interlath or intralath carbides observed in this steel [19]. Austenitizing conditions, continuous cooling rate, and isothermal quenching time and temperature have a significant influence on acicular formation. However, the controversy on whether displacive-controlled or C atoms diffusion-controlled mechanism during acicular ferrite formation remains [7].

The microstructure revealed granular bainite sites delimited by polygonal grain boundary regions originated from the primary austenite. MA constituents are formed in carbon-enriched regions and their origin is attributed to the partitioning of carbon contained in austenite during its transformation to ferrite. Dispersed MA in the matrix may be related to the rapid diffusion process that occurs after shear. In regions where carbon segregation occurs, mainly in the grain boundaries, the formation of the MA constituent is identified, as shown in Figures 4g and 4h.

Figure 5 shows an optical micrograph of the cycle 2 sample cooled at 30°C/s as an example of the overall microstructure. The micrograph demonstrates the characteristics of AF, granular bainite (GB) and upper bainite (UB).
Figure 4. Cycle 1 at different cooling rates: (a) 10°C/s, (c) 20°C/s and (e) 30°C/s. Cycle 2 at different cooling rates (b) 10°C/s, (d) 20°C/s and (e) 30°C/s. MA morphologies are shown in (g) thermomechanical cycle 1 at 10°C/s cooling rate showing a bainite field associated with elongated MA. (h) thermomechanical cycle 2 at 10°C/s cooling rate showing a region with bainite associated with elongated MA and acicular ferrite associated with equiaxed MA.

Figure 5. Optical micrograph from showing examples of acicular ferrite (AF), granular bainite (GB) and upper bainite (UB). Vickers HV₅ hardness measurements were taken from every sample. For each thermomechanical cycle, faster cooling rates promoted higher hardness, as the microconstituents formed at lower temperatures. When raising austenite deformation from 25% in cycle 1 to 40% in cycle 2, hardness was slightly lower on the latter, although within the margin of error. The exception was at 30°C/s, where the cycle 2 sample displayed 10 HV below the cycle 1 sample. This occurred because at 30°C/s there was the largest difference in the transformation start temperatures between the cycles 1 and 2. Although PF was identified in cycle
2 cooled at 10°C/s, the quantity was insufficient to promote hardness differences from the cycle 1 sample cooled at the same rate.

To confirm if B and AF are forming simultaneously, an interrupted dilatometry test was performed as per thermomechanical cycle 1 at 10°C/s and interrupted by quenching. Based on the transformation start temperature obtained by dilatometry, the test was interrupted at 690°C where some amount of transformation was expected to have taken place. The microstructure was analyzed by optical microscopy and Vickers microhardness tests were taken from different microconstituents, as seen on Figure 6a-c. Three different microconstituents were identified as acicular ferrite (AF), bainite formed before quenching (B1), bainite formed during quenching (B2) and the microhardness tests resulted in 223, 236 and 247 HV100, respectively.

![Figure 6. Micrographs obtained after thermomechanical cycle 1 and interruption at 690°C. (a) indentation aimed at acicular ferrite, (b) Vickers indentation at bainite formed prior to the cooling interruption (B1) and (c) Vickers indentation at bainite formed during the cooling interruption (B2).](image)

### 2.4 Local Misorientation

KAM data were extracted for each sample for a comparison on the level of local misorientation. The variation of the local misorientation is an indication of the level of plastic strain and the dislocation density within the microstructure \(^{20,21}\). In general, KAM angles above 1° characterize deformed grains with high dislocation density, while recrystallized grains are represented by angles below 1° \(^{22}\). Figure 7 shows KAM maps and local misorientation angles distribution, comparing the influence of the deformation schedule on the misorientation for the 3 cooling rates.
Figure 7. (a,d,g) Graphs showing the KAM distribution for each test piece after cooling rates of 10°C/s, 20°C/s and 30°C/s, respectively. Thermomechanical cycle 1 is presented in blue circles and cycle 2 is presented in red squares on the graphs. (b,e,h) KAM maps for cycle 1 in cooling rates of 10°C, 20°C and 30°C/s. (c,f,i) KAM maps for cycle 2 in cooling rates of 10°C, 20°C and 30°C/s.

2.5 Discussion

According to the results exposed on Table 1, the transformation starting temperatures increase for lower cooling rates and also when higher deformation levels below Tnr were applied. The deformation of austenite is responsible for raising the transformation start temperatures, due to the increase in the stored energy. This will promote an increment in the driving force for transformation. The coupling between a higher level of deformation in austenite and lower cooling rate, favored a diffusion based transformation due to an increasing quantity of nucleation sites, enabling the transformation to PF. This result is consistent with the findings from other authors [23, 24]. The length variation versus the temperature curve most likely did not register the PF transformation because the volume fraction of this microconstituent was below the detection limit of the instrument. The transformation to ferrite enables the migration of carbon to the neighboring austenite, promoting the stabilization of the latter and delaying the transformation of to bainite. As the cooling rate increases there is insufficient time for the diffusion of carbon and austenite transforms to bainite.

The bainitic microconstituents in all samples were identified as granular bainite (GB) and upper bainite (UB). These types of bainite can be categorized as BI and BI I, respectively, as per the classification criteria established by Ohmori et al [25]. GB presented irregular grain boundaries and the absence of carbide particles between the bainite sub-units. GB is formed first and the partitioned carbon will stabilize the remaining austenite that will generate second phases at the end of the cooling process, such as degenerated pearlite and MA. The cooling conditions also allowed the formation of UB showing a lath type morphology associated with cementite precipitated from carbon enriched austenite.

The austenite condition prior to the cooling process can play an important role on the nucleation of AF and/or B. As the prior austenite grain size becomes smaller, the restriction to the growth of bainitic displacive transformation intensifies. Additionally, when strain is imposed to austenite, deformation bands and higher dislocation density will further suppress the B laths. In contrast AF is restricted by other adjacently nucleated AF instead of the austenite grain boundaries and the austenite deformation substructures[26]. This leads to AF having small grain sizes in all trans-formed microstructures.

Gourgues et al. studied in detail the acicular ferrite, bainite, and martensite microstructures found in three low al-loy steels. EBSD was used to assess crystallographic features of these microstructures. In each area studied by EBSD mapping,
crystallographic packets defined as clusters of points sharing the same crystallographic orientation were compared with morphological packets observed in the corresponding micrograph, as illustrated in Figures 4a, b, d and h, and Figure 5. Microtexture studies suggested that acicular ferrite and upper bainite grow with Nishiyama–Wassermann relationships with the parent austenite phase, whereas lower bainite and martensite consist of Kurdjumov–Sachs relationships with the parent phase. In summary, EBSD analysis showed that the acicular microstructure consisted of highly intricate, highly misoriented plates having internal low angle boundaries 27).

Studies have shown that increasing the deformation in austenite leads to a maximization of AF 4,26,28). The higher amount of strain applied in cycle 2, however, did not reflect in the increase of the AF volume fraction. A possible explanation for this observation is the precipitation of Nb on the recrystallized austenite during the long period of time between the roughing pass and the finishing pass (Figure 1). As a result, there was not enough Nb in solution to activate the solute drag effect and promote the formation of sufficient intragranular nucleation sites for AF.

The observation of elongated MA constituents, as depicted on Figure 4g, give an indication of the presence of B, whereas equiaxed MA is associated with AF, as indicated within the dashed polygon on Figure 4h. This behavior was observed in other works 29,30). The explanation for this difference in morphology is due to the transformation process, where bainite is formed in an organized manner with parallel lath packages and with similar crystallographic orientations 31), thus the remaining austenite present between the laths will transform in MA with an elongated and coarse morphology. On the other hand, acicular ferrite grows randomly in different directions resulting in an equiaxed and fine MA shape, as presented in Figure 4h. The presence of coarse MA constituent can have a detrimental effect on toughness, therefore, the refinement of MA constituent through austenite deformation can mitigate this negative effect on toughness 32).

The interrupted dilatometry test revealed morphological and mechanical differences between B and AF. The bainite formed in high temperature (B1), prior to the rapid cooling, was characterized as having a granular morphology, exhibiting irregular grains, while the acicular ferrite presented a much finer morphology. Both microconstituents were found to originate simultaneously at 690°C, contrary to what other authors have found in their studies 28), where AF is formed at higher temperatures and/or lower cooling rates than B. After rapid cooling, bainite (B2) was presented as a mixture of upper and lower bainite, having elongated laths accompanied with cementite. Bainite types B1 and B2 were identified in the continuously cooled samples by the SEM micrographs and KAM maps.

The KAM maps shown on Figure 7 revealed that cycle 1, in general, presented higher local misorientation angles as the cooling rate increased, this implying on the formation of microconstituents with higher strain and dislocation levels when the start of transformation occurred at lower temperatures. This suggests that the predominant kind of bainite found on the continuously cooled samples were the B2 type. Cycle 2, however, presented higher KAM levels when cooled at 20°C/s. The KAM distribution demonstrated a shift from the highest frequencies towards lower local misorientations when the deformation increased from 25% in cycle 1 to 40% in cycle 2 at all cooling rates (Figures 7a, d and g). This shift was less prominent for the samples cooled at 20°C/s because the transformation temperatures were practically the same for both cycles, as shown on Table 1. As mentioned previously, at 30°C/s the dilatometry test registered the largest difference between the transformation temperatures in cycles 1 and 2. The lower hardness found on cycle 2 for this cooling rate agrees with the KAM results, as shown on Figure 7g.

The KAM distribution at 10°C/s in Figure 6a showed a high frequency for low misorientation angles when the deformation below Tnr increased (cycle 2). This result shows that the combination of lower cooling rate and higher austenite deformation favored the formation of low dislocation density microconstituents, enabling some degree of nucleation via diffusion, such as PF. The presence of PF in cycle 2 caused strain relief, lowering the peak frequency to 0.15°. This observation agrees with the microstructure showed on Figure 4b.

CONCLUSIONS

Dilatometric tests for continuous and interrupted cooling schedules, paired with OM and SEM images, along with microhardness measurements, demonstrated the simultaneous formation of AF and B. Additionally, the precipitation of Nb on the recrystallized austenite during the long period of time between the roughing pass and the finishing pass may have hindered the formation of favorable intragranular nucleation sites for AF after the second deformation pass was applied. These findings suggest that maximizing the volume fraction of AF is an inefficient route for improving toughness in the industrial production of this type of steel. In this scenario other approaches should be pursued for improving the steel mechanical performance.

The KAM maps and local misorientation distributions revealed a decrease in the internal strain as the percentage of deformation in the austenite increased from 25% to 40%, leading to the identification of polygonal ferrite at the lowest cooling rate. This observation, along with the hardness results, suggests that cycle 1 is favorable for specifications where toughness requirements are less stringent and tensile properties are the main concern.
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