The microstructural description in bainitic steel is commonly ambiguous, and the interpretations of results that originate from applied methods are usually user dependent. In consequence, a manifold description of bainite makes it difficult to reveal structure–property relationships. This is why a novel classification and quantification routine for bainitic microstructures in wire rod steel is presented. The classification is based on electron probe microanalysis (EPMA), electron backscatter diffraction (EBSD), and nanohardness of the same local area. Microstructural constituents with different characteristics (carbon concentration, misorientation, and nanohardness) are classified into low-, intermediate-, and high-temperature morphologies. After the classification, quantification is conducted by combining scanning electron microscopy (SEM) analysis, dilatometry, and X-ray diffraction (XRD). The bainite quantification reveals a homogeneous microstructure for cooling along the lower bainite regime. Increasing the manganese content causes a lower sensitivity to change in the cooling parameters. Combining nanohardness and EBSD is suitable to describe microscopic microstructural properties, whereas the quantification of bainite is eligible to explain differences in the microscopic microstructural properties. The used approach avoids overcomplexity and manifold terminologies of bainitic microstructures that are commonly found in the literature.

1. Introduction

Bainite is known as probably the most complex microstructure in steel with similarities to other microstructures depending on the transformation temperature and the alloying concept. A simplified classification of bainite was introduced by Takahashi and Bhadeshia by differentiating lower from upper bainite based on the occurrence of intralath or interlath cementite.[1] However, the development of alloying concepts and new processing routes caused more varieties in the bainitic microstructure. In need of a new classification approach, Zajac categorized different types of bainite according to the misorientation angle at the phase boundary.[2,3] In the last decades, the different appearances of bainite have caused a vast list of terminologies. Bramfitt and Speer compiled a list of 30 terms extracted from various publications, all referring to different types of bainite.[4] Since then, the list can be extended by further terms to describe bainite.[5–9] One conclusion of the previous work is that the classification of different bainite microstructures depends highly on the expert and therefore causes ambiguous interpretations.

Another trend that has evolved in recent years is the increasing application of high-resolution techniques with results reaching to an atomic level. It is true that high-resolution methods (such as transmission electron microscopy (TEM), atom probe tomography (APT), and atom force microscopy (AFM)) provide useful information at the nanometer scale. But the localized information cannot always be generalized and applied to the bulk to an atomic level. It is true that high-resolution methods (such as transmission electron microscopy (TEM), atom probe tomography (APT), and atom force microscopy (AFM)) provide useful information at the nanometer scale. But the localized information cannot always be generalized and applied to the bulk.
material. For instance, continuous cooled wire rod steel is commonly exposed to external cooling effects (by air or forced air cooling in a Stelmor conveyor)\[19\]. This causes a temperature gradient in the radial direction from the core to the outer surface depending on several factors, for example, the wire rod diameter and the ring density on the cooling conveyor\[17\]. Consequently, a microstructural and hardness gradient can be observed in the same direction. In this case, the experimental setup requires adjustments to deliver more representative results, instead of relying only on high-resolution techniques.

In this work, different methods are combined to get a more representative analysis of bainite in wire rod steel. The applied alloying concept, referred to as carbide-free bainite,\[12-14\] yields an incomplete transformation for the applied cooling parameters with retained austenite as secondary phase. This alloying concept is known for an outstanding combination of strength and ductility.\[5\] The retained austenite occurs either as films separating bainitic lath or as blocks between sheaves of bainite.\[15\] At room temperature, the secondary phase is meta-stable. This means that external events can trigger a transformation of austenite to martensite, known as the transformation-induced plasticity (TRIP) effect.\[16\] Austenite blocks commonly inherit a carbon gradient toward the core. Cooling below the \(M_s\) temperature partly transforms these blocks into martensite, while the outer rim remains austenitic to some extent.\[13\] Therefore, the mixture of martensite and austenite is commonly referred to as a martensite–austenite (M–A) island.\[17\]

2. Experimental Section

2.1. Laboratory Melts

In total, three microalloyed steels were produced as laboratory melts alloyed with substantial differences in manganese content (Table 1). The steels contained a medium carbon content with 0.19–0.23 and >1 wt% Si to delay carbide precipitation in austenite.\[18\] Manganese of 1.5–2.5 wt% was used to adjust the hardenability.\[19\] This concept is commonly applied to generate carbide-free bainite. A combined utilization of boron and titanium delays the ferrite/pearlite transformation to widen the process window for bainite.\[20\] Molybdenum has the same effect with delaying the ferrite/pearlite transformation to widen the process window for bainite.\[20\] Consequently, a microstructural and hardness gradient can be observed in the same direction. In this case, the experimental setup requires adjustments to deliver more representative results, instead of relying only on high-resolution techniques.

In this work, different methods are combined to get a more representative analysis of bainite in wire rod steel. The applied alloying concept, referred to as carbide-free bainite,\[12-14\] yields an incomplete transformation for the applied cooling parameters with retained austenite as secondary phase. This alloying concept is known for an outstanding combination of strength and ductility.\[5\] The retained austenite occurs either as films separating bainitic lath or as blocks between sheaves of bainite.\[15\] At room temperature, the secondary phase is meta-stable. This means that external events can trigger a transformation of austenite to martensite, known as the transformation-induced plasticity (TRIP) effect.\[16\] Austenite blocks commonly inherit a carbon gradient toward the core. Cooling below the \(M_s\) temperature partly transforms these blocks into martensite, while the outer rim remains austenitic to some extent.\[13\] Therefore, the mixture of martensite and austenite is commonly referred to as a martensite–austenite (M–A) island.\[17\]

2.2. Thermomechanical Treatment

All the samples passed through the same austenitization treatment with initial heating of 3 K s\(^{-1}\) to 1200 °C. This temperature was held for 10 min until cooling with 1 K s\(^{-1}\) to a hot deformation step at 900 °C that initiated compression of the sample (\(\varphi = 0.3\), \(\dot{\varphi} = 10\) s\(^{-1}\)). In the following, three different cooling regimes according to the cooling conveyor in a wire rod mill were defined to generate different mixtures of bainite. The cooling parameters in regime I consisted of fast initial cooling with 5 K s\(^{-1}\) to 400 °C and subsequent cooling in the bainite phase field of 0.3 K s\(^{-1}\). Slower cooling of 2 K s\(^{-1}\) from 900 to 500 °C with simulated air cooling in the bainite phase field of 1 K s\(^{-1}\) is denoted as regime II. Regime III was cooled as regime II, but the cooling rate in the bainite phase field was reduced to 0.3 K s\(^{-1}\). Cooling rates of regimes I–III were confirmed for feasibility by prior tests on the cooling conveyor of the wire rod mill at ArcelorMittal Duisburg Long Products. A hot deformation step at 900 °C simulated the final rolling step but it has to be noted that actual area reduction and deformation rate during processing in a wire rod mill are beyond the limits of the TTS in the laboratory. For instance, a lower degree of deformation and a lower rate of deformation in the TTS are expected to promote a larger prior austenite grain size relative to industrial processing.

2.3. Applied Methods for Bainite Classification

The quantification of phase fractions in the three steels was conducted in two steps: an identification (classification) phase and a quantification step (Figure 1). In the first step, morphological features were described by a comprehensive analysis of different experiments to extract information in the same local area. The heat-treated samples were further processed to extract secondary samples for metallographic preparation and mechanical testing. Samples were ground and polished to a surface finish of 1 μm. Oxide polishing suspension (OPS) made of colloidal silica with a 0.25 μm particle size was used to obtain a surface with minimal roughness. This final step was chosen instead of electropolishing to satisfy the surface requirements of electron probe microanalysis (EPMA) and nanohardness measurements despite a small risk of a possible TRIP effect initiated by mechanical preparation. Microhardness indenters were used as markers to locate the same spot for the different experiments.

EPMA measurements in a Schottky field-emission gun electron microprobe, JEOL JXA-8530 F (JEOL Ltd., Tokyo, Japan),
revealed segregation of alloying elements during cooling with the predefined regimes. This method reveals, for example, carbides or stable retained austenite films in regions that incorporate high carbon concentrations. The instrument was operated at 15 kV accelerating voltage and at a beam current of 100 nA. Map and line scans were recorded with a step size of 100 and 300 nm, respectively. Hydrocarbon cracking adsorbed on the surface of the sample is known to cause carbon contamination of the measured surface area. This problem was minimized by conducting an acquisition procedure prior to the measurements to obtain reliable carbon concentrations.\(^{[22]}\)

Subsequent electron backscatter diffraction (EBSD) measurements provided crystallographic information of the morphological features. The experiment was conducted at 20 kV and a step size of 80 nm in a field emission scanning electron microscope (type Zeiss Sigma) with an EBSD detector.

Nanoindentation was performed with a Berkovich diamond tip in an iNano nanoindenter (NanoMechanics Inc., TN), with the area function of the tip calibrated prior to indentation on fused silica.\(^{[23]}\) The area of interest was probed by a \(10 \times 10\) grid (in total 100 indents) with 10 \(\mu\)m indentation spacing. The experiments were conducted to a target depth of 100 nm in all cases such that the plastic zones of the individual indents did not overlap. The results of these three methods—EPMA, EBSD, and nanoindentation—were combined to elaborate a classification scheme.

In the second step, the classification system was applied as guideline for the scanning electron microscopy (SEM) analysis. Instead of marking phases on a pattern of lines, phases were cut out by polygons to also account for the space that was otherwise missed in between the lines of the line intercept method. For the SEM analysis, samples were etched with 2\% nitric acid solution (nital) to reveal the microstructure. For all samples, a central spot on the cross-section of the TTS samples was probed under SEM.

For each condition, the metallographic procedure was repeated, with electropolishing in A2 solution (40 V for 10 s) as a surface finish, instead of using OPS. These samples were analyzed in X-ray diffraction (XRD) under Co K\(\alpha1\) radiation to obtain the fraction of retained austenite. The samples were rotated along both the \(\phi\) and \(\psi\) axes to minimize texture effects on the phase quantification.

In addition, the cooling regimes were applied in dilatometer experiments. The length change–temperature data were transformed via the lever rule into a transformed fraction over temperature plots. This provided the transformation start and finish temperature. The resulting data were combined with the phase fractions obtained from the SEM analysis. Ultimately, the phase fractions can be described by the temperature range of the phase transformation.

2.4. Mechanical Testing

The TTS samples were processed by electrical discharge machining (EDM) to obtain secondary samples for tensile and Charpy V-notch testing. For the tensile test, two cylindrical specimens were
machined per condition with a cylindrical shape of 5 × 25 mm² between the screw threads. The test was conducted using the tensile test machine Zwick 4204 at a constant speed of 0.4 mm min⁻¹ (strain rate of 0.00025 s⁻¹).

From another TTS sample per condition, three subsized Charpy V-notch samples were machined with a rectangular cross-section of 2.5 × 10 mm² and 55 mm in length. Both tests were conducted at room temperature.

3. Results

A combination of carbon map (EPMA), force-depth behavior (nanohardness), band contrast, and misorientation boundaries (EBSD) was used for steel 1 in different cooling regimes.

Regime III produces a high degree of inhomogeneity. Therefore, a larger surface area was analyzed to attain more representative results (Figure 2). In the granular-type morphology, large areas of carbon depletion are visible, whereas a uniform carbon distribution can be observed in the shape of islands with a large scattering in size. Again, the band contrast reveals a link of highly distorted areas and high carbon concentration. Larger islands with high carbon concentration contain a carbon gradient from the border to the core of the island. The grain boundaries show only a few high-angle boundaries but dominantly low-angle boundaries. Regime I and regime II were analyzed accordingly (for the images, the reader is referred to the Supported Information of the online version).

In regime I, carbon-enriched areas were observed along carbon-depleted lath, while some grains showed a homogeneous carbon distribution. The carbon-enriched areas coincide with regions of low distortion (dark gray scale values in the band contrast). Otherwise, carbon depletion can be linked to light gray scale values and therefore, microstructural constituents with low distortion. High-angle boundaries are prevalent in regime I. Consequently, the microstructure of regime I contains a homogeneous lath-type structure with a small fraction of highly distorted areas, typical for morphologies transformed at lower temperatures (i.e., fresh martensite).

In regime II, the carbon map reveals areas of carbon separation by diffusion and regions of uniformly distributed carbon. The former corresponds to areas of low distortion in the band contrast with embedded block-type retained austenite with high carbon concentration. Areas of uniform carbon distribution appear in dark gray scale values as an indicator for transformation products that originated from low temperatures. The coarser carbon-depleted areas result in an increase of the number of low-angle boundaries at the cost of high-angle boundaries.

In addition to EPMA maps, line scans were obtained to monitor the elemental changes across phase boundaries for steel 1 in regimes I–III (Figure 3). In regime I, the carbon distribution corresponds either to moderate fluctuations or regions of high carbon concentration with high distortions. Regime II contains in addition carbon-depleted areas next to high-carbon areas with up to 0.6 wt% C. Alternating carbon depletion and high carbon concentration can be observed in regime III, with maximum concentrations of up to 0.9 wt% C. For other alloying elements, no diffusion can be identified from the EPMA results, as it can be seen from regime II.

Nanohardness in regime I reflects the homogenous carbon distribution by a narrow hardness distribution with a mean hardness and standard deviation of 4.44 ± 0.74 GPa (Figure 4).

Figure 2. Left: Carbon distribution by EPMA of steel 1 in regime III (1-500-0.3) shows strong localized carbon concentrations surrounded by carbon-free areas. Regions of elevated carbon with boundaries of high carbon indicate M–A islands. The same area was probed by nanohardness with a 10 × 10 grid of indentations (indicated by red arrows). The red rectangle marks the region of the EBSD measurement. Upper right: The band contrast of this area represents a low degree of distortion (light gray values) in the carbon-depleted areas (retained austenite is colored in green). Lower right: The same region obtains a high amount of low-angle boundaries in carbon-depleted areas and only a few high-angle boundaries (in red) in areas of uniform carbon distribution. A larger area was analyzed due to the strong microstructural inhomogeneity.
For regimes II and III, peak broadening can be detected with 4.69 ± 1.16 GPa in regime II and 4.55 ± 1.47 GPa in regime III. The right column of Figure 4 shows examples of microstructural categories (with fcc austenite in green) overlapped with the corresponding nanohardness indent. The indents are classified into different microstructural constituents for steel 1, separated after the cooling regime. The lowest hardness was measured for bainitic ferrite, in regimes II and III, with a corresponding mean value of 2.64 ± 0.81 GPa for regime III. The absence of granular bainite in regime I results in a higher hardness for carbon-supersaturated lath of bainitic ferrite with a mean value of 3.88 ± 0.04 GPa. Dark gray scale values in the band contrast represent fresh martensite. These regions yield a high nanohardness above 5 GPa, as it can be seen for regime III with 5.53 ± 0.52 GPa. Retained austenite as block or film contributes to an intermediate range of hardness. During loading, a “pop-in” in the early stage of loading is seen as the result of the homogeneous nucleation of dislocations under the indenter tip when no sources exist due to a damage-free surface (the interpretation of pop-ins will be further specified in the discussion). During unloading, an “elbow” behavior can be observed. In the literature, “pop-out” or elbow behavior can be correlated with an expansion of volume below the indenter tip due to a phase transition. In case of an extensive or very rapid expansion, pop-out is commonly seen for high loading rates and high maximum loads, while for a lower degree of expansion, rather an elbow-like behavior can be observed for low loading rates and lower maximum loads during indenter release. Pop-outs and elbows are both signs of indentation-induced transformation. Pop-in behavior during loading was observed in retained austenite of steel 1 in regime I (Figure 5). Another indent that coincided with retained austenite yielded a gradient change of slope during unloading (elbow behavior).

Additional information can be extracted by the spatial correlation of nanohardness with the observed morphology. It becomes clear that dark gray scale values correspond to martensite and...
Figure 4. Left: Broadening of nanohardness distribution in b) regime II and c) III based on steel 1, whereas nanohardness values of a) regime I are observed in a narrow hardness distribution (each hardness distribution was fitted with a lognormal distribution function). Right: Nanohardness as a red triangle with mean value (±standard deviation) of selected microstructural constituents according to (a–c) in steel 1 after cooling in different regimes (hardness of martensite in regime I is based only on one measurement). Green regions indicate fcc austenite; dark areas indicate highly distorted martensite (scale bar applies for all images; red, blue, and black will be used again in Figure 5 to assign the three microstructural constituents).

Figure 5. Load–displacement behavior of selected morphologies. “pop-ins” during loading in the vicinity of retained austenite and martensite represent homogeneous nucleation of dislocations. For instance, a) block-type retained austenite in regime I as a solid line indicates homogenous nucleation of dislocations during loading. An “elbow” during unloading is caused by phase transformation. For instance, elongated retained austenite in regime I as dashed line during unloading causes volume expansion below the indenter probably due to martensite transformation. A high scattering of load–displacement behavior was observed for b) regime II and c) III.
bright gray values to bainitic ferrite. In addition to retained austenite, the former shows occasionally an elbow behavior as an indicator of martensite transformation. Only bainitic ferrite shows an absence of such discontinuities. Furthermore, cooling regimes II and III resulted in a significant scatter of load-displacement curves in each microstructural class, whereas the curves almost overlap in regime I.

The previous findings were used to elaborate a classification scheme. Afterward, the classification was applied on steel 1 in different regimes and on three different steels with different hardenability levels. For the quantification of microstructures,
three classes are defined as follows: 1) high-temperature, 2) intermediate-temperature, and 3) low-temperature morphologies, according to differences in the morphology, carbon distribution, distortion, and hardness (Table 2).

In addition, dilatometer experiments were conducted to track the evolving phase transformation after interrupting the cooling at different temperatures. In the SEM micrographs after interrupting the cooling of regime II (Figure 6), these regions can be differentiated by the etching response. No or low etching response corresponds to low-temperature morphologies, whereas coarse regions with pronounced topographic effect relate to the first class. In case of a clearly visible lath shape, the region is marked as intermediate morphology.

The results of the phase fraction analysis for different cooling conditions and three different steel compositions are shown in Figure 7. Each pie chart contains the retained austenite fraction obtained from XRD. The fraction of retained austenite lies in the range of 3.6% in regime II of steel 3 and 14.7% for regime III of steel 1. A change in cooling regime causes from regime I to regime III an increase in high-temperature morphologies (in brown) on the cost of intermediate morphologies. Regime III displays a mixture of low- and high-temperature morphologies with a low fraction of intermediate morphologies.

The effect of manganese was analyzed in regime I and II. In both regimes, the fraction of low-temperature morphologies (in blue) increases. This increase in regime I reduces the amount

Figure 8. a–c) Effect of cooling regime in steel shows primarily intermediate-temperature morphologies in regime I (0.3 K s⁻¹ from 400°C), whereas in regime II (1 K s⁻¹ from 500°C) and regime III (0.3 K s⁻¹ from 500°C) high-temperature morphologies dominate the microstructural composition. d–f) Increasing the manganese content from 1.5 to 2 and 2.5 wt% causes in regime II higher fractions of low-temperature morphologies, but at the cost of high-temperature morphologies. The same tendency was observed in regime I.
of intermediate-temperature morphologies, where no high-temperature morphologies are observed. In regime II, the rise of blue fractions is more severe and diminishes high-temperature morphologies.

The phase fractions are assigned to the dilatometer data (transformed fraction over temperature) for cooling in regimes I–III, as shown in Figure 8a–c. The curves were obtained from the length change data via the lever rule. Cooling in regime I generates a homogeneous microstructure originated from a narrow temperature range. At higher transformation temperatures, the microstructure is composed of different morphologies formed at a broader temperature range. A steep transition of high- to low-temperature morphologies can be seen in regime III. It has to be noted that the phase fractions add up to 100%, indicated by the y-axis. But the diagram refers to the transformation start (0%) and finish temperatures (100%) in the shown temperature range. Retained austenite is not considered in the curve as it is assumed stable at room temperature. Thus, the overall phase fractions are reduced by the amount of retained austenite.

The effect of hardenability by increased manganese content is plotted in Figure 8d–f. The fractions of high- and intermediate-temperature morphologies (in green and blue) reduces and diminishes at 2.5 wt% Mn in steel 3. For this steel, regimes I and II visually almost overlap with a high fraction of low-temperature morphologies. In regime I, increasing the manganese content correlates to increased fractions of low-temperature morphologies on the cost of intermediate-temperature morphologies.

Table 3 provides an overview of transformed volume fractions and the according transformation start temperatures. The effect of manganese in regime I in steel 1, 2, and 3 shows a clear decrease of intermediate start temperatures, while the low-temperature morphologies begin to transform in the same range of $\approx 370^\circ C$. The effect of manganese is more severe in regime II by decreasing the transformation temperatures significantly and by shifting the transformation curve to lower temperatures.

The results of mechanical testing in different cooling regimes of steel 1 are summarized in Table 4. Regime I stands out with high strength of 953 MPa [yield stress (YS)] and 1274 MPa [ultimate tensile strength (UTS)]. This causes a high yield ratio of 0.75. In addition, regime I yields the highest impact energy among the tested conditions. Regimes II and III cause an early onset of plastic deformation and lower tensile strength and a deterioration of impact energy. The ductility can be increased by regime III with 11.6% uniform elongation (UEl) and 14.7% total elongation (TEl), respectively.

The mechanical properties of steel 2 and 3 with different manganese contents are separated into regimes I and II. In the former, the increase of manganese causes higher strength but lower impact energy. Uniform elongation and total elongation are increased in steel 2, while a manganese content of 2.5 wt% causes a deterioration of ductility. In regime II, a broader range in yield stress (YS: 785–1035 MPa) and tensile strength (UTS: 1157–1552 MPa) can be observed. Elongation values appear again higher in steel 2, whereas the impact energy increases slightly with increasing manganese content.

## 4. Discussion

### 4.1. Microstructure–Property Relationship

The impact of the phase fractions on the mechanical properties at room temperature depends on the cooling parameters and the chemical composition.

### Table 3. Bainite fractions in steel 1 (regimes I–III), steel 2 (regimes I–II), and steel 3 (regimes I–II) with transformation start temperatures of high-, intermediate-, and low-temperature morphologies (last column). In regime I, two classes can be observed, while high-temperature morphologies occur in regime II. An increase of manganese causes lower transformation temperatures, with a more severe effect in regime II than in regime I.

| Regime | Low [T] [%] | Intermediate [T] [%] | High [T] [%] | Bs [°C]:low-intermediate–high |
|--------|------------|---------------------|-------------|-------------------------------|
| Steel 1 I | 4.9 | 95.1 | – | 373–487 (–) |
| II | 31.1 | 16.4 | 52.5 | 390–414–528 |
| III | 30.0 | 9.2 | 60.8 | 448–459–517 |
| Steel 2 I | 8.1 | 91.9 | – | 369–395 (–) |
| Steel 3 | 80.8 | 19.2 | – | 371–384 (–) |
| Steel 2 II | 85.8 | 9.5 | 4.7 | 401–435–469 |
| Steel 3 | 96.7 | 1.4 | 1.9 | 368–379–394 |

### Table 4. Mechanical properties according to cooling regime of steel 1 and according to manganese content of steel 2 and 3 for cooling regimes I and II.

| Regime | Yield strength [MPa] | Tensile strength [MPa] | Yield ratio [-] | UEl [%] | TEI [%] | CIV [J] |
|--------|---------------------|-----------------------|----------------|-------|--------|--------|
| Steel 1 (1.5 wt% Mn) | | | | | | |
| I | 953 | 1274 | 0.75 | 3.8 | 12.2 | 21 |
| II | 785 | 1157 | 0.68 | 5.1 | 7.5 | 9 |
| III | 709 | 1133 | 0.63 | 11.6 | 14.7 | 5 |
| Steel 2 (2.0 wt% Mn) | | | | | | |
| I | 992 | 1394 | 0.71 | 4.8 | 15.5 | 18 |
| Steel 3 (2.5 wt% Mn) | | | | | | |
| I | 1068 | 1486 | 0.72 | 3.0 | 6.4 | 16 |
| II | 989 | 1444 | 0.68 | 3.0 | 8.7 | 14 |
| Steel 3 (2.5 wt% Mn) | | | | | | |
| I | 1035 | 1552 | 0.67 | 4.3 | 7.8 | 15 |

$^{4}$UEI, TEI, and CIV indicate uniform elongation, total elongation, and Charpy V-notch toughness (at room temperature), respectively. CIV values are based on the average of three subsized samples.
4.1.1. Effect of Cooling Regime

Slow cooling in the lower bainite phase field beginning from 400 °C (regime I) produces primarily one type of bainite transformed in a narrow temperature range with 4% low-temperature morphologies. The low amount of brittle morphologies surrounded by a lath-shape morphology with high-angle misorientations causes a high yield point during tensile testing. The absence of granular morphologies in this regime provides a relatively high tensile strength. In this context, regime I showed the highest impact energy among the tested steels and the EBSD results revealed a high degree of high-angle misorientations as effective barriers against crack propagation.[6]

At higher temperatures, a simulated air-cooling route beginning from 500 °C (regime II) produces a broader temperature window and subsequently a mixture of morphologies dominated by coarse high-temperature morphologies. This type of bainite contains coarse bainitic ferrite with low-angle boundaries. The low strength of this particular morphology causes deterioration of the yield stress. Together with a decrease of tensile strength, a lower yield ratio of 0.68 was observed, as an indicator of an increasingly inhomogeneous microstructure. The mixture of morphologies seems disadvantageous for ductility. Compared to regime I, this regime obtains slightly more retained austenite but the increase of low-temperature morphologies with uniform carbon distribution provides less deformability during tensile testing. The high degree of coarse morphologies with low-angle boundaries can be linked to low resistance against impact loads.

A further decrease of the cooling rate in the upper bainite phase field at 500 °C (regime III) introduces more retained austenite (in agreement with the retained austenite measurements in Table 5) and high-temperature morphologies at the cost of the lath-type bainite from intermediate temperatures. With a high amount of granular bainitic ferrite, yielding already occurs at 700 MPa. This microstructure represents the highest degree of observed microstructural inhomogeneity, which manifests in a low yield ratio of 0.63. The broad scattering of nanohardness confirms this tendency. The lack of intermediate-temperature morphologies yields a hardness gradient from low-temperature to high-temperature constituents. Under loading, this heterogeneity appears critical for crack nucleation. Moreover, the observed low-angle misorientations are responsible for the deterioration of impact energy. On the other hand, regime III benefits from a high degree of soft bainitic ferrite and retained austenite to provide better ductility.

4.1.2. Effect of Manganese Content

The hardenability level can be controlled by the manganese content. In regime I, the increase of manganese from 1.5 to 2.5 wt% produces a microstructure dominated by low-temperature morphologies. The high nanohardness in this microstructural constituent correlates to a higher overall strength (UTS = 1486 MPa). In contrast, the microstructure is not suitable to provide high ductility and impact toughness. The former can be explained by a lack of softer phases with a reduced fraction of retained austenite and bainitic ferrite. Low temperature morphologies such as fresh martensite can be regarded as brittle and therefore, the impact toughness deteriorates.

With increasing manganese content, the difference in properties from regime I to II becomes less significant. For instance, the difference in yield strength between regime I and II for steel 1 is significantly higher, with 168 MPa compared to 33 MPa in steel 3. This makes steel 3 less sensitive to changes in the cooling parameters. Thus, steel 3 is more suitable for larger wire diameters, which are usually prone to axial temperature gradients and microstructural heterogeneities.

4.2. Comparison of Characterization Methods

Different methods were used to characterize the microstructure of wire rod steels. Each method contributes differently to the final phase quantification. For instance, in high-temperature morphologies, carbon maps by EPMA provide a clear distribution of carbon on the sample surface in addition to the precise measurements of carbon concentrations via line scans of carbon. In contrast, transformation at lower temperatures causes lath-type morphologies with a lath width on the submicron level. This exceeds the resolution of EPMA to resolve fine differences in carbon distribution. On the other hand, EPMA records line scans that can be correlated with a misorientation analysis by EBSD. Although the absolute carbon concentration in lath-type regions seems underestimated due to an overlap of bainitic ferrite and retained austenite films, EPMA is a powerful tool in extension to the averaged information of carbon concentration (provided by XRD).

Nanohardness is able to obtain mechanical properties of a single microstructural morphology. TEM observation in silicon steels proved that the elbows and pop-outs occurring during unloading can be linked to a phase transformation.[25] The volume expansion by transformation causes an uplift of the indenter tip. In case of pop-out, this uplift takes place abruptly, promoted by higher maximum loads and increased (un-)loading rates. Otherwise the elbow-like gradual change in the unloading curve is the result of a phase transformation with a lower volume expansion, for example, at lower maximum loads and loading rates. And in fact, the discontinuities were observed in the

---

Table 5. Retained austenite (RA) fraction, lattice constant (a_i) and carbon concentration (C_f) in steel 1 (regimes I–III), steel 2 (regimes I–II), and steel 3 (regimes I–II) obtained by XRD measurements. Cooling in regime III yields a high fraction of retained austenite with lower stability (low C_f), while an increase of manganese decreases the fraction of retained austenite and C_f. The carbon concentration was calculated according to the equation after Dyson and Holmes.[19]

| Steel  | RA fraction [%] | a_i [Å] | C_f [wt%] |
|-------|----------------|---------|-----------|
| Steel 1 |         |        |           |
| I      |  8.9 | 3.6166 |  1.2     |
| II     | 10.1 | 3.6119 |  1.0     |
| III    | 14.7 | 3.6078 |  0.9     |
| Steel 2 |         |        |           |
| I      |  8.2 | 3.6100 |  1.0     |
| II     |  7.5 | 3.6071 |  0.7     |
| Steel 3 |         |        |           |
| I      |  5.6 | 3.6041 |  0.8     |
| II     |  6.4 | 3.6028 |  0.8     |
| Steel 3 |         |        |           |
| I      |  5.6 | 3.6041 |  0.8     |
vicinity of martensite and the retained austenite—i.e., where a TRIP effect would be expected—and not near the bainitic ferrite. Discontinuities are seen in the indentation load–displacement curves both in the loading data (as pop-ins) and in the unloading data (as elbows). In the literature, pop-ins during the testing of similar steels have been linked to a phase change via a TRIP effect,[26–28] and in some cases this has also been confirmed by TEM.[27,28] However, such pop-ins are also commonly associated with the homogenous nucleation of dislocations underneath the sharp indenter tip,[29–31] and therefore in this work it cannot be unambiguously said to which mechanism these features belong. However, the additional “elbow-ing” in the unloading curve has also been shown to be associated with a phase transformation.[29,32] This, in conjunction with the fact that these discontinuities were observed in the vicinity of martensite and the retained austenite—i.e., where a TRIP effect would be expected—and not near the bainitic ferrite, therefore, strongly implies they are a result of a TRIP effect also occurring here. A more pronounced elbow was observed in the martensitic morphology of regime III compared to the retained austenite. Thus, it can be assumed that retained austenite facilitates dislocation movements during phase transformation, which can be seen by a damped elbow in contrast to an indentation-induced transformation with adjacent martensite. In summary, nanohardness is a powerful tool to gain further insights into the microstructural homogeneity, given a sufficient number of indents. But the full potential of nanohardness is reached by coupling this technique with EBSD and EPMA. The local information of the local carbon concentration, nanohardness, and misorientation provides complementary information on the microstructural constituents.

5. Conclusion

The carbide-free bainite concept was tested on three steels with different manganese contents and three different cooling regimes, according to the process window of a wire rod cooling conveyor. Different characterization methods were combined to reveal the effect of varied cooling parameters and hardenability level on the microstructure. In summary, the findings are as follows. 1) The change of cooling regime causes significant differences in the bainite morphology. The comprehensive use of EBSD, EPMA, and nanohardness is suitable to monitor and categorize these differences. Merging the results of the adjusted line-intercept method, XRD, and dilatometry yields a quantification of the microstructural features. 2) A classification based on EPMA or nanohardness alone is not sufficient. Only a combination of these techniques with EBSD provides enough information for classifying bainite in wire rod steel. The results were used to classify the microstructure into three classes: low-, intermediate-, and high-temperature morphologies. 3) The cooling rate in the bainite phase field has an impact on the homogeneity of the final microstructure. At 400 °C, a relatively low cooling rate to 0.3 K s⁻¹ causes a homogeneous microstructure primarily composed of intermediate-temperature morphologies. In contrast, simulated air cooling of 1 K s⁻¹ at 500 °C causes a mixture of all three microstructural classes. Cooling from 500 °C with 0.3 K s⁻¹ develops a granular microstructure with primarily low- and high-temperature morphologies with a steep hardness gradient. 4) Nanohardness measurements are suitable to reveal microscopic properties of microstructural features. The retained austenite shows linkages to elbows in the load–displacement curves during unloading as an indicator of martensite transformation beneath the indenter tip, whereas pop-ins during loading cannot be unambiguously interpreted. 5) Regarding the macroscopic mechanical properties, high-temperature morphologies contain relatively high amounts of ferrite and retained austenite, which in turn have a beneficial effect on ductility. A steep hardness gradient for a mixture of low- and high-temperature morphologies facilitates crack propagation and thus, yields low impact energies. 6) An increase of manganese content from 1.5 to 2.5 wt% has an impact on the resulting microstructure and properties. For 2.5 wt% Mn, changes in the cooling regime become more negligible. Thus, manganese causes a lower sensitivity to the cooling regime. This makes higher manganese contents attractive for larger wire diameters, which are prone to temperature gradients in axial direction during cooling.

Acknowledgements

The work was done in the framework of a collaboration project with ArcelorMittal Maizières, Research and Development Bars and Wires, as part of the knowledge-building program at ArcelorMittal. Open access funding enabled and organized by Projekt DEAL.

Conflict of Interest

The authors declare no conflict of interest.

Keywords

carbide-free bainite, classification, microstructures, wire rod steel

Received: August 20, 2020
Revised: September 23, 2020
Published online: October 25, 2020

[1] M. Takahashi, H. K. D. H. Bhadeshia, Mater. Sci. Technol. 1990, 6, 592.
[2] S. Zajac, S. Komenda, P. Morris, P. Diericks, S. Matera, F. Penalba Diaz. Quantitative Structure–Property Relationships for Complex Bainitic Microstructures. Final Report. EUR Technical Steel Research – Physical Metallurgy and Design of New Generic Steel Grades EUR-21245-EN, Luxembourg, 2005.
[3] S. Zajac, V. Schwinn, K. H. Tacke, Mater. Sci. Forum 2005, 500, 387.
[4] B. L. Bramfitt, J. G. Speer, Metall Trans A 1990, 21, 817.
[5] K.-i. Sugimoto, T. Iida, J. Sakaguchi, T. Kashima, ISIJ Int. 2000, 40, 902.
[6] F. G. Caballero, H. Roelofs, S. Hasler, C. Capdevila, J. Chao, J. Cornide, C. Garcia-Mateo, Mater. Sci. Technol. 2013, 28, 195.
[7] F. G. Caballero, H. K. D. H. Bhadeshia, Curr. Opin. Solid State Mater. Sci. 2004, 8, 251.
[8] X. Y. Long, F. C. Zhang, J. Kang, B. Lv, X. B. Shi, Mater. Sci. Eng. A 2014, 594, 344.
[9] T. Sourmail, C. Garcia-Mateo, F. Caballero, L. Morales-Rivas, R. Rementeria, M. Kuntz, Metals 2017, 7, 31.
[10] I. Jain, S. Lenka, S. K. Ajmani, S. Kundu, J. Thermal Sci. Eng. Appl. 2016, 8, 1129.
[11] P. Janssen, Wire J. Int. 2014, 47, 60.
[12] F. G. Caballero, H. K. D. H. Bhadeshia, Mater. Sci. Forum. 2003, 426, 1337.
[13] C. Hofer, H. Leitner, F. Winkelhofer, H. Clemens, S. Primig, Mater. Character. 2015, 102, 85.
[14] C. Hofer, F. Winkelhofer, H. Clemens, S. Primig, Mater. Sci. Eng. A. 2016, 664, 236.
[15] L. Guo, H. Roelofs, M. I. Lembke, H. K. D. H. Bhadeshia, Mater. Sci. Technol. 2017, 34, 54.
[16] B. P. J. Sandvik, H. P. Nevalainen, Metals Technol. 1981, 8, 1213.
[17] A. Lambert, J. Drillet, A. F. Gourgues, T. Sturel, A. Pineau, Sci. Technol. Weld. Join. 2013, 5, 168.
[18] P. Jacques, F. Delannay, X. Cornet, P. Harlet, J. Ladriere, Metall. Mater. Trans. A. 1998, 29, 2383.
[19] K.-I. Sugimoto, R. Kikuchi, S.-I. Hashimoto. Steel Res. 2002, 73, 253.
[20] K. Zhu, C. Oberbillig, C. Musik, D. Loison, T. Iung, Mater. Sci. Eng. A 2011, 528, 4222.
[21] S. Y. Han, S. Y. Shin, C.-H. Seo, H. Lee, J.-H. Bae, K. Kim, S. Lee, N. J. Kim, Metall. Mater. Trans. A 2009, 40, 1851.
[22] P. T. Pinard, A. Schwedt, A. Ramazani, U. Prahl, S. Richter, Microsc. Microanal. 2013, 19, 996.
[23] W. C. Oliver, G. M. Pharr. J. Mater. Res. 1992, 7, 1564.
[24] M. Aarnst, Microstructural Quantification of Multi Phase Steels-MICRO-QUANT, 2009.
[25] V. Domnich, Y. Gogotsi, S. Dub, Appl. Phys. Lett. 2000, 76, 2214.
[26] B. B. He, M. X. Huang, Z. Y. Liang, A.H.W. Ngan, H. W. Luo, J. Shi, W. Q. Cao, H. Dong. Script Mater. 2013, 69, 215.
[27] T.-H. Ahn, C.-S. Oh, D. H. Kim, K. H. Oh, H. Bei, E. P. George, H. N. Han, Script Mater. 2010, 63, 540.
[28] Z. Xiong, G. Casillas, A. A. Saleh, S. Cui, E. V. Pereloma, Sci. Rep. 2017, 7, 17397.
[29] R. Rao, J. E. Bradby, S. Ruffell, J. S. Williams, Microelectron. J. 2007, 38, 722.
[30] C. A. Schuh, Mater. Today 2006, 9, 32.
[31] R. Navamathavan, S.-J. Park, J.-H. Hahn, C. K. Choi, Mater. Character. 2008, 59, 359.
[32] J.-I. Jang, M. J. Lance, S. Wen, T. Y. Tsui, G. M. Pharr, Acta Mater. 2005, 53, 1759.
[33] D. J. Dyson, B. Holmes, J. Iron Steel Inst. 1970, 469.