

Local Hard Zones on TMCP Large Diameter Line Pipes: What do we know about their metallurgy and microstructure?

Hervé Marchebois, Bruce Cowe, Thierry Cassagne

TotalEnergies, Pau, France
E-mail: herve.marchebois@totalenergies.com

ABSTRACT

The manufacturing and field experience of steel plates used to manufacture line pipes produced by Thermo-mechanically Controlled Processing (TMCP) are well defined in industry standards and literature. Compared to the Quenched & Tempered heat treatment process, TMCP plates are designed with a leaner chemical composition combining micro-alloying elements, precipitation, recrystallization and phase transformation during rolling and accelerated cooling. Technical challenges and process improvements moved older generation TMCP pipes from coarse microstructures and presence of non-metallic inclusions and/or mid-thickness segregation, to finer, homogenized microstructures and improved properties typically present in modern TMCP pipes.

Despite such an improvement, local hard zones (LHZ) have recently been experienced in the Oil & Gas industry on large diameter line pipes manufactured from TMCP plates. These LHZ must be distinguished from regular hard spot formation mechanisms known for years and highlighted in industry standards such as API[†] Spec. 5L / ISO[‡] 3183.

The current paper deals with a thorough analysis of these newly recognized LHZ with Scanning Electron Microscope (SEM) and Electron Back Scattering Diffraction (EBSD) investigations through the wall thickness of pipes. Internal diameter (ID) surface, intermediate zone and bulk metal microstructures showed an increase of a strong misorientation while approaching the ID. Thus, LHZ is characterized by the presence of lath and especially lower bainite type microstructures associated to high local hardness above the NACE MR 0175 / ISO 15156 limits for sour service applications.

Key words: local hard zone (LHZ), sulfide stress cracking (SSC), Thermo-Mechanically Controlled Process (TMCP), linepipe, hardness

[†] American Petroleum Institute (API), 1220 L St. N.W. Washington, DC 20005-4070, USA.

[‡] International Organization for Standardization (ISO), ISO Central Secretariat, Chemin de Blandonnet 8, CP 401-1214 Vernier, Geneva, Switzerland.

© 2022 Association for Materials Protection and Performance (AMPP). All rights reserved. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means (electronic, mechanical, photocopying, recording, or otherwise) without the prior written permission of AMPP.

Positions and opinions advanced in this work are those of the author(s) and not necessarily those of AMPP. Responsibility for the content of the work lies solely with the author(s).

BACKGROUND

The manufacturing and field experience of high strength low alloy (HSLA) steel plates produced by Thermo-Mechanical Controlled Process (TMCP) are well defined in industry standards e.g. American Petroleum Institute (API) and literature. The TMCP method consists of a well-prescribed rolling pass schedule followed by accelerated cooling that leads to a fine-grain microstructure with the desired mechanical properties of the produced plates. From a metallurgical point of view, TMCP utilizes the complex interrelationship between (micro) alloying elements, precipitation, recrystallization and phase transformation during rolling and accelerated cooling to systematically adjust the microstructure and crystallographic texture. Due to the economical benefits of TMCP manufacturing over quench and tempered processes, development of the processes continued over the preceding decades. By further research and development, the steel manufacturers were able to attain a good combination of required strength and toughness properties by optimizing technologies such as micro-alloying and TMCP.

Micro-alloying elements such as niobium, vanadium and titanium can enhance the mechanical properties of steels. Solute may then precipitate in the form of carbides or nitrides. Fine dispersion of particles can retard grain boundary motion by a pinning effect, which promotes the stability of a fine microstructure.¹ Moreover, these alloying elements may also elevate the recrystallization stop temperature.

Other typical alloying elements such as nickel, chromium, molybdenum and copper are needed to further adjust the required mechanical properties. Beyond high strength and elongation values, low-temperature toughness may be improved by grain refinement and reducing the carbon content; the latter being particularly helpful in enhancing weldability which can be particularly beneficial with modern low heat input, high cooling rate welding processes used for line pipe fabrication. HIC susceptibility can be improved by reducing impurity concentrations through the control of non-metallic inclusion quantity and shape in the steel.

Where H₂S containing effluent is a concern, the steels particularly need to resist Hydrogen Induced Cracking (HIC) and Sulfide Stress Cracking (SSC). Further issues arise because significant changes occur in mechanical properties due to pipe-forming, such as the change of yield strength and a decrease on impact toughness due to work hardening.^{2,3} Dedicated requirements through specifications and detailed analysis are thus defined to ensure integrity during service.

RECENT INCIDENTS AND INDUSTRY EXPERIENCE

As discussed above, TMCP leads to considerable economic advantages such as reduced alloying element content and faster rolling and heat treatment process with respect to phase transformation points as well as enhanced weldability. Therefore, TMCP has been accepted in API 5L / ISO 3183⁴ without additional requirements for severe sour service as compared to QT manufactured pipes from which limits of use have been derived.

More recently, this TMCP process resulted in detrimental local variations of mechanical properties and recent industry issues (among them⁵) showed that API and NACE compliant linepipe made via TMCP manufacturing process may have hidden hardness variations on pipe ID, so-called local hard zones (LHZ). These anomalies are very difficult to detect as both standard QA/QC and NDE methods do not deal with this matter and current API 5L / ISO 3183⁴ standard does not address this problem.

In case LHZ are present (as it has been observed on X60 and X65 grades) this may lead to rapid SSC failures in severe sour environments; typically, in SSC Region 3 as per NACE MR0175/ISO 15156⁶ diagram that defines regions of environmental severity with respect to SSC of carbon and low alloy steels.

Hardened areas on pipe surfaces are known to be initiation points for sulfide stress cracking (SSC) in sour service applications. Due to that the relevant standards limit hardness of sour service pipes up to 248 Vickers (i.e. 22 HRC), which is generally proven during manufacturing and weld procedure qualification, and during production and fabrication by hardness tests on cross sections. Recently, randomly distributed hardened areas were found on inner pipe surfaces in which SSC developed during operation.

Thus, the current paper deals with a thorough investigation of LHZ recorded on a pipe subject to SSC.

MICROSTRUCTURAL ASPECTS

Due to the specificity of the TMCP manufacturing, quite complex microstructures can be obtained, ranging from ferrite to martensite with the presence of residual austenite.⁷

The description of the microstructures is sometimes not easy, and some confusion exists. The last part of the heat treatment process consists of a rapid cooling performed on a strained microstructure (rolling), generally performed from the austenite or from the (ferrite + austenite) domains. Depending on the composition (micro alloying elements and the thermal/strain profile), the microstructure is tailored to obtain the requested mechanical properties (yield stress and toughness). The formation of fine grains of ferrite with low levels of pearlite is obtained for the grades with highest deformability. The presence of bainite (ferrite + carbides arranged in laths) is also possible, as well as small islands of martensite + austenite, depending on the process conditions, for example cooling rate and final cooling temperature.

An illustration of the type of microstructures encountered is given in Figure 1.⁸

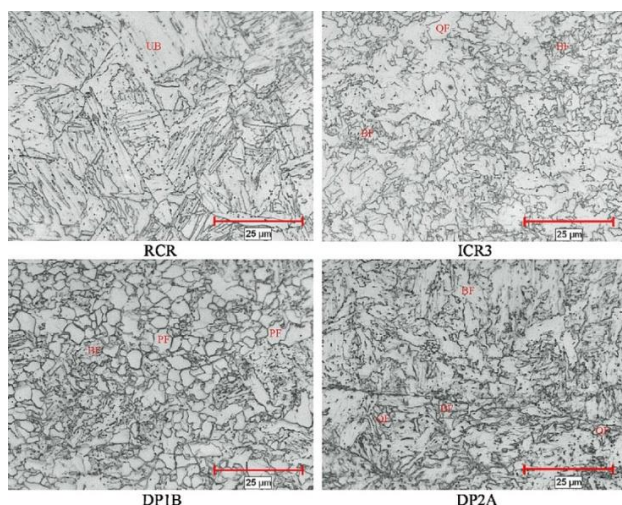


Figure 1: Optical micrographs showing microstructures obtained via different thermomechanical processing (same steel) – RCR= recrystallization Controlled Rolling, ICR = Isothermal controlled rolling, DP = dual phase, UB=upper bainite, B= Bainitic ferrite, PF=polygonal ferrite, QF= Quasi-Polygonal ferrite⁸

In the case of the presence of bainite, the difference between lower and upper bainite is hardly distinguished in the literature dedicated to pipeline materials, however the difference between the two structures relies on accepted criteria, such as lath misorientation and location of carbides. In the case of the samples studied in this paper, upper bainite+lower bainite have been found in the local hard zones (250 to 260 HV). A gradient of microstructure has been observed in these particular areas, that may influence the hydrogen embrittlement (HE) sensitivity of the material, at the crack initiation as well as the crack propagation stages.

HE susceptibility of TMCP steel phases

Generally speaking, it is recognized that the highest HE sensitivities are associated to structures far from thermodynamic equilibrium which gives the following ranking, presented in the basic literature on hydrogen embrittlement: martensite>>bainite>>ferrite (“>” meaning “more sensitive to HE than”). This classification is very rough and is not appropriate when considering the microstructures developed by the TMCP process. As shown for example by Carneiro et al.⁹, banded ferrite-pearlite microstructures can be far more sensitive to HIC than what is called BF (homogeneous bainitic ferrite, fine grained ferrite with very fine carbides).

The HE sensitivity of these structures is often addressed by their properties regarding H solubility and diffusivity. The experiments are most often performed under cathodic polarization at controlled potential or controlled current density. Both HIC (Hydrogen Induced Cracking without applying an internal stress) and HISC (Hydrogen Induced Stress Cracking) type experiments are conducted. In the latter case, fracture mechanics tests have been used to compare the effects of hydrogen on different kind of pipeline materials.

Diffusion and concentration in TMCP structures

Dunne et al.¹⁰ conducted an interesting discussion based on the results obtained after cathodic charging and measuring the total hydrogen (diffusible + thermal desorption) and the diffusible hydrogen obtained at room temperature. The experiments were performed on a API 5L X70, after different heat treatments. As shown in the *Table 1*, the levels of diffusible and residual hydrogen are quite different depending on the microstructural characteristics.

Table 1: Diffusible and residual hydrogen in different microstructures¹⁰

microstructure	Diffusible hydrogen (ppm)	Residual hydrogen (ppm)	DH/RH (%)
FGB (quasi polygonal ferrite+granular bainite)	3.57±0.4	1.77±0.4	33
EFP (equiaxed ferrite + pearlite) (ferrite grain size 14 µm)	2.9±0.3	1.5±0.1	34
BFP (banded ferrite + pearlite) (ferrite grain size 9 µm)	3.61±0.8	3.02±0.07	46
CGHAZ (coarse grained simulated HAZ, BF bainitic ferrite laths)	6.29±0.8	1±0.1	14

The authors performed also measurements on EFP structures with various ferrite grain size (15, 50 and 120 µm). They show that hydrogen at the triple junctions of grain boundaries can only be released after thermal desorption highlighting their trapping behaviour. Grain boundaries are also fast hydrogen diffusion path which lowers the hydrogen diffusion coefficient obtained for the large grain structure. In addition, the residual hydrogen is also linked to the interfaces between ferrite and pearlite (fine cementite lamellae). The high amount of diffusible hydrogen observed for the FGB and CGHAZ is ascribed to the presence of dislocations, grain and lath/plate boundaries and

internal stresses, which promote hydrogen uptake. The BF laths present in the CGHAZ structure act as easy path for hydrogen diffusion too.

Szost et al.¹¹ recorded the hydrogen distribution in carbide free nanostructured bainite (containing retained austenite) compared to nano-precipitation hardened martensite. With respect to retained austenite, the authors showed that the B/RA interfaces are more important in the absorption and “trapping” of hydrogen than the phases themselves. They also pointed out that the coherency of the interfaces is an important factor.

Hydrogen Induced Cracking

The occurrence of blistering has been studied by Dunne et al.¹⁰, after cathodic charging. An attempt to correlate internal cracking to hydrogen diffusion and trapping characteristics was proposed. A clear ranking could be made between the banded ferrite+pearlite (the more prone to cracking), the equiaxed ferrite+pearlite and the ferrite+granular bainite (FGB) (the less prone to cracking).

An extensive study of the cracking paths was conducted by Saleh et al.¹², using Scanning Electron Microscope (SEM) and Electron Back-Scattering Diffraction (EBSD) to establish the influence of parameters such as grain orientation, etc. In the FGB microstructure, small intragranular as well as intergranular cracks were developed. The hydrogen distribution in the structure is homogeneous due to the presence of hydrogen located at dislocations, grains/laths boundaries.

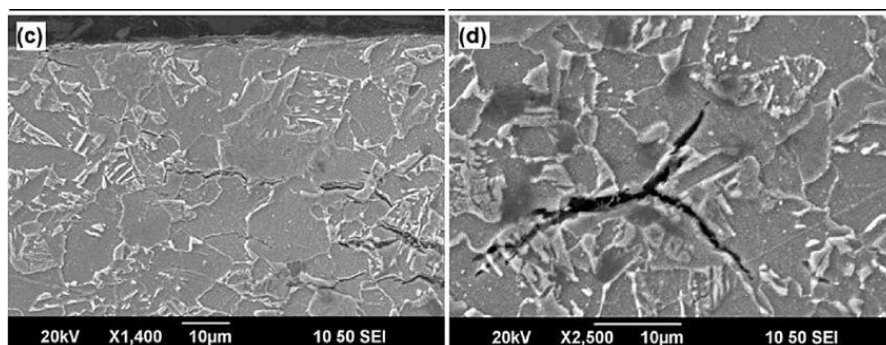


Figure 2: small internal cracks generated by electrolytic charging of a X70 steel (FGB)¹²

Hydrogen Induced Stress Cracking

Arafin et al.¹³ studied bainitic structures in connection with the near neutral pH stress corrosion cracking phenomena, at different applied potentials, and in particular at low potentials (cathodic protection) where the cracking process is related to hydrogen embrittlement. They performed SSRT experiments on X80 and X100 pipe materials, composed predominately of bainites with different amounts of M/A phases. The authors discussed the influence of diffusible hydrogen, considered as detrimental, since it is able to concentrate at the crack tips. Diffusible hydrogen is involved in the propagation process. Preferential crack initiation was observed at bainite lath boundaries, present in the bainitic X100 structure. The X80 material contained ferrite and granular bainite and was less prone to crack initiation.

The above example is discussed as the TMCP material investigated in this paper shows the local hard areas constituted of predominately lath bainitic structures whereas the bulk material is composed of predominately of ferrite and granular bainite.

The effect of hydrogen on fracture toughness was also addressed by several authors in the frame of a project related to hydrogen transportation, where API 5L X65 ferritic bainitic steels were investigated and compared to other grades.^{14,15} The authors tested the fracture of 3 point bending

notched specimens strained at low rate. The experiments were conducted in air as a reference and under applied cathodic current at different current densities.

From the work of Capelle¹⁶ on the force-load line measurement associated to the plastic fracture energy, the fracture toughness of API X52 depends on the hydrogen content in the metal. The authors also highlighted the influence of the stress on the absorption of hydrogen, with the known consequences in the strained areas at the notch, or crack tips.

The comparison of different microstructures was performed, using this method, by Chatzidouros et al.^{14,15} thanks to the method developed by Capelle¹⁶. They studied API X52 and X70 materials, each of them presenting banded structures, ferrite+pearlite in the case of X52, bainite + pearlite for X70 and compared them to X65, which displayed a more homogeneous microstructure (ferrite+bainite+pearlite islands + martensite with retained austenite (M/A)). The average hardness of X65 was 200HV (180 HV for ferrite, 220 HV for bainite, 235 HV for pearlite), the average for X52 was 154 HV and for X70 212HV. The results obtained were quite different between the three materials, X65 showing the most important reduction of fracture toughness with the presence of hydrogen as compared to air. This result was ascribed to the difference in hydrogen availability (diffusible and reversible) among the different structures. The banded microstructures were supposed to contain higher concentration of reversible hydrogen, leading to possible crack deviation and lower propagation.

TMCP behavior in H₂S containing solution

With respect to the previous paragraphs, hydrogen from H₂S containing environment is produced by the cathodic reaction associated to corrosion in the H₂S environments, which makes the analysis more difficult since surface effects and the scales formed (iron sulfide with various stoichiometries) may modify the hydrogen concentration (local effects, time dependent effects, etc.).

Hydrogen induced cracking in H₂S containing solution

Within the different papers giving results of HIC tests performed following the NACE TM0284 standard, the work from Park et al.¹⁷ is often cited, since it gives some insight on the influence of microstructural parameters, keeping the same chemical composition. The microstructures were obtained by varying the SCT (start cooling temperature) and FCT (final cooling temperature), as can be seen in Figure 3, accompanied by the micrographs illustrating the four microstructures.

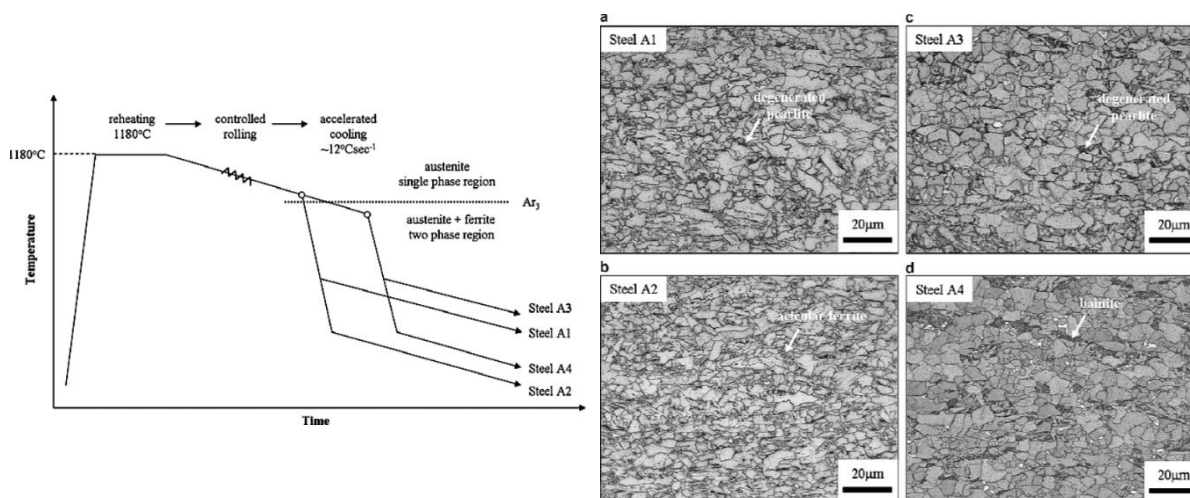


Figure 3: TMCP parameters and corresponding microstructures – A1 ferrite (F) + degenerated pearlite (DP), A2 F + acicular ferrite (AF), A3 F + DP, A4 F + bainite (B).¹⁷

It is worth noting that in all cases M/A islands were detected (average size < 5µm). The concentration of M/A islands is higher when the FCT is the lower. After the TM0284 standard HIC test (solution A), the evaluation of cracking was performed by Ultrasonic testing (CAR, Crack Area Ratio). The results were the following: CAR=0 for A1 and A3, CAR=0.05% for steel A2 and CAR=0.6% for steel A4. The fractographic analysis revealed that agglomerates of M/A constituents were found at the fracture surface, indicating a detrimental influence on the cracking process. All the microstructures were fine and homogeneous, showing the beneficial influence of DP (location of reversible hydrogen at the interface between ferrite and cementite), and the detrimental influence of M/A clusters. The difference observed between F+AF and F+B was attributed to the higher toughness of AF, as compared to B, thus impeding crack propagation. The interest of promoting the formation of AF was also shown by Shi¹⁸, working on X80 with reduced Mn content. The heat treatment parameters and significant microstructural differences are shown in *Figure 4*.

Table 1 Chemical compositions of the experimental steel											mass%
C	Si	Mn	S	P	Mo	Cu	Cr	Ni	Al	Nb+V+Ti	Fe
0.046	0.14	1.53	0.001 4	0.005	0.20	0.31	0.30	0.10	0.061	0.132	Balance

Table 2 Processing parameters in the TMCP for the experimental steel

Microstructure	Rolling temperature for different interpass reduction/°C								Accelerated cooling	
	60-80	45-60	30-45	24-30	16-24	11-16	8-11	Start and end temperatures/ °C	Cooling rate/ (°C·s ⁻¹)	
	mm	mm	mm	mm	mm	mm	mm			
M1	1 079	1 051	989	899	861	830	781	700→300	25	
M2	1 109	1 053	975	913	875	—	778	706→389	21	
M3	1 116	1 035	976	899	858	826	774	697→RT	>50	

M1: F+B

Figure 4: Steel composition, heat treatment parameters and microstructures of three steels meeting API 5L X80 requirements.¹⁹

The three steels displayed cracks after the TM0284 standard test (solution A), M1 and M2 exhibited almost the same behavior whereas M3, with the presence of martensite was much more affected by HIC damage.

Sulfide Stress cracking

With respect to the SSC process, both initiation and propagation stages may be influenced by microstructural effects. Generally, the results obtained by tests conducted in constant load conditions following the NACE TM0177 standard are discussed. In a few papers SSRT tests in the NACE solution are used to have a more quantitative tool to evaluate the microstructures than a pass/fail response.

The initiation process is most often attributed to surface defects, such as inclusions present at the surface of the specimen.

To help understanding microstructural effects, very detailed microstructural analyses are needed, which are only presented in a few papers.