

Research Article

The Effect of Microstructure on Stress-Strain Behaviour and Susceptibility to Cracking of Pipeline Steels

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Received 19 August 2011; Revised 21 October 2011; Accepted 21 October 2011

Academic Editor: Gerhard Sauthoff

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The effect of microstructure on the stress-strain behaviour of pipeline steels was studied. Slow strain rate ($2 \times 10^{-6} \text{ s}^{-1}$) tests were conducted on grade X65 and X100 steels in silicone oil and hydrogen carbonate/carbonate solution. The as-received grade X100 steel at 75°C showed serrated stress-strain curves. The magnitude of the serrations depended upon the strain rate and test temperature. Annealing at 600°C or above removes the serrations, but this increased the susceptibility to transgranular cracking in hydrogen carbonate/carbonate solution at potentials below -800 mV (sce). The removal and reformation of banding in pipeline steels were also studied. Ferrite/pearlite becomes aligned during the hot rolling stage of pipe manufacture and causes directionality in crack propagation and mechanical properties. Heat treatments were carried out which show that banding in grade X65 and X100 can be removed above 900°C . This depends on homogenisation of carbon which also depends on temperature, time, and cooling rate.

1. Introduction

The strength and toughness requirements of pipeline steels may be achieved by appropriate steel production techniques. Different strengthening methods are used by manufacturers to satisfy codes. Mechanical performance and susceptibility to environment-assisted cracking depend on microstructure, and so heat-treated pipeline steels with uniform fine-grained microstructure such as bainite are generally more resistant to stress corrosion cracking, SCC, than those consisting of pearlite and ferrite [1]. Some cracks that are hydrogen-induced have been shown to be associated with the boundary between ferrite and pearlite [2]. Such work suggests that homogenisation of composition and phases in steels improve their resistance to cracking.

Pearlite-ferrite bands in carbon steels are associated with the segregation of substitutional alloying elements which raise or lower the temperature (A_{r3}) for the formation of proeutectoid ferrite upon cooling [3]. The segregation occurs during solidification of steel and gives rise to longitudinal bands during hot rolling [3–5]. If the A_{r3} is raised by the solute, then proeutectoid ferrite nucleates first in the solute-rich regions. On the other hand, if the A_{r3} temperature is

lowered by the solute, then the nucleation of the proeutectoid ferrite begins in the solute lean regions. In either case, carbon atoms, which diffuse rapidly, are rejected from the proeutectoid ferrite, thus producing carbon-rich regions of austenite, which transform finally to pearlite [3, 4]. In pipeline steels, manganese lowers the A_{r3} temperature and pearlite forms in the regions of higher manganese. Hence band removal in pipelines steels depends on homogenisation of either carbon or manganese. Ferrite and pearlite bands cause directionality in crack propagation [2] and mechanical properties [6].

High-pressure transmission pipelines are often buried in the ground, and a cracking environment may be generated as a result of conversion of ground water to high pH solution during cathodic protection [7]. Hydrogen carbonate/carbonate solutions with a pH range between 9.6–12.3 have been reported [8, 9] to cause cracks that have led to the catastrophic failure of a number of buried pipelines when associated with poorly applied cathodic-protection systems. The high pH solution causes IG cracking whereas a low pH ~ 6.5 ground water promotes TG cracking. Cracking of buried pipes is therefore a safety consideration for evaluating new pipeline steels.

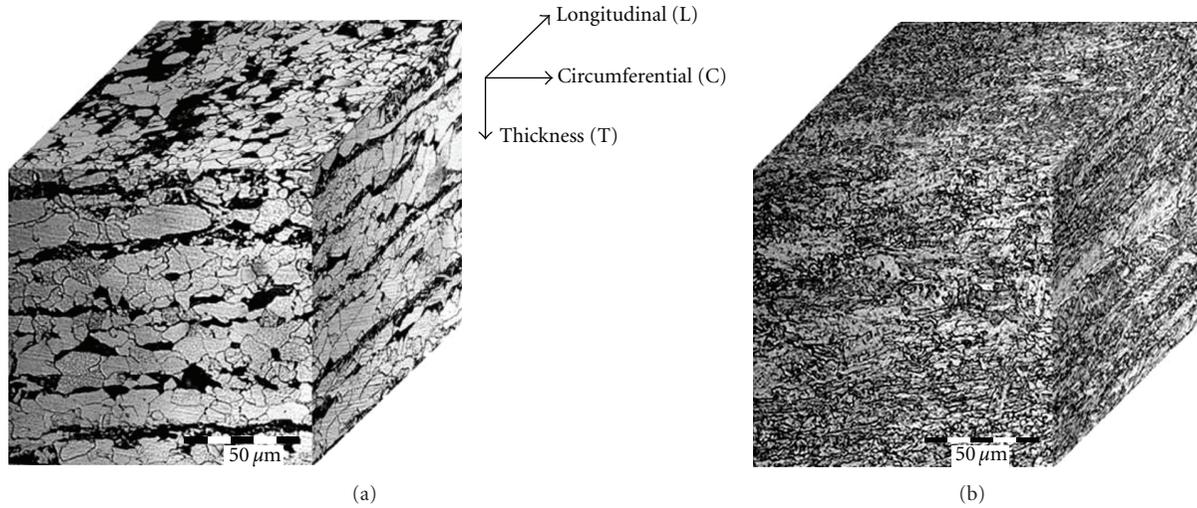


FIGURE 1: Microstructure (magnification 458 times) of (a) grade X65 and (b) grade X100.

The aim of the present work was to study the effect of removal of load-serrations on the resistance of the grade X100 steel to cracking in hydrogen carbonate/carbonate solution using the slow strain rate technique and also to examine the formation and removal of microstructural banding in grade X65 and X100 pipeline steels.

2. Experimental

The two pipeline steels tested were supplied in accordance with API-5L specifications and have the compositions given in Table 1. The microalloying and controlled rolling of the fully killed steel achieves high strength by grain refinement. The microstructures of the steels viewed in three different planes: short transverse (thickness), longitudinal, and circumferential are shown in Figure 1. The top view in the figure is the circumferential face of the pipe. The grains in the short transverse and the longitudinal directions are elongated due to rolling. Microstructures of the steels were revealed by mounting 10 mm cube blocks in bakelite, wet-grinding with silicon carbide papers, polishing on diamond cloths down to a $1\ \mu\text{m}$ finish, and finally etching with 2% nitric acid in alcohol (Nital). The average band centre-to-centre spacing and grain size were measured by counting the number of bands or grains across each sample using a travelling microscope.

Slow strain rate test (SSRT) specimens, 90 mm long, were machined to produce parallel gauge lengths of 12.7 mm and of 2.5 mm in diameter, polished to P1200 grit silicon carbide and then cleaned with ethanol. The specimens were cut from the wall of fabricated pipes with their major axes transverse to the longitudinal axis of the pipes. The major banding axis was perpendicular to the specimen surface.

Environment-assisted cracking tests were conducted in 1 M sodium hydrogen carbonate - 0.5 M sodium carbonate solution (pH \sim 9.5 at room temperature) under potentiostatic control. The specimens were contained in glass cells with external electric heating and were sealed at each

TABLE 1: Chemical composition of the steels (balance iron) in wt%.

Steel Grade	C	Mn	Si	Al	P	S
X65	0.07	1.36	0.19	<0.07	0.013	0.002
X100	0.06	1.82	0.16	0.03	0.011	0.002

end with rubber bungs that permitted the insertion of the specimen, a platinum counterelectrode, a condenser, and a thermocouple for temperature control. A conducting salt bridge led from the solution to an external saturated calomel reference electrode (sce). The solution volume was $85\ \text{cm}^3$. Tests were conducted at a strain rate of $2 \times 10^{-6}\ \text{s}^{-1}$, a temperature of 75°C , and over an applied potential range from -800 to $-1100\ \text{mV}$ (sce). Before SSRT, some specimens were heat-treated at various temperatures in a preheated, nitrogen-purged, tube furnace and then different modes of cooling were applied. The samples were cleaned with ethanol and dried before heat treatment.

The average crack velocity was determined from a metallographic cross-section, by measuring the deepest secondary crack in any one specimen and dividing it by the time for plastic deformation for tests anodic to the free corrosion potential ($-890\ \text{mV}$ (sce)) where SCC occurs. For cracks formed at potentials cathodic to the free corrosion potential, which may be the result of hydrogen embrittlement, the time from UTS to fracture was used in the calculation of the average crack velocity as here the onset of cracking occurs after necking.

3. Results and Discussion

3.1. Serrations. Tests conducted at a strain rate of $2 \times 10^{-6}\ \text{s}^{-1}$ and a temperature of 75°C gave serrated stress-strain curves for the as-received grade X100 steel (Figure 2). The serrations are characterised by a series of sudden load drops followed by an increase with a gradient similar to that of the elastic portion of the curve. The load drop in

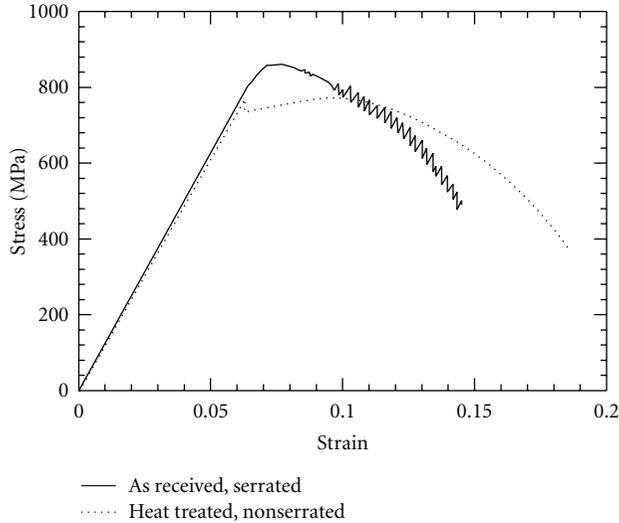


FIGURE 2: Typical SSRT stress-strain curves of grade X100 pipeline steel strained to fracture in silicone oil at 75°C.

most cases occurs soon after necking and its magnitude gradually increases to fracture. The observed serrations are similar to the reported [10–12] “Type-B serrations” which rise and fall about the general level of the stress-strain curve. This dynamic strain ageing phenomenon most likely involves the locking of dislocations followed by either unlocking of dislocations to allow them to move and/or the creation of new dislocations to allow the further deformation [13, 14]. The load increases until unlocking and/or new dislocation formation occurs, and the dislocations become separated from obstacles such as solute atoms resulting in a load drop. This process occurs many times and causes serrations in the stress-strain curve during plastic deformation [15, 16].

It has been suggested [15] that interstitial atoms such as carbon and nitrogen interact with the strain field of dislocations in steel and dislocations can be locked in position by the formation of an interstitial “atmosphere” of either carbon or nitrogen in the vicinity of dislocations but may be freed by increasing stress or temperature causing further deformation. In an attempt to further understand the nature of the serration mechanism, a number of SSRTs were conducted and the results show that the serrations are independent of whether tests are conducted in silicone oil or carbonate solution at 75°C since both give serrated curves. The serrations are affected by strain rate (Figure 3) and the size of the maximum load drop due to serrations decreases as the strain rate increases over the range tested. A test at a strain rate of $2.3 \times 10^{-5} \text{ s}^{-1}$ and a temperature of 75°C produced no serrations which agrees with previous work [16–18] and indicates that serrations occur at a matching combination of strain rate and temperature for a low carbon steel. The strain rate for the onset of serrations at 75°C is approximately $1 \times 10^{-5} \text{ s}^{-1}$. SSRT for the grade X65 steel at 75°C gives nonserrated stress-strain curves at $2 \times 10^{-6} \text{ s}^{-1}$ and this is probably due to difference in microstructure and composition.

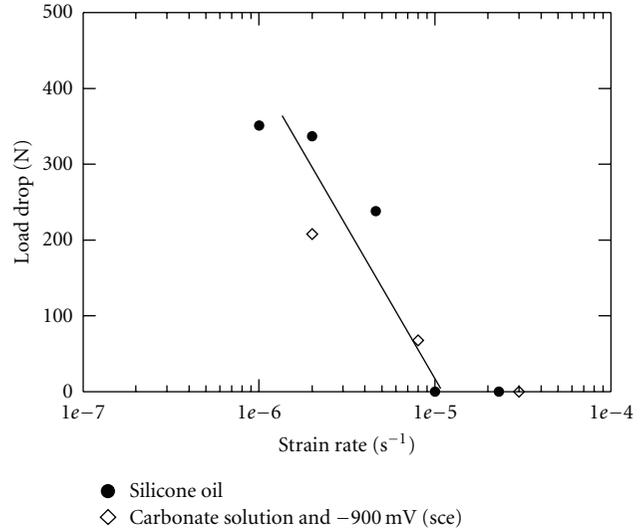


FIGURE 3: Effect of strain rate on maximum load drop due to serrations for tests in both silicone oil and carbonate solution at 75°C.

The serrations in the as-received material were completely removed by air cooling after 30 minutes at 600°C (Figure 2) and the resulting microstructure is shown in Figure 4(a). This heat treatment removes carbon and possibly nitrogen from solution and thus reduces the obstacles to dislocation movement within grains [13].

The process of removal of carbon from solution is quite noticeable at a slightly higher temperature of 700°C (Figure 4(b)). The serrations are also removed by annealing at temperatures higher than 600°C; however, a significant change in mechanical properties (Figure 5) then occurs. The change in microstructure due to the heat treatment at 600°C is a result of the removal of carbon from solution through the formation of globular carbides (sorbite). The change increases the susceptibility of the grade X100 steel to transgranular cracking (Figure 6) in the carbonate solution within a potential range of -800 to -1100 mV (sce) . At potentials anodic to the free corrosion potential, -890 mV (sce) , the cracking may be designated as SCC whilst at more cathodic potentials hydrogen embrittlement is most likely. The change in SCC cracking mode from intergranular at larger anodic potentials to transgranular as a pipeline steel approaches the free corrosion potential has been reported earlier by Li et al. [19].

3.2. Banding. Removal of the pearlite/ferrite bands in grade X65 steel depends upon the heat treatment temperature and the rate of cooling (Figure 7). A range of cooling rates was tried: furnace cooling is the slowest and water quenching the fastest. Oil and water quenching from 900°C gives a bainitic microstructure whereas air cooling from 1175°C gives a Widmanstätten structure. Air cooling from 1000°C and fan cooling from 950°C both give slightly banded pearlite/ferrite microstructures. A heavily banded pearlite/ferrite structure is obtained by furnace cooling from 1000°C. The nonbanded

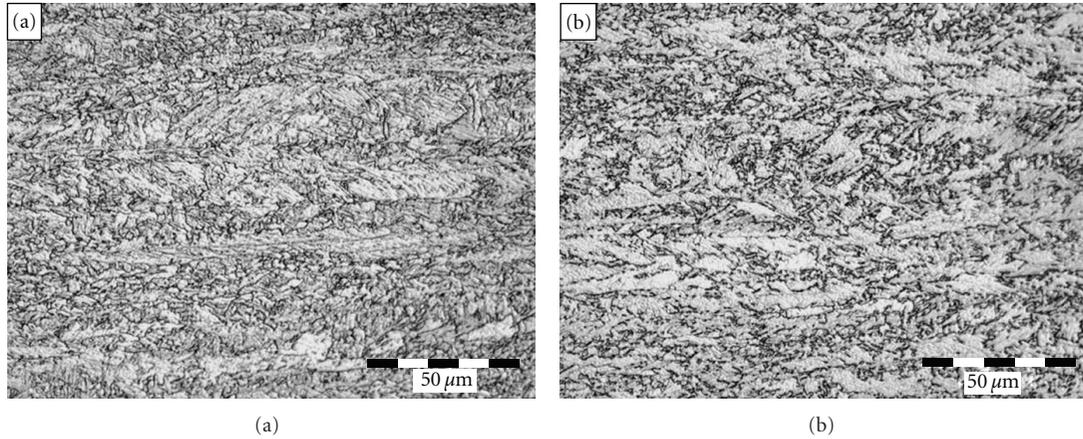


FIGURE 4: Microstructures of the longitudinal section of the grade X100 air cooled after 30 min. at (a) 600°C and (b) 700°C.

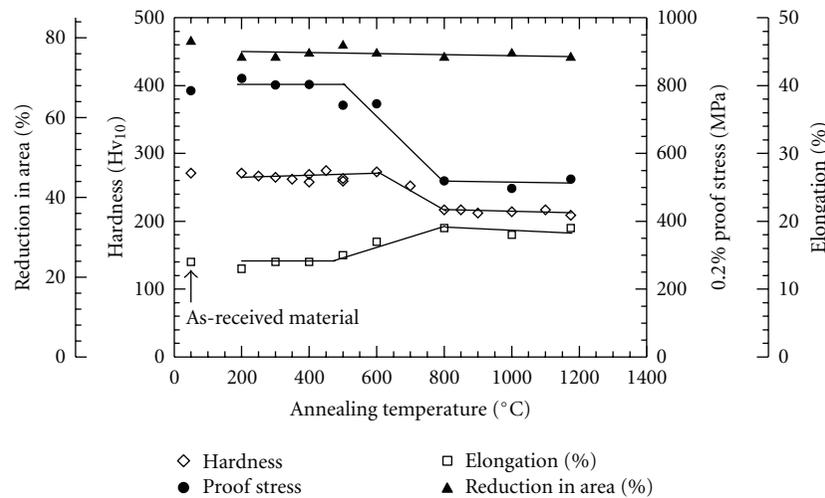


FIGURE 5: Mechanical properties of heat-treated grade X100 line pipe steel. The proof stress and the ductility were measured in silicone oil at 75°C. Average hardness was measured on the longitudinal section at room temperature.

microstructures are obtained by transformation of austenite to a nonequilibrium microstructure by using a faster cooling rate. The faster cooling rate suppresses the enrichment of austenite by carbon diffusion at the austenite/ferrite interface during cooling and this may lead to the precipitation of carbides within the ferrite. This is a process of band removal by hindering the segregation of carbon to manganese-rich regions.

Another approach is to remove banding by homogenisation of manganese through diffusion, as the concentration of Mn in the pearlite band is higher than that of the ferrite band. The result of an attempt to eliminate Mn segregation through diffusion is shown in Figure 8. This demonstrates that as the soak time increases the pearlite centre-to-centre spacing increases. This is probably due to elimination of intermediate bands of lower concentration through diffusion. Other heat treatments considered in an attempt to eliminate banding are summarised in Table 2.

These results indicate that band formation and removal from the pipeline steel depends on heat treatment tem-

perature, soak time, and cooling rate. Slow cooling from austenitic temperatures gives intense banding (provided the average grain size is less than approximately twice the average band spacing) and the intensity decreases with increasing cooling rate. Banding is completely removed by quenching or air cooling from a higher temperature (Figure 7). The fast cooling suppresses the diffusion of carbon from the region of low manganese concentration to the region of higher manganese concentration during the proeutectoid ferrite formation. Band removal by homogenisation of manganese through diffusion requires a longer time and a higher temperature than for carbon (Figure 8 and Table 2). An almost complete homogenisation of manganese was achieved by furnace cooling after 5 hours at 1175°C, and this removed banding completely through grain growth. The average grain size was refined by air cooling after 20 minutes at 950°C without the return of banding (Figure 9(b)).

These results are consistent with the trends in the literature concerning the removal of banding through quenching or air cooling from higher temperature and agree with

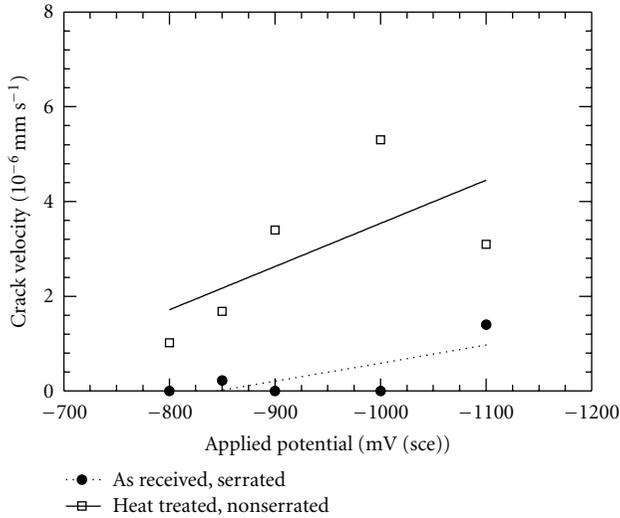


FIGURE 6: Crack velocities of the as-received grade X100 compared with that of specimens air cooled after 30 min at 600°C.

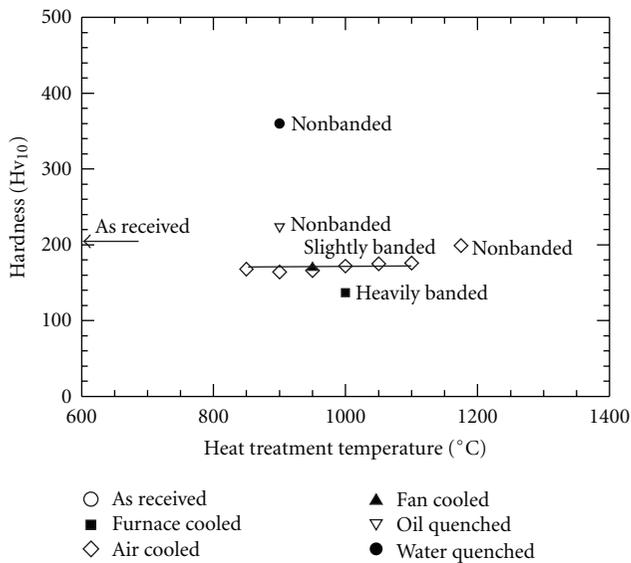


FIGURE 7: Effect of heat treatment on average hardness and banding in the longitudinal section for the grade X65 pipeline steel. Heating time for each heat treatment was 20 minutes.

the work of Wilms [20] and Grossterlinden et al. [21]. Although the homogenisation of manganese through high-temperature diffusion agrees with earlier work [22–24], in this study homogenisation was achieved at a lower temperature. Homogenisation at temperatures greater than 1200°C may result in overheating the steel, and therefore the removal of banding above this temperature was considered impractical [21]. In this study, band removal through high-temperature homogenisation followed by normalisation was reexamined with a third heat treatment involving furnace cooling after 20 minutes at 1000°C. Widely spaced traces of bands reappeared (Figure 10) indicating that the homogenisation of manganese is not complete. In a slow cooling

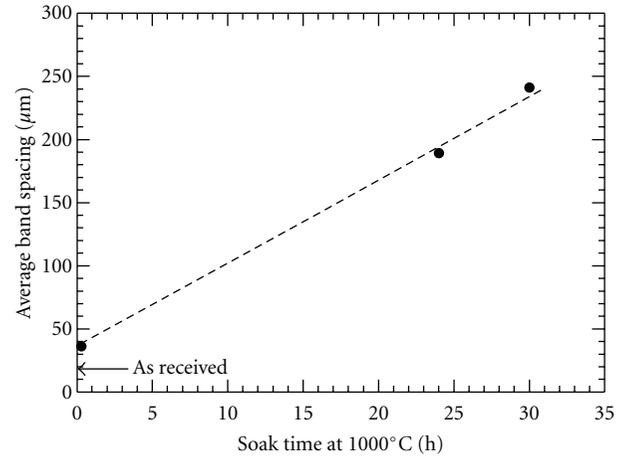


FIGURE 8: Effect of soak time on the average centre-to-centre spacing of pearlite bands on the longitudinal section of grade X65 steel; specimens were furnace cooled.

process, little differences in concentration of manganese favour segregation of carbon but the threshold difference has not been established. When these findings from the grade X65 were applied to the grade X100 steel similar results were produced, although here the average band spacing is smaller than that of the grade X65 steel. The results suggest that a key to band removal is knowledge of average band spacing and the diffusion coefficient of manganese in steels. A time for manganese homogenisation was based on the diffusion coefficient of manganese in a carbon steel [20] but the present work suggests that the diffusion rate of manganese in the grade X65 pipeline steel is slightly lower.

A comparison of SSRT fracture surfaces of banded and nonbanded specimens demonstrates that banding causes fracture surface ovality (ratio of two perpendicular diameters on fracture surface). The ovality is more severe in the as-received material than in the heat-treated specimens because the rolling effect is partially removed by the annealing. The grains are equiaxed and the carbon distribution is fairly uniform because of air cooling from higher temperatures. It is the pearlite bands that contribute to the ovality rather than the segregation of the manganese. The pearlite/ferrite banding creates a layered arrangement of hard and soft regions and, during cross-sectional reduction, it is easier to squeeze the softer ferrite bands than the pearlite bands. So, the reduction in area is greater in a direction perpendicular to banding than parallel to it. Therefore, the elimination of the segregation is important in the removal of directionality to achieve isotropy in reduction area.

4. Conclusions

- (1) Slow strain rate tests on as-received grade X100 at 75°C give a serrated load-extension curve. The serrations at 75°C and a strain rate of $2 \times 10^{-6} \text{ s}^{-1}$ were removed by air cooling after 30 minutes at 600°C (simulated tempered region of HAZ). This

TABLE 2: Effect of heat treatment on pearlite bands in the “LT” face of grade X65 steel.

Temperature (°C)	Soak time	Mode of cooling	Microstructure	Grain size (μm)	No. of treatments
950	60 h	FC	Banded with average spacing* (AS) 201 μm	24	1
1175	1 h	FC	Slight traces of banding with large pearlite areas and some Widmanstätten	29	1
1175 then 1000	1 h then 20 min	FC	Banded with AS 60 μm	12	2
1175	5 h	FC	Not banded with ferrite/pearlite and some Widmanstätten	50	1
1175 then 950	5 h then 20 min	FC then AC	Not banded (ferrite/pearlite)	9	2
1175 then 950 then 1000	5 h then 20 min then 20 min	FC then AC then FC	Banded with AS 130 μm	10	3
1200 then 1000	20 min then 20 min	FC	Banded with AS 52 μm	13	2

*As-received average pearlite band spacing and average grain size: 19 and 6 μm , respectively.

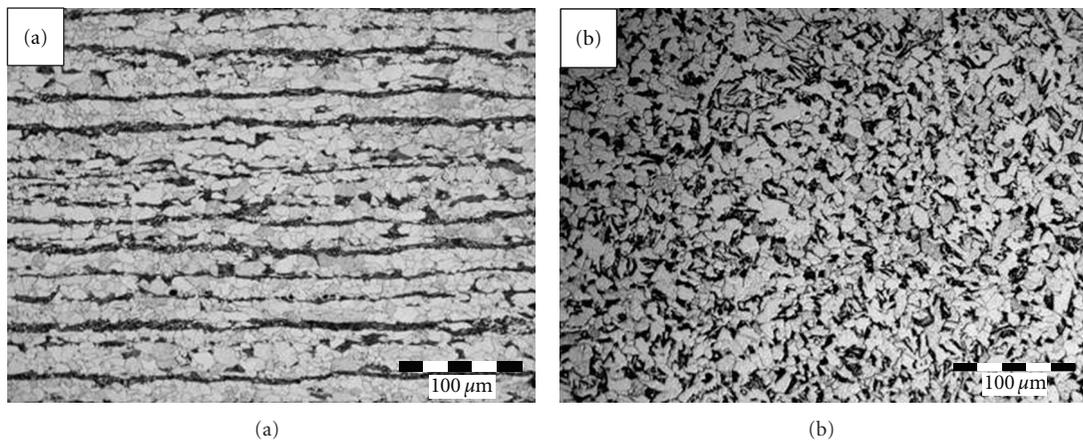


FIGURE 9: (a) Banding in the as-received grade X65 steel (b) was removed by furnace cooling after 5 hours at 1175°C and then air cooled after 20 minutes at 950°C.

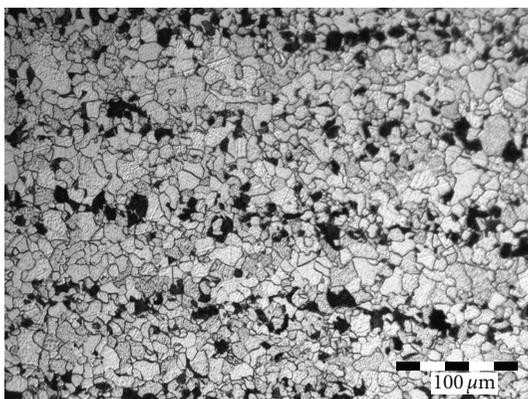


FIGURE 10: X65 steel furnace cooled after 5 hours at 1175°C, then air cooled after 20 minutes at 950°C and again furnace cooled after 20 minutes at 1000°C.

heat treatment increases the susceptibility to transgranular cracking at potentials more negative than -800 mV (sce) when compared with the as-received microstructure. The removal of the serrations and the increase in susceptibility are due to precipitation of carbides from solution as a result of the heat treatment.

- (2) Furnace cooling after 5 hours at 1175°C eliminated banding in grade X65 pipeline steel but increased the average grain size. This was refined without the return of banding by air cooling after 20 minutes at 950°C. Thus, banding was removed at a relatively low temperature but with a longer soak time. Quenching in oil or water from 900°C removes banding but the resulting microstructures are harder than the as-received ones.

Acknowledgment

This paper was supported by Petroleum Technology Development Fund (PTDF), Nigeria, to whom the authors express their gratitude.

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