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Effects of Martensite-Austenite Constituents on Mechanical Properties of Heat Affected Zone in High Strength Pipeline Steels-Review

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To improve oil and gas pipelines reliability through the minimization of defects in the welded joint, many researches were conducted to control local brittle zones in weld joints, mainly Martensite-Austenite constituents (M-A). The latter may present toughness problems for the Heat Affected Zone (HAZ) in American Petroleum Institute (API) grade steels. Given the complexity of this problem due to several parameters involved in the behavior of M-A constituents and consequently the performance of the HAZ, the recent literature was reviewed. The formation, quantity and morphology of M-A in the HAZ of multi-pass welds were elucidated. The inherent potency of M-A constituents in deteriorating toughness and initiating fracture was highlighted. The purpose is to give a state of the art and extract the points not treated yet, related to the interaction between M-A constituents, metallurgical design, welding thermal cycle and toughness in HAZ of pipeline steels. In this article, a particular attention was given to the yet not clarified behavior of Niobium (Nb) and Titanium (Ti) carbonitride, in the case of two pass welding and their ability to control Austenite grain growth.

1. Introduction

To satisfy increasing demand in energy, larger pipe diameters and high pressure are recognized as the most economical solutions in oil and gas transportation. High quality steel pipelines should be used to improve transportation efficiency, reliability and safety. In response, new API grade steels (X70-X120) are profitable since they present high mechanical properties and sufficient resistance to sour services. Generally, pipes are made from plates using seam and field girth welding. During welding, the microstructure of the base plate is modified. The combination of high strength and toughness in API grade steels can be deteriorated due to the thermal cycle effects. Previous work by Abdelbaki et al. (2015) has shown that sources of failure in welded joints accounts for up to 60 % of the probable causes, this result can make the weld the weakest point in the pipeline. In the HAZ, the matrix phases may transform to austenite and the precipitates will coarsen and/or dissolve during heating. The matrix will then retransform to martensite, ferrite, and/or bainite with new precipitate distributions upon cooling (Spanos et al., 1995). In addition, small amounts of high carbon martensite and retained austenite can be expected to form Local brittle zones. The latter have been defined by Bhadeshia (2011) as small regions of relatively hard mixtures of phases which form in the HAZ of multi-pass welds.

Several studies were conducted separately in different API grade steels to understand the formation mechanism of M-A constituents, their relation to the thermal cycle and their ability to alter the fracture mechanism. However, due to the complexity of the problem caused by a superposing effect of the thermal cycle, microstructure and alloy elements, there is still a need for a simultaneous analysis of previous results to clarify the effect of API grade steel development (microstructure, mechanical properties, and alloy design) on the HAZ response to the presence of M-A constituents. This work presents a review about the formation and effects of M-A constituents on HAZ performance for different microstructures of API grades. It is thought that this will provide a better understanding of their interaction with welding thermal cycle and alloy elements thus addressing for future studies.

2. Development of high strength pipeline steel

The need to reach high strength and good toughness in pipeline steels has enhanced progress of API grade steels (X70- X120). High mechanical properties were obtained due to the combination of metallurgical design development and thermo-mechanical controlled process (TMCP) implementation. In the seventies, application of thermo-mechanical processing and (V, Nb) addition gave rise to X70 grade steel. Later on, carbon content reduction combined with thermo-mechanical processing followed by accelerated cooling (Acc) and Ti addition lead to the development of high strength steel (X80) with good weldability. Since the 1990's, a new generation of steels X100 was developed. The use of TMCP followed by an accelerated cooling and direct quenching, coupled with (Mo, Ti) addition enabled better yield and tensile limits. The development of X120 in the last decade was achieved due to a better use and control of alloying design and grain size. The reduction of carbon percentage in different API grade steels has a positive effect in field weldability and on the reduction of the amount of hard phases such as M-A constituents. However, it causes a reduction in strength. Recently, Ti, Mo, Cr, Ni, Cu and B additions promote grain refinement, precipitation strengthening and better Control of microstructural transformations.

3. Welding of high strength steel

In single pass welds, the HAZ is divided into four distinct zones based on the microstructure changes at different distances from the welding line (Wang et al., 2015), defined as: coarse grained (CGHAZ), fine grained (FGHAZ), intercritical (ICHAZ) and sub-critical (SCHAZ) heat affected zones (a, b, c and d in figure 1). The use of high heat input can improve welding efficiency and save cost. However, the coarse grained heat affected zone CGHAZ undergoes an extensive heating due to the high peak temperature. The prior austenite grains become 5 to 6 times larger than that of the base plate (Spanos et al., 1995). Consequently, the CGHAZ exhibits the lowest toughness values due to the coarsening effect. In two-pass welds, CGHAZ is altered due to the subsequent reheating cycle. New sub-zones of the HAZ are characterized by small and discontinuous features appearing. Figure 1 shows four regions: subcritical (SC CGHAZ), intercritical (IC CGHAZ), supercritical (S CGHAZ) and unaltered (U CGHAZ) of two-pass weld joint.

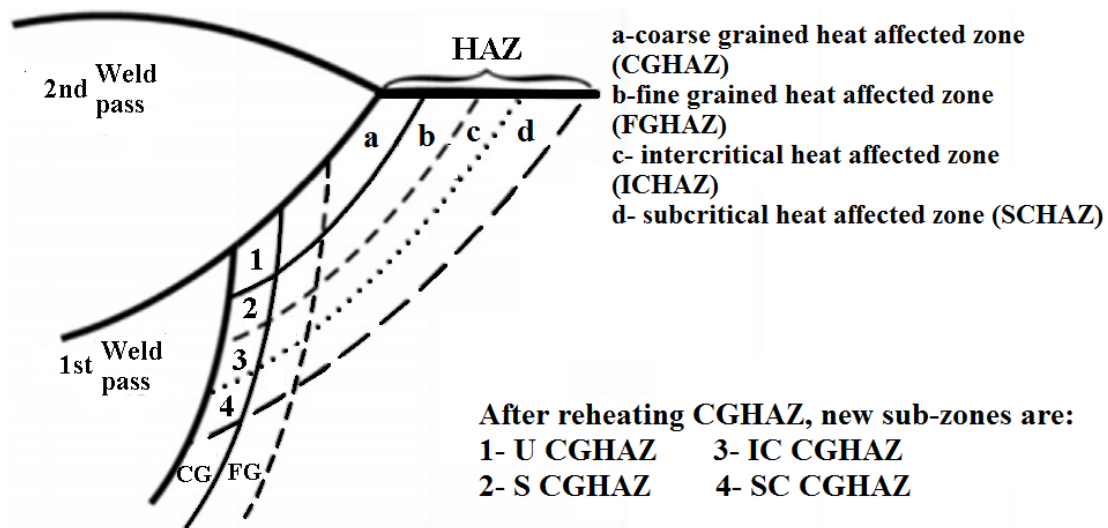


Figure 1: Sub-zones of HAZ in two pass welding

Intercritically reheated coarse grained HAZ (IC CGHAZ) is formed by reheating CGHAZ to a temperature range between Ac_1 (temperature at which austenite begins to form during heating) and Ac_3 (temperature at which transformation of ferrite to austenite is completed during heating) called intercritical temperatures ($Ac_1 < T < Ac_3$). Fraction of the matrix transforms to austenite. In the cooling stage, austenite transforms to different microstructures. The nature, quantity and distribution of the resulting products, are function of the chemical composition of the base plate and the thermal cycle experienced during welding. The HAZ toughness is influenced too. Several studies have considered the effect of the first and second pass peak temperatures (T_{p1} , T_{p2}) and of second thermal cycle cooling rate between 800 °C and 500 °C ($t_{8/5}$) on the structure of M-A constituents. The duplication of testing in situ in real welds is a major difficulty. In order to reproduce these zones

and study the effect of M-A constituents, simulation using a thermal cycle simulator (e.g. Gleeble simulator) has been widely used. It gives the opportunity to manage the parameters cited above and produce acceptable results compared to real HAZ. Specimens are subject to thermal cycles T_{p1} , T_{p2} and $t_{8/5}$ and varying cooling rate according to the field situation. Table 1 summarizes some welding simulation studies carried out to investigate the effect of thermal cycle on M-A and HAZ toughness.

Table 1: Summary of several studies on the effect of the welding cycle on M-A and HAZ toughness

Reference	Steel	Variable Parameter	Results
(Li et al., 2014) (Davis and King 1993)	X100 HSLA	T_{p1}	1- High values of T_{p1} promote austenite grain growth. 2- Coarsening of austenite grain size assist M-A coarsening and HAZ toughness decrease.
(Zhu et al., 2014b) (Li et al., 2011) (Moeinifar et al., 2010) (Li et al., 2015a)	X70 X70 X80 X100	T_{p2}	1- Decrease of toughness with a reheat in intercritical domain. 2- Increasing T_{p2} near the A_{c3} makes M-A more dispersed and improves toughness. 3- Toughness of ICGHAZ is lower than that of CGHAZ. 4- M-A constituents are observed in necklace
(Davis and King 1993)	HSLA	$t_{8/5}$	1- Low cooling rates limit the M-A formation and promote ferrite and pearlite microstructures. 2- Low toughness is related to the M-A formation in IC CGHAZ.

It is shown, first of all, that deleterious effect is associated with high values of T_{p1} which promote growth of austenite grain and control the M-A morphology. Reheating to a temperature T_{p2} near the A_{c1} makes M-A coarser and near-connected. This situation leads to lower toughness values of IC CGHAZ compared to CGHAZ. A long cooling time has a good effect in promoting the amount of ferritic and pearlitic microstructure and limits the formation of M-A. In practical welding, long cooling times are achieved by post weld heating which is expensive.

M-A constituents are small islands constituted of a mixture of high carbon Martensite and retained austenite. During a second pass welding, part of matrix reverts to austenite. Generally, the reverted zones appear firstly along prior austenite grain boundaries and bainite laths boundaries (Bhadeshia, 2014). During the cooling stage, the austenite transforms to ferritic and bainitic products which exhibit a low solid solubility of carbon. This situation leads to the diffusion of carbon to the untransformed austenite. Just below the Martensite start temperature (M_s), the austenite transforms to high carbon martensite. Since M_s is lower than room temperature, residual Austenite is retained. The composition of a high carbon martensite and retained austenite forms M-A constituents. It is well known that in IC CGHZ, M-A constituents have different size, shape and distribution: thin and stringer M-A between bainite laths, coarse and near-connected M-A constituents form at prior austenite grains and blocky M-A form within the grains (Li et al., 2015a). The hardness of M-A constituents is function of carbon concentration (Mohseni et al., 2014). Carbon concentration of M-A constituents was found to be independent from the average concentration in steel (Bhadeshia, 2011).

4. Interaction of metallurgical design and M-A constituents

During multi-pass welding, the chemical composition influences microstructural transformations. The reduction of carbon content and thus of carbon equivalent in modern API grade steels has a positive effect in decreasing hardenability, and consequently preventing M-A constituent formation (Liessem and Erdelen-Peppler 2004). An extensive use of alloy elements offsets, however, the reduction of carbon content and stabilizes the carbon equivalent. In addition, alloy elements influence transformation pattern in different manners. The efforts to minimize the amount of M-A constituents lead researchers to reduce the balance of alloying elements which promote the M-A formation. Meester (1997) reported that the tendency to reduce these elements is significant in decreasing order: B, N followed by carbide forming Nb, V, Mo and Cr.

Nb, V and Ti are known to have two major advantages. Firstly, it promotes Nb- V- Ti carbonitride (C, N) precipitation hardening. Second advantage is the ability to limit austenite grain growth due to the undissolved Nb- V- Ti (C, N) during first cycle of multi-pass welding. This gives rise to grain refinement and limits the coarsening of M-A during intercritical reheating. In the event of high heat input welding, the carbonitride can dissolve due to high peak temperature. Ti carbonitrides are known to have more thermal stability than Nb ones. Results reported by (Zhu et al., 2014a) showed that a near-stoichiometric Ti/N ratio promote toughness of CGHAZ by grain refinement. This effect didn't extend to ICGHAZ where no correlation was found between

M-A morphology and Ti/N ratio. Some studies reported that addition of Nb can alter the thermal stability of Ti carbonitride by formation of complex (Ti, Nb) (C, N) which is less thermo-stable (Moon et al., 2007). Neither the standards nor the published works give any Ti/Nb ratio which gives the most effective pinning effect on grain growth. The API 5L (2005) standards specify a level of $(Ti + Nb + V) \leq 0.15\%$.

The pinning force can be expressed as (Dong et al., 2016):

$$F_z = \frac{1}{K_z} * \frac{f}{R} \quad (1)$$

Where F_z is the pinning force, K_z is the dimensionless Zener coefficient, f is the volume fraction of particle and R is the equivalent radius of precipitates. The volume fraction of precipitates can be calculated as:

$$f = \frac{N}{S * D} * \frac{4\pi}{3} * R^3 \quad (2)$$

Where N is the number of precipitates per area, S is the specific area for estimation, D is the precipitates equivalent diameter and R is the precipitates equivalent radius. The volume fraction of precipitates decreases with austenitizing temperature due to the dissolution of the particles.

Eq(1) and (2) show greater pinning force for a higher precipitates volume fraction. But the volume fraction decreases with increasing austenitizing temperature. More efforts are still needed to clarify the behavior of complex Ti and Nb carbonitride according to the ratio Ti/Nb during welding for different configurations of alloy elements. Steel makers and operators should take in consideration this effect during production and welding of API grade steels.

5. Impact toughness and fracture mechanisms associated with M-A constituents

A number of researches have shown that M-A constituents are the predominant cause in lowering the HAZ toughness of multi-pass welds due to their crack susceptibility and hardness (Li et al., 2015b). Charpy impact toughness is the most common test used to evaluate toughness and fracture mechanisms in HAZ. Generally, to evaluate the effect of M-A constituents in the IC CGHAZ, Charpy impact toughness tests can be performed at room temperature to evaluate the absorbed energy for different intercritical temperatures. Many researchers showed an increase in the absorbed energy with increasing T_{p2} temperature within intercritical domain (Zhu et al., 2014b). In the literature, it has been mentioned that impact toughness decrease with increasing fraction of the M-A constituents. Nevertheless, the impact toughness of the IC CGHAZ strongly depends on the matrix grain boundaries, the area percentage, shape, morphology and connectivity of grain boundaries of M-A constituents (Davis and King, 1994). A moderate first cycle peak temperature can make the M-A constituent more dispersive and increase toughness. Comparing with the CGHAZ, the toughness of the IC CGHAZ is consistently lower even with some improvement in the high temperature of intercritical domain (Zhu et al., 2014b). Lambert-Perlade et al. (2004) found that both M-A constituents and coarse upper bainite had a deleterious effect on the toughness. Isolated M-A particles do not have such a detrimental effect on toughness at room temperature. Bott et al. (2005) paradoxically found that the impact toughness of HAZ in SAW process tandem technique was better than the base plate. This can be assigned to smaller size of M-A constituents and larger spacing between them coupled to the fact they are not interconnected. According to published data, the presence of M-A constituents increase the susceptibility of cleavage cracks (Mohseni et al., 2014). Four mechanisms were proposed in the literature and were collected by (Davis and King, 1994). Many results showed that the mechanism by which M-A constituents decrease toughness in IC CGHAZ appear to be due to brittle cracks resulting from debonding of M-A constituents or between two closely separated M-A particles (Mohseni et al., 2014). Luo et al. (2018) found that microstructure with slender M-A exhibit lower toughness comparing to a microstructure with blocky M-A with a core-shell structure with Martensite forming the "shell" and austenite forming the "core". This is contradictory to the finding by (Li and Baker, 2010) which throws a shade of confusion. More studies should therefore be conducted to understand the effect of shape, morphology and distribution of M-A constituents in their fracture mechanisms, with respect to the surrounding microstructure and the M-A constituent's hardness.

6. Discussion

A considerable number of recent researches have established a close relation of the deteriorating effect on HAZ toughness of multi-pass welds to M-A constituents. However, in some cases, a detailed characterization of these components has not been considered. A complete description of M-A constituents should consider different aspect simultaneously: the volume fraction of M-A in microstructure, their hardness, the amount of austenite in M-A, shape and morphology of M-A constituents and the surrounding microstructure. Incomplete description can lead to contradictory results between published results. A microstructure composed mainly from acicular

ferrite and small uniform distributed amount of M-A is known to give a reasonable strength-toughness performances. A minimum volume of M-A fraction is needed to play as a strengthening mechanism. Hardness of M-A is related to the amount of carbon in M-A. For the same level of carbon, M-A can be less deleterious if there is a sufficient volume fraction of M-A. The confusing result reported in section 5 about the most dominant factor in brittle crack initiation being due to blocky or slender M-A in IC CGHAZ may be attributed to not considering of the difference in M-A Hardness.

High heat input welding alter toughness by a coarsening effect on austenite grain which in turn has a same effect on M-A. An effective way to reduce this coarsening is a design of base plate containing carbonitride with strong pinning ability. This can be done with the appropriate use of Ti, Nb and V alloying elements. On the other hand, Nb is widely recognized to promote formation of M-A constituents (Li et al., 2001) by increasing hardenability. Yan and Bhadeshia (2015) reviewed different proposed mechanisms of the way that niobium enhances hardenability during casting and hot rolling prior to pipe construction. In the case of multi-pass welding, the given information are mere qualitative observations relating the amount of M-A to the percentage of Nb in base plate. It is proposed that further studies should be conducted to develop quantifying techniques and parameters to evaluate this effect.

7. Conclusions

The foregoing review has focused on the development of API grade steel and the HAZ response to the presence of M-A constituents. A close examination of the considered studies has clearly shown the considerable influence of M-A constituents on the mechanical properties and the toughness in particular. The number of parameters involved, however, and the interconnections between them makes the treatment of the M-A constituents a complex issue.

The following conclusions can be formulated:

- Carbon content reduction and a better management of alloying elements, enhance mechanical properties in developed API grade steel. However, an obtained new metallurgical design stabilizes the carbon equivalent, without reducing the amount M-A constituents in weld joints.
- A better optimization approach of metallurgical design must take in consideration the selection of total alloying elements resulting in lower carbon equivalent and preventing high heat input undesirable effect in the HAZ.
- M-A constituents control the fracture behavior of ICCGHAZ and reduces toughness value locally.
- The use of Ti and Nb in Pipeline steel can change the behavior of the resulting carbonitride during welding and alter the pinning effect on austenite grain growth, a specified Ti/Nb ratio in steel can be helpful in optimizing the chemical composition of API grade steels.
- The use of high heat input in welding is economically beneficial but it promotes excessive M-A formation.
- The presence of M-A constituents within the HAZ is not a sufficient condition for their deleterious effect. Their shape, morphology, distribution, the matrix phases and grains refinement influence their behavior.

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