

STEEL ALLOY DESIGNS FOR CONTROL OF WELD HEAT AFFECTED ZONE PROPERTIES

F.J. Barbaro¹, Z. Zhu¹, L. Kuzmikova¹, H. Li¹ and J.M. Gray²

¹University of Wollongong, NSW, Australia.

²Microalloyed Steel Institute, LP, Houston, Texas, USA.

Keywords: Titanium, Niobium, Precipitation, HAZ, TMCP, Weldability, Mechanical Properties, Toughness, HTP Steel, Thermal Simulation

Abstract

This paper reviews the current technology related to the control of weld HAZ properties in high strength steels. The debate related to the appropriate balance of Ti and N is addressed and it is concluded that a stoichiometric ratio provides optimum fracture toughness. Although the control of the Ti/N ratio is difficult, this paper now provides guidance in terms of alloy design to achieve optimum fracture toughness in the HAZ.

The enhanced resistance to grain coarsening behaviour in higher Nb steels has now provided another incremental improvement in the weldability of these steels for critical applications, such as high pressure gas transmission pipelines. The role of Mo in the alloy design of these new steels is a further activity to be undertaken in the design concept to further improve the performance and safety of modern steel structures.

Introduction

Modern high strength steels are manufactured using tailored alloy designs and processed using advanced thermomechanically controlled processes. The final mechanical properties of the steel are controlled through development of specific microstructures which are characterised by a fine ferrite grain size and a dispersion of alloy precipitates that optimise strength, ductility and toughness.

The process of welding inherently subjects the steel to elevated temperatures, which significantly modifies microstructures and therefore the mechanical properties. The region immediately adjacent to the weld, where the temperature approaches the melting point, can be significantly degraded by the extreme grain coarsening that can occur in the austenite phase region. Although many parameters control the microstructure and mechanical properties of the HAZ, it is control of the austenite grain size that is critical in terms of the performance of these steels.

Ti microalloying produces a dispersion of stable TiN precipitates which retard austenite grain coarsening by a grain boundary pinning action [1-5], as shown in Figure 1. Other methods of HAZ microstructural control have been developed [6] but Ti microalloying, following complete deoxidation, to ensure formation of nitride precipitation, is currently the dominant method employed.

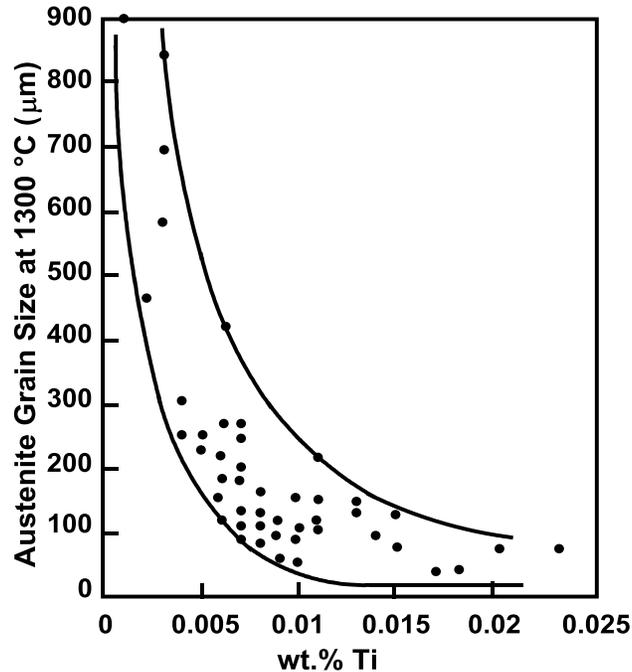


Figure 1. The effect of Ti content on austenite grain size at 1300 °C [4].

It is true to say that the use of Ti in steels is by far the most complex and least understood of all the microalloying elements. Ti is a strong oxide, sulphide, nitride and carbide former and it is the chemical balance of impurities and processing that determines the final form of the Ti compounds and the resulting properties.

A very fine dispersion of TiN precipitates is required to exert a grain boundary pinning action in opposition to the driving force for grain growth. Maximum pinning force (Z) has been shown by Zener [7], to be directly proportional to the volume fraction of precipitates (f) and inversely proportional to the precipitate size or radius (r), as defined in Equation (1). Therefore, a large volume fraction of small precipitate is beneficial for grain size control.

$$Z \propto f/r \quad (1)$$

The approach to achieving the optimum dispersion of precipitates is the basis of ongoing debate amongst steelmakers and specifiers. Different approaches have been proposed to control the precipitate dispersion but the reported results are generally clouded by the influence of secondary effects associated with base alloy design and processing, including casting conditions.

This paper details the technology of TiN precipitation and recent research that clarifies the current debate regarding appropriate alloy designs of Ti bearing high strength steel grades. The issues associated with the current Ti alloy design are discussed and an alternative incremental approach using increased levels of Nb to improve the weld HAZ properties of these steels is presented.

Background

An addition of Ti, following deoxidation and in the presence of N, results in the formation of TiN precipitates which have high thermal stability [8]. As discussed above, the effectiveness of such precipitates is dependent on their size distribution and volume fraction and is related to the solubility product data, as shown in Figure 2.

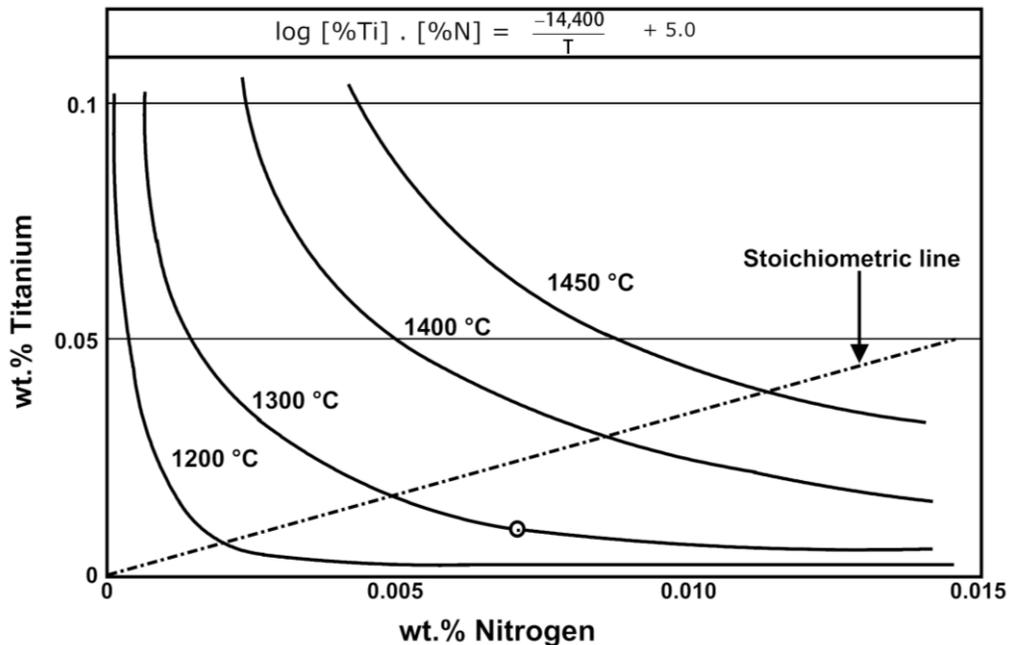


Figure 2. Solubility isotherms for TiN in austenite [9].

As the concentration of Ti and N increase, the temperature at which precipitation forms increases and results in the formation and/or growth of large TiN precipitates. Large TiN precipitates which are primarily cuboidal in shape are not only ineffective in grain boundary pinning [7] but also provide sites for the initiation of cleavage fracture [10,11]. An optimum dispersion of TiN precipitates can be achieved by limiting the overall concentration of both Ti and N, to reduce the temperature of formation and minimise thermally activated diffusion processes that lead to precipitate growth, as shown in Figure 2. In other words, a large number of very small precipitates are required to achieve maximum grain size control. The grain coarsening temperature increases when there is a maximum volume fraction of small pinning particles.

Therefore, a certain volume fraction of precipitation is required to effect grain boundary pinning and a minimum level of Ti is essential to ensure optimum grain size control. Unfortunately, however, current steelmaking practice struggles to accurately control the final level of N and so a specific stoichiometric ratio can be difficult to achieve. A non-stoichiometric ratio not only results in a non optimal size distribution but also results in either excess Ti or N, which generates the formation of TiC in the former and excess N in the latter. There is also controversy about the role of excess N as some claim that free N is detrimental to toughness [12-15] while others suggest beneficial effects as the N retards precipitate dissolution [16].

It should however be emphasised that the overall performance of the steel is defined by the range of Ti and N levels as well as the casting conditions and the slab cooling rates. The latter processing conditions are also critical, as the as-cast precipitates, especially the detrimentally large precipitates, are difficult to modify because of their high thermal stability.

In terms of procurement of steel for critical applications, such as pipelines, it is appropriate to evaluate steel performance and production capability, with respect to weld HAZ properties. Reliance solely on an alloy design or specification is insufficient when specific mechanical property performance is essential to ensure structural integrity and public safety.

More recently, the work of Zhu et al. [17] has now demonstrated, using detailed quantitative metallography, a more complete understanding of the influence of Ti/N ratio on weld HAZ microstructure. This work utilised a set of samples from commercial API 5L X70 grade pipe, which were produced using identical processing conditions and contained the same base chemistry, but most importantly a range of Ti/N ratios, for critical evaluation of weld HAZ microstructure and mechanical properties.

As experienced by previous investigators, the inherent difficulty in accurately evaluating a narrow weld HAZ necessitated the use of a Gleeble weld thermal simulator to provide sufficient, suitably sized specimens containing the critical CGHAZ region of the welds. The peak temperature and cooling rate employed directly reflected real welding conditions to ensure that the microstructures and importantly the austenite grain size were identical to those of the real weld.

Metallographic examination revealed the simulated microstructures to primarily consist of an aligned bainitic ferrite with small amounts of interlath M-A islands. Hardness values were consistent with the peak hardness values recorded in HAZ profiles from the real welds and so confirmed the suitability of the weld simulation conditions.

Although qualitative evaluation of the austenite grain size indicated minor variations in average grain size, the significant observable difference was revealed using quantitative metallographic techniques. The results presented in Figure 3 confirm that a distinct difference in distribution of grain size was observed which suggested that both an increased volume fraction of fine austenite grains and less coarsened austenite grains were present in the steel with a Ti/N ratio close to the stoichiometric combination of Ti and N. This was further supported by the results of Charpy impact tests, shown in Figure 4, which demonstrated a “measurable” improvement in impact toughness.

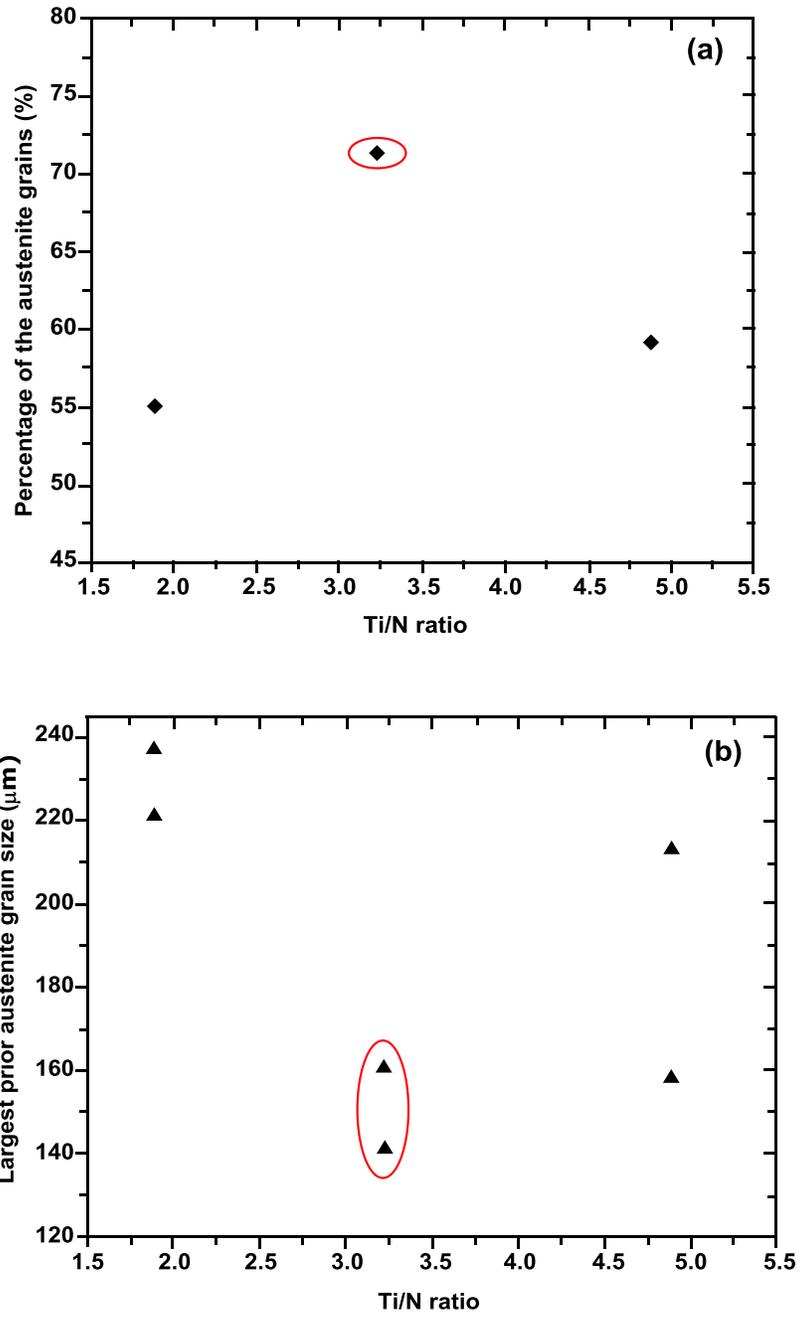


Figure 3. Results of statistical analysis of prior austenite grains size in steels with different Ti/N weight ratios; (a) percentage of fine grains (smaller than 80 μm), (b) largest grain size [18].

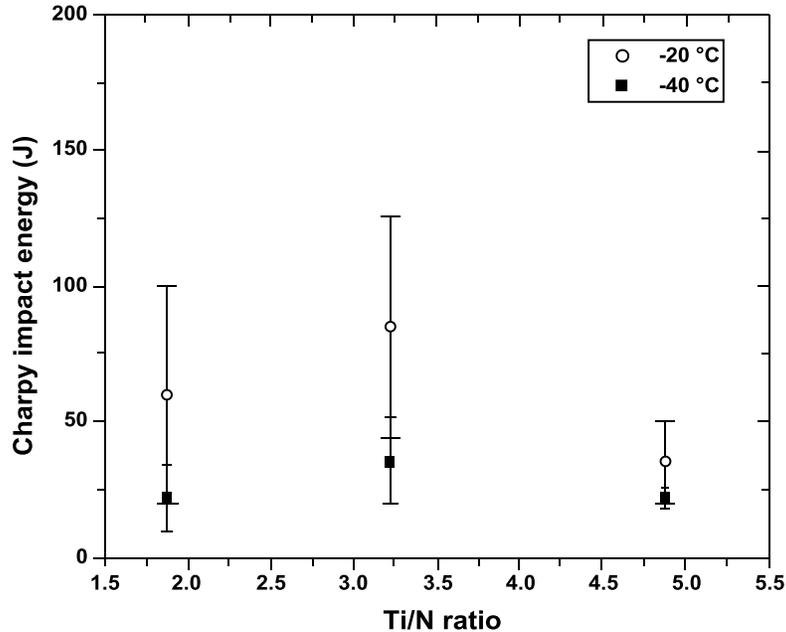


Figure 4. Mean Charpy impact energy of the simulated CGHAZ of samples with different Ti/N weight ratios. Each point is an average of three measurements [18].

Such a fine difference in microstructure and toughness performance would not have been discernible in evaluation of real welds because of the difficulties in evaluating and testing the sharp gradient of microstructures immediately adjacent to the weld fusion line.

It is reasonable to assume that based on these results, the practice of first tier steel producers [19] and thermodynamic considerations, that the optimum combination of Ti and N exists around the stoichiometric ratio, at levels that restrict initial formation of TiN in the liquid steel, ie above the solidus.

High Temperature Processed Steel

A new alloy design, although reported many decades ago [20], has only now been commercially developed and successfully utilised in a number of large international pipeline projects [21,22]. The steel design typically contains a low C content, microalloyed with Ti but with a Nb content in the range 0.08-0.11 wt.%, which enables the full capability of Nb microalloying to be effectively utilised in steel manufacture and service performance [23].

It has been clearly demonstrated that Nb provides a number of benefits to the mechanical properties of steels: grain size refinement; lowering the γ to α transition temperature (A_{r3}), precipitation hardening and retardation of austenite recrystallisation [24,25].

It is pertinent to point out the salient features of the higher Nb alloy design and the improvements to processing conditions that also provide economic benefits to the steelmaker. Key features of this higher Nb alloy design include:

Reduced C Content:

- (a) Minimises elemental partitioning, ie segregation, during casting, as schematically shown in Figure 5 [26]. Avoidance of the peritectic reaction eliminates the involvement of enriched interdendritic liquid during transformation and furthermore provides an extended solidification range in delta ferrite that enhances homogenisation of the newly formed solid prior to further transformation on cooling;
- (b) Maintains and enhances the solubility of the Nb to assist in the benefits outlined above, Figure 6 [26]. Most notable is the delayed precipitation of carbonitrides that enhances the hot ductility and increases the operating window for hot rolling, and therefore reduces mill operating loads. The consequential benefits to available mill uptime and maintenance downtime should also be considered;
- (c) Reduces carbon equivalent and therefore improves the weldability for reduced susceptibility to hydrogen assisted cold cracking.

Controlled Ti Addition which provides control of N thus allowing optimum NbC precipitation, and avoids the formation of mixed carbonitride precipitates which occur over a larger temperature range.

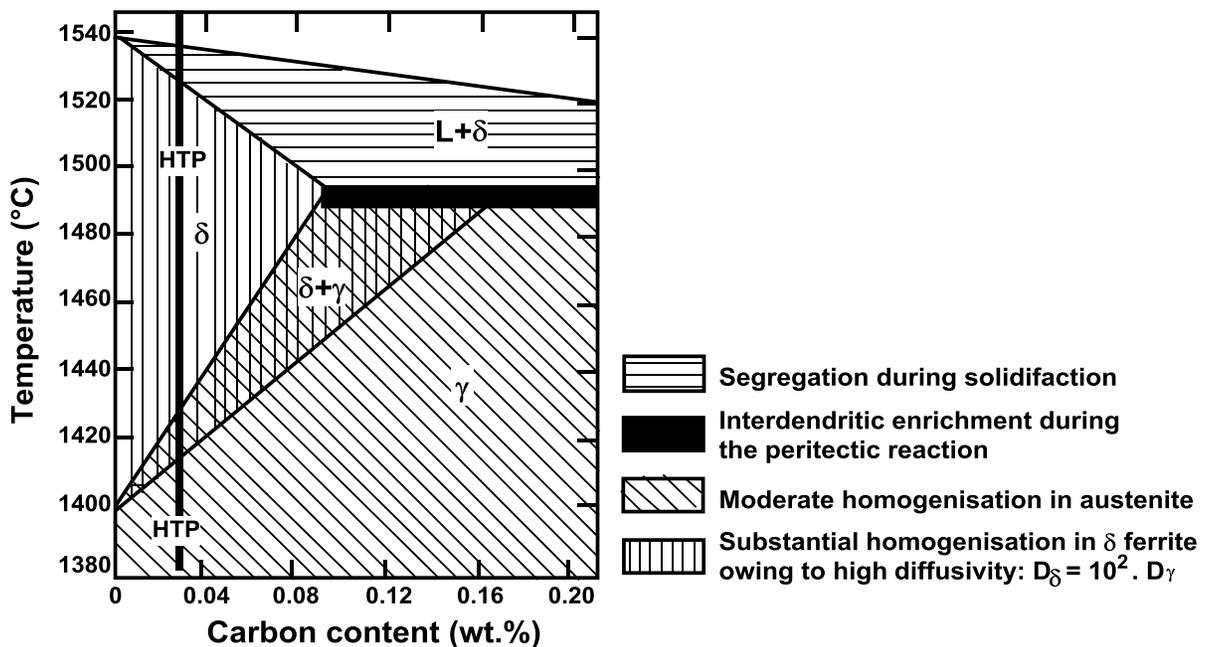


Figure 5. Part of the Fe-C diagram with classification of the segregation severity.

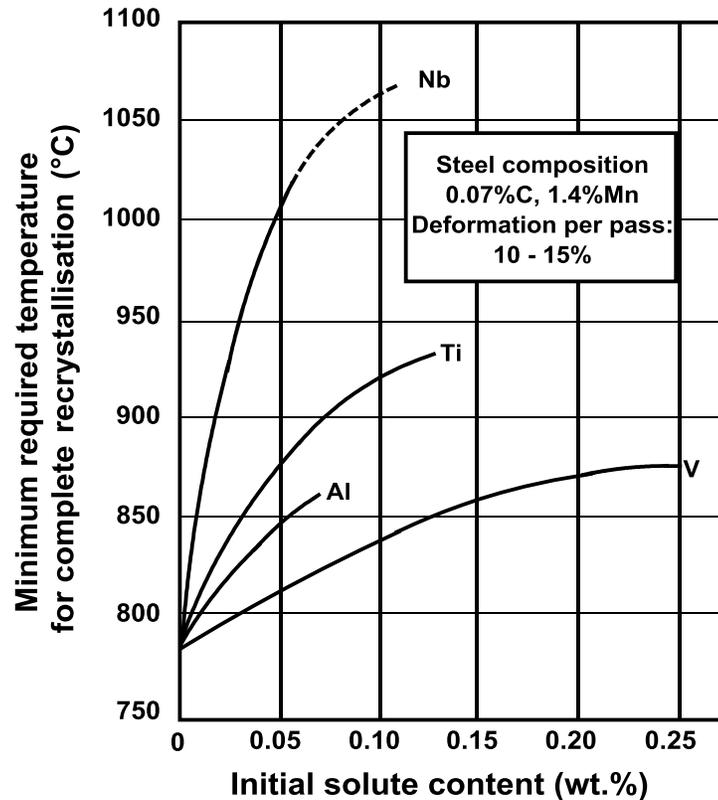


Figure 6. Retardation of recrystallisation by the microalloying elements.

This alloy design therefore offers a number of economical processing advantages over traditional grades, particularly API high strength grades, as pointed out by Ouaisa et al.[27]:

- (a) Continuous casting can be successfully achieved over an increased slab width range providing greater tonnage throughput at the slab making operation stage;
- (b) Hot ductility is improved and so slab-cracking issues are avoided. This not only avoids the necessity for slab inspections and reconditioning but also provides the opportunity to reduce energy consumption by direct hot charging;
- (c) Increased thermal processing window enables more significant slab width reductions at the sizing press;
- (d) Finish rolling temperature is increased, ie T_{nr} is raised, and so entry to the finishing mill is increased by roughly 100 °C and so provides latitude for increased reductions and/or reduced mill loadings, thus minimising mill wear and tear.

In summary, the alloy design permits steel mills with low tolerable rolling forces to produce high strength steel grades with enhanced toughness [26]. The weldability performance of these steels therefore needs to be more thoroughly understood.

Despite the wide ranging research [28-30] carried out on the role of Nb in steel, there is relatively limited work on the effect of increased Nb levels on weld HAZ grain size control in these low C steels. Therefore, this experimental work was conducted to investigate the effect of Nb and C content on austenite grain coarsening during thermal cycles similar to those experienced in the HAZ of a typical fusion weld. The chemical composition of the studied steels is presented in Table I.

Table I. Chemical Composition (Key Alloy Elements) of the Investigated Steels, wt.%

Steels	C	Mn	Si	Mo	Cr	Nb	Ti	V	N
Steel 1	0.140	1.20	0.27	0.002	0.020	0.001	0.0140	0.006	0.0021
Steel 2	0.085	1.44	0.30	0.010	0.030	0.033	0.0160	0.066	0.0060
Steel 3	0.047	1.59	0.23	0.150	0.031	0.055	0.0077	-	0.0031
Steel 4	0.050	1.61	0.16	0.002	0.240	0.110	0.0120	0.003	0.0048

Three different Nb bearing steels were selected, along with a Nb free steel, all of which contained Ti, and subjected to four different peak temperatures (1050, 1150, 1250 and 1350 °C) using a Gleeble 3500 thermalmechanical simulator. The heating rate, dwell time and cooling rate, which are presented in Figure 7, were selected to duplicate typical thermal conditions experienced during actual weld fabrication, but in any case were identical for each steel.

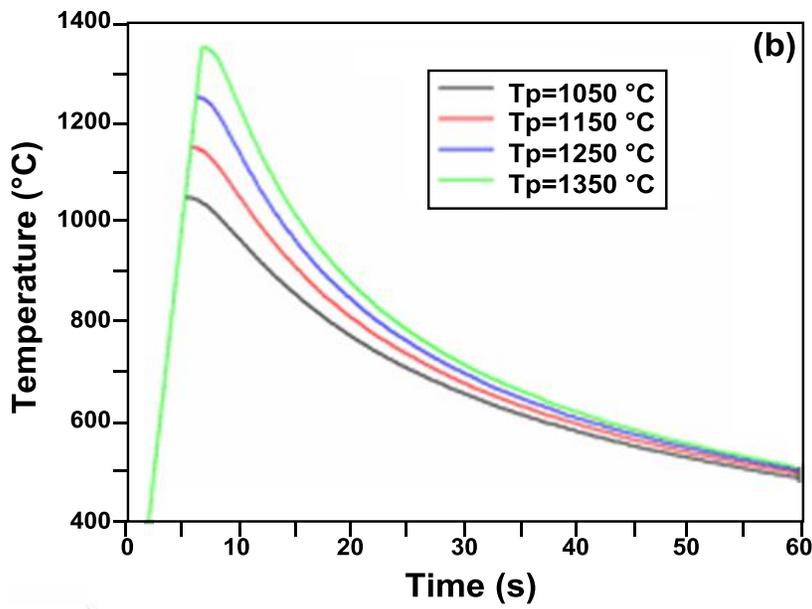
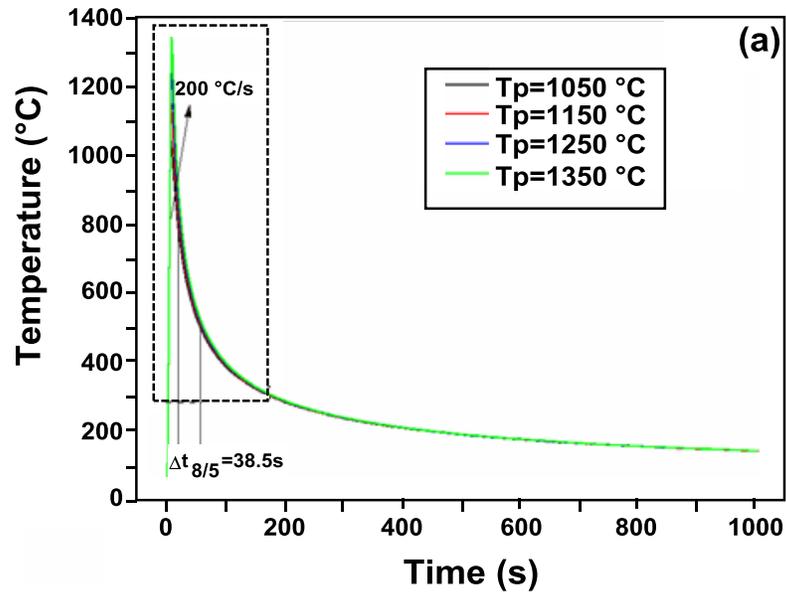


Figure 7. Thermal cycles used for the HAZ simulation of the studied steels; (a) overview, (b) close-up. T_p is peak temperature.

The average austenite grain size values of the four investigated steels are presented in Figure 8. The measured grain size of the samples subjected to a thermal cycle with a peak temperature of 1050 °C was fairly uniform with an average value of ~10 µm. Increasing the peak temperature to 1150 °C resulted in significant grain growth in the Nb free steel. A peak temperature of 1250 °C stimulated grain growth in all four steels but interestingly, the high Nb steel experienced a rapid increase in grain size, in comparison to 1150 °C. As the peak temperature increased to 1350 °C, significant coarsening occurred in all the steels except the high Nb steel, where only a slight increase occurred.

Overall, the Nb-containing steels exhibited greater austenite grain size control compared to the Nb free grade while the high Nb, low C steel demonstrated remarkable control at the peak temperature of 1350 °C.

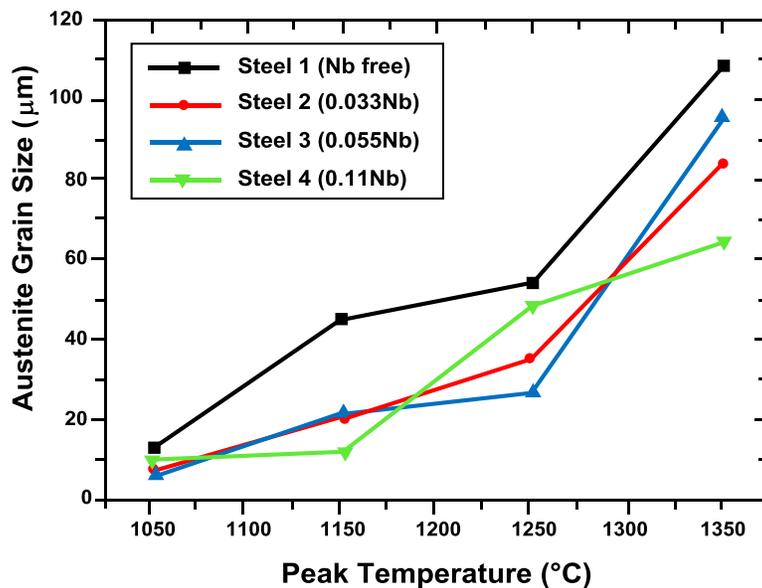


Figure 8. Grain size measurements of the investigated steels at different peak temperatures.

Charpy data from weld thermal simulation trials, where the $\Delta t_{8/5}$ cooling time was varied to simulate differences in weld heat input, is shown in Figure 9. The data confirmed that the extent of toughness degradation in the higher Nb steel was significantly less than that in the more conventional Nb bearing steel 3. Further evidence of the remarkable performance of the high Nb steel is revealed in Figure 10 where the Charpy impact transition temperature is approximately 20 °C lower than Steel 3 which can be attributed to the finer grain size and uniform CGHAZ microstructure as shown in Figure 11.

These results demonstrate the ongoing technical development that underpins the increasing versatility of steels in our society and the diligent use of microalloying to improve steel performance.

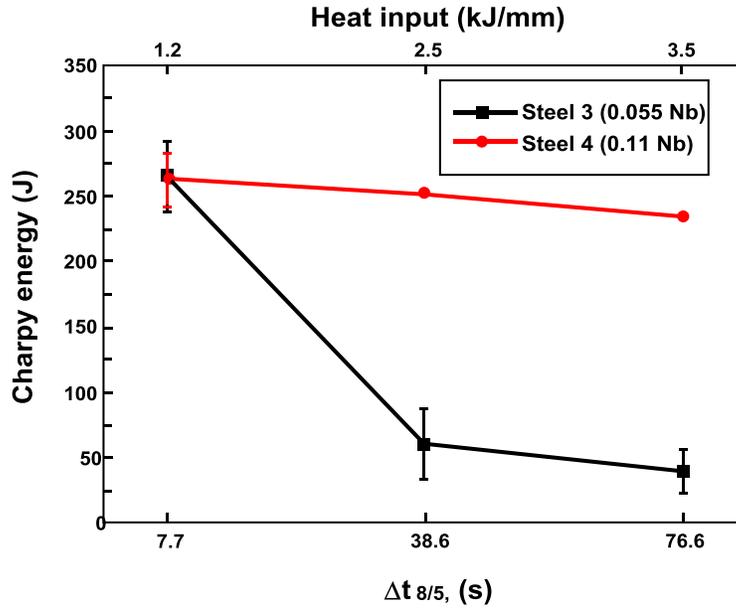


Figure 9. Comparison of Charpy impact toughness (at -20 °C) for Steels 3 and 4 as a function of ($\Delta t_{8/5}$) weld cooling time, which can be correlated with weld heat input.

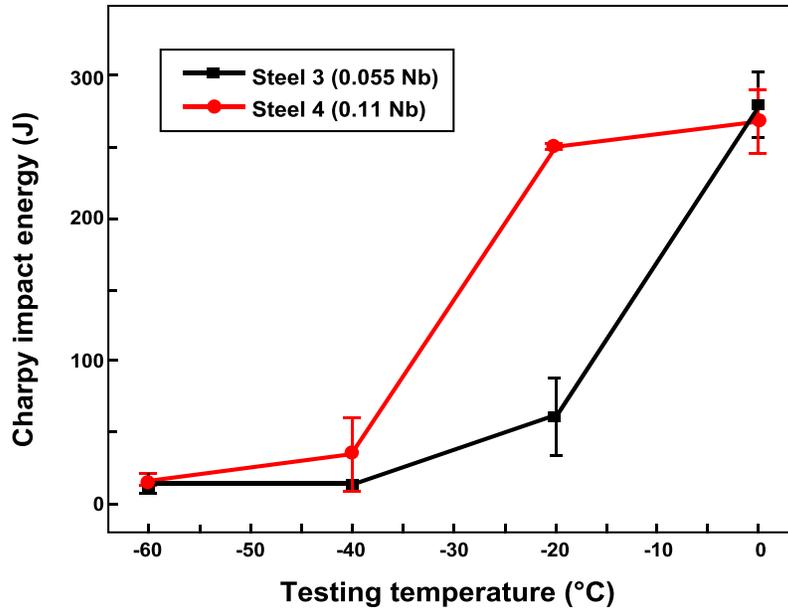


Figure 10. Ductile brittle transition curves for Steel 3 and Steel 4.

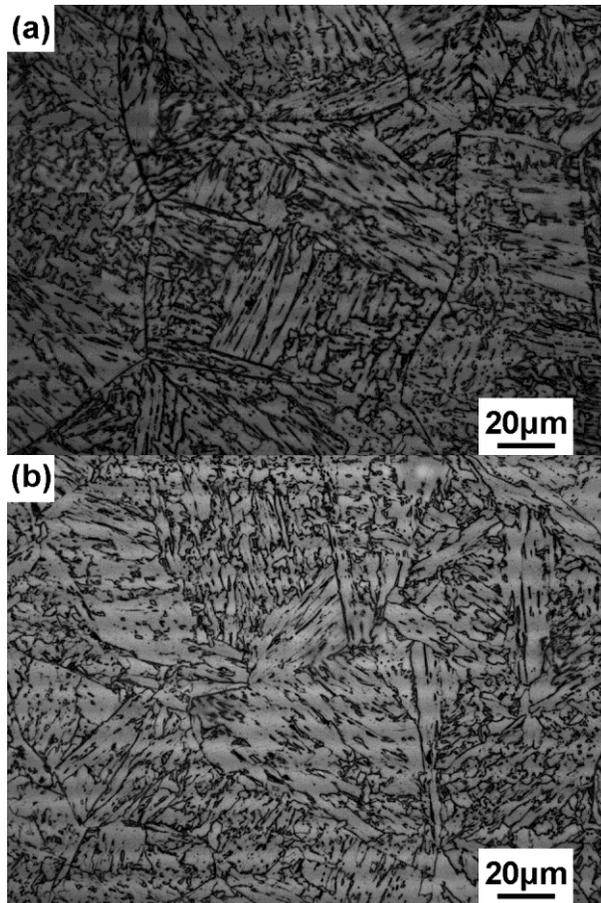


Figure 11. Representative micrographs of simulated CGHAZ;
(a) Steel 3 (0.055 Nb), (b) Steel 4 (0.11 Nb).

Conclusions

This paper has reviewed the current technology related to the use of Ti in control of weld HAZ properties in high strength steels. The additional results presented have served to highlight the influence of Nb content on HAZ grain coarsening characteristics and weldability.

Interestingly, all investigated steels contained a deliberate addition of Ti which is known to restrict austenite grain coarsening at high temperatures but the role of higher Nb and low C in this work has demonstrated an unexpected beneficial effect.

Nb-Ti microalloyed steels are known to contain complex precipitates which can include (Ti,Nb)(C,N), some can exist as a Ti-rich core and Nb-rich shell structure [31] and others as separate precipitates.

It is also well known that the dissolution of Nb rich precipitates in these steels occurs at a lower temperature than that of Ti, particularly when TiN exists. The fact that the high Nb steel demonstrated superior austenite grain size control at 1350 °C and enhanced Charpy impact toughness in simulated coarse grained HAZ samples, presents an exciting opportunity that could further incrementally improve the weldability and structural integrity of fabricated structures produced from high strength steels.

The mechanism for this performance could be related to a number of different effects which include, but are not limited to:

- Solute drag effects from the presence of Nb in solid solution, from dissolution of NbC and NbCN;
- The role of the Nb rich shell surrounding TiN precipitates, forming a barrier to the coarsening of TiN precipitates;
- Segregation of Nb to austenite grain boundaries [32] and/or decreasing grain boundary energy [33], which can thus retard grain growth.

It is clear that these recent results present an exciting opportunity to further improve the reliability of modern high strength steels and warrants further investigation which is the subject of ongoing studies at the University of Wollongong.

Acknowledgements

This work was funded by the University of Wollongong. The in-kind support from CBMM Technology, Suisse is gratefully acknowledged. One of the authors (Zhixiong Zhu) is also grateful for the financial support from the China Scholarship Council. The Energy Pipelines CRC (EPCRC), supported through the Australian Government's Cooperative Research Centres Program, is also acknowledged for its in-kind support.

References

1. F. Pickering, "Titanium Nitride Technology," *Proceedings of the 35th Mechanical Working and Steel Processing Conference (1993)*, Pittsburgh, Pennsylvania, USA, 477- 494.
2. T. Gladman and D. Dulieu, "Grain-Size Control in Steels," *Metal Science*, 8 (1) (1974), 167-176.
3. A. Batte, P. Boothby and A. Rothwell, "Understanding the Weldability of Niobium-bearing HSLA Steels," *Proceedings of the International Symposium Niobium (2001)*, Orlando, Florida, 2 - 5 December, 2001, 931-958.
4. P. Bateson et al., "Development of TMCR Steel for Offshore Structures," *Proceedings of Structural Materials in Marine Environments (1994)*, London, 226-243.
5. T. George and J. Irani, "Control of Austenitic Grain Size by Additions of Titanium," *Journal of Australian Institute of Metals*, 13 (2) (1968), 94-106.

6. F.J. Barbaro and P. Krauklis, "Intragranular Ferrite in Inoculated Low-carbon Steels," invited review paper, *Materials Forum*, 23 (1999), 77-104.
7. C.S. Smith, "Grains, Phases and Interfaces: as Interpretation of Microstructure," *Metallurgical Research and Technology*, 15 (4) (1948), 1-37.
8. B. Loberg et al., "The Role of Alloy Composition on the Stability of Nitrides in Ti-Microalloyed Steels during Weld Thermal Cycles," *Metallurgical and Materials Transactions A*, 15 (1) (1984), 33-41.
9. S. Matsuda and N. Okumura, "Effect of Distribution of Ti Nitride Precipitate Particles on the Austenite Grain Size of Low Carbon and Low Alloy Steels," *Transactions of the Iron and Steel Institute of Japan*, 18 (4) (1978), 198-205.
10. D. Fairchild, D. Howden and W.A.T. Clark, "The Mechanism of Brittle Fracture in a Microalloyed Steel: Part I. Inclusion-induced Cleavage," *Metallurgical and Materials Transactions A*, 31 (3) (2000), 641-652.
11. D. Fairchild, D. Howden and W.A.T. Clark, "The Mechanism of Brittle Fracture in a Microalloyed Steel: Part II. Mechanistic Modeling," *Metallurgical and Materials Transactions A*, 31 (3) (2000), 653-667.
12. K.S. Bang and H.S. Jeong, "Effect of Nitrogen Content on Simulated Heat Affected Zone Toughness of Titanium Containing Thermomechanically Controlled Rolled Steel," *Materials Science and Technology*, 18 (2002), 649-654.
13. J.S. Smaill, S.R. Keown and L.A. Erasmus, "Effect of Titanium Additions on the Strain-aging Characteristics and Mechanical Properties of Carbon-manganese Reinforcing Steels," *Metallurgical Research and Technology*, 3 (1976), 194-201.
14. K.S. Bang, C. Park and S. Liu, "Effects of Nitrogen Content and Weld Cooling Time on the Simulated Heat-affected Zone Toughness in a Ti-containing Steel," *Journal of Materials Science*, 41 (18) (2006), 5994-6000.
15. S.C. Wang, "The Effect of Titanium and Nitrogen Contents on the Microstructure and Mechanical Properties of Plain Carbon Steels," *Materials Science and Engineering A*, 145 (1) (1991), 87-94.
16. J.K. Choi, "Development of High Strength and High Performance Steels at POSCO through HIPERS-21 Project," *Proceedings of the 1st International Conference on 'Super-high Strength Steels' (2005)*, 2nd-4th November, Rome, Italy: Associazione Italiana di Metallurgia.
17. Z. Zhu et al., "Influence of Ti/N Ratio on Simulated CGHAZ Microstructure and Toughness in X70 Steels," *Science and Technology of Welding and Joining*, 18 (1) (2013), 45-51.

18. Z. Zhu et al., "Role of Ti and N in Line Pipe Steel Welds," *Science and Technology of Welding and Joining*, 18 (1) (2013), 1-10.
19. G.F. Bowie, R.M. Smith and F.J.Barbaro, "The Influence of Micro Titanium Additions on the Weld Heat Affected Zone Hardness of C-Mn Structural Steels," *Proceedings of the International Conference Welding 90 (1990)*, Geestacht, 219.
20. J.M. Gray, "Transformation Characteristics of Very-Low-Carbon Steels," ed. F.J. Barbaro, (Wollongong, 1969).
21. L. Ji et al., "Research on Key Technology and Production Quality of X80 Linepipe for the 2nd West-East Gas Pipeline," *Proceedings of the International Seminar on X80 and Higher Grade Line Pipe Steel (2008)*, Xi'an, China.
22. W.J. Fazackerley, P.A. Manuel and L. Christensen, "First X-80 HSLA Pipeline in the USA," *Proceedings of the International Symposium on Microalloyed Steels for the Oil and Gas Industry (2006)*, Araxá, Brasil.
23. K. Hulka and J.M. Gray, "High Temperature Processing of Line Pipe Steels," *Proceedings of the International Symposium Niobium (2001)*, Orlando, Florida, 2 - 5th December, 2001.
24. H. Mohrbacher, "Grain Size Control by Niobium Microalloying in Gear Steel during High Temperature Carburizing" (NiobelCon bvba, 2970-Schilde, Belgium).
25. S. Vervynckt et al., "Study of the Austenite Recrystallization - Precipitation Interaction in Niobium Microalloyed Steels," *The Iron and Steel Institute of Japan International*, 49 (6) (2009), 911-920.
26. K. Hulka, P. Bordignon and J.M. Gray, "Experience with Low Carbon HSLA Steel Containing 0.06-0.10 Percent Niobium," *Proceedings of the International Seminar The HTP Steel Project (2003)*, Araxá, Brasil.
27. B. Ouaisa et al, "Investigations on Microstructure, Mechanical Properties and Weldability of a Low-carbon Steel for High Strength Helical Linepipe," *Proceedings of the 17th Joint Technical Meeting (2009)*, Milan, Italy.
28. E. Palmiere, C. Garcia and A. De Ardo, "Compositional and Microstructural Changes which Attend Reheating and Grain Coarsening in Steels Containing Niobium," *Metallurgical and Materials Transactions A*, 25 (2) (1994), 277-286.
29. L. Cuddy and J. Raley, "Austenite Grain Coarsening in Microalloyed Steels," *Metallurgical and Materials Transactions A*, 14 (10) (1983), 1989-1995.

30. K.A. Alogab, D.K. Matlock and J.G. Speer, "Abnormal Grain Growth During Heat Treatment of Microalloyed-Carburizing Steel," *Proceedings of the Conference on New Developments on Metallurgy and Applications of High Strength Steels*, Buenos Aires, Tenaris Center for Industrial Research, 2008.
31. A. Craven et al., "Complex Heterogeneous Precipitation in Titanium-niobium Microalloyed Al-killed HSLA Steels--I.(Ti, Nb)(C, N) Particles," *Acta Materialia*, 48 (15) (2000), 3857-3868.
32. E. Essadiqi and J.J. Jonas, "Effect of Deformation on the Austenite-to-ferrite Transformation in a Plain Carbon and Two Microalloyed Steels," *Metallurgical and Materials Transactions A*, 19 (3) (1988), 417-426.
33. B. Vynokur, "Influence of Alloying on the Free Energy of Austenitic Grain Boundaries in Steel," *Materials Science*, 32 (2) (1996), 144-157.