THE ROLE OF NIOBIUM IN LOW CARBON BAINITIC HSLA STEEL

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ABSTRACT

With higher strength, weight reduction can be achieved. Besides the required tonnage, formability and weldability have an important influence on the costs of a construction. Furthermore, sufficient toughness is a pre-requisite to guarantee structural integrity. A low carbon bainitic microstructure offers a good combination of these properties for steel with yield strength above 500 MPa.

Thermomechanical rolling followed by accelerated cooling is the standard production route for steel with high strength and good toughness. This processing route relies on niobium microalloying, which promotes a finer grain size via austenite conditioning. With reduction of the carbon below 0.05 %, higher amounts of niobium (typically up to 0.10 %) can be used effectively. With this alloy design the recrystallization stop temperature is shifted to higher levels. Furthermore, the transformation gets retarded and thus bainite formation is promoted, and also strength increase via niobium carbide ferrite precipitates becomes important.

The balance of niobium in solid solution and as precipitate is shown over the rolling process as well as the final microstructure as result of the processing conditions. Properties of low carbon bainitic plate, strip and pipe are shown. Especially the optimization of the heat affected zone toughness for various welding conditions is being discussed. Even though large diameter pipes are dominating the actual application, but this alloy design has also a big potential in many other constructions.

KEYWORDS

Low carbon bainite, acicular ferrite, high strength low alloy steel, thermomechanical rolling, niobium microalloying, heat affected zone toughness

INTRODUCTION

High strength low alloy (HSLA) steel has been defined as a material where the properties (strength, fracture toughness, formability and weldability) are balanced in such a way, that the reliability and overall costs are optimized [1]. Thus, their strength is considered ‘high’ only compared to mild steel and their domain is in the yield strength range of 350 to 700 MPa.

Figure 1 shows the development of pipeline steel as an example of HSLA steel development. The application as large diameter pipe was a frontrunner and driving force in the development of HSLA steel, because the pipe operators asked for higher transportation capacity already in the 1960’s for economical reasons. Since higher operating pressure simultaneously asks for improved toughness and higher impact energy in order to avoid brittle and long running ductile cracks, the increase in strength had to be assisted by better toughness data, which could be fulfilled only by refining the microstructure and reducing the amount of second phases, such as inclusions and pearlite. Therefore

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a high cleanliness, represented e.g. by a low sulphur content, and a low carbon content are prerequisites in HSLA steel.

Furthermore, the economy of steel production has been also improved by substituting the heat treatments (normalizing, quenching plus tempering) by controlled rolling processes and so guaranteeing the properties already in the as rolled condition. Several specific rolling and cooling regimes have been developed to achieve this goal.

Actually HSLA steel is widely spread and besides large diameter pipes many other steel constructions and also automobiles apply this concept, guaranteeing steel to maintain its position as most relevant metallic material.

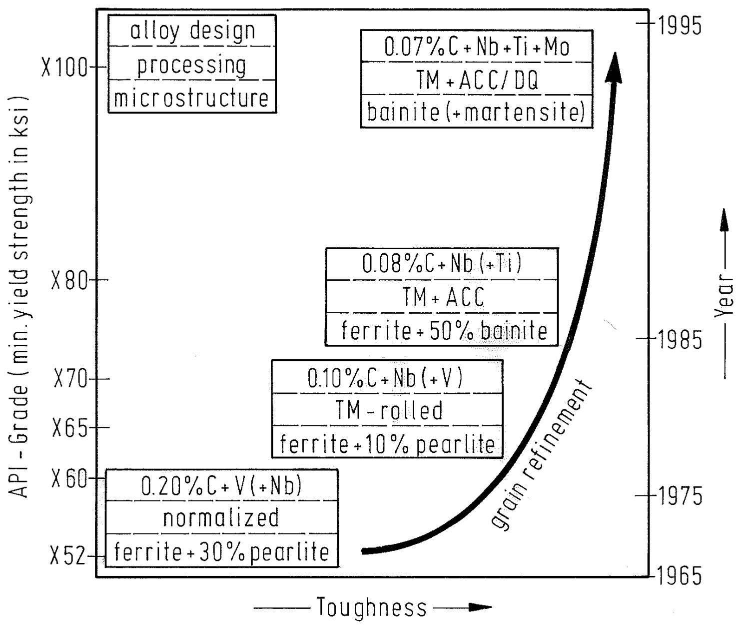

Fig. 1: Pipe steel development

The metallurgical mechanism in the development of HSLA steel is to maximize grain refinement, since this is the only means to improve both, strength and toughness of steel. Furthermore, by substituting carbon by other strengthening mechanism, a further increase in toughness, formability and weldability is obtained.

The majority of high strength low alloy steels are actually produced by thermomechanical rolling, a process, which is characterized by a final deformation in the lower austenite region without recrystallization. By this means an enhanced number of nucleation sites exists for the γ to α transformation and so a fine-grained microstructure is guaranteed, fulfilling the demands in strength and safety considerations of a construction.

Up to a yield strength level of about 500 MPa, one typically relies on ferrite plus pearlite microstructures, while higher strength steels need to have bainitic constituents and steel with about 700 MPa yield strength is typically 100% bainitic. Figure 2 shows schematically the microstructure development of austenite during thermomechanical rolling and the resulting microstructure after transformation: dependent on the alloy content and the applied cooling rate, the volume fraction of the microstructural constituents (ferrite, pearlite, bainite, martensite) is different, and there exists one condition, where 100 percent fine grained lower bainite is being formed. In case of low carbon steel, there will be very little or even no cementite precipitated in the bainite and this carbide-free bainitic constituent is often called acicular ferrite.

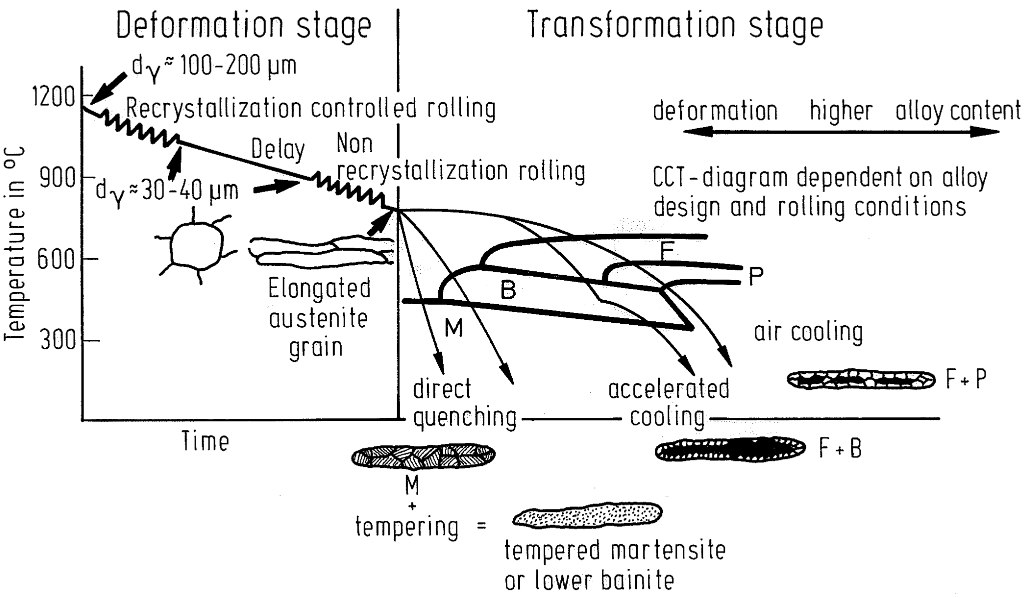

Fig. 2: Schematic presentation of thermomechanical rolling with in-line cooling

Figure 3 [2] underlines the great benefits obtained, when using a bainitic microstructure, which exhibits a much finer effective grain size and a higher dislocation density. The characteristic features of the investigated ferrite and bainite were obtained from thermomechanically rolled and accelerated cooled plate. With higher amounts of bainite one achieves higher strength by further grain refinement and higher dislocation density and this without impairing toughness, when the finer grain size will compensate the toughness deterioration by dislocation hardening.

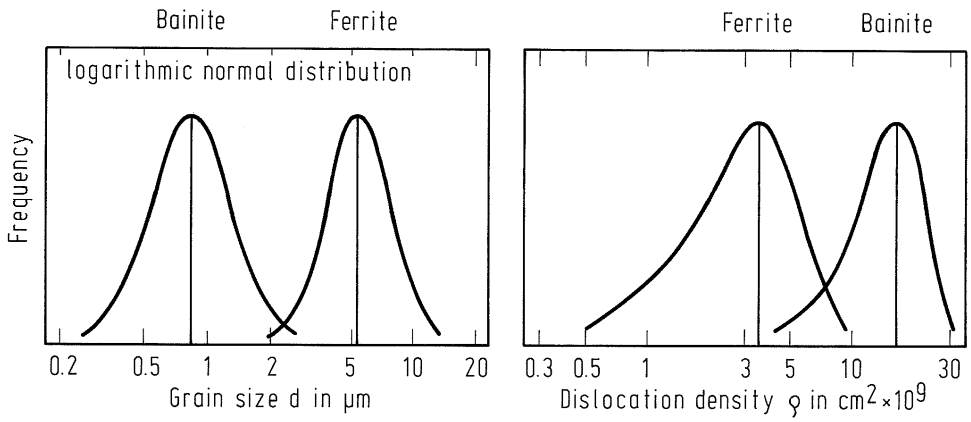

Fig. 3: Ferrite and acicular bainite in thermomechanically rolled and accelerated cooled plate

1. MICROSTRUCTURE-PROPERTY RELATIONSHIPS

In order to compare the mechanical properties of steels with various microstructures, research has been carried out, where either the cooling rate or the chemical composition of steel has been varied to produce different microstructures. The final goal of this investigation was to develop a chart,

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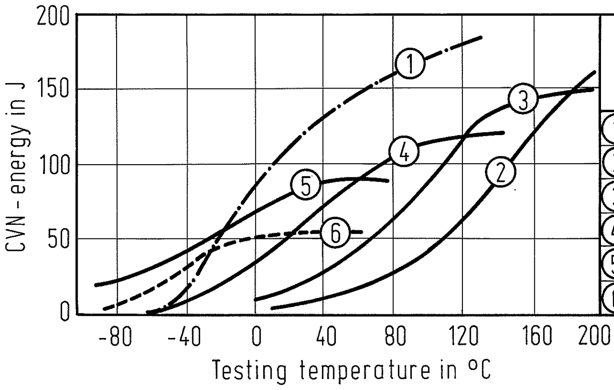
which characterizes bainitic microstructures by optical metallography in such a way, that it allows a correlation with the mechanical properties. This approach has been summarized elsewhere [3].

One example is given in figure 4: It is well known that the strength (or hardness) increases with a higher cooling rate, when the microstructure changes from ferrite plus pearlite to bainite and martensite. While the upper shelf energy becomes lower with higher hardness, there is not such a simple correlation regarding the impact transition temperature: one observes the worst and the best transition temperature within the bainitic microstructures and these two extremes are the granular and the fine acicular bainite. As result of a lower transformation temperature, there is a continuous transition from granular bainite via coarse grained to fine grained acicular bainite and these differences can be distinguished by optical microscopy. This proposed classification is different to this between upper and lower bainite, which needs the higher magnification of an electron microscope.



	Cooling rate $t_{8/5}$ in s	Microstructure	HV10
1	20000	ferrite + pearlite	290
2	2500	granular bainite	320
3	300	granular bainite	330
4	100	coarse acicular bainite	380
5	30	fine acicular bainite	420
6	10	martensite	450

Fig. 4: Microstructure and mechanical properties as result of the cooling rate from 930°C (0.18%C, 0.7%Si, 0.9%Mn, 0.9%Cr, 0.45%Mo)

The correlation of toughness and microstructure is also shown in another diagram, figure 5, where the manganese content has been increased to lower the transformation temperature at a given cooling rate. It underlines that the ‘effective grain size’ determines the toughness: in a ferritic plus pealitic microstructure the effective grain size is that of the ferrite grain and in the acicular

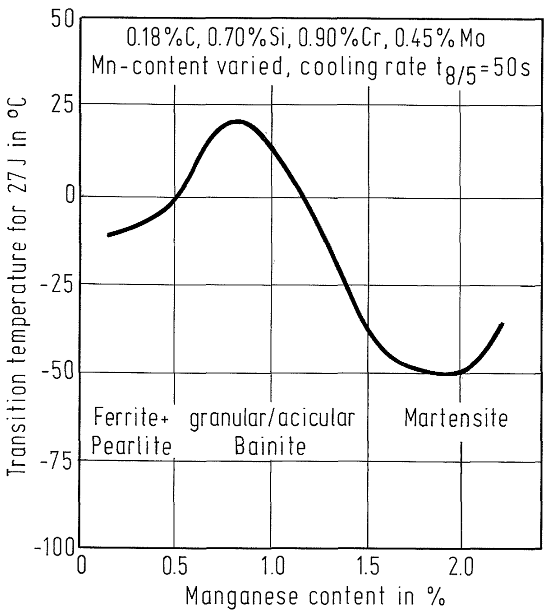


Fig. 5: Influence of microstructure on toughness

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microstructures (acicular bainite and martensite) it is the width of the needles. In a granular bainite, however, no clear needles can be distinguished, and the effective grain size correlates with the austenite grain, which is naturally larger than that of a ferrite grain. Therefore, the poorest toughness is obtained with granular bainite and the best toughness with fine acicular bainite. Even though the martensite shows an even finer needle width, the hard particles in the martensite cause a certain deterioration of toughness.

2. THE ROLE OF NIOBIUM TO INFLUENCE THE MICROSTRUCTURE

When the amount of solute niobium is increased, retardation of austenite recrystallisation (austenite conditioning) is observed at significantly higher temperatures, Figure 6 [4], thereby allowing the benefits of thermomechanical rolling to be achieved already at higher temperatures. Low carbon contents and the fixing of nitrogen with titanium, an element with higher affinity for nitrogen than niobium and thus preventing niobium carbo-nitride formation, allow a higher niobium content to be easily dissolved during reheating for rolling, Figure 7 [5,6].

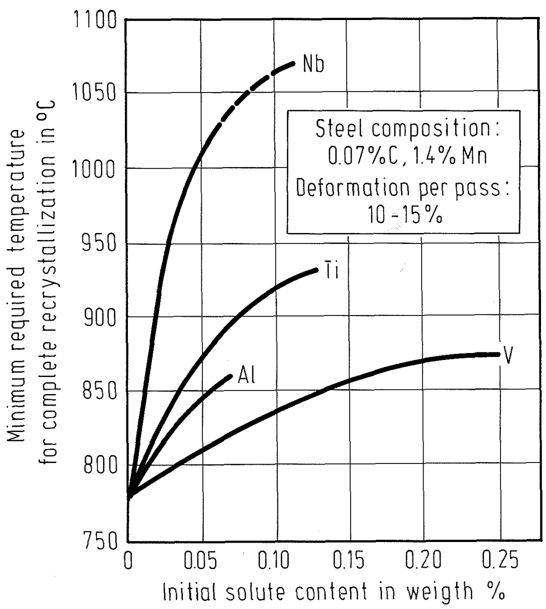

Fig. 6: Retardation of recrystallisation by microalloying

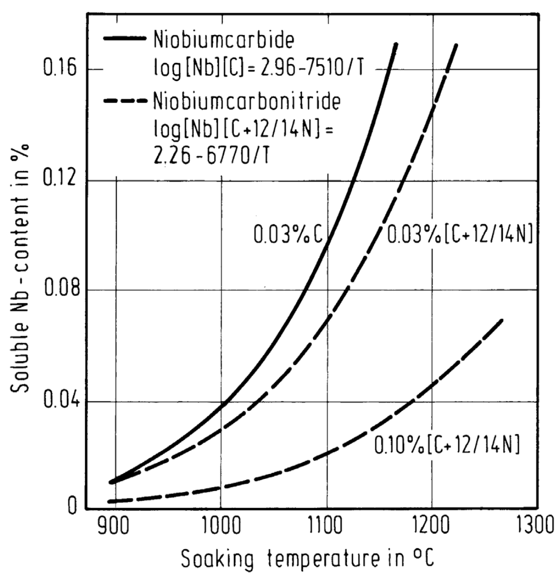

Fig. 7: Solubility of niobium carbide and niobium carbo-nitride

During rolling in austenite a portion of the niobium will precipitate strain induced, especially on dislocations. The chemical extraction technique allows investigating the status of niobium at finish rolling temperature. Besides the equilibrium condition, results for a conventional pipe steel with about 0.10 %C and a 0.03 %C - 0.10 %Nb steel are shown in Figure 8 [7]: The conventional pipe steel, processed on a plate mill with a typical finish rolling temperature below 800 °C, allows only a small portion of niobium to remain in solid solution and the amount of 'soluble' niobium is close to the equilibrium condition. A higher 'soluble' niobium content of almost 0.02 % is observed in hot strip, where the final deformation passes are continuous, asking for higher rolling speed and shorter inter-pass times, and the finish rolling temperature is about 100 °C higher than typical for plate rolling. In contrast to the conventional pipe steel the 0.03 %C - 0.10 %Nb, Ti-treated steel shows a 'soluble' niobium content as high as 0.04 % for typical finish rolling temperatures in a plate mill, with even higher values for hot strip processing.

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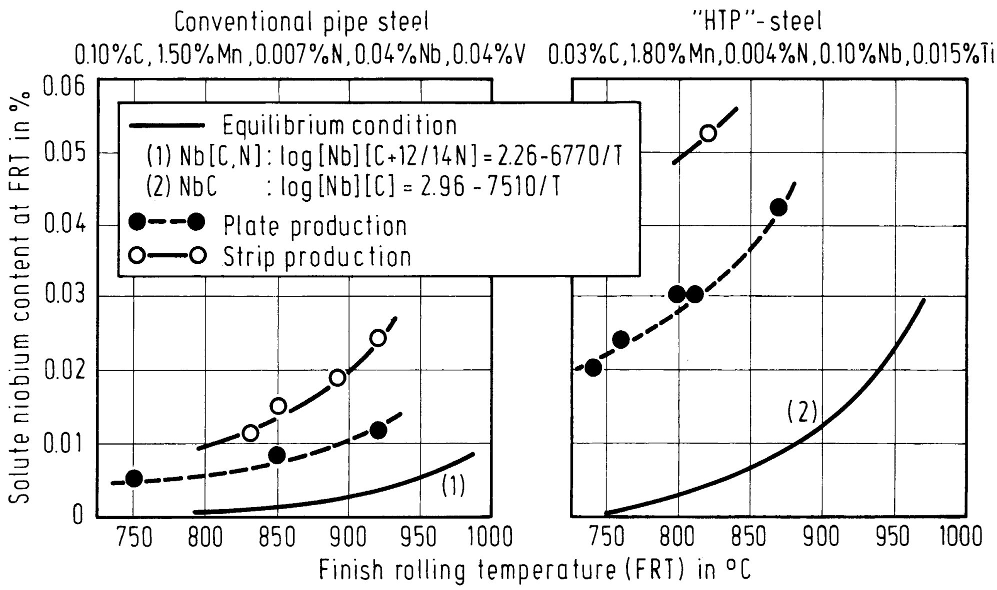

Fig. 8: Niobium in solid solution at different finish rolling temperatures for two pipe steels, determined by the chemical extraction method

Niobium in solid solution has also an effect in lowering the γ to α transformation temperature: Figure 9 [8] confirms, that solute niobium has the strongest effect in lowering the transformation temperature among commonly used microalloying elements, and this effect gets even more pronounced with a higher cooling rate. The lowering of transformation temperature has a pronounced effect on the microstructure of thermomechanically rolled plus accelerated cooled plates by refining the grain size of polygonal ferrite and by increasing the volume fraction of bainite.

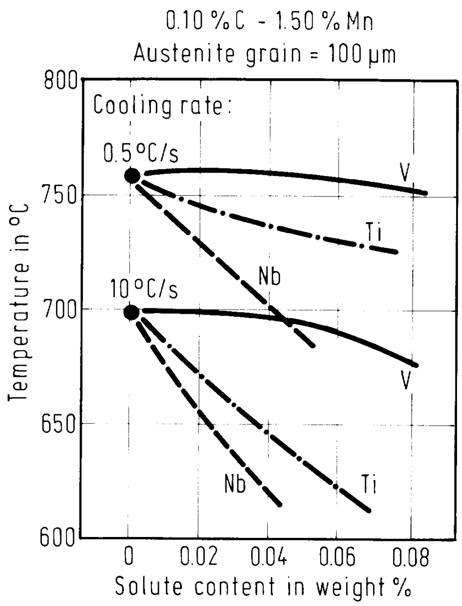

Fig. 9: Ar_3 temperatures of microalloyed steels with equal austenite grain size

Since all diffusion controlled processes - including the γ to α transformation - are retarded more effectively, the bigger the difference in the atom size of the base element compared to any other element in solid solution is, it makes sense to compare the atom radii of the elements of interest:

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The niobium atom is 15.6 % bigger than iron, while the difference of the two other microallying elements titanium or vanadium is lower, +14.8 % and +6.2 %, respectively. Since the difference in the atom size of molybdenum to iron, +9.4 %, is also lower than that of niobium, the higher effectiveness of solute niobium compared to molybdenum to increase hardenability becomes obvious.

Niobium in solid solution at the finish rolling temperature is available for the formation of niobium carbide precipitates in ferrite, which have the appropriate size for additional strength increase via precipitation hardening. This strength increase amounts to about 100 MPa for 0.03 % niobium in solid solution at finish rolling temperature, when the particle size of the ferrite precipitates is 1.5 to 2 nm [9]. These small particles are difficult to determine and just particles with a size of 4 nm and more can be found by transmission electron microscopy. Figure 10 shows an example [10] and in this work it has been confirmed, that these particles are cubic NbC not containing any other element than niobium and carbon having a lattice parameter of 0.446 nm.

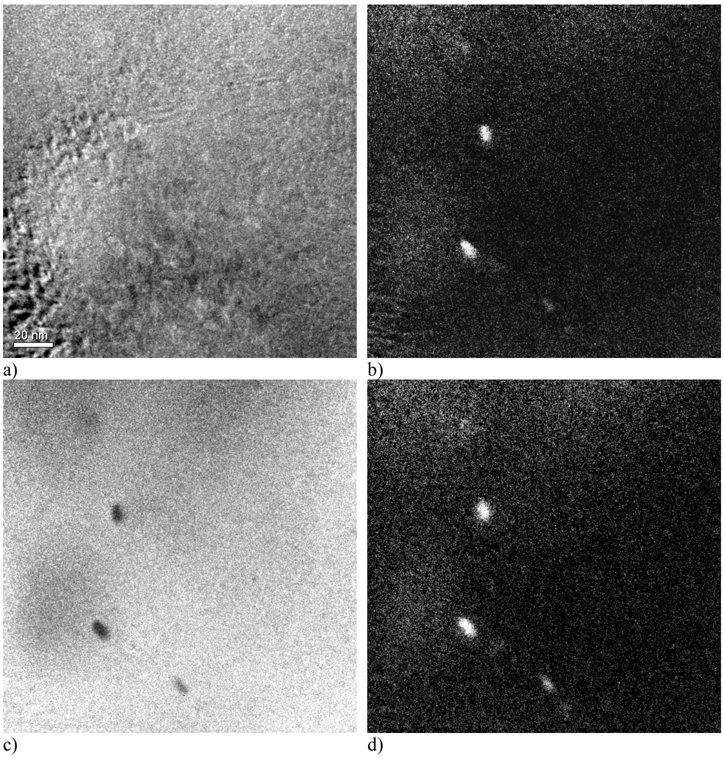

Fig. 10: Bright field TEM of niobium carbide ferrite precipitates (a) and elemental distribution images of Nb (b), Fe (c), and C (d)

3. MECHANICAL PROPERTIES OF LOW CARBON BAINITIC STEEL

Two major demonstration trials with participation of steel companies all over the world have been carried out using clean steel with 0.03 %C – 0.10 %Nb and different levels of manganese and other alloying elements like Cu, Cr and Ni. [11,12]. Since this alloy concept allows thermomechanical rolling at higher temperatures than usual, it is also called HTP steel (HTP = high temperature processing). Within these trials a big variety of rolling schedules and cooling conditions have been applied including finish rolling in the two-phase region and different cooling regimes.

All plate or hot strip had in common, that the toughness was excellent and similar for various strength levels and various processing routes regarding both, a very low transition temperature from

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ductile to brittle fracture, determined by the Batelle drop weight tear (BDWT) test and a high Charpy - V- notch energy. Figure 11 shows one example and these outstanding results, indicating that no brittle fracture occurred at test temperatures above –40 to –50 °C and the Charpy –V- notch energy was above 300 J at testing temperatures above –80 to –90 °C.

That there exists no influence of the finish rolling temperature on the toughness is of course not unlimited: it recently has been defined [13] that the ductile to brittle fracture transition temperature in the BDWT test is being deteriorated, when the finish rolling temperature is above 920 °C, even though no deterioration is observed in Charpy –V- notch testing. Such finish rolling temperature is however about 150 to 200 °C higher than in traditional thermomechanical rolling.

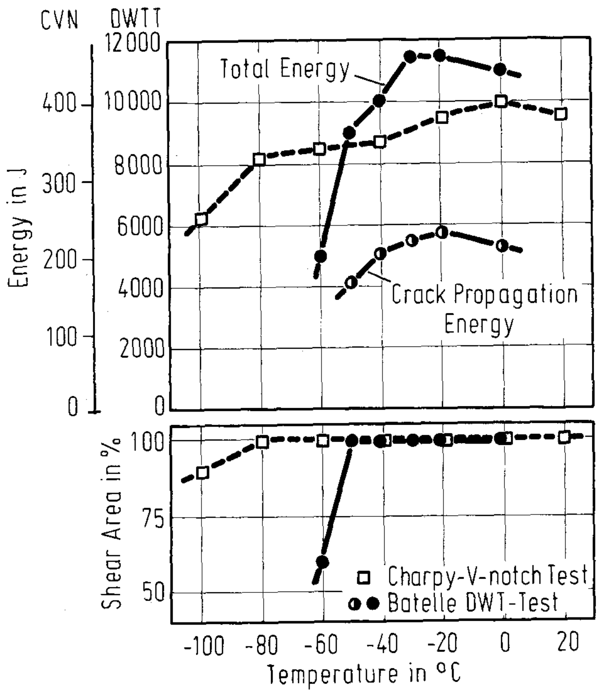

Fig. 11: Results of CVN and BDWTT impact test versus testing temperature of 18 mm HTP plate (FRT = 820 °C)

The processing conditions have a remarkable influence on the yield and tensile strength:

Figure 12 shows results of plates, where different cooling regimes have been applied, as well as the results of hot strip production. After air-cooling, the microstructure consists of polygonal ferrite and very small amounts of pearlite and/or bainite and HTP steel with about 1.75 %Mn results in yield strength of about 520 MPa. When after transformation, e.g. at a temperature of 550 °C, slow cooling is applied, ferrite precipitates are promoted, giving an additional strength increase by precipitation hardening. Under practical conditions this strength increase is about 40 to 50 MPa. Accelerated cooling after rolling further refines the ferrite grain size and promotes a higher volume fraction of bainite to be formed. The correlated yield strength increase amounts to about 100 MPa and pipe steel X 80 can be achieved easily this way.

It has been shown, that both, a higher cooling rate and a higher alloy content, especially a higher manganese content, promote the formation of finer grained bainite and thus add to strength increase [12]. The excellent strength – toughness combination of HTP steel is also applied in the production of thick wall plate, and e.g. 50 mm plate shows yield strength of 520 MPa after accelerated cooling (cooling rate 8°C/s), when the manganese content is 1.75 %.

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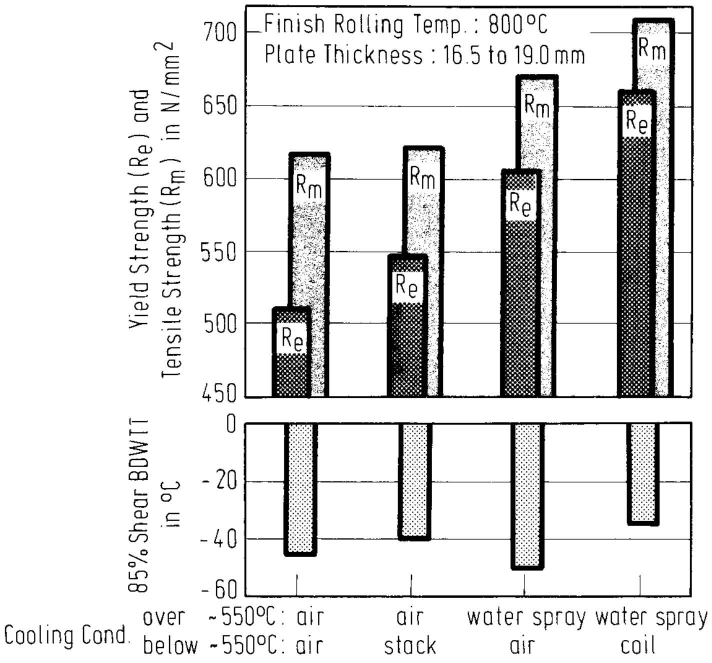

Fig. 12: Influence of cooling regime on the mechanical properties of HTP plate/skelp

Such low carbon steel has a comparably high transformation start temperature, which is further increased by the fact that the transformation starts from deformed austenite. Thus traditional finish rolling temperatures for HSLA steel (e.g. 720 to 750 °C) result in the fact, that the finish rolling temperature is already in the two phase region $\alpha + \gamma$ and not any more in the metastable austenite. This is connected with a remarkable increase in yield and tensile strength as result of cold deformation of the already formed ferrite as shown in figure 13. Different to HSLA steel based on higher carbon content, this cold deformation has no negative impact on the toughness and no impaired Charpy –V data are observed than reported above.

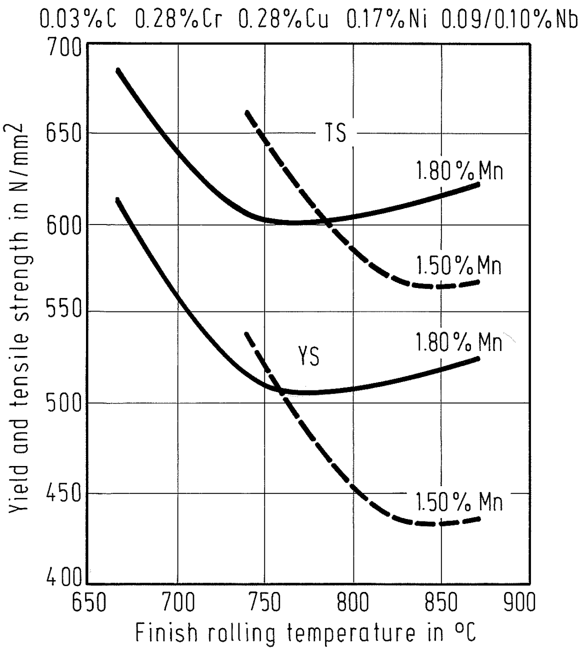

Fig. 13: Strength increase of HTP steel by rolling in the two-phase region

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4. TECHNOLOGICAL PROPERTIES

Figure 14 shows the time – temperature – transformation diagram of HTP steel after reheating to a peak temperature of 1350 °C, simulating the grain coarsened heat affected zone (HAZ) and helping to classify the <u>*weldability*</u> of this steel. The diagram is characterized by a wide range of cooling conditions resulting in bainitic microstructures, ranging from < 3 °C/s to >100 °C/s, covering the cooling conditions of various welding processes leading from high heat input welding, via submerged arc welding to manual field welding. Even if higher cooling rates than 100 °C/s occur, typical for electron beam or laser welding, the hardness in the low carbon martensite remains on a level below 300 HV. Consequently the toughness in the heat-affected zone is excellent for many welding processes, as shown in figure 15 and by far superior to toughness data observed in the HAZ of conventional HSLA steel.

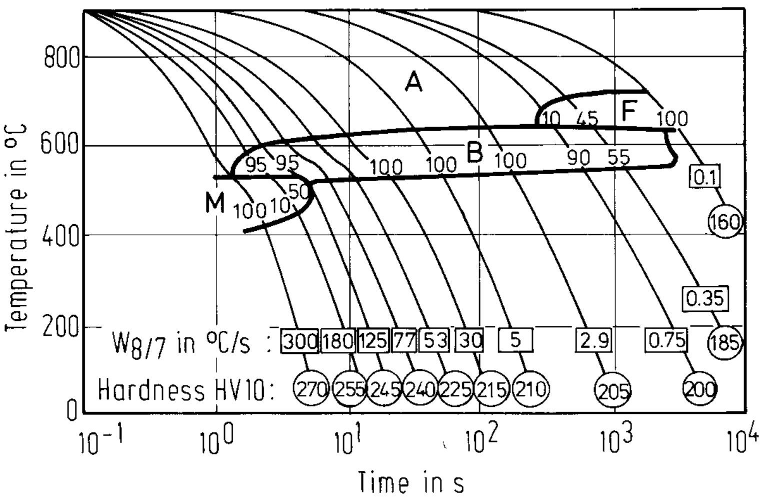

Fig. 14: Transformation behaviour for simulated HAZ (peak temperature 1350 °C) of HTP steel with 0.03 %C – 0.10 %Nb and 1.75 %Mn

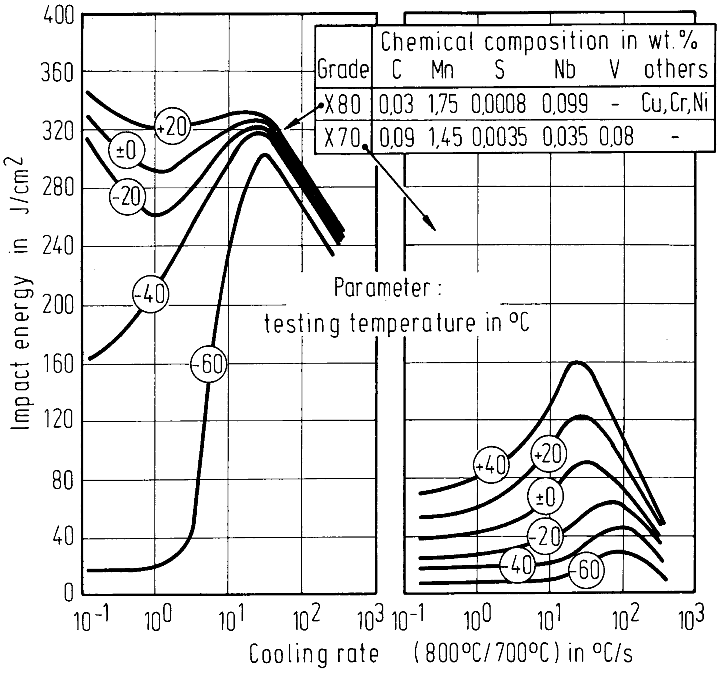

Fig. 15: Charpy –V – notch impact energy of two pipe steels in the grain coarsened heat affected zone; welding simulation with a peak temperature of 1350 °C

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New oil and gas pipelines transporting material from deeper wells often ask for resistance against *hydrogen induced cracking (HIC).* This failure originates by the recombination of atomic hydrogen to H_2 at elongated inclusions and its propagation follows hard phases (e.g. pearlite) in the microstructure. With clean steel being a pre-requisite, the low carbon helps avoiding a banded microstructure, since any interdendritic enrichment of alloying elements during solidification is naturally reduced. The iron – carbon phase diagram in figure 16 explains schematically the reduced segregation with low carbon steel, since:

1. the small interval of liquidus to solidus temperature reduces any segregation already during solidification and
2. the big interval in the δ-ferrite allows a remarkable reduction of the crystal segregation during cooling after solidification

HSLA steel with about 0.03 %C can tolerate manganese content of 1.70 % maintaining to be HIC resistant as shown in figure 17. Using accelerated cooling after rolling, even the tensile properties of heavy wall X 70 pipes can be guaranteed with this alloy design.

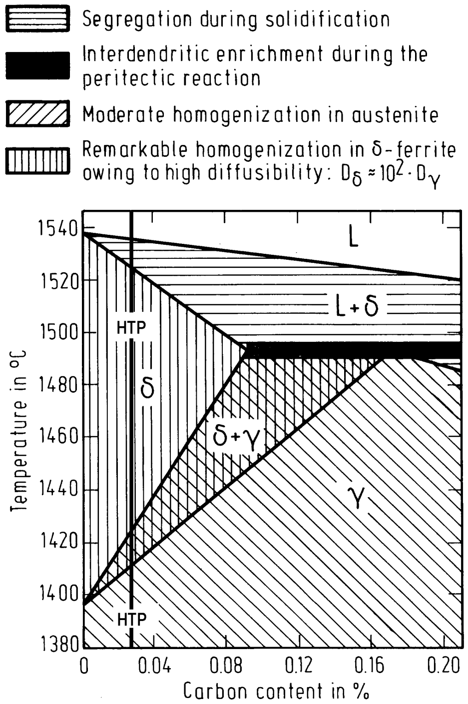

Figure 16: Part of the Fe-C diagram with classification of the segregation severity

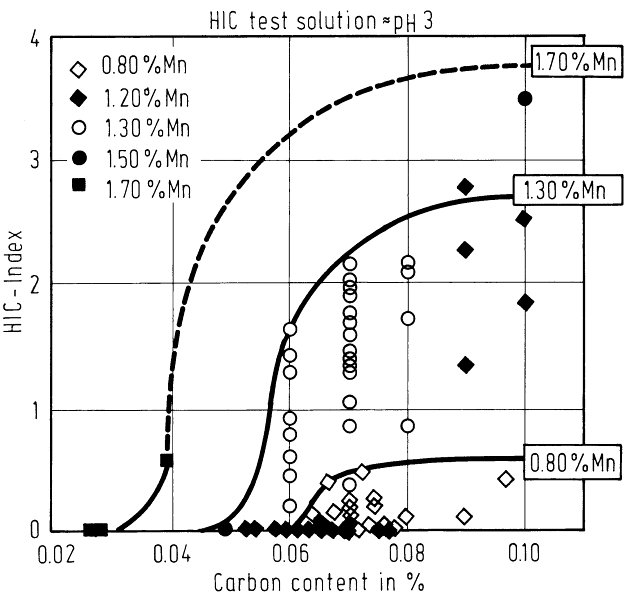

Figure 17: Influence of carbon and manganese on HIC resistance

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Since the stress-strain curve of such bainitic steels do not show any Lüders plateau, any cold deformation results in an increase of the yield strength. This is especially relevant in pipe steel production, where ferrite-pearlite steel typically exhibit a yield strength drop after pipe forming as result of the Bauschinger effect. Also for many automotive parts good *cold formability* is demanded. While bending operations are mainly ask for excellent elongation data, the process hole expansion is mainly correlated to the homogeneity if the microstructure and also there the low carbon bainite offers the best values among various possible microstructure with hole expansion ratios of about 150 % [14].

5. CONCLUSION

Fine acicular bainitic microstructures show an excellent combination of strength and toughness. Such microstructures can be achieved via thermomechanical rolling in combination with accelerated cooling processes by tuning the chemical composition accordingly. Very positive results can be achieved with low carbon (0.03 %) and comparably high niobium contents (0.10 %). With such low carbon content, the final microstructure becomes very homogeneous, helping to improve HIC resistance and hole expansion behaviour. Furthermore, impact toughness in the base metal and the heat-affected zone for a big variety of welding processes becomes outstanding. With the reduced carbon level also an increased niobium content becomes effective, allowing austenite processing to occur already at higher temperatures and assisting in promoting a bainitic microstructure with fine NbC precipitates for additional strength increase.

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