

## HIGH STRENGTH PIPELINE STEELS WITH OPTIMIZED HAZ PROPERTIES

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### Abstract

Large diameter linepipes for long-distance transport of natural gas are produced using either heavy gauge coil or heavy plate material. The combination of high strength and excellent low-temperature toughness of modern linepipe steels is a direct result of the thermomechanically controlled processing (TMCP), which leads to a fine grain size after rolling. Because TMCP allows attainment of high levels of strength by reducing the grain size, it was possible to lower the C content and to improve the weldability significantly. Addition of Nb is an effective measure to inhibit recrystallization during finish rolling, which is the key to obtaining a fine-grained microstructure. The temperature threshold below which recrystallization is severely retarded between rolling passes depends on the level of the Nb addition and accumulated strain. Thus, the level of the Nb content can be adjusted to the limitations of the rolling mill. Double submerged arc welding during the production of large-diameter linepipes leads to severe changes in the microstructure of the heat-affected zone (HAZ), including grain coarsening by more than one order of magnitude, as well as different transformation products. Because of the large austenite grain size close to the fusion line, the phase transformation during cooling is retarded and C-rich particles can form. This can have a negative effect on the toughness in the HAZ. An experimental investigation was carried out at Salzgitter Mannesmann Forschung GmbH (SZMF), in which the Nb content of laboratory heats was varied between 0.02% and 0.10%. These heats were thermomechanically rolled to a wall thickness of 25 mm and subsequently used for double-layer submerged arc welding trials. The processing parameters during rolling and welding were held constant in order to ensure that the effect of the alloying elements could be isolated. The fusion line toughness was determined at -20 °C as well as -40 °C and the microstructure was investigated by high-resolution scanning electron microscopy. It was found that high levels of toughness in the heat-affected zone could be achieved across the full range of Nb studied.

## **Introduction**

Modern high-strength heavy plates used in the production of UOE pipes are generally produced by thermomechanical rolling, followed by accelerated cooling (TMCP/AC). This processing route results in a microstructure that consists predominantly of bainite. The combination of high strength and high toughness of these steels is a result of the microstructure realized by TMCP and is strongly influenced by the steel composition and rolling and cooling conditions. Continuous alloy and process development has made it possible to achieve high strength levels at C levels below 0.1% in combination with Mn levels below 2.0%, which led to a significantly improved weldability. These steels are typically microalloyed with Ti and Nb, in order to inhibit grain growth during reheating and recrystallization during rolling, which leads to a fine grain size after TMCP. The temperature threshold below which recrystallization is severely retarded between rolling passes depends on the level of the Nb addition and accumulated strain. Thus, the level of the Nb content can be adjusted to facilitate heavy plate production [1]. The level of other alloying elements strongly depends on the desired strength level and the wall thickness.

For the production of UOE pipes, submerged arc welding (SAW) in two passes is the most economic solution. However, the thermal cycles during high heat input SA welding induce a significant modification of the microstructure in the heat affected zone (HAZ) and, therefore, can lead to a critical change of local mechanical properties in terms of toughness and strength.

In order to investigate the impact of Nb on the HAZ properties, welding trials were carried out on laboratory heats with Nb contents of up to 0.105% using high heat input SAW. Charpy V-notch tests, as well as hardness tests, were performed to clarify the correlation between the Nb content and HAZ properties. Finally, the fusion line microstructure was investigated in terms of the volume fraction of C-rich constituents by high resolution scanning electron microscopy, to correlate the microstructure in the HAZ with the HAZ toughness.

### **Requirements on Linepipe Steels and HAZ Properties**

High strength linepipe steels offer an economic advantage compared to lower strength grades as they allow a reduction in wall thickness of the pipe at the same operating pressure. This leads to savings with regard to raw materials, transportation and field welding. Conversely, they allow an increase of the operating pressure at the same wall thickness. Since the first application of X80 large-diameter pipes more than 25 years ago, the combination of mechanical properties required by customers has become increasingly complex, due to the strong worldwide interest to develop remote natural gas resources in hostile environments. These can include pipeline operation under arctic conditions, ie. at temperatures of -40 °C or below, or in areas with ground movement. High deformability and low-temperature toughness are therefore critical requirements in order to ensure pipeline safety. The optimization of the toughness has therefore been a strong focus of development, both in the case of the base metal and the HAZ.

In pipe manufacturing, special attention has to be paid to the welding as an essential aspect for the whole pipeline safety. From the economic point of view, multi-wire submerged arc welding in two passes (DSAW) is the most sensible welding technique for medium and high wall thicknesses. However, besides the economic advantage, the DSAW process leads to various challenges in terms of weld properties. The high heat input induces long cooling times which are associated with unfavorable microstructure modifications and reduced HAZ toughness [2]. The essential parameter for characterization of the HAZ properties is the cooling condition after welding, or more exactly the cooling time between 800 °C and 500 °C ( $t_{8/5}$  time). In general, the impact energy of the HAZ decreases and the transition temperature is shifted to higher values with increasing  $t_{8/5}$  time [3,4]. For DSAW of large diameter pipes, heat input values above 4 kJ/mm are employed, which corresponds to a  $t_{8/5}$  time in the HAZ above 50 seconds. Figure 1 shows an example of a numerically calculated temperature-time cycle for the HAZ of the second weld layer. Apart from the cooling conditions, the microstructure and toughness of the HAZ are strongly influenced by the peak temperature. With regard to the resulting microstructure modifications, the HAZ can be divided into four different zones [5]:

- Sub-critically reheated HAZ (SCHAZ) – temperature below  $A_{c1}$ ;
- Inter-critically reheated HAZ (ICHAZ) – temperature between  $A_{c1}$ - $A_{c3}$ ;
- Grain-refined HAZ (GRHAZ) – temperature above  $A_{c3}$ , with no extensive grain growth;
- Coarse-grained HAZ (CGHAZ) – temperature above  $A_{c3}$ , with significant grain growth.

Generally, the critical part of the weld in terms of toughness measurement is represented by the CGHAZ. Here, a significant austenite grain coarsening occurs which retards the phase transformation during cooling, as the nucleation density is reduced [6]. As a result, coarse upper bainite can be formed. To inhibit this unfavorable  $\gamma$ -grain coarsening, Ti and Nb are added, which form stable precipitates that exert a pinning force on grain boundaries [6]. However, during high heat input SA welding, partial or complete dissolution of these precipitates takes place in regions with peak temperatures above 1200 °C combined with low cooling rates, so that the  $\gamma$ -grain coarsening cannot be suppressed, resulting in a coarse microstructure. This is one reason why multi-wire SAW is critical in terms of HAZ toughness. In addition, C-rich constituents between bainite lath boundaries can be formed during cooling. The volume fraction, size and morphology of these C-rich constituents have a negative effect on the toughness in the HAZ of SA welds [7,8]. This is even more critical in the case of the area of the weld overlap between the first and second layer, where the peak temperature during SA welding is between  $A_{c1}$  and  $A_{c3}$ , see Figure 2. In this intercritically reheated coarse-grained (ICCG) HAZ, only a certain fraction of the material transforms to austenite. Because of the higher solubility, these  $\gamma$ -phase portions have substantially higher C content than the matrix [6]. This stabilizes the intercritical austenite by shifting the  $A_{r3}$  temperature to lower values, which promotes the formation of M-A constituents to a significant extent.

Because of these interactions between heat input, cooling time and resulting microstructures during DSA (Double Submerged Arc) welding of large diameter pipes, special attention has to be paid to the HAZ properties, in particular to the absorbed impact energy values. Requirements concerning acceptable CVN values are given by specifications such as DNV-OS-F101 and API 5L for a broad range of steel grades and wall thicknesses. More than these standards, customer requirements define the desired weld properties. These requirements strongly depend on the loading and environmental conditions in the field and can therefore differ from the specifications.

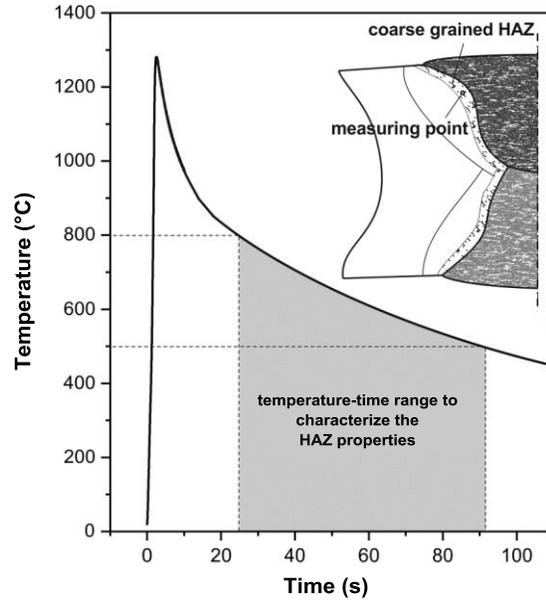


Figure 1. Temperature-time behavior in the CGHAZ of SA welds (outside layer) determined with FE simulation.

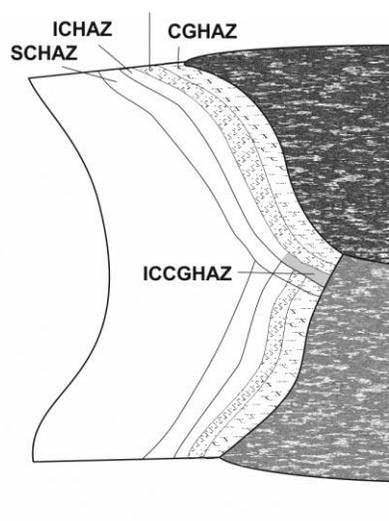


Figure 2. HAZ microstructure regions within two pass SA weld.

As mentioned above, Nb is an essential alloying element to generate a favorable fine-grained base metal microstructure. However, concerning the HAZ properties some inconsistent aspects have been reported in the literature. Thus, many researchers have investigated the influence of Nb content on HAZ toughness. Some investigators have found that low additions of Nb decrease toughness [8,9] while others [10] have demonstrated that there is no significant effect of Nb. Kawano et al. [11] have found that small additions of Nb (0.02%) lower the toughness in low C steel, especially for high heat input welds. Hulka and Heisterkamp [12] have shown that for a submerged arc welded 0.08%C-1.5%Mn steel (heat input of 2 kJ/mm) increasing Nb content up to a level of 0.08%Nb results in a deterioration of HAZ toughness, while further increase of Nb content leads to improved toughness. Similar results were presented by Bersch and Kaup [13] for a 0.09%C- steel for various welding processes, see Figure 3. The authors pointed out that first a reduction in toughness was observed, followed by an improved toughness behavior with increasing Nb content for both the multilayer technique, as well as double submerged arc welding.

The metallurgical phenomena in the HAZ are connected with the solubility of Nb-carbonitride precipitates [14]. If the temperature during welding reaches a level at or above the solubility of Nb carbonitride precipitates, dissolution will occur with the consequence of austenite grain growth. Additionally, it was mentioned in [15] that at intermediate cooling rates the solute Nb will depress the  $\gamma \rightarrow \alpha$  transformation by decreasing the  $A_{r3}$  temperature [16]. This promotes the formation of upper bainite as well as the development of martensite-austenite (M-A) constitutes. Thus, Koseki [9] showed that Nb increases the M-A area fraction in low (0.03%C) and high (0.16%C) C steels. However, Shams [17] has demonstrated for a 0.07%C – 1.4%Mn steel grade that the extent of  $A_{r3}$  temperature reduction depends strongly on the dissolved Nb content and the cooling conditions, see Figure 4. Nb content in solution by up to 0.05% lowers the  $A_{r3}$  temperature significantly, while a further increase in dissolved Nb content tends to increase the  $A_{r3}$  temperature.

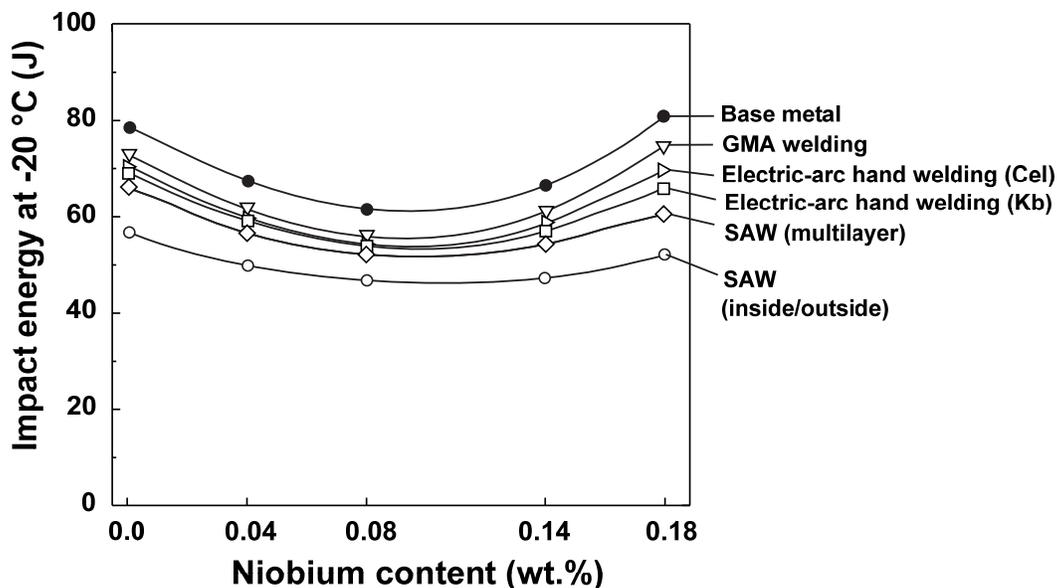


Figure 3. HAZ impact energy values depending on the Nb content for 0.09%C- steel [13].

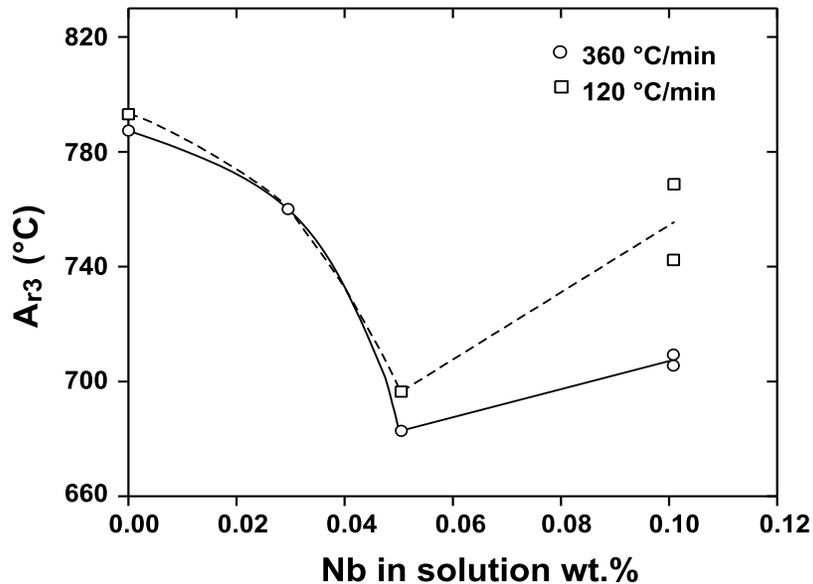


Figure 4. Effect of Nb in solution on  $A_{r3}$  temperature of 0.07 C- 1.5 Mn steel [17].

### Alloy Design for Optimized HAZ Properties

The improvement of the HAZ toughness of heavy wall linepipes at low temperatures has been the focus of alloy development at SZMF for several years. Solutions for optimizing HAZ properties have been presented recently for Grades X65 to X80 [6]. It was found that the  $CE_{IIW}$  does not correlate well with the HAZ toughness and that the Pcm-value is a more suitable measure. Microstructural investigations have shown that the decrease of toughness is related to an increase in the volume fraction of C-rich constituents. These can include M/A-constituents, bainite and pearlite. The volume fraction of these C-rich constituents in a given position of the HAZ is related to the base metal composition if the welding parameters and the wall thickness are held constant. Higher levels of HAZ toughness were achieved by reducing the C and Si content, which naturally led to a drop in base metal strength. Strategies to compensate for this drop were identified that did not affect the HAZ properties significantly. Especially a slight increase of the Mn content from 1.6% to 1.8% did not have a detrimental effect. The influence of a variation in the level of Nb was, however, not investigated. For this reason, laboratory trials with a systematic variation of the Nb content between 0.02% and 0.10% were carried out.

Four 100 kg laboratory heats were produced by vacuum induction melting, see Table I. The carbon equivalent (IIW) and Pcm values were constant for all four heats. The ingots were sectioned into coupons and were rolled on a two-high rolling mill down to a wall thickness of 25 mm. Final rolling temperatures above the  $A_{r3}$ -temperature were selected for all three compositions in order to ensure a predominantly bainitic microstructure after accelerated cooling, which was interrupted above the martensite-start temperature at around 450 °C.

Table I. Composition of the Investigated Laboratory Heats (wt.%)

Steel	C	Si	Mn	Cr	Nb	Ti	CE <sub>IW</sub>	Pcm
0.022 Nb	0.04	0.1	1.8	0.1	0.022	0.014	0.36	0.14
0.046 Nb	0.04	0.1	1.8	0.1	0.048	0.014	0.36	0.14
0.071 Nb	0.04	0.1	1.8	0.1	0.071	0.014	0.36	0.14
0.105 Nb	0.04	0.1	1.8	0.1	0.105	0.014	0.36	0.14

The same reheating temperature was selected for all four steels, above the equilibrium dissolution temperature of Nb(C,N) precipitates based on thermodynamic calculations. Once these are dissolved, only Ti(N,C) particles can inhibit grain coarsening, because these are stable over the whole range of feasible reheating temperatures.

### DSA Welding of Optimized HAZ Alloys: Welding Conditions and Destructive Tests

In order to determine the effect of a variation of the Nb content on the HAZ properties for high heat input welding, welding trials were carried out using multi wire double submerged arc welding (DSAW) which is extensively used for longitudinal welding in large diameter pipe production [18]. For both inside and outside welding, high heat inputs above 5 kJ/mm were achieved at welding speeds typically used in production. The welding parameters as well as bevel preparation were chosen based on mill production values. In the welding trials, a slightly basic flux was used in combination with filler wires following the TiB concept.

The toughness of the HAZ, as the key parameter, was determined by Charpy V-notch tests. In order to obtain statistically relevant results regarding the correlation between chemical composition of the base metal and the HAZ toughness, these Charpy tests were performed at two significant temperatures (-20 °C/-40 °C) on nine samples for each temperature. The notch positions were selected according to DNV-OS-F101 [19] and ISO 3183 [20], respectively, Figure 5. However, it can be assumed that the notch position according to DNV-OS-F101 represents the more critical case, because a higher fraction of the notch is located in the brittle coarse-grained HAZ, Figure 6. In addition, hardness tests were carried out according to DNV-OS-F101, in order to investigate the effect on the hardness in the vicinity of the weld.

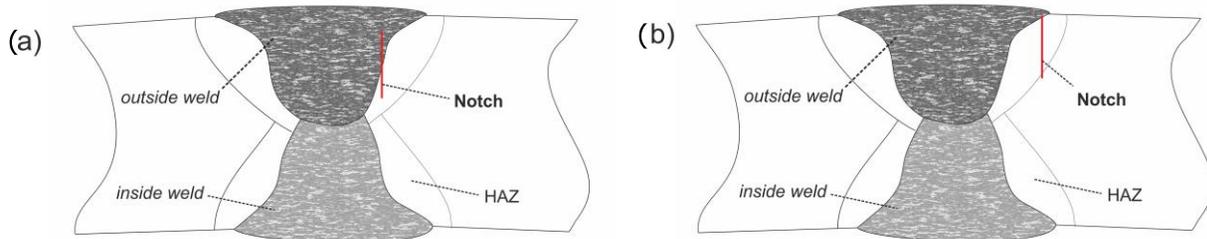


Figure 5. Notch positions according to; (a) DNV-OS-F101 and (b) ISO 3183.

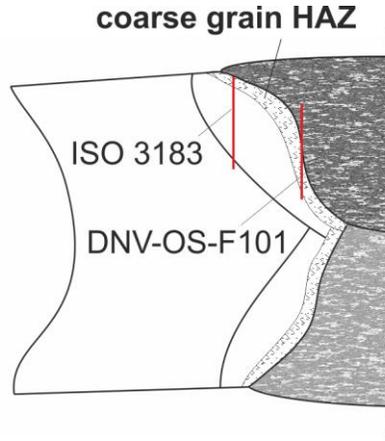


Figure 6. Schematic representation of the impact of the notch position on the portion of coarse grain HAZ sampled by the Charpy notch.

### Results and Discussion

The base metal was characterized in the as-rolled condition by light-optical microscopy and tensile tests in the transverse direction. An example of the microstructure that was realized by accelerated cooling is shown in Figure 7. In all four cases, a predominantly bainitic microstructure was achieved. The results of the transverse tensile tests are shown in Figure 8. The yield strength and tensile strength were found to increase continuously with increasing Nb content. The steels with Nb levels of 0.022% and 0.048% achieved values that are in the range of Grade X70, while the steels with the higher Nb content were at the X80 strength level.

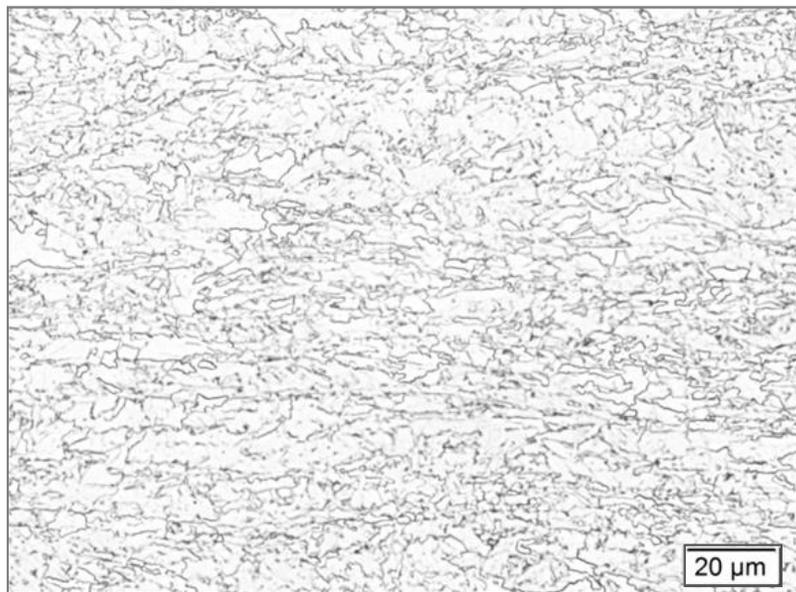


Figure 7. Microstructure of the laboratory rolled plate with 0.105%Nb.

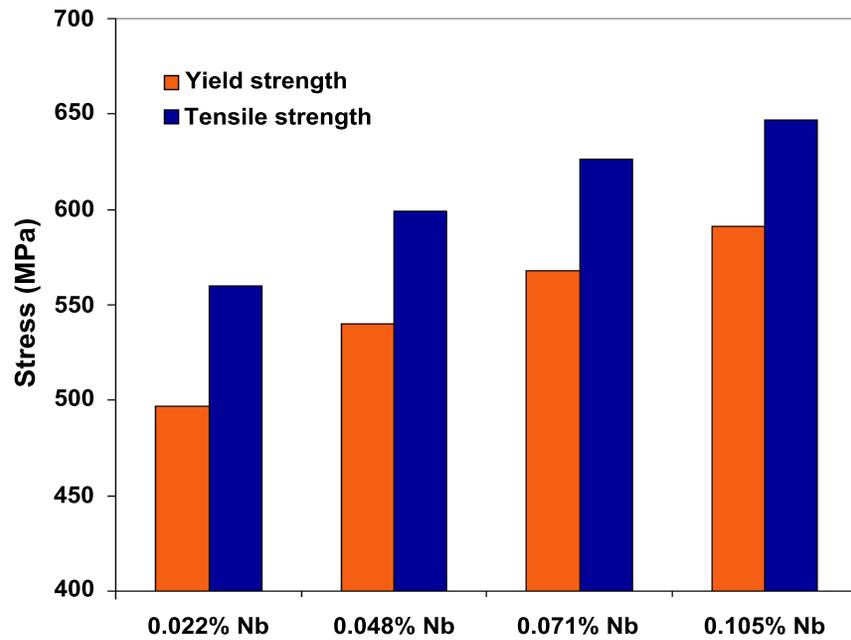


Figure 8. Results of transverse tensile tests on the base metal in the as-rolled+AC condition.

Figure 9 shows an example of a produced weld seam. All welds show typical shapes with no anomalies.

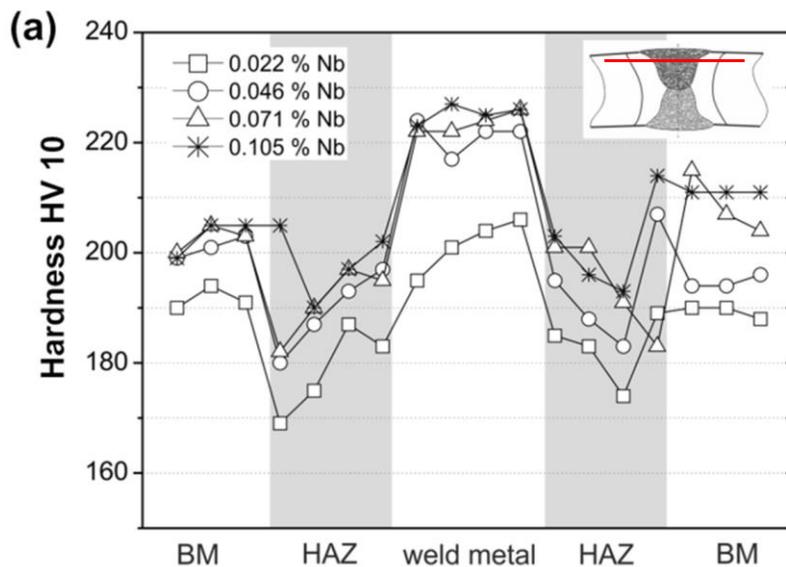


Figure 9. Cross section of SA weld, Nb content 0.022%.

In Figure 10, the hardness profiles of the SA welds, depending on the Nb content for the different areas of the weld, are presented. A typical hardness curve was found for thermomechanically rolled (+AC) microalloyed steels with a hardness reduction of the HAZ and an increasing hardness of the weld metal compared to the base metal. In general, a rise in base metal hardness was observed with increasing Nb content up to 0.071%. However, a further increase in Nb content did not have a significant impact on the hardness. This is in contrast to the base metal strength values, where a rise in Nb content was connected with continuously increasing strength properties.

The lowest hardness was found at the transition from the base metal to the HAZ independent of the Nb content. In this area of the weld, a peak temperature during welding below 600 °C occurred, which leads to softening.

Based on Figure 10(b), which shows the hardness profiles of the overlap areas of the inside/outside welds, it appears that welding of the second (outside) layer leads to the lowest HAZ hardness. This behavior is well known for DSA welding of thermomechanically rolled steels and is caused by the annealing of the HAZ during welding of the second layer.



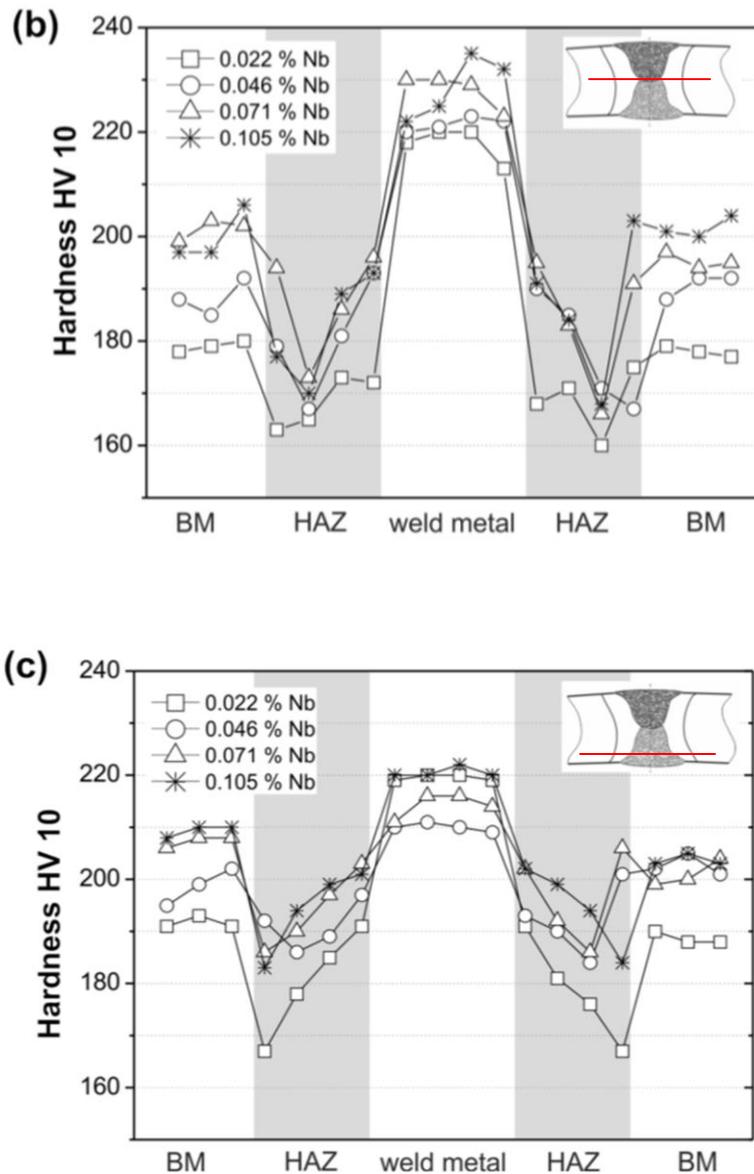
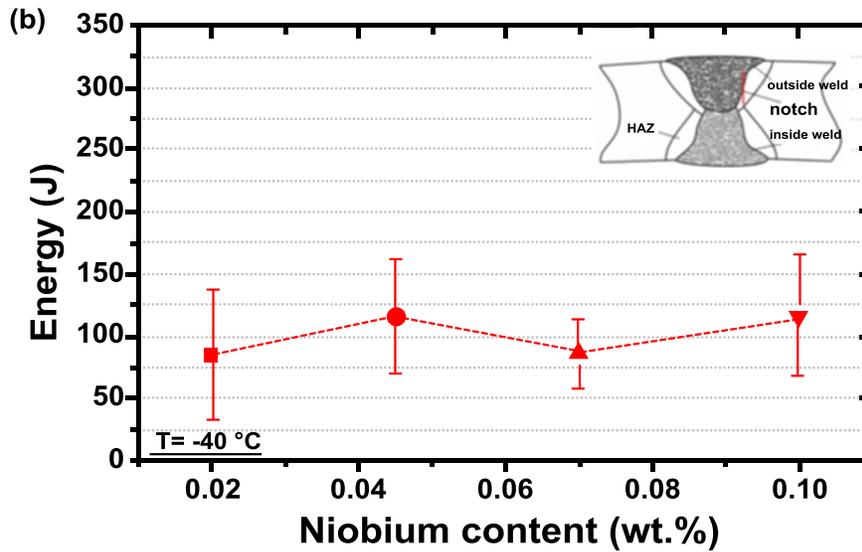
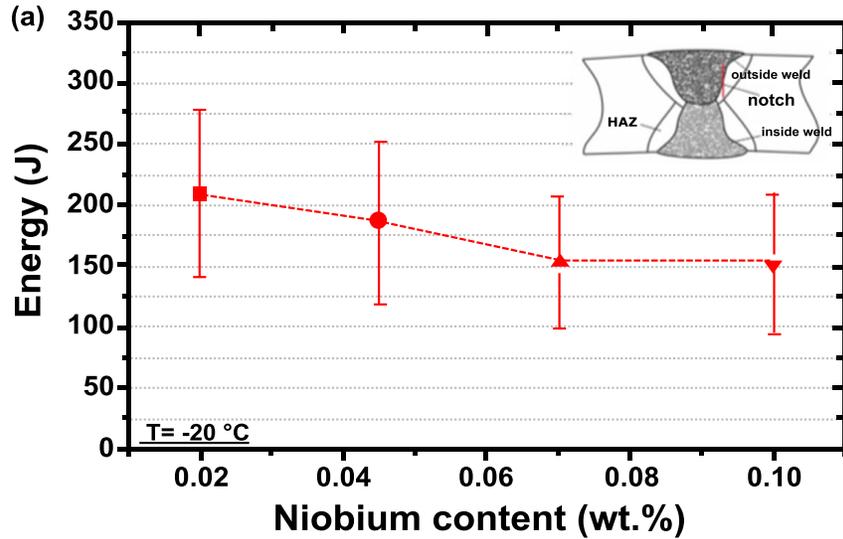


Figure 10. Hardness distribution of the inside/outside SA welds; (a) outside weld, (b) overlap area, (c) inside weld.

The weld metal showed an inconsistent behavior with regard to the hardness depending on the Nb content. Thus, for the outside weld, base metal with a low Nb content of 0.022% led to the lowest hardness, while a further increase up to 0.105%Nb in the base metal resulted in comparable weld metal hardness in all cases, Figure 10(a). In contrast, for the overlap area (Figure 10(b)) as well as for the inside layer (Figure 10(c)) the impact of Nb is not pronounced. This result is surprising in light of the extensive dilution during welding (around 60% to 70%). It appears that other parameters such as the composition of the filler wire play a major role for the weld metal hardness. In this context, it can be summarized that a high Nb content does not have a significant effect on the weld metal hardness.

The results of Charpy V-notch tests are presented in Figure 11. A slight reduction in impact energy with increasing Nb content was found for the notch position according to DNV-OS-F101 at -20 °C, see Figure 11(a). However, all mean values are relatively high and exceed the toughness requirements in [19,20] considerably.



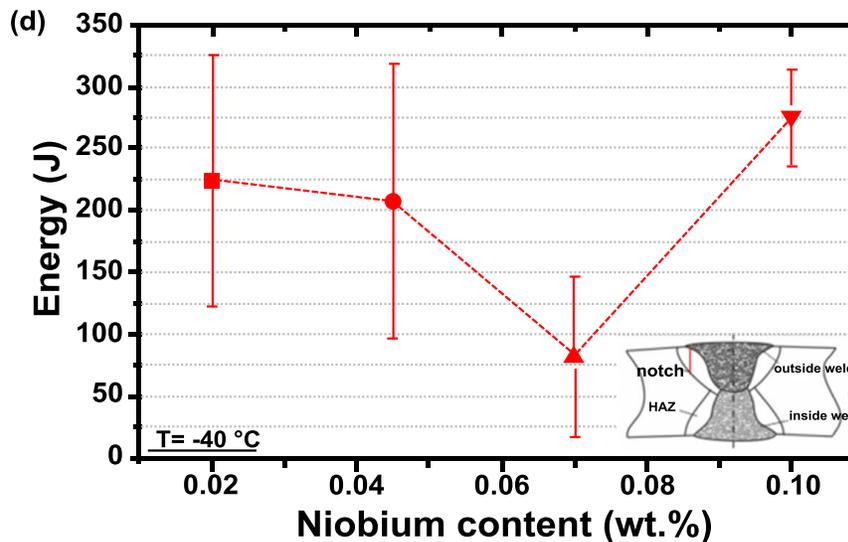
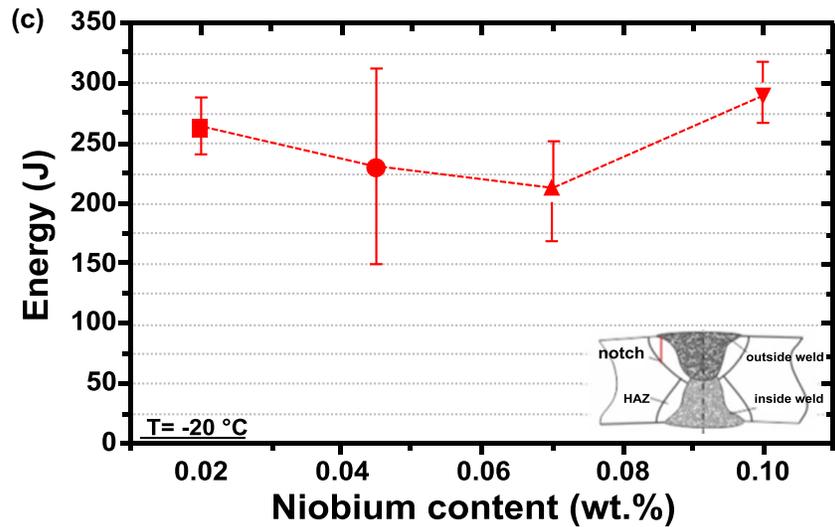


Figure 11. Evolution of Charpy impact energy depending on the Nb content for test temperatures; (a) -20 °C and (b) -40 °C, notch position according to DNV-OS-F101, as well as (c) -20 °C and (d) -40 °C, notch position according to ISO 3183.

At a test temperature of -40 °C and a notch position at the fusion line according to DNV-OS-F101, an increase of the Nb content did not have a negative impact on the toughness, see Figure 11(b). For a notch position according to ISO 3183, for both test temperatures (-20 °C/-40 °C), an increase of the Nb content up to 0.071% led to a reduction in impact energy. However, a further increase in Nb content to 0.105% led to the highest impact toughness values for both test temperatures. This trend of high impact toughness values at a high Nb content (0.105%) was observed previously in similar investigations [12,13].

The decrease of the impact energy at 0.071%Nb could be related to unfavorable weld geometry. Generally, the notch position specified by ISO 3183 results in a notch that is located in the HAZ including sub-critically reheated HAZ and a low fraction of the base metal, see Figure 12(a). The fractions of base metal and sub-critically reheated HAZ potentially lead to a mechanical support effect. However, the notches for most of the specimens with 0.071%Nb at -40 °C were positioned completely in the HAZ with a lower fraction of sub-critically reheated HAZ and no base metal fraction, ie. little support effect is expected, see Figure 12(b).

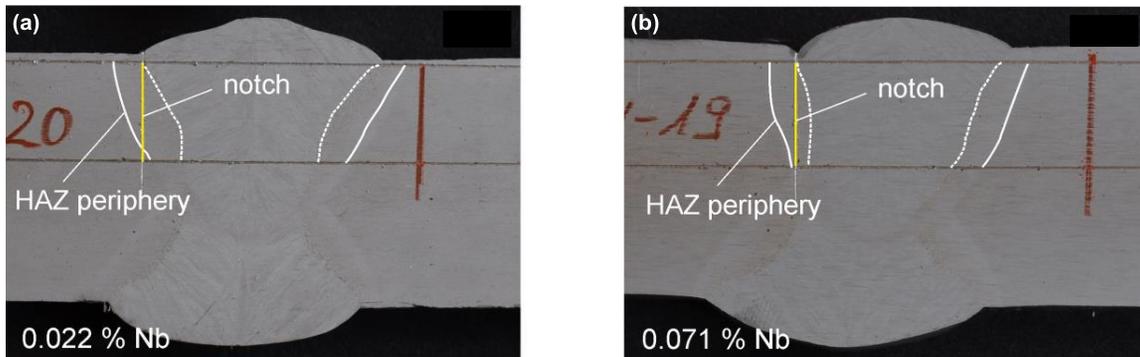


Figure 12. Difference in weld geometry leads to deviating notch position in the HAZ periphery.

It was found that the scatter of the measured impact energy values for the notch position according to DNV-OS-F101 is comparable for all alloy types and is nearly independent of the test temperature. For the notch position according to ISO 3183, the CVN test results for alloys with 0.046%Nb and 0.071%Nb show a high scatter at both test temperatures. In contrast, the high Nb steel (0.105%) exhibits low scatter at -20 °C and -40 °C, which is evidence that the observed high level of impact energy for this steel is genuine.

The comparison in Figure 13 shows that the notch position affects the impact values considerably. As discussed above, the notch position according to DNV-OS-F101 tends to be more critical in all cases in terms of HAZ toughness measurement. Thus, differences in notch position result in changes above 25% at a test temperature of -20 °C and above 40% at a test temperature of -40 °C, respectively. Because of the discussed critical interaction between notch position and weld geometry for the steel with 0.071%Nb, the difference at -40 °C represents an exception which, therefore, should not be taken into account in this context.

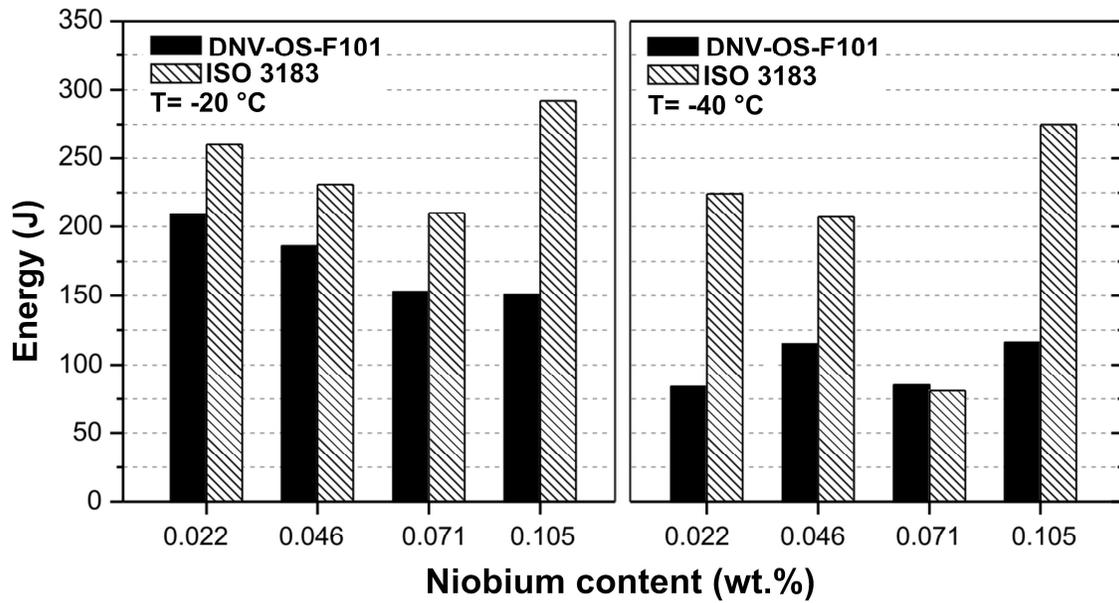


Figure 13. Effect of the notch position on CVN test results for test temperature of -20 °C (left) and -40 °C (right).

In summary, it can be stated that a Nb content of 0.105% does not have a negative impact on the toughness of the HAZ compared to a material with low Nb content (0.02%), see Figure 14. On the contrary, a trend towards higher toughness was observed.

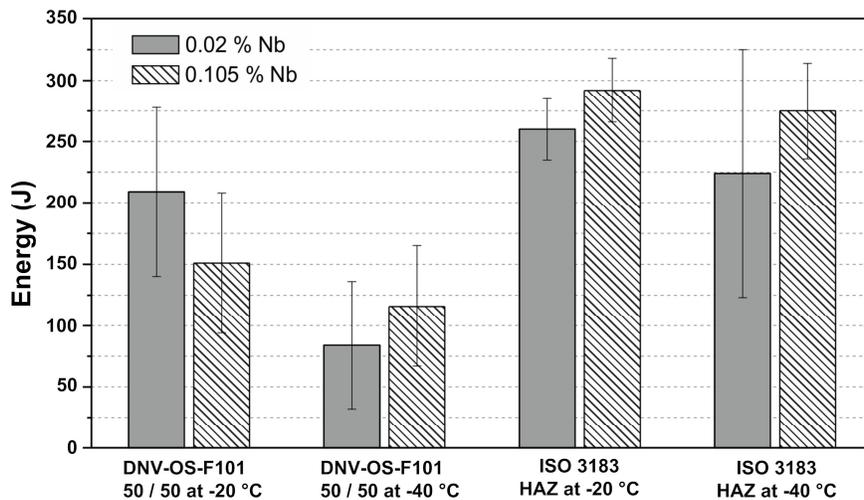


Figure 14. Comparison of the Charpy impact energy of the investigated steels containing low and high levels of Nb.

The microstructure in the CGHAZ was investigated by high-resolution scanning electron microscopy (SEM) of cross sections of the welds produced using the laboratory heats, in order to relate the HAZ-toughness to the microstructure. The samples were etched for a few seconds using a solution of nitric acid. Fifteen images were taken per sample at a magnification of 2000x within the CGHAZ in the position where the notches of the FL50/50 Charpy specimens intersect the fusion line. These were subsequently used for point analysis in order to measure the volume fraction of C-rich microstructure constituents. An example of microstructure, as observed by SEM, is shown in Figure 15. The microstructure consists of M/A-constituents and pearlitic and C-rich bainitic islands in a low-C bainitic matrix.

The average volume fraction of C-rich constituents in the CGHAZ and standard deviation obtained from the point analysis of 15 SEM images per weld are shown in Figure 16. It was found that the mean volume fraction of C-rich constituents was below 2.5% in all cases. This is lower than the level reported for any of the steels in a similar investigation [6] and is believed to be a result of the low C content of only 0.04%. In addition, the Nb content did not affect the volume fraction of the C-rich constituents significantly. A minor increase of the volume fraction of M/A-constituents with increasing Nb content was concomitant with a decrease of the volume fraction of pearlitic/bainitic islands.

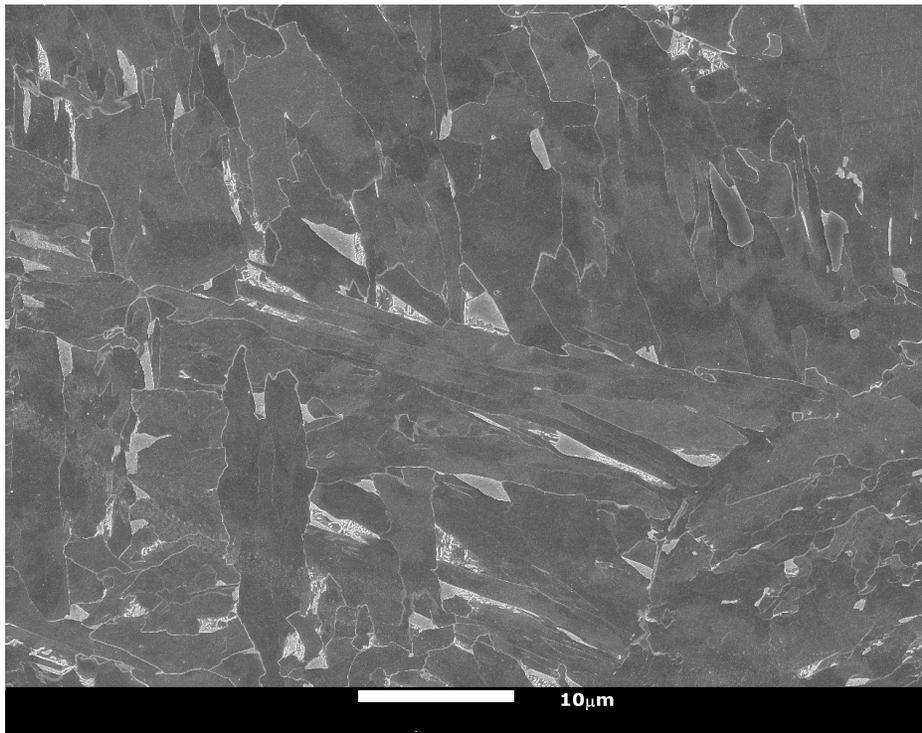


Figure 15. Example of the microstructure in the CGHAZ.

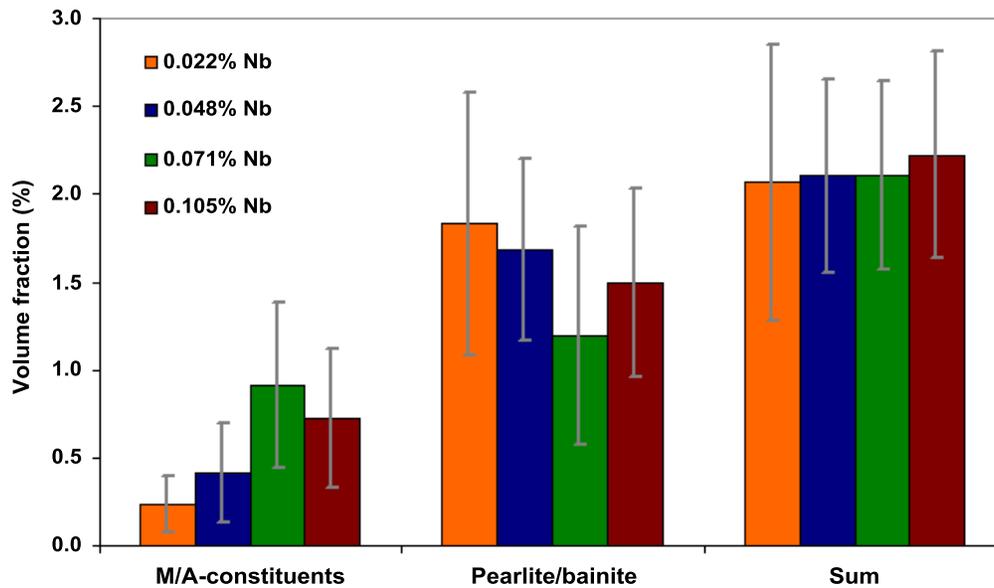


Figure 16. Volume fraction of C-rich constituents in the HAZ.

The impact of the different Nb contents with regard to the sensitivity to test temperature variations is shown in Figure 17. For the notch position according to DNV-OS-F101, all alloys exhibit decreased CVN values at the lower test temperature (-40 °C). The 0.022%Nb containing alloy demonstrated the highest drop (>50%), while the 0.105%Nb containing alloy type showed the lowest drop (around 20%). The highest CVN values at -40 °C were obtained at Nb contents of 0.046% and 0.105%, respectively.

For the notch position according to ISO 3183, a minor drop (<20%) of impact energy with decreasing test temperature was observed. However, the steel with 0.071%Nb constitutes an exception and could be related to the above mentioned unfavorable weld geometry (Figure 12). Again, the lowest drop was found for the steel containing 0.105%Nb.

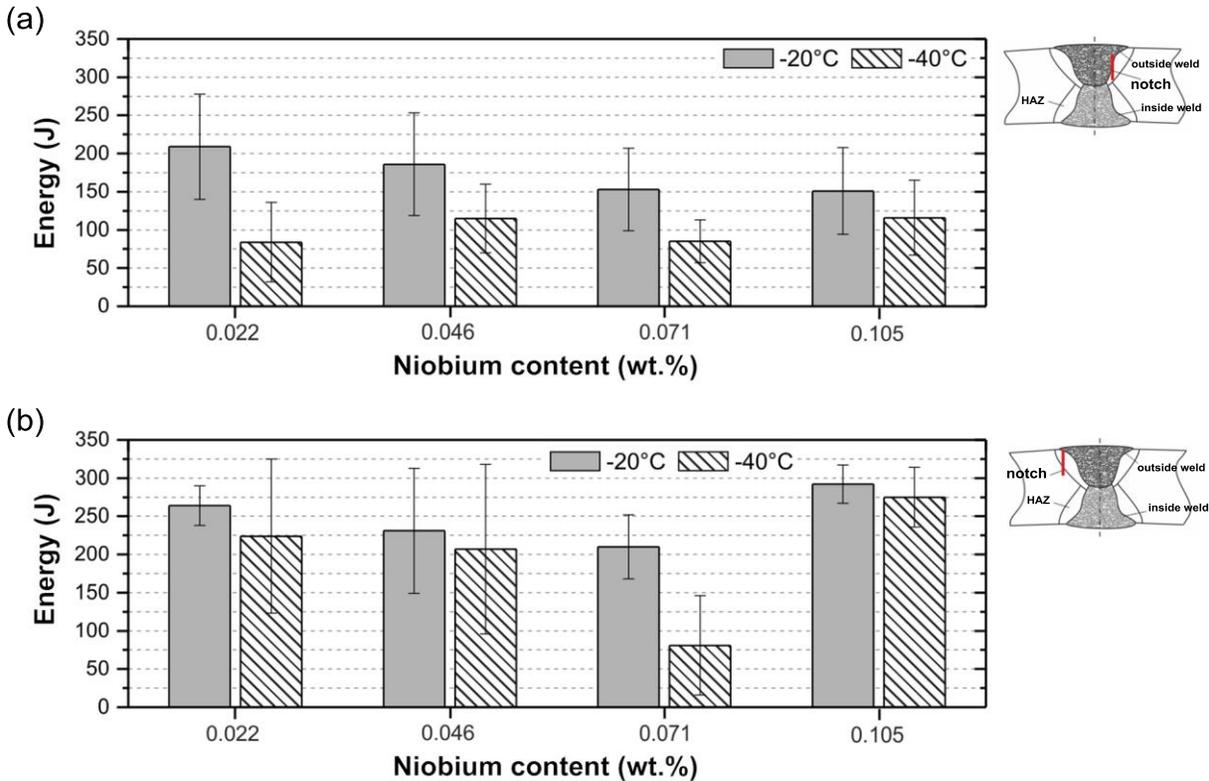


Figure 17. Influence of the test temperature on the CVN test results of the investigated steels with different Nb contents; (a) notch position according to DNV-OS-F101, (b) notch position according to ISO 3183.

### Summary

Laboratory DSA welding trials were carried out on four laboratory heats with a Nb content between 0.022% and 0.105% with the aim to investigate the effect of Nb on the HAZ toughness of steels suitable for large diameter pipes. Tensile tests in the as-rolled condition showed that the strength increased from the X70 to the X80 level above a niobium content of 0.07%.

SA welding trials with high heat input (>5 kJ/mm) were carried out in order to quantify the impact of Nb on the HAZ properties under production conditions. It was demonstrated within this investigation that the influence of Nb up to 0.105% on HAZ toughness of SA welds is not pronounced. The impact energies for the 0.07%Nb steel were a bit lower than the others, the reasons for which were not clear, though possibly due to unfavorable weld geometry. In addition, the impact of the test temperature on the HAZ toughness was analyzed depending on the Nb content. The lowest decrease in CVN values between -20 °C and -40 °C was observed in the case of a Nb content of 0.105%. The microstructure of the HAZ was investigated by scanning electron microscopy and did not vary significantly depending on the Nb content. Finally, it was shown that the notch position has a significant effect on the observed impact energy.

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