Temper embrittlement of 9%Ni low carbon steel and nondestructive inspection by magnetic Barkhausen noise

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Abstract. 9% Ni and low carbon steel is used in cryogenic services in oil and gas industries. The final mechanical properties are adjusted by quenching and tempering heat treatments. However, the un-correct tempering may cause temper embrittlement, with drastic decrease of toughness at cryogenic temperatures. In this study, specimens tempered at 350°C, 400°C and 450°C showed very low toughness at low temperature (-196°C) due to temper embrittlement. Specimens slowly cooled from the tempering temperature (565°C, 585°C and 605°C) also showed toughness reduction in comparison with specimens tempered at the same temperature and cooled in water. The brittle fracture was characterized by intergranular cracks and cleavage. Magnetic Barkhausen Noise (MBN) inspection was conducted to verify if this technique can be used to detect the temper embrittlement in 9Ni steels. The root mean square (RMS) of the MBN signal was higher in specimens as quenched and in specimens tempered in the temper embrittlement range (350°C-500°C) than in specimens which were correctly tempered (565°C-605°C and water cooled). Comparing specimens tempered at 565 and 585°C range and slowly cooled with those which were water cooled, the RMS(MBN) was higher in the former group, which presented the lower toughness. However, the MBN inspection could not separate specimens tempered at 605°C slowly and rapidly, which can be related to the higher austenite volume fraction measured in the specimen slowly cooled.

1 Introduction

9% Ni low carbon steels were developed in 1940's by International Nickel Company as an option to replace materials used in cryogenic applications (such as austenitic stainless steels, nickel alloys and aluminum alloys). In 1954, 9% Ni steel was recognized by the ASME code

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as a material used in cryogenic applications until temperatures as low as -196°C , through heat treatments according to normative specifications. The use of this steel reduced productions costs and problems with welding, in addition to producing lighter constructions, while mechanical properties, as mechanical resistance and toughness, are maintained satisfactory in temperatures well below 0°C . [1]

Standards recommend tempering in the $565\text{--}605^{\circ}\text{C}$ range after quenching or double normalizing treatments. [2–4]. This treatment produces a microstructure of ferrite and iron carbides (tempered martensite), with retained austenite. This combination provides very high impact toughness Charpy at -196°C . Although one of the objectives of the tempering treatment is to increase the toughness of the steels, un-correct tempering temperature selection may cause a brittle behaviour, decreasing severely the low temperature toughness [5]. Temper embrittlement of 9%Ni steels is reported to occur in the $370\text{--}540^{\circ}\text{C}$ range. [1]

The microstructural changes arising from the heat treatments have a direct relation to the electromagnetic properties of steels [6]. The results of Barkhausen noise measurements are responsive to metallurgic parameters variations such as phase transformation, grain size, precipitates, and crystallographic texture [7, 8].

In this work, the temper embrittlement phenomena of a 9% Ni low carbon steel was investigated. MBN inspection was performed as an attempt to identify non-destructively the heat treatments condition in which temper embrittlement is observed.

2 Experimental

The 9% Ni low carbon steel used in this work was from a seamless pipe, with chemical composition shown in Table 1. Specimens were roughly machined to dimensions close to standard Charpy specimen ($11\times 11\times 55\text{mm}$) to be heat treated. Table 2 describes the heat treatments and specimens identification.

Table 1. Chemical composition (%wt.) of 9% Ni low carbon steel studied.

C	Mn	P	S	Si	Ni	Cr	Cu	Al	V	Nb	Mo	Co
0,061	0.520	0.005	<0.001	0.208	9.590	0.056	0.383	0.014	0.002	0.011	0.016	0.009

Table 2. Heat treatments performed.

Identification	Heat treatment description
Q	Water quenching from 800°C
QT350	Quenched and tempered at 350°C
QT400	Quenched and tempered at 400°C
QT450	Quenched and tempered at 450°C
QT500	Quenched and tempered at 500°C
QT565	Quenched and tempered at 565°C (water cooling after tempering)
QT585	Quenched and tempered at 585°C (water cooling after tempering)
QT605	Quenched and tempered at 605°C (water cooling after tempering)
QT565-F	Quenched and tempered at 565°C (furnace cooling after tempering)
QT585-F	Quenched and tempered at 585°C (furnace cooling after tempering)
QT605-F	Quenched and tempered at 605°C (furnace cooling after tempering)

Specimens quenched and tempered at 350°C, 400°C, 450°C and 500°C were water cooled to room temperature after tempering. Specimens quenched and tempered at 565°C, 585°C and 605°C were either water cooled or slowly cooled in furnace (samples identified with the letter F) after tempering. After heat treatments the samples were machined to final dimensions and roughness of Charpy specimens with V-notch (10x10x55 mm). Then, the Charpy samples were submitted to magnetic Barkhausen noise (MBN) inspection. Then, Charpy impact tests at -196°C were performed in an universal pendulum with maximum energy of 300 J. The specimens fractured were observed in the scanning electron microscope (SEM). Microstructures of some specimens were observed in the SEM with specimens etched with nital 2% and Vickers hardness were measured with 10 kgf load.

Magnetic Barkhausen noise (MBN) inspection was performed with a computer controlled equipment called “Barktech”, developed by the Laboratory of Dynamic and Instrumentation of University of São Paulo (LADIN-USP). Barktech was responsible for the generation and control of excitation current and acquisition and filtering of the voltage signals generated by the MBN sensor. This sensor was customized for the material and specimens dimensions. The parameters for MBN inspection were 10 Hz of excitation frequency and excitation amplitude of 1.5 V.

The MBN signal was statistically treated by calculating the root mean square (RMS) according to Eq. (1):

$$RMS_{MBN} = \sqrt{\frac{\sum_{i=1}^n (V_i - V_a)^2}{n-1}} \quad (1)$$

where V_i is the voltage value of a pulse and V_a is the average voltage value of all n pulses of the Barkhausen signal. Since the MBN signal is centered in amplitude, that is, it has zero mean, the RMS_{MBN} value is equivalent to the standard deviation and represents a measure of the average signal of the fluctuations around the mean.

Fig. 1 shows schematically the magnetic Barkhausen noise (MBN) in the hysteresis loop curve.

Fig. 1. Schematic showing the region of the hysteresis loop where the magnetic Barkhausen noise (MBN) is measured.

Austenite volume fraction was analysed by magnetization saturation measurements. The magnetization saturation (m_s) was measured by plotting magnetization (M) versus $1/H$, where H is the applied magnetic field. The m_s value was obtained by extrapolation of $1/H=0$. A detailed description of the method is described in [9].

The austenite volume fraction (AVF) could be calculated by Eq. (2):

$$AVF = 1 - \frac{m_s}{m_{s(i)}} \quad (2)$$

where $m_{s(i)}$ is the intrinsic magnetization saturation of the sample with 100% of martensite (produced by quenching and cold deformation).

3 Results and discussion

Fig. 2 shows the variation of impact toughness and lateral expansion with heat treatments in specimens water cooled after tempering. Average hardness values were also inserted in Fig.2. Tempering at 400°C caused severe embrittlement in the 9% Ni steels, as expected from previous works [1,5,9]. In 9% Ni low carbon steels the embrittlement is not coincident with any hardening increase. The lowest toughness was obtained in samples tempered in the range between 350°C and 450°C, while the hardness always decreased with the increase of tempering temperature.

Fig. 2. Variation of impact energy and lateral expansion with tempering temperature. Vickers hardness data are also included.

The observation of the fracture surface by scanning electron microscope (SEM) of specimen QT400 reveals a brittle aspect with cleavage facets and intergranular cracks (Fig. 3(a-b)).

Fig. 3. Fracture surface of specimen QT400 in center: (a) low and (b) high magnification.

Table 3 compares the toughness and hardness of specimens water cooled with those cooled in furnace after tempering. In this comparison, specimens QT605F, QT585F and QT565F undergone some embrittlement because they were slowly cooled in the range of temperatures of temper embrittlement. The hardness values of all specimens are very similar, which indicates once more that the temper embrittlement of 9Ni is not related to hardening.

Table 3. Toughness and hardness of specimens water cooled and slowly cooled after tempering.

Property	QT565	QT565F	QT585	QT585F	QT605	QT605F
Toughness (J)	212	144	201	169	221	113
Hardness (HV10)	245	242	242	238	240	239

The observation of the fracture surface of specimen QT585F (furnace cooled after tempering) reveals a predominantly brittle aspect, but with a layer of ductility in the lateral part of the specimen. Fig. 4(a) shows the brittle fracture aspect in the central part of this specimen, where it can be observed an intergranular crack and cleavage facets. On the other hand, the fracture of specimen QT585 (water cooled after tempering) was completely ductile, with dimples (Fig. 4(b)) nucleated in non-metallic inclusion.

Fig. 4. Fracture surface of (a) QT585F showing cleavage and intergranular cracking in the center of specimen, indicating low toughness; (b) QT585 with dimples indicating high toughness.

Fig. 5 shows the microstructural characterization by SEM of specimen quenched and tempered at 400°C (QT400). The temper embrittlement of 9%Ni steel is attributed to intergranular Fe₃C carbides and/or intergranular phosphorus segregation [5,10].

Fig. 5. Microstructure of specimen QT400.

Figs. 6(a–b) show the microstructural characterization by SEM of specimens QT585 and QT585F, respectively. The specimen slowly cooled (QT585F) has more and coarser precipitates (carbides), which suggests additional precipitation during the slow cooling.

Fig. 6. Microstructure of specimen: (a) QT585 and (b) QT585F.

Fig.7(a) compares the MBN signal *versus* time of specimen as quenched with specimen tempered at 400°C. Fig.7(b) compares the MBN signal *versus* time of specimen tempered at 400°C with the one tempered at 565°C (QT565). Fig.7(c) compares the MBN signal *versus* time of specimen tempered at 565°C and water cooled (QT565) with the specimen tempered at the same temperature and slowly cooled in furnace (QT565F). As will be discussed, except for the specimens tempered at 605°C, the RMS_{MBN} signal was higher in the specimens which suffered temper embrittlement and in the as quenched specimen (Q).

Fig. 7. Comparison of MBN signal against time of specimens (a) QT400 and Q; (b) QT400 and QT565; (c) QT565 and QT565F.

Fig.8 shows the variation of the RMS_{MBN} with tempering temperature for specimens which were water cooled after tempering. Hardness and impact toughness variations were also included. Fig.9 compares the RMS_{MBN} of specimens tempered and water cooled with those specimens tempered and slowly cooled in furnace. The results in Figs. 8 and 9 show

that the RMS_{MBN} was higher in the specimens which suffered temper embrittlement and in the as quenched specimen (Q), exception to the specimen QT605F, which suffered temper embrittlement due to the slow cooling but had RMS_{MBN} (0.118) very similar to that of specimen QT605 (0.115). This probably occurs due to the higher austenite volume fraction (paramagnetic phase) measured in specimens QT605F and QT605 (see Table 4), which provokes the decrease of the RMS_{MBN} . In a previous work [11], the temper embrittlement of a supermartensitic 12%Cr tempered at 500°C could be detected by MBN analysis, but the embrittlement effect of specimens tempered at 620°C and slowly cooled could not be detected because this sample contained a significant higher amount of austenite. Tempering above the A_1 temperature of the steel and slow cooling were the factors responsible for the high austenite content. As a consequence, the RMS_{MBN} measured was very low and similar to that of specimens which did not suffer embrittlement. This is the same behaviour observed in specimen QT605F of the present work.

Fig. 8. Variation of RMS_{MBN} , impact toughness and hardness with tempering.

Fig. 9. Influence of cooling after tempering on RMS_{MBN} . Toughness values were also included.

Table 4. Austenite Volume Fraction measured by magnetic methods.

Specimen	QT565	QT565F	QT585	QT585F	QT605	QT605F
AVF	0.093	0.069	0.090	0.076	0.072	0.154

4 Conclusions

The classic temper embrittlement of 9%Ni low carbon steel occur when this material is tempered in the 350 – 450°C range. Specimens which undergone embrittlement could be distinguished by magnetic Barkhausen noise (MBN). The root mean square of MBN signal (RMS_{MBN}) of specimens as quenched and those tempered in the 350 – 450°C were significantly higher than those correctly heat treated in the 565 – 605°C range.

Specimens which were tempered in correct temperatures (565 – 605°C), but slowly cooled in furnace, also undergone some toughness reduction in comparison to water cooling. In this case, the MBN measurement was effective to separate furnace cooled from the water cooled samples if the tempering temperature was 565°C or 585°C. However, no difference was observed in the MBN of specimens tempered at 605°C slowly and rapidly cooled. This behaviour was attributed to the high austenite content of the specimen slowly cooled from 605°C, which resulted in a low MBN signal, similar to the specimen water cooled.

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