

# The influence of thermal treatments on acoustical-spectral parameters

P. PETCULESCU\*, R. ZAGAN, M. BORMAMBET

"OVIDIUS" University of Constanta, Mamaia Ave. 124, 900527, Constanta, Romania

In this paper, the authors aim to analyze the influence of thermal treatments on the microstructure of cryogenic material welds. The changing of physical-mechanical and acoustical-spectral parameters is analyzed when a thermal treatment and a thermo-mechanical treatment are applied. These welds are analyzed having as reference the sample B without being exposed to a thermal treatment that is also the witness sample. One noticed that due to the applied thermal treatment, the weld constituents are composed of austenite + ferrite + Cr carbide precipitate with different volume fractions and different microstructures (grain sizes). There are shown welds that present a high hardness, a higher transparency of acoustic energy, different resonance frequency and Spectral Peak Ratio (SPR), all these depending on the scope of using the weld. The experimental measurements presented here were obtained by the direct contact method using silicon gel as the coupling medium with the pulse-echo technique, with a 5MHz transducer (Nortec). The instrumentation consisted of an IPR-100 signal generator, an A/D-90 converter and SMC-4 step by step motors from Physical Acoustic Corporation. The signals received at the transducer from the sample were sent to an oscilloscope, where their amplitude and velocity was read directly and to the sampler. Signals from the sampler were transmitted to the spectral analyzer where the spectrum of the concerned signal was photographed.

(Received May 29, 2007; accepted December 4, 2007)

*Keywords:* Ultrasounds, Thermal treatments, Seam weld, Attenuation, Microstructures, SPR

## 1. Introduction

The industrial usage of steels at low temperatures leads to different studies, which tend to increase the installation capacity and security in this field. Cryogenic steels can be used in aeronautic, food, chemical, petrochemical and transportation industries to manufacture equipments, claddings, and pipes and also to joint them by welding in order to obtain, stock and transport fluids which reach low temperatures while using them. Steels and additional materials which must correspond to an accumulated operating time are required taking into consideration their chemical composition and mechanical features such as high failure stress associated with a good tenacity, a bigger fatigue limit, a good weldability, etc. Our scope is to obtain crystalline structures closer to the perfect ones, the control of dislocations density and their displacement, the alloying with elements, which form substitution solid solutions, the grain sizes increase. The main alloying element in cryogenic steels is Ni because it manifests a strong positive influence on the fragility threshold, 6%Ni lowering the transition temperature at -200 °C, the resilience is excellent and the plasticity is high. In many cases, physical, chemical, mechanical and technological properties of metallic materials depend on the applied thermal treatments (reanneal treatment, annealing treatment, hardening, slackening); for austenitic stainless steels the applied thermal treatments give a better corrosion resistance.

## 2. Experimental

### 2.1 Theory

By applying Fast Fourier Transform FFT [1] and Wavelets Transform WT [2,3] it was possible to transfer the echoes from the time domain into the frequency domain. Thus, one could determine the peak frequency  $f_p$ , the resonance frequency  $f_0$  [4,5] and the ratio between the amplitudes of the extreme frequencies from the spectrum (Spectral Peak Ratio SPR). In order to assess the grain size from the austenitic samples in the seam weld (SW) we used the value of the ultrasonic attenuation determined by two methods, such as:

$$\alpha_1 = 7.768 - 1.337 \cdot 10^{-6} \cdot f_p + 4.8 \cdot 10^{-27} \cdot f_p^4$$

from USIS [6,7] where  $f_p$  - is the peak frequency SW being specific to the thermal treatment applied to the sample, by applying the scattering relation from the Rayleigh domain, or the grain size was determined by applying SPR after Kumar [8]:

$$\alpha_2 = S_L \cdot D^3 \cdot f_p^4$$

where  $S_L$  - is the scattering coefficient ( $S_L = 13 \cdot 10^{-8}$  [N/m<sup>2</sup>]);

$D$  - is the grain size in [ $\mu$ m];

When the samples were examined with an angular transducer, the ultrasonic beam propagated through three different elastic media: the couplant  $Z_1$  (glycerin), the base material  $Z_2$  and the seam weld  $Z_3$ . In order to find a correlation between the ultrasonic attenuation, the relative

transmission coefficient  $T_r$  (the relative transparency) [9, 10] we determined these parameter by applying the following relation:

$$T = \frac{4 \cdot Z_1 \cdot Z_3}{Z_2 + \frac{Z_1 \cdot Z_3}{Z_2}} \quad (3)$$

leading to

$$T_r = \frac{T}{100} \quad (4)$$

where 100% is the incident ultrasonic beam,

### 3. Experimental

The experimental measurements presented here were obtained by the direct contact method using silicon gel as the coupling medium with the pulse-echo technique. The instrumentation consisted of an IPR-100 signal generator, an A/D-90 converter and SMC-4 step by step motors from Physical Acoustic Corporation. The signals received at the transducer, of 5 MHz fundamental frequency (Nortec), from the sample were sent to an oscilloscope, where their amplitude and velocity was read directly and to the sampler. Signals from the sampler were transmitted to the spectral analyzer where the spectrum of the concerned signal was photographed.

The accuracy in time of flight (TOF) was better than 1 ns and the maximum scatter in the ultrasonic velocity was  $\pm 2.5 \text{ ms}^{-1}$ . The Vickers micro-hardness measurements were carried out at 29.42 daN load on all samples. An average hardness value from five measurements taken in each condition is reported; the maximum scatter was  $\pm 5\text{HV}$ .

#### Samples

The measurements were performed on the austenitic steel sample 10TiNiCr180 with the following chemical composition [%]: C-0.034, Si-0.69, Mn-1.70, Cr-18.55, Ni-9.41, Mo-0.28, Cu-0.35, Ti-0.0018, Nb-0.049, Al-0.068, W-0.085, V-0.061, C-0-0.24.

The welding is made with an electrode E19.9NbB22 (SREN 1600-2000) which has the following chemical composition: C =0.035%,  $M_n = 1.3\%$ ,  $S_i = 0.4\%$ ,  $C_r = 19.8\%$ ,  $N_i = 10\%$ ,  $N_b = 0.35\%$ .

The equivalent chemical composition of the seam weld was determined and the following values were found:  $C_{re} = 20.43\%$ ,  $N_{ie} = 11.61\%$  and the ratio is  $C_{re}/N_{ie} = 1.76$ .

Introducing these values in Schaeffer diagram, it was noticed that the seam weld structure is composed of austenite and 11% ferrite.

Sample B- manual metal arc welded sample with coated electrodes without being exposed to a thermal treatment.

Sample 2- manual metal arc welded sample with coated electrodes to which it was applied a hardening + stress-relieving treatment.

Sample 2p- manual metal arc welded sample with coated electrodes to which it was applied a hardening + annealing + stress-relieving treatment.

Sample 21- manual metal arc welded sample with coated electrodes to which it was applied a thermo-mechanical treatment + stress-relieving treatment.

Sample 21p- manual metal arc welded sample with coated electrodes to which it was applied a thermo-mechanical treatment + annealing + stress-relieving treatment.

#### Thermal treatments

The thermal treatment of hardening - heating temperature = 1100C, maintaining time = 25-30 min., depending on the sample thickness and water quenching.

Annealing treatment - heating temperature = 400 C, maintaining time=60 min and the quenching is lent.

Stress-relieving treatment - heating temperature =180 C, maintaining time=120 min and the quenching is lent in the oven.

Thermo-mechanical treatment - heating temperature = 1100C, maintaining time = 30 min., deformation 30% (for 1100 °C- 850 °C) and water quenching.

### 4. Results and discussion

Sample B is made of cryogenic austenitic material type 10 TiNiCr180 without a thermal treatment applied after the welding; one consider sample B as the reference (witness) sample. Samples 2 and 2p are a first set of samples being exposed to a specific thermal treatment, and samples 21 and 21p are another set of samples being exposed to a thermo-mechanical treatment.

The aim of examining the seam welds of the two set of samples (2 and 2p and 21 and 21p) is to study the influence of thermal treatments on the inner microstructure, on the changing of physical-mechanical and acoustical-spectral parameters. All these changes will be reported to the reference sample, sample B.

Samples 2 and 2p from the first set of samples submitted to a hardening and stress-relieving treatment (sample 2) and then to an annealing treatment (sample 2p) present a constant in the ratio  $v_T/v_L$  (where  $v_T$  is the ultrasonic shear velocity and  $v_L$  ultrasonic longitudinal velocity) and also in the Poisson ratio  $\sigma$  comparative to the reference, whereas the grain size decreases gradually presenting a relative variation of 25% comparative to the reference (Fig. 1a).

Concerning the hardness and Young modulus, after equaling the values in the reference sample, the hardness presents a light decrease in sample 2 followed by an increase exceeding the reference value with 13.7% in the relative value. Young modulus, after a significant decrease in sample 2, increases exceeding the reference value in sample 2p (Fig. 1b).

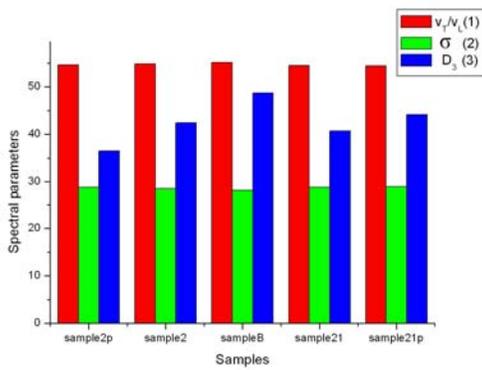


Fig. 1 a

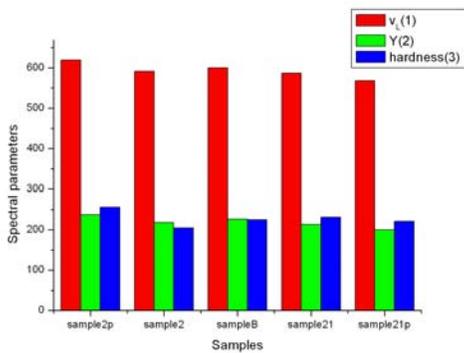


Fig. 1 b

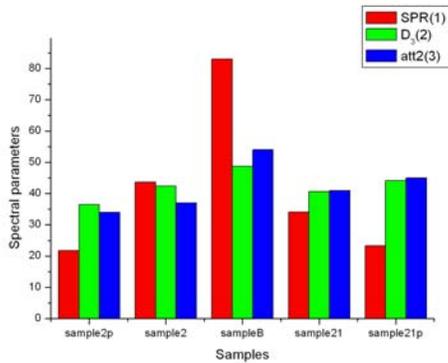


Fig. 1 c

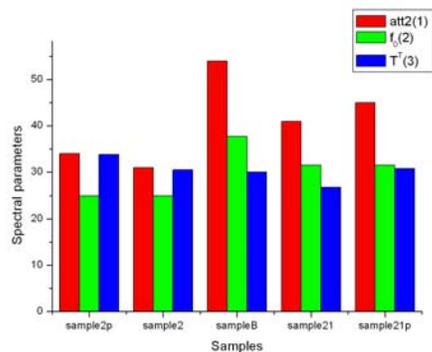


Fig. 1 d

A combination of acoustical and spectral parameters but presented in a different way is shown in Fig. 1c. The value of SPR decreases significantly at sample 2p submitted to hardening, annealing and stress-relieving treatments (73.8%), which shows that the extreme values of signal amplitudes are lower comparative to the ones of the reference sample; implicitly, the ultrasonic attenuation will decrease (37%) and also the grain size. In Fig. 1d there are shown parameters like ultrasonic attenuation, resonance frequency  $f_0$  and ultrasound transparency  $T^T$  in the shear waves that characterize the ultrasonic energy transmitted to the interface between the base material and the weld.

One notice that for the first set of samples (2 and 2p) the resonance frequency  $f_0$  remains constant but its value is lower then the reference with 33.6% and ultrasonic transparency increases with 12% due to the ultrasonic attenuation and grain size decrease.

Samples 21 and 21p from the second set of samples, submitted to a thermo-mechanical treatment (water quenching) and stress-relieving (sample 21) and then annealing (sample 21p) present a constant in the ratio  $v_T/v_L$  and also in the Poisson ratio  $\sigma$  comparative to the reference, whereas the grain size decreases presenting a relative variation of 16.4% comparative to the reference (sample 21) and then increases with 10% (Fig. 1a).

The velocity of longitudinal waves decreases gradually, step by step, comparative to the reference in the same way as Young modulus (Fig.1b) and the hardness increases first (sample 21) and then decreases to the reference value.

Spectral Peak Ratio (SPR) presents an emphasized decrease from 59% for sample 21 to 72% for sample 21p (Fig. 1c).

Ultrasonic attenuation decreases significantly with 24% comparative to the reference and then presents a light increase of 12% comparative to sample 21.

After resonance frequency remains constant (Fig. 1d), there appears a decrease of ultrasound transparency and then an increase almost reaching the reference level (see sample 21p).

A combination of physical-mechanical and acoustical-spectral parameters having a different aspect, is shown in Fig. 2 for all analyzed samples. One noticed an identical shape for the ultrasonic attenuation factor and for the grain size due to the existence of a correlation between these three parameters.

From the hardness graph, one can notice that in the case of manually welded steel sample, applying of an annealing treatment after a thermal treatment of hardening (sample 2p) would lead to an increase of hardness and applying of a thermo-mechanical treatment of annealing (sample 21p) would lead to a decrease of hardness; these observations are considered comparative to the reference sample (sample B).

After analyzing the physical-mechanical and acoustical-spectral parameters of the two sets of samples and after applying the thermal treatments and comparing them with the reference sample, our aim is to study the metallographic aspect of these samples (see Fig. 3).

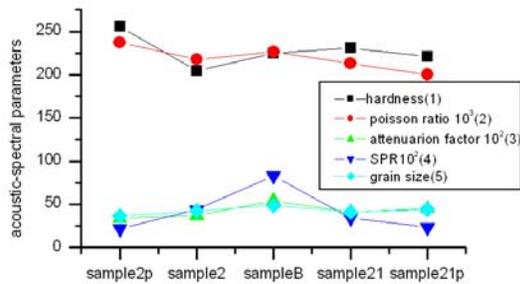


Fig. 2

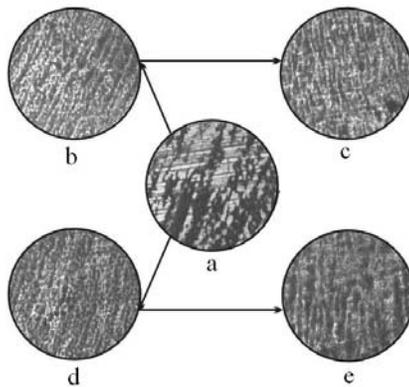


Fig. 3

The metallographic aspect of sample B is shown in Fig. 3a. The metallographic aspect of sample 2 submitted to a thermal treatment of hardening is shown in Fig. 3b. One noticed here a casting-solidification structure having a dendrite aspect having a strong orientation of the grains to the right. The predominant light zones are due to austenite presence; less dark zones, interpolated between the light zones, show the presence of ferrite  $\delta$  and the few black dots are due to the presence of some Cr carbide precipitates at the grain limits.

After applying a thermal treatment of annealing to the sample 2 (Fig. 3c) one obtains a structure of casting-solidification having a dendrite aspect and in the metallographic aspect appear some changes in grains orientation direction, the quantity of austenite decreases (bright zones), a denser group of ferrite  $\delta$  constituents appears and Cr carbide precipitates have bigger sizes.

Concerning the second set of samples submitted to a thermo-mechanical treatment (deformation at heat), the weld has a structure of casting-solidification having a dendrite aspect. In sample 21 one can notice a grains orientation after the deformation direction, bright zones (austenite) are reduced in size and in number and one can notice an increase of dark zones (ferrite  $\delta$ ) and Cr carbide precipitates, having smaller sizes, are situated at grain limits. In Fig. 3e one can see the metallographic aspect of sample 21p after a thermo-mechanical treatment (water quenching) and an annealing treatment that shows a new orientation of the crystals, an increase of ferrite  $\delta$  by elongation, a decrease of austenite and Cr carbide

precipitates are numerous (may be forming a group) but smaller in size.

It is known that the velocity of ultrasonic propagation is in correlation with the material density and with Young modulus as physical-mechanical properties and with the grain size and attenuation as acoustic properties. Therefore, any change in the material microstructure leads to a change of these parameters. In the metallographic aspect of sample 2p, at the end of hardening and annealing treatment, the fraction of the volume increases in ferrite  $\delta$  and Cr carbide precipitates are globular at the grain limits and lenticular inside the grains. Due to the increase of the volume fraction of ferrite  $\delta$ , both hardness and Young modulus present an increase comparative to the reference sample. In the same time, one can notice a decrease of the grain size and of the ultrasonic attenuation and an increase of the ultrasonic propagation velocity. These increased is due to the decrease of the ultrasonic propagation time through a small grain. Also, an increased ultrasound transparency appears. One believe that all these changes are due to grains reorientation toward a favorable position for the ultrasonic propagation, to an increase of the volume fraction in ferrite and to some precipitates in Cr carbide aleatory distributed.

One believe that the decrease of the resonance frequency  $f_0$  comparative to sample B is due to the increase of volume fraction in ferrite and of precipitates in Cr carbide or in general to a compactization of the three phases (austenite, ferrite, Cr carbide).

In the case of the second set of thermo-mechanical treatment of sample B, through samples 21 and 21p, one notice in Fig. 1b an increase followed by a decrease of the hardness and of Young modulus. Also, from the metallographic aspect one can notice an increase of volume fraction of ferrite  $\delta$  having the form of a dense fabric, of vertical, equidistant strings (toothed).

## 5. Conclusions

For the sample 2, in the seam weld, where  $SPR=0.473$ , the grain size is  $42.40\ \mu\text{m}$ , its structure being composed of austenite + ferrite + Cr carbide.

For the sample 2p, in the seam weld, where  $SPR=0.217$ , the grain size is  $39\ \mu\text{m}$ , its structure being composed of austenite + ferrite + Cr carbide.

Knowing the ultrasonic attenuation values in the seam weld, one can conclude that in the weld there is a structure having a dendrite aspect of austenite + ferrite that leads to an increased attenuation compared to the base structure, this being also confirmed by the metallographic analysis.

Regarding the metallographic analysis of the welds in all analyzed samples, one obtains a structure of casting-solidification having a dendrite aspect, where the austenite and the ferrite have a relative fine granulation in the composition (decreasing the risk of cracking at heat). Also, one noticed the presence of some Cr carbide precipitate. In the case of samples deformation (distortion) at heat in the seam weld, one noticed a grains orientation after the deformation direction.

For the welded samples where a hardening treatment was applied, the structure is composed of austenitic polyedric grains with macle strips and with a decreased quantity of Cr carbide precipitate at the grains limit.

For hardened and annealed samples, after applying the annealing treatment, one obtain the same structure, that is composed of austenitic polyedric grains but one noticed an increase of the quantity of Cr carbide precipitate inside and especially at the grains limit and also a number of frequencies in the spectrogram.

The experimental measurements presented here were obtained by the direct contact method using silicon gel as the coupling medium with the pulse-echo technique, with a 5 MHz transducer (Nortec). The instrumentation consisted of an IPR-100 signal generator, an A/D-90 converter and SMC-4 step by step motors from Physical Acoustic Corporation, a sampler and a spectral analyzer.

### Acknowledgment

We gratefully acknowledge funding for this work from the Romanian Education Ministry, through grant RELANSIN no. 2075/01.10.2004.

### References

- [1] L. Cohen, 'Time-Frequency Distributions: A Review' - Proc. IEEE, 941-981 (1989).
- [2] A. Abbate, J. Koay, J. Frankel, S. C. Schoeder, 'Signal detection and noise suppression using a wavelet transform signal processor: application to ultrasonic flaw detection', IEEE Trans. Ultrason., Ferroelect., and Freq. Contr. **44**(1), 14-25, (1997).
- [3] P. M. Gammell, 'Analogue implementation of analytic signal processing for pulse-echo systems', Ultrasonics, 279-283 (1981).
- [4] R. Zăgan, P. Petculescu, N. Peride, G. Prodan, 'Comparison between ultrasonic and wavelets analysis for characterization stainless steel alloys', The Fifth World Congress of Ultrasonics, Paris 2003.
- [5] R. Zăgan, P. Petculescu, N. Peride, 'Analyse of frequency spectrum in austenitic steel sample', European Conference of Physics, Dresda, 2006.
- [6] P. Petculescu, R. Ciocan, V. Bokas, 'Investigation of the austenitic structures by ultrasonic spectral analysis', 7 th ECNDT Copenhaga, 1998 and in Journal of Nondestructive Testing and Ultrasonic **3**(11), (1998).
- [7] P. Petculescu, R. Ciocan, D. Ciobanu, 'Automated system and automated method by ultrasonic investigation', patent no.118612B, RO.
- [8] Kumar, T. Jayakumar, R. Baldev, 'Ultrasonic spectral analysis for microstructural characterization of austenitic and ferritic steels', Philosophical Magazine A **80**(11), 2469-2487 (2000).
- [9] P. Petculescu, 'Ultrasounds Fundamentals Application' (in Roum.), edited by Ovidius University Press, ISBN: 973-614-024-5, 2002.
- [10] P. Petculescu: 'Nondestructive Methods for the Fast Determination of The Acoustic Parameters and Their Applications', The e-Journal of Nondestructive Testing, **11**(6), 2006.

\*Corresponding author: petculescu@univ-ovidius.ro