

# Damage investigation on welded tubes of a reforming furnace

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*In this work the creep damage of radiant tubes of a reforming furnace has been investigated. The considered furnace contains a battery of tubes constructed by butt welding three spun cast pieces, made of ASTM 608 HP-Nb alloy. They are designed to operate at temperatures of about 900°C, pressures of about 30 bars and times of the order of 100000 h. Tubes were inspected during the plant stops scheduled every two years, in order to identify and replace the damaged ones with the aim to ensure conditions of safe operation in the furnace. They were selected through a criterion based on measures of the internal diameter deformation performed in situ by Laser Optic Tube Inspection System (LOTIS). For a verification of this method, optical and scanning electron microscopy observation, Vickers microhardness and creep tests have been carried out on samples taken from tubes put out of service.*

**Keywords:** Superalloys - Welding - Non-destructive testing - Mechanical testing - Metallography - Electron microscopy

## INTRODUCTION

During service at high temperature and for long time, metallic materials undergo to different levels of creep phenomena, which lead first to slow deformations and finally to crack nucleation and propagation. These conditions are particularly dangerous in the case of piping and pressure devices. In order to verify every compliance with safety conditions and working reliability, the operational practice is based on a series of control activities. Investigations on life conditions of pressure vessels are addressed both to identify premature failures and avoid, even if their safe working can still be fully guaranteed, to put them out service for attainment of the design life, previously evaluated on the basis of conservative criteria.

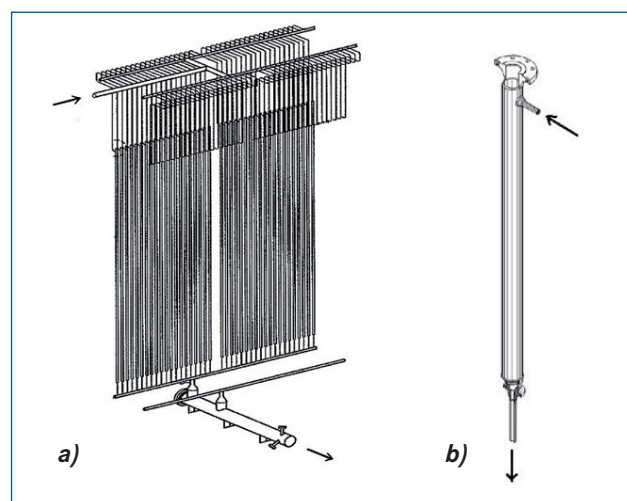
In this work we investigate the radiant tubes of a furnace for the hydrogen production from methane and water vapor by means of the endothermic reforming reaction that take place thanks to a granular Ni-based catalyst.

The furnace here considered is a chamber, coated inside by refractory material, containing 4 parallel rows of 44 vertically hung radiant tubes (Fig. 1). These tubes are constructed by butt welding three spun cast pieces, made of a Ni-Cr-Fe alloy. Radiant tubes, heated from the outside by

means of a system of gas burners, operate at temperature of about 900°C and pressure of 33 bar, for times of the order of 100000 hours.

Service temperature and tensile circumferential stress, due to the internal pressure, give rise to a severe creep conditions that led to diameter expansion [1]; inside tubes there are carburizing conditions, however unforeseen catastrophic failure may be accelerated by local oxidative phenomena [2].

Two examples of tube damage are given here: the yellow arrows in figure 2a indicates a tube section that underwent creep expansion; figure 2b shows a large longitudinal crack across a butt welded joint.



**Fig. 1 – Assembly of a tubes row (a) and detail of a single tube (b). The arrows indicate the gas flows entering and leaving the tube.**

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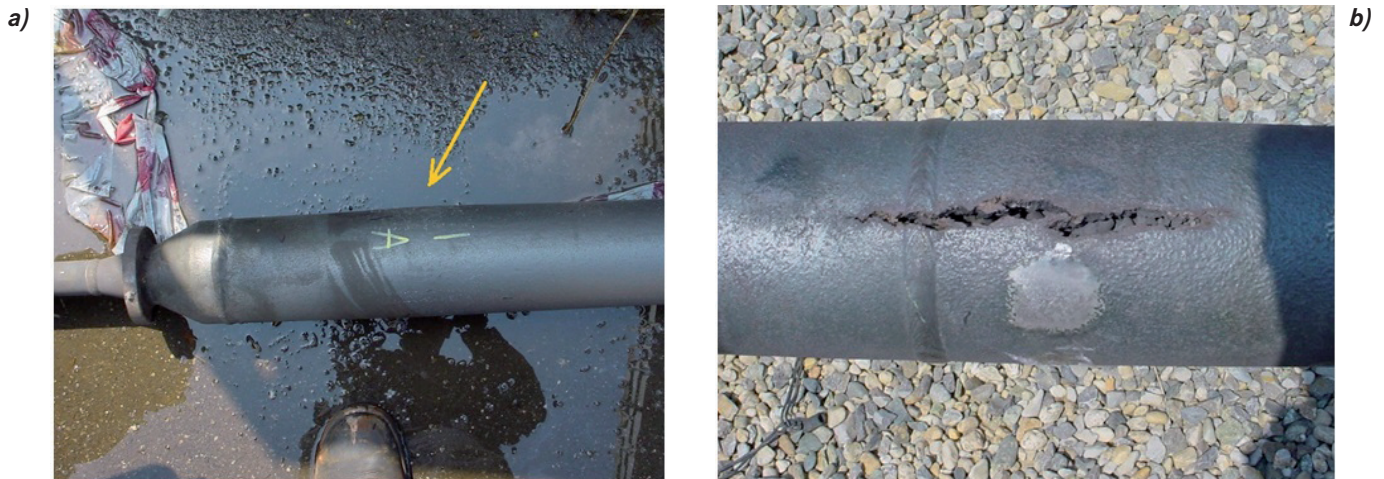
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**Fig. 2 – Examples of radiant tube damage: a) creep expansion of a tube section; b) large longitudinal crack across a butt welded joint.**

Therefore good mechanical properties and corrosion resistance at high temperature are requested for radiant tube alloys [3]. These materials have seen significant improvements during the last fifty years: in the 60s and 70s the alloy 25Cr-20Ni-Fe-0.4C, designated as HK-40, was the most utilized, while in the following decades the HP-40 alloy (25Cr-35Ni-Fe-0.4C-1.5Si) has been widely considered [4]. The high concentrations of Cr and Ni give great mechanical strength and corrosion resistance at service temperature, the presence of Si improves carburization resistance. The mechanical reinforcement of the HP-40 alloy is obtained by a dispersion of carbides particles with high hardness [5] whose stability is decisive for creep behavior [6]. Moreover, because it has been tested that microalloying elements are able to stabilize a fine dispersion of carbides, starting from the 90s HP-40 alloys have been developed with addition of Nb (about 1%) [7], Ti (up to about 0.8%) [8] and Y (about 0.3%) [9] to improve their creep behavior.

Tubes life is shortened by creep damage, being characterized by progressive microstructural changes [10], as carbides transformations, microcracks nucleation and formation of voids, typical of the final creep stage that precedes fracture [11, 12].

Unfortunately, during the furnace scheduled stop, tubes have to be decommissioned to cut metallographic samples and perform microscopic observations and mechanical test. However, because each single tube is very expensive and its damage conditions are not necessarily representative of the other tubes, many experimental works have been addressed to non destructive methods, such as eddy current and ultrasonic measurements [13, 14, 15]. Moreover, being these investigations affected by many uncertainties, studies on the relationship between microstructural degradation and mechanical properties are largely quoted in literature [2, 11, 16]. In this respect it is worth noting that the residual life evaluation depends on actual service temperatures (measured by optical pyrometers with an error of  $\pm 20$  °C), mechanical properties values after long time service, actual stress state and ther-

mal cycles associated to the furnace start-up and shut-down. So adequate means of investigation giving reliable information about tubes damage are required.

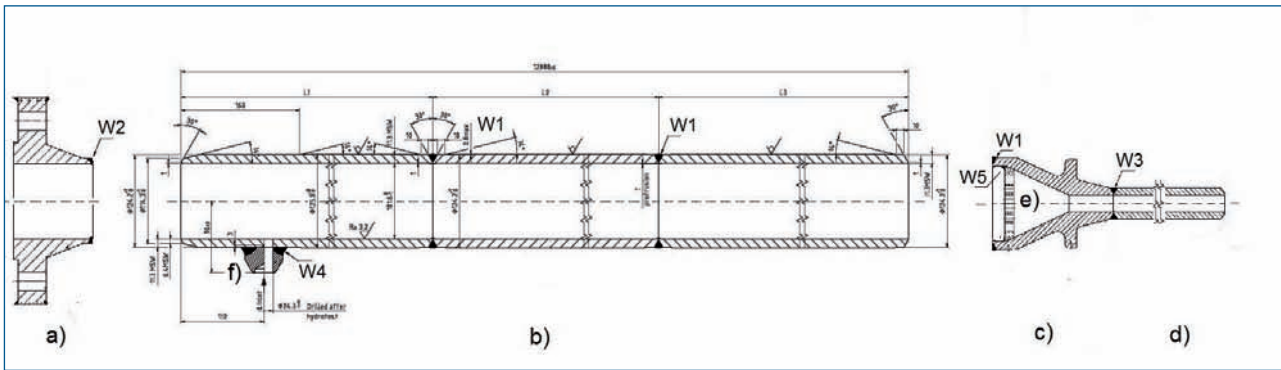
Nowadays reforming furnaces rely on the Laser Optic Tube Inspection System (LOTIS), a non-destructive control technique based on creep deformation measures performed in situ by driving a laser probe inside tubes [17]. It is very promising for the development of a criterion for decommissioning tubes that would be no longer safe until the next scheduled stop [18]. So furnaces have to be regularly inspected during each stop, in order to identify the damaged components and replace them. The radiant tubes are visually observed with the aim of identifying macroscopic damage; creep deformations are detected through LOTIS measures in order to select the tubes to put out of service for safety reasons.

In previous works [18, 19] laboratory tests were carried out to correlate mechanical properties and microstructural changes in tubes decommissioned after long time service. In this paper the experimental investigations have been extended to the butt welds that join the tube pieces. Samples for creep tests and microstructural observations were cut from various sections also including welds.

## MATERIALS AND METHODS

As shown in the working drawing (Fig. 3), radiant tubes are constructed in three butt welded pieces (W1 welds); the first one (located in the upper vertical position inside the furnace) is welded to the upper flange (W2), the third one is welded to the reducer (W1), in turn welded to the outlet tube (W3); other welds concern the gas inlet nozzle (W4) and the catalyst support grid (W5).

Radiant tubes have an internal diameter of 101.6 mm, a nominal thickness of 10.5 mm and an overall length of 12.8 m. The design conditions are pressure of 32.7 bar and temperature of 950°C, while the operating temperature is 900°C.



**Fig. 3 - Working drawing: a) upper flange, b) radiant tube constructed in three pieces, c) reducer, d) outlet tube, e) catalyst support grid, f) gas inlet nozzle.**

The three pieces of the radiant tube (b), the connection of the gas outlet (c) and the catalyst support grid (e) are made of the ASTM 608 HP-Nb alloy, a HP alloy modified by Nb micro additions; the upper flange (a) and the gas inlet nozzle (f) are made of the ASTM A182 Type F22 steel; the gas outlet tube (d) is made of the ASTM B407 UNS NO8811 Incoloy.

The composition of the ASTM 608 HP-Nb alloy is given in table 1. The microstructure of the spun cast pieces is characterized by radial austenitic dendrites, that are well delineated by carbide particles precipitated in the interdendritic spaces.

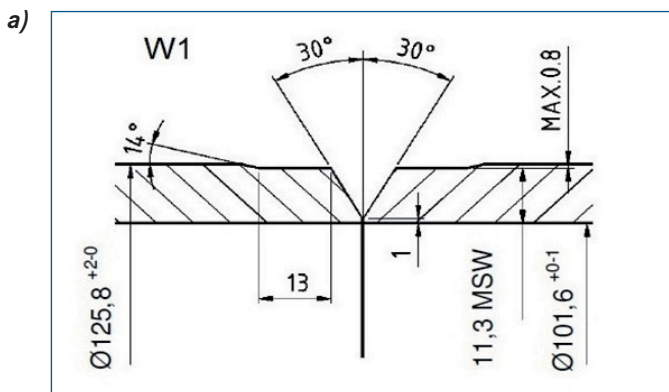
Welds are performed by means of GTAW, utilizing pure Ar as shielding gas. Fig. 4 shows the edge preparation for W1, W2 and W3 butt welds. Welding parameters are given in table 2.

Alloy	C	Si	Mn	Cr	Ni	Nb	Ti	Fe
A 608 HP-Nb	0.45	1.5	1	25	35	1.5	add.	bal.

**Table 1 - Composition given by the manufacturer of the alloy.**

	Root pass	Subsequent passes	Final pass
Current	DC	DC	DC
Electrode polarity	negative	negative	negative
Intensity (A)	140-170	100-150	100-130
Voltage (V)	14-16	14-16	14-15
Welding speed (cm/min)	4-7	6-9	5-8
Tungsten electrode	Type WS2 Ø 2,4 mm		
Filler material	UTP A2535Nb (25Cr-35Ni-1.2Nb) Ø 2,4 mm rod		

**Table 2 - Process parameters of the W1 welds.**



**Fig. 4 - Edge preparation for butt welding of tube pieces (a), welded joint (b).**

Creep deformation of tubes is due to the hoop stress caused by the internal pressure. The most damaged zones are in the lower part, near the catalyst support grid, where the gas temperature, starting from the inlet value of 500°C, reaches values around 900°C.

Creep deformation was evaluated, during the programmed furnace stop, through LOTIS measures of the internal diameter ( $D_i$ ), inserting in the upper flange a probe with the laser source and the receiving system. The probe, mechanically driven inside the examined tube, rotates up to 1800 r.p.m. and generates a helical map of internal surface, with an accuracy of 0.05 mm; so the deformation can be measured with a precision of 0.05%, being the internal diameter about 100 mm.

The deformation of a tube section ( $\epsilon$ ), on the basis of the measured internal diameter ( $D_i$ ) and referred to the nominal diameter ( $D_o$ ), equal to 101.6 mm, is given by the following equation:

$$(1) \quad \epsilon = (D_i - D_o) / D_o$$

Following a criterion developed in previous work [18, 19], the tubes where deformation reached values greater than  $\epsilon = 1.5\%$  were put out of service and utilized to take the experimental samples.

The decommissioned tubes were visually investigated for a first inspection of their damage conditions.

Creep tests were performed on samples cut longitudinally from tube put out of service after about 100000 hours and, for comparison, from samples taken from a tube in the as cast condition. Samples with welded joint were also machined.

For microhardness test and microscopic observations samples were metallographically prepared with the usual techniques and etched by a solution containing 15% of glycerol (100% concentration), 45% of  $\text{HNO}_3$  (65% concentration) and 40% of  $\text{HCl}$  (37% concentration).

Considering that the results of LOTIS measurements showed inhomogeneous deformations along the tubes axis, experimental surveys were carried out on samples cut longitudinally both from sections with high deformation (1.5-2.0%) and from undeformed sections, in order to put in comparison their microstructure and creep strength. Samples of welded joints were also cut.

Metallographic samples were observed optically and by scanning electron microscopy (SEM) with energy dispersive X-ray spectroscopy (EDS).

## RESULTS AND DISCUSSIONS

### *LOTIS measurements*

The internal diameter creep expansion along each tube was accurately recorded through LOTIS measures, performed in situ with the aim of selecting the tubes to put out of service when  $\epsilon > 1.5\%$ . Results can be summarized as 78% of tubes with diameter deformations less than 1.5%, while in the remaining 22% were recorded deformations exceeding the established limit [18].

Moreover LOTIS measurements showed that diameter deformation increases from the upper flange, where in general it is negligible, up to the lowest zone of the tube. In the decommissioned tubes diameter deformation values of 1.5-2.0% were recorded near the catalyst grid.

### *Visual inspections*

The tubes put out of service after 114000 hours show highly oxidized surfaces along all their length and zones located in the lower part which have undergone creep deformations detectable also by visual inspection. These zones are mainly near the catalyst grid, where the diameter expansion is clearly observable (fig. 2a).

### *Creep tests*

Results of creep test refer to samples in the as cast condition and samples cut longitudinally from both undeformed and deformed ( $\epsilon=1.5-2.0\%$ ) zones of a decommissioned tube. Results of samples with welds are also considered (cross-weld test). The test temperature (T) was in the range

between 920 and 980°C. In order to achieve acceptable rupture times (between 200 and 1000 hours), the test stress was assumed in the range 22-30 MPa, greater than the hoop stress at the design pressure ( $\sigma = 17.5$  MPa).

For a useful comparison, in fig. 5 the creep test results are plotted on a Larson Miller diagram ( $\sigma$  vs. LMP), being  $\sigma$  test stress and  $\text{LMP} = T (C + \log t) / 1000$ , with T (K) test temperature, t (h) rupture time and  $C=22.9$  a constant characteristic of the considered material [10].

Moreover both average and minimum curves characteristics of the A 608 HP-Nb alloy are given in the same diagram as reported in the manufacturer catalogue [20].

Creep tests results can be summarized as follows:

- the experimental point of the as supplied material (blue triangle) is on the minimum curve of the manufacturer catalogue,
- results of samples taken from undeformed zones (yellow rhombus) are similar to the as supplied material,
- results of samples taken from deformed zones (blue circle) are well below those of the undeformed material,
- samples cut transversely to the welded joint (red circle) behave in agreement with the previous ones.

The experimental diagram  $\sigma$  vs. LMP shows that specimens with welds have behavior similar to the base material with the same diameter deformation. However they differ in creep ductility: rupture elongation in creep test resulted around 3-5% for samples with welds, while it was 20-30% for samples of base material.

The residual life of decommissioned tubes can be obtained thanks to the curve extrapolated from experimental data: entering with the hoop stress  $\sigma = 17.5$  MPa, the corresponding LMP value, equal to about 32.6, allows to calculate the rupture time at a given temperature. For example, with temperatures of 900, 920 and 940°C, the corresponding rupture times are respectively 10, 3.2 and 1 year. Then rupture time after long time service is strongly dependent on temperature changes that are in the range of measurement errors typical of pyrometer systems used in reforming furnaces.

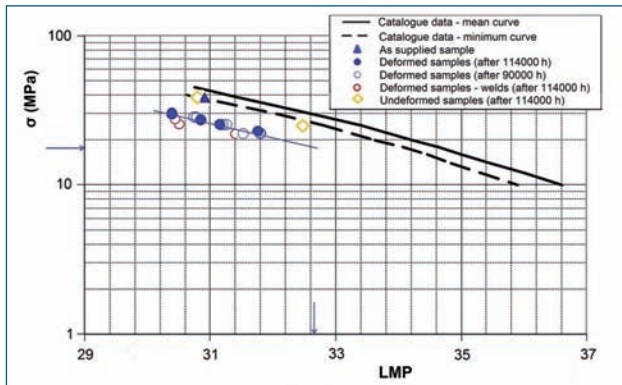
### *Optical microscopy and Vickers microhardness test*

After long time service, the A 608 HP-Nb alloy is characterized by microstructure modifications that can be related to the degree of diameter expansion.

The radiant tube walls undergo creep caused by circumferential stress and temperature and each zone has specific metallurgical features.

Some considerations can be drawn with reference to the optical micrographs shown in figure 6 at the respective stages of a typical creep curve.

- The as cast microstructure is characterized by austenitic dendrites, well outlined by a network of coarse carbides precipitated in the interdendritic spaces.
- The microstructure of samples cut from undeformed zones of a decommissioned tube is nearly similar to the one of as cast alloy, so it can be referred to the first creep stage.
- The microstructure of samples cut from deformed zone



**Fig. 5 – Creep test results: Larson Miller diagram.**

( $\epsilon = 1.5-2.0\%$ ) is characterized by a precipitation of fine secondary carbides inside the austenitic grains, optically observable at high magnification, and by nearly circular isolated microvoids.

Isolated microvoids appear during the steady state creep. In particular the absence of aligned microvoids, formed by the coalescence of two or more microvoids [21], can be considered as an indication that the final creep stage has not yet started.

In welded sections, no evidence of particular structures was detected at the boundary between base metal and molten zone (fig. 7). The latter appears to be characterized by very fine carbides aligned with the heat flow direction.

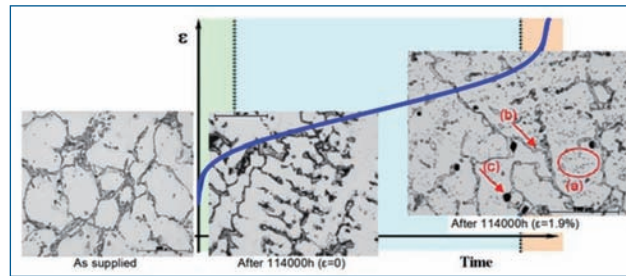
Regarding microhardness survey, it was noticed a remarkable softening after 114000 hours of service compared to values of about 400HV measured in the as cast material: microhardness values through base metal and molten zone are around 180 HV (fig. 8).

#### SEM and EDS investigations

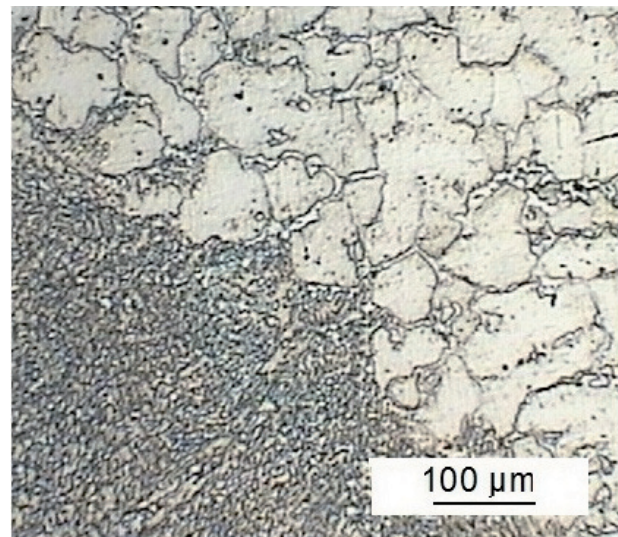
Microstructural changes in samples taken from deformed zones of a decommissioned tube respect to the as cast alloy are investigated by SEM observations and EDS measurements too.

In the as cast alloy, two kind of precipitates at the austenitic grain boundaries are clearly observable: a grey phase, with greater volume fraction, and a phase in the form of small white particles. The EDS spectra allow to recognize the grey phase as Cr carbide, while the small white particles are rich of Nb and Si.

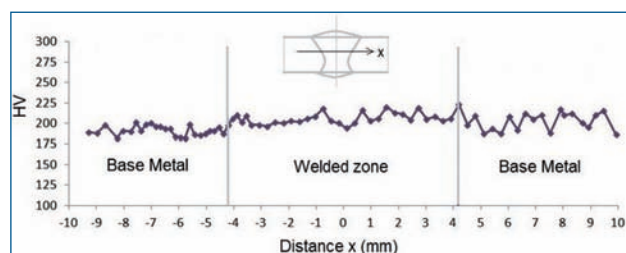
In samples taken from deformed zones, grey precipitates are located both inside grains and at their boundaries (fig. 9). These particles are recognized as Cr carbide through EDS measurements. The HP alloys underwent phase transformations due to thermal activation during service at temperatures between 1123 and 1325 K and primary Cr carbides  $M_7C_3$ , instable at high temperature, transform into intragranular and intergranular  $M_{23}C_6$  precipitates [23]. Another change regards the white precipitates at grain boundary that in deformed samples have dimension and volumetric fraction greater than in the as cast alloy; more-



**Fig. 6 – Microstructures of the ASTM 608 HP-Nb alloy, in relation to the respective level of creep deformation. In the case of material after 114000 hours of service it can be outlined: a) fine carbide precipitation inside grains, b) primary carbide at grain boundary, c) nearly circular microvoids.**

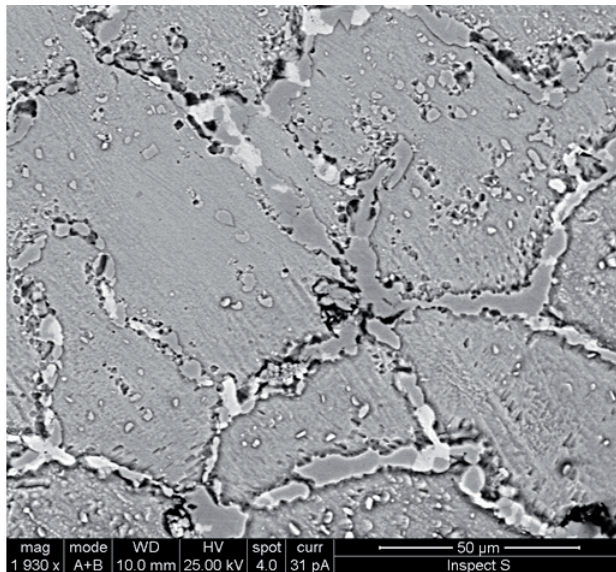


**Fig. 7 – Optical micrograph of the boundary between molten zone and base metal in a welded joint (tube decommissioned after 114000 hours).**



**Fig. 8 – Vickers microhardness (200g -15s) in a welded section (tube put out of service after 114000 hours).**

over EDS measurements give high concentrations of Nb, Ni, Si. As quoted in literature [33, 34], Nb carbides are not stable in the range of service temperatures, between 700 and 900°C, transforming into Ni-Nb silicates known as G-phase. Unlike the Nb carbides that give good creep properties, the G-phase coarsening is followed by reduction of creep resistance [3]: this negative effects can be



**Fig. 9 – SEM micrograph of a sample taken from a deformed zone (tube decommissioned after 114000 hours).**

ascribed to the interface between G-phase and matrix that becomes a preferential site for creep damage [7]. Luckily this phenomenon can be overlooked in the HP grades microalloyed with small quantities of Ti, that reduce the G-phase volume fraction [7].

## CONCLUSIONS

The experimental investigations carried out on radiant tubes, made of ASTM 608 HP-Nb alloy and installed in a reforming furnace, have confirmed that LOTIS measurements of the internal diameter give good indications on the degree of creep damage and allow to select the tubes to put out of service.

Creep tests and metallographic observations have shown that samples taken from undeformed section are free of damage and have characteristics similar to those of the as cast alloy, while samples taken from deformed section (in the range 1.5-2%) show microstructural changes and decay of creep strength. In any case, welds do not affect further the strength properties of the reforming tubes: the hardness values through them are almost similar to those of base material; furthermore, in the same conditions of diameter deformation, creep behavior of samples with welds and sample of base material is comparable.

The residual life of samples taken from deformed section is very sensitive to small temperature changes, in the range of measurement errors, becoming dangerously close to two years, that is the time between two scheduled stops of the furnace. Thus is confirmed the validity of putting out of service tubes when the internal diameter deformation is greater than 1.5%.

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