ANALYTICAL DETERMINATION AND VALIDATION BY FINITE ELEMENTS METHOD OF HYDROGEN WELD OF CARBON STEEL AFTER POST-HEATING

by

Jose L. MESEGUER-VALDENEBRO^{a*}, Antonio PORTOLES^a, and Eusebio MARTINEZ-CONESA^b

 ^a Department of Applied Physics and Materials Engineering, ETSII, Technical University of Madrid, Madrid, Spain
 ^b Faculty of Architecture and Building Engineering, Universidad Politecnica de Cartagena, Cartagena, Spain

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The objective of this work is to determine analytically the amount of hydrogen residual in a weld after having carried out post-heating for a certain period of time in order to reduce the risk of cold cracking due to the presence of hydrogen in the weld and its validation by the finite element method. Post-heating is a variable present in the welding procedures and therefore, it is mandatory in those welds that require it. This work can be helpful to determine both numerically by the finite element method and analytically the post-heating suitable in a welding process depending on that process, the welded material and the base material. In this work, the phase transformation and time difference of the phase transformation between the weld metal and base metal are not considered. The diffusivity values are those used by the reference method that analytically calculates the residual hydrogen in a carbon steel weld. There are two values of hydrogen diffusivity (minimum value and maximum value) in this way the diffusivity values that represent all types of carbon steel are collected. The least amount of hydrogen in the weld is with a post-heating to 200 °C, producing a decrease in hydrogen in the weld at a higher speed than with the rest of temperatures below this.

Key words: hydrogen-assisted cracking, diffusible hydrogen, steel, finite elemente method

Introduction

The term post-heating refers to heating carried out on a weld once it has been completed. The post-heating temperature can be equal to or greater than the preheating temperature before starting to weld. The term post-heating is different from post-weld heat treatment, since the objective of the post-weld heat treatment is to relieve stresses in the welded joint and the objective of post-heating is to reduce the hydrogen in the weld [1].

Post-heating is not mandatory in design codes but it is usually a requirement of the customer or because it is a particular design specification. The application of post-heating requires control of the time and the temperature that is applied on the weld bead.

One of the most severe manifestations of hydrogen cracking is hydrogen-assisted cracking (HAC), also known as cold cracking [2, 3], so it is necessary to perform a post-heat-

^{*}Corresponding author, e-mail: jlmeseguer507@gmail.com



Figure 1. Combination of the three factors necessary for there to be a risk of cold cracking or HAC

ing after welding in order to reduce the risk of hydrogen cold cracking [4-8].

Background

Cold cracking or HAC is caused by the combination of three factors:

- the presence of hydrogen,

residual stresses during the cooling of the weld, and

- hard micro-structures in both the weld metal and the HAZ.

Figure 1 shows the combination of the three factors that lead to a risk of cold cracking.

There are two factors that increase the probability of cold cracking:

the temperature of the weld is between 50 °C and 150 °C: the probability of cracking is then at a maximum [9] and

 the cold cracking of a weld is delayed for hours and sometimes days, the cracks being hardly detectable.

Studies carried out to determine hydrogen in a weld

More than 1500 studies have been carried out to determine the residual hydrogen in a weld once the welding has been completed. In this work we will mention the most relevant [4-8].

The work carried out by Padhy and Komizo [10] reviews the state of the art on hydrogen diffusivity in steel welds.

In the 1950's, one of the first works to determine the importance of hydrogen in welding was carried out by Grant and Lunsford [11], where the cold cracking in carbon steel was investigated. From this, studies were carried out to determine the minimum preheating temperature that a weld should have in order to reduce the cold cracking of the steel. These studies were carried out by Ito and Bessyo [12], Suzuki *et al.* [13], Satoh *et al.* [14], Yurio-ka *et al.* [15, 16] and in European regulations EN 1011-2 [1] and the American AWS D1.1 [17]. On the other hand, there are standards that have arisen with the need to experimentally determine hydrogen in a weld, such as IIW / ISO 3690: 2012 [18], ANSI / AWS A4.3: 1993 [19], BS 6693: 1988, JIS Z 3118: 2007, JIS Z 3113: 1975, DIN 8572: 1981 Part 1, AS / NZS 3752: 2006, GOST 23338-91 and BIS IS 11802: 1986. These standards include the glycerin method, the mercury method, the hot gas scanning chromatography method and the vacuum extraction method.

There are other, analytical, methods that make an estimation of the hydrogen that can be left in a weld once the post-heating is applied [20-22]. Finally, there are numerical methods that use the calculation by finite elements to determine the hydrogen residual in a welded joint [23-26].

Diffusion of hydrogen in welding

Bailey *et al.* [21] assess the diffusion of hydrogen in a ferritic steel, as shown in fig. 2, where it is observed that the hydrogen diffusion is between an upper limit (dashed line) and a lower limit (continuous line).

For microalloyed steels and low carbon steels, the hydrogen diffusion curve is defined by reference [27] and the diffusion of steel with martensitic and ferritic microstructure is defined by reference [28].

In the case of assessing the diffusion of hydrogen during the welding process, where temperatures higher than those shown in fig. 3 are reached, it is recommended to use the aforementioned figure, where the effect of hydrogen diffusion in a weld can be evaluated for a single pass or multipass and for minimum and maximum values of diffusivity.



Figure 2. Diffusion curves for ferritic steels [21]



Figure 3. A scatterband for hydrogen diffusion coefficients in microalloyed and low carbon structural steels [24, 28]

A study conducted by Nelson and Stein [29] determines the diffusion of hydrogen for iron in alpha phase, low alloy carbon steel 4130 in accordance with ASTM A29, and stainless steel AISI 304, by means of the following analytical expressions:

Iron, alpha phase:

diffusion

$$D = 2.33 \cdot 10^{-3} \,\mathrm{e}^{(-6680/\mathrm{R}T)} \tag{1}$$

saturation concentration

$$C_{\rm sof} = 3.45 \cdot 10^{-2} \, p^{1/2} {\rm e}^{(-27600/{\rm R}T)} \tag{2}$$

For standard 4130 low alloy carbon steel:

- diffusion

$$D = 3.53 \cdot 10^{-3} \,\mathrm{e}^{(-12600/\mathrm{R}T)} \tag{3}$$

- Saturation concentration

$$C_{\rm sat} = 1.85 \cdot 10^{-3} \, p^{1/2} {\rm e}^{(-27100/{\rm R}T)} \tag{4}$$

For tempered 4130 low alloy carbon steel:

diffusion

$$D = 3.56 \cdot 10^{-3} \,\mathrm{e}^{(-7950/\mathrm{R}T)} \tag{5}$$

saturation concentration

$$C_{\rm sat} = 229 \cdot 10^{-3} \, p^{1/2} {\rm e}^{(-27200/{\rm R}T)} \tag{6}$$

For AISI 304 stainless steel:

diffusion

$$D = 2.72 \cdot 10^{-2} \,\mathrm{e}^{(-54400/\mathrm{R}T)} \tag{7}$$

saturation concentration

$$C_{\rm sat} = 8.6 \cdot 10^{-3} \, p^{1/2} {\rm e}^{(-9600/{\rm R}T)} \tag{8}$$

The ASTM G 148-97 standardizes an experimental method that determines the diffusivity curves of hydrogen in a metal [29].

In the study conducted by Feng *et al.* [30], equations are shown that determine the solubility of hydrogen in carbon steel:



Figure 4. Post-heating application for a certain time *t* [20]



Figure 5. Types of joints and the position of the parameter L; (a) single V butt, (b) double V butt, (c) fillet weld (both sides), (d) single V at fillet, and (e) one-sided fillet joint [20]

$$=159e^{-23.54/RT}$$
 (9)

Feng *et al.* [30] show the solubility data for A106 grade B with a content of 0.185:

- 150 °C = 29.25 mol/m³ MPa^{1/2}

- 175 °C = 1.26 mol/m³ MPa^{1/2}

 $200 \text{ °C} = 3.64 \text{ mol/m}^3 \text{ MPa}^{1/2}$

The concentration of hydrogen saturation in steel is used as an input in the simulation by finite elements.

Analytical determination of hydrogen in a weld

In [20] the hydrogen in the weld is determined by the area under the curve shown in fig. 4.

The analytical expression that determines the diffusion coefficient is:

$$T = \int_{0}^{t} D \mathrm{d}t \tag{10}$$

where T is the area under the curve. If the descent time is not known, the estimated area would be the area of a rectangle.

There is another geometric parameter, L, that depends on the thickness of the welded piece, differentiating between a fillet joint and a butt joint, fig. 5, where the joints figs. 5(a) and 5(b) are butt joints and the joints figs. 5(c)-5(e) are fillet joints [20].

Once the type of joint is determined, the % of the residual hydrogen is according to the following chart.

The value of T is obtained by multiplying the diffusivity, D, by the duration of the post-heating carried out, corresponding to the abscissa axis of fig. 6. The geometric factor, L, corresponds to the thickness of the butt joint. The, L, squared is divided by, T, and the result of the quotient is introduced on the abscissa axis of fig. 6. To determine the residual hydrogen at the centre of the butt weld, the curve shown in fig.



residual for the butt joint [21]

the centre of the butt weld, the curve shown in fig. 6 is used. To do this, the adjustment equation shown below is used, which allows the residual hydrogen in the weld to be determined:

Remaining hydrogen =
$$-542.69 \left(\frac{T}{L^2}\right)^6 + 1807 \left(\frac{T}{L^2}\right)^5 - 2310.5 \left(\frac{T}{L^2}\right)^4 + 1355 \left(\frac{T}{L^2}\right)^3 - 229.08 \left(\frac{T}{L^2}\right)^2 - 171.12 \frac{T}{L^2} + 100$$
 (11)

Case study

Analytical model

A practical case for a butt joint, in a single V, 30 mm thick and with a post-heating temperature of 80 °C, 150 °C, and 200 °C for five hours (18000 seconds) would be as we shown in tab. 1.

	Values (80 °C)	Values (150 °C)	Values (200 °C)
$D_{\min} \left[\mathrm{cm}^{2} \mathrm{/s}^{-1} \right]$	2 · 10 ⁻⁷	9 · 10 ⁻⁶	$1.8 \cdot 10^{-5}$
$T_{\min} [\mathrm{cm}^2] = D_{\min} \cdot t$	$2 \cdot 10^{-7} \cdot 18000 = 3.6 \cdot 10^{-3}$	$9 \cdot 10^{-6} \cdot 18000 = 0.16$	$1.8 \cdot 10^{-5} \cdot 18000 = 0.32$
T_{\min}/L^2	$3.6 \cdot 10^{-3}/1.5^{2} = 1.60 \cdot 10^{-3}$	$0.16/1.5^2 = 7.20 \cdot 10^{-2}$	$0.32/1.5^2 = 1.44 \cdot 10^{-1}$
Residual hydrogen [%] (minimum)	99.73	86.94	73.77
$D_{ m max} \left[m cm^2/s^{-1} ight]$	$1.5 \cdot 10^{-5}$	$3 \cdot 10^{-5}$	5 · 10 ⁻⁵
$T_{\rm max} [{\rm cm}^2] = D_{\rm max} \cdot t$	$1.5 \cdot 10^{-5} \cdot 18000 = 0.27$	$3 \cdot 10^{-5} \cdot 18000 = 0.54$	$5 \cdot 10 - 5 \cdot 18000 = 0.9$
$T_{\rm max}/L^2$	$0.27/1.5^2 = 0.12$	$0.54 / 1.5^2 = 0.24$	$0.9/1.5^2 = 0.4$
Residual hydrogen [%] (maximum)	78.07	58.14	38.75

Table 1. Percentage of residual hydrogen in weld for different temperatures

The *L* is equal to half the thickness of the plate, that is, 3/2 = 1.5 cm.

On the other hand, the distance, x, that the hydrogen will travel during a period of time, t, determined for diffusivity, D, will be:

$$x = 2(Dt)^{1/2}$$
(12)

For this particular case, the minimum and maximum distance covered will be as we shown in tab. 2.

Table 2. Percentage of residual hydrogen in weld for different temperatures

0			•
	Values (80 °C)	Values (150 °C)	Values (200 °C)
X_{\min} [cm]	0.12	0.80	1.13
$X_{\rm max}$ [cm]	1.03	1.46	1.89

The analytically obtained results in tab. 2 can be used for validation by the finite element method (FEM).

Dissociated hydrogen in the electric arc of the weld

In [22] the atomic hydrogen dissociated in the electric arc is determined, obtaining the following system of equations:

$$(n_{H})^{2} = \exp\left(\frac{-53844.6}{T} + 14.49\right) n_{H_{2}} \left(0.95 + n_{H} + n_{H_{2}}\right)$$

$$0.05 = n_{H_{2}} + \frac{1}{2}n_{H}$$
(13)

Solving the system of eq. (13), the moles of atomic hydrogen nH and moles of molecular hydrogen nH₂ are cleared as a function of the temperature at which the weld metal T [K].

For the particular case, the temperature of 2800 K is considered: nH = 0.03274 mol and $nH_2 = 0.03363$ mol.

The percentage of dissociation is equal to:

$$\frac{\left(\frac{0.03274}{2}\right)}{0.05} \times 100 = 32\%$$

Therefore, the maximum amount of hydrogen that welding can have is 32% of the hydrogen present in the filler metal, that is, of every 100 mg of hydrogen in the filler metal, 32 mg of hydrogen is present in the weld.

This paragraph is introduced at this point in case it is intended to carry out an experimental validation knowing the amount of hydrogen in the electrode in the previous moment to being welded.

Numerical analysis of the diffusion of hydrogen in post-heating by finite elements

The numerical model using finite elements to determine the hydrogen in a welded joint is the following:

- The diffusion coefficients of hydrogen at the temperature to which the post-heating is applied are according to tab. 1.
- Once the diffusion coefficients of hydrogen at the post-heating temperature are determined, eq. (15) simulates the hydrogen diffusion based on the analogy of the Fourier heat conduction differential equation and Fick's second law, eq. (16):

$$\rho c_{p} \left(\frac{\partial \vartheta}{\partial t} \right) = \left(\frac{\partial}{\partial x} \lambda \frac{\partial \vartheta}{\partial x} + \frac{\partial}{\partial y} \lambda \frac{\partial \vartheta}{\partial y} \right) + \dot{q}_{E}$$
(14)

$$\frac{\partial HD}{\partial t} = \frac{\partial}{\partial x} D_{\text{eff}} \frac{\partial HD}{\partial x} + \frac{\partial}{\partial y} D_{\text{eff}} \frac{\partial HD}{\partial y}$$
(15)

In eq. (15) it is assumed that there is no heat source. Therefore, substituting the temperature ϑ for the HD hydrogen concentration, substituting the thermal conductivity λ for the diffusion coefficient, D_{eff} , and equalizing the density, ρ , and the specific heat, c_{ρ} , to the unit, the simulation can be carried out by the transient thermal module of ANSYS for the diffusion of hydrogen in the post-heating process. In eq. (16), HD represents the concentration of hydrogen

in the nodes for each time instant in each of the directions for a diffusion value of the hydrogen that depends on the post-heating temperature.

The ANSYS has a particular way of evaluating the migration effect of hydrogen:

$$J = \frac{[D]C\Omega}{RT} \nabla \sigma_{H}$$
(16)

where [D] is the diffusivity matrix, considering the values of D_{eff} , C – the molar concentration of hydrogen, Ω – the molar volume, R – the universal constant of the gases, T – the temperature at which the post-heating is carried out, and σ_H – the hydrostatic tension that, for the case of coupled thermal-diffusion, is equal to the unit.

Unlike the analytical method, in the analysis by finite elements it is necessary to know the hydrogen concentration that the welding has before starting the post-heating, since this is the boundary condition or initial condition, also known as the Dirichlet condition [25, 26], assuming a uniform distribution of hydrogen in the weld. It is considered that the welding metal has initial conditions of the hydrogen concentration in the welding of 1, which represents the unit of hydrogen in the weld bead. The initial concentration of hydrogen in ANSYS is represented as an elementary unit that can also be expressed in 100, as a %. The base metal is considered to have no starting hydrogen concentration. The post-heating will be carried out for five hours, which is the time used in the analytical method and both results will then be compared.

- In the finite elements model, ANSYS proposes the diffusive model:

$$J = -[D]\nabla C + \frac{[D]C\Omega}{kT}\nabla\sigma_{H} - \frac{[D]CQ}{kT^{2}}\nabla T - \frac{[D]CZe}{kT}\nabla V$$
(17)

where [D] is the diffusion matrix, C – the hydrogen concentration, C_{sat} – the hydrogen saturation concentration, Ω – the atomic volume of hydrogen, Q – the heat of mass transport, Z – the atomic charge number, e – the elementary charge, k – the Boltzmann's constant, T – the absolute temperature, σ_H – the hydrostatic stress, and V – the voltage.

Equation (17) is reduced to the first term since non-e of the other mass transport phenomena are considered. The C_{sat} is the entry data of the steel and considered as a property of the material.

Finite element analysis of hydrogen diffusion in welding

The geometry of the model corresponds to the fig. 7.

Performing the meshing of the joint is shown in fig. 8. The mesh density is slightly greater in the bead than in the base metal because that is where the initial concentration of





Figure 7. Modelling of the weld to be evaluated in ANSYS – dimensions in mm

Figure 8. Meshing the joint

hydrogen is applied. The mesh size is adequate, having performed a prior mesh sensitivity analysis.

At the initial time in the filler metal a concentration of 1 and a concentration of 0 is applied to the base metal with a post-heating temperature of 80 °C, 150 °C, and 200 °C.

The analysis performed is carried out with the transient structural analysis module of ANSYS and therefore, the boundary conditions are gravitational loading and fixing at the base of the weld to converge the model, as shown in the following figure. The calculation module has an associated diffusivity analysis macro.

The structural loads applied to the model are the fig. 9. The thermal and concentration loads applied to the model are the fig. 10.



Figure 9. Structural loads applied in the model

Figure 10. The thermal and concentration loads applied to the model

The initial concentration is applied to the weld bead with a value equal to 1. For the case of 200 °C, that temperature is applied for five hours. The convection applied is equal to 50 W/m^3 .

Numerical results by finite elements

The results obtained by ANSYS are the fig. 11.

Cases 1, 2 and 3 correspond to the maximum diffusivities for temperatures of 80 °C, 150 °C, and 200 °C, respectively. The same happens with Cases 4, 5, and 6 for the minimum diffusivities for temperatures of 80 °C, 150 °C, and 200 °C, respectively.

Discussion of results

In fig. 12 it is observed that the weld that contains the most residual hydrogen is that which has a post-heating of 80 °C for the minimum diffusivity value, and the case where there is least hydrogen residual in the weld is that which has a post-heating at 200 °C with the highest diffusivity value.

Table 2 shows the distance at which hydrogen diffuses for each of the cases studied. Qualitatively, there is a relationship between the results in tab. 2 and the results observed in fig. 12, where it can be seen that the width of the dissipation of hydrogen in the weld is proportional to the diffusivity value used and that it coincides with the results shown in tab. 2.

Figure 13 shows the residual hydrogen in the centre after welding for five hours for all cases studied.

The curves that have a greater slope, *i.e.*, that have less residual hydrogen after five hours of post-heating, are those that have a post-heating temperature of 150 °C and 200 °C with a maximum diffusivity. The slope of the curve for the post-heating at 200 °C with the mini-

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Figure 11. Detail of results of the post-heating welding

mum diffusivity is very similar to the curves with post-heating at 80 °C and 150 °C for the maximum diffusivity. The most unfavorable situation, *i.e.*, where there is the most residual hydrogen, occurs in the cases where the diffusivity is minimal for temperatures of 80 °C and 150 °C. For the most favourable cases, *i.e.*, for post-heating at 150 °C and 200 °C for maximum diffusivity, it is observed that a difference of 50 °C implies a 14% decrease of the residual hydrogen.



Comparison of the residual hydrogen analytically calculated with respect to the hydrogen obtained by FEM

Comparison of the residual hydrogen analytically calculated with respect to the hydrogen obtained by FEM is shown in fig. 13.



Figure 13. Comparison between both methods for calculating the residual hydrogen in weld

The maximum error between the two methods is 10% for D_{max} is 150 °C. In the residual cases the error is lower. The FEM model is acceptable in estimating the residual hydrogen in the centre of the weld.

Conclusions

- The residual hydrogen is analytically calculated in a 30 mm thick butt weld after the post-heating has been applied from a reference method, and the results obtained are validated by finite element calculation, using the transient structural module. The maximum error obtained between the analytically calculated residual hydrogen and the hydrogen calculation by the FEM is 10%.
- The finite element calculation allows the variation of the hydrogen residual in the weld over time to be checked.
- Using the FEM, the effect of a drop in the post-heating temperature on the residual hydrogen in the weld can be simulated.
- The optimum post-heating temperature for five hours on a 30 mm thick butt weld of carbon steel is 200 °C.
- The calculation method presented does not take into account the number of welding passes since this study evaluates hydrogen once the welding is finished.
- A mesh sensitivity analysis has been carried out and for hydrogen diffusivity processes using the FEM, the use of coarse meshes gets a satisfactory result.
- The least amount of hydrogen in the weld is with a post-heating to 200 °C, producing a decrease in hydrogen in the weld at a higher speed than with the rest of temperatures below this.

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