JMEPEG (1994) 3:612-618

Postweld Electrotransport Treatment

B. Mishra, D.L. Olson, and S.A. David

Hydrogen damage susceptibility of high-strength steel weldments and in other advanced materials, such as intermetallics, is a problem that restricts their application in many structural uses. A high-temperature (~500 °C) postweld treatment is often applied to remove the diffusible hydrogen and make the material more resistant to hydrogen damage. An electrotransport technique, using a direct current, can be applied at room temperature to move hydrogen from the underbead region, which is crack susceptible. The method has been used to electrorefine many metals and alloys containing both interstitial and substitutional solutes. This paper reviews the electrotransport theory and applies it to estimate the time of electrotransport treatment for lowering the hydrogen content from the underbead region by 50%. Thus, this theoretical work forms the basis for developing a new practice and provides sample calculations. Under a combined influence of the diffusional and electrotransport at room temperature using an electric field of 0.10 V/cm, ~30 min is required to remove 50% of the hydrogen from a starting concentration of 5 ppm. Theoretically, the treatment will also generate a heat of 0.95 kcal/s, which may not cause an excessive temperature increase in the workpiece. The electrotransport technique thus has the potential for a low-cost, low-temperature practice for mitigating hydrogen damage susceptibility.

Keywords

electrotransport, hydrogen diffusion, steels, welding

1. Introduction

THE introduction of hydrogen in metals and alloys can lead to catastrophic failures under certain conditions and severely restrict the life of structural and other engineering components (Ref 1, 2). Hydrogen is particularly harmful when it is picked up by weldments in high strength steels. Hydrogen damage has long been recognized as a potential source of welded structural failure. Therefore, postweld heat treatment is commonly used to reduce the susceptibility to hydrogen damage for a steel with a high carbon equivalent. Generally, the practice is to heat steel weldments to 500 °C for a number of hours determined by 1 h/in. of steel thickness. This method of lowering the susceptibility of weldments to hydrogen damage is very expensive due to both the cost of energy and the influence of the excessive annealing time on productivity. The increasing use of high strength steels in structural applications to reduce weight has been limited by the constraints on meeting the necessary preheat and postheat treatment requirements.

Electrotransport of hydrogen under the influence of an electrostatic force field is a phenomenon that has been well researched (Ref 3) over the last four decades in many metal and alloy systems, including iron. In simple terms, if a direct current of controlled magnitude is applied across a solid, hydrogen can be made to move from anode to cathode, thus offering a method for lowering the hydrogen concentration from the high hydrogen content sites. This movement can redistribute the amount of hydrogen in the metal and eliminate hydrogen damage susceptible zones. Such a method is analyzed in this paper, and a distinct scenario is presented where electrotransport can be successfully used to eliminate hydrogen damage susceptibility in welded steel components. Sufficient lowering in hydrogen concentration can be achieved using practical values of current density without excessive heating of the component. In addition, recognize that the concentration gradient driven hydrogen transport will supplement the lowering of hydrogen in sensitive areas, such as the underbead region of the weld. The theoretical analysis presented in this paper shows the feasibility of applying electrotransport principles to a new technology with potential gains in terms of performance and cost saving.

2. Background Information

2.1 *Hydrogen in Weldments*

Weld hydrogen comes from many possible sources, such as the moisture in the electrode coating; wet, rusted, or coated weld surfaces; the introduction of hydrocarbons into the weld pool; and poor shielding gas control. Figure 1 shows the effect of welding method on the possible weld hydrogen level (Ref 4). The available hydrogen can be easily introduced into the weld pool because liquid steel has a high solubility for hydrogen. As the weld pool drops in temperature and solidifies, the resulting solid weld metal becomes supersaturated with hydrogen, which causes concentration gradients that allow for hydrogen transport. Some of the hydrogen escapes from the weld into the atmosphere, but a significant fraction diffuses into the near heat affected zone, which transforms into austenite during the weld thermal cycle. Since austenite has a much larger hydrogen solubility than ferrite or martensite, a high hydrogen concentration develops in this thin austenitic region. This underbead region, after becoming charged with hydrogen during cooling, transforms to harder microstructural constituents, such as martensite and/or bainite. This localized high hydrogen concentration is in a matrix of martensite that causes cracking. The resulting

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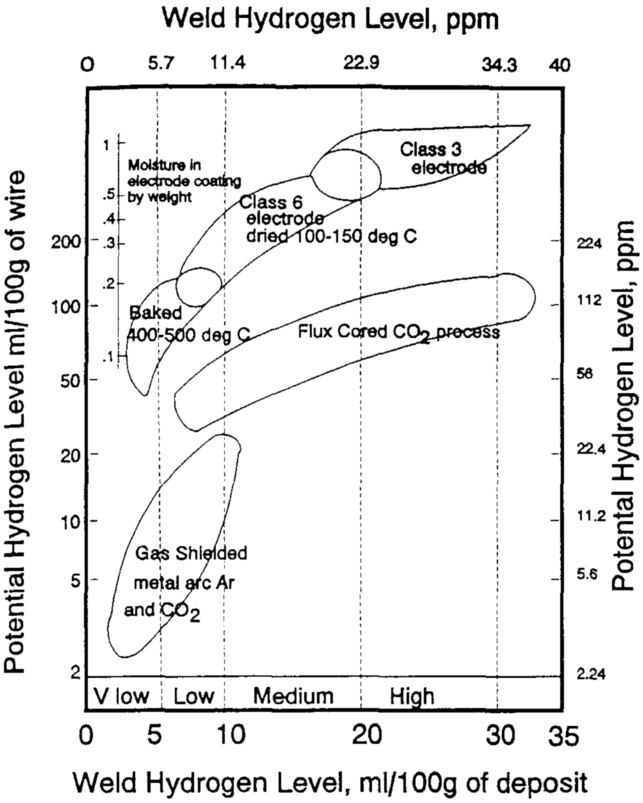

Fig. 1 Relationship between the possible and actual hydrogen levels in welds as a function of different processes and drying procedures (Ref 4)

crack is an underbead crack. A schematic diagram of the underbead cracking and the need to lower the hydrogen content from the crack susceptible zone is shown in Fig. 2. Hydrogen cracks can also occur in the weld metal, but this metal seems to be less susceptible, except for very high hydrogen contents, and often develops a cracking pattern known as chevron cracking, which aligns with the solidified weld structure (Ref 5). There is a transition from heat-affected zone cracking to weld metal cracking as weld metal tensile strength increases (Ref 6). The critical cracking stress decreases with increasing total hydrogen content in carbon steels (Ref 7), highlighting the need for lowering the hydrogen content in high strength steels.

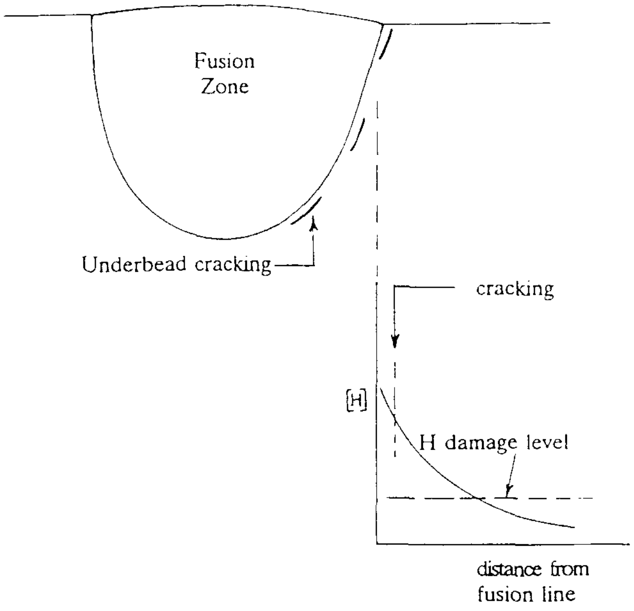

Fig. 2 Schematic diagram of underbead cracking and associated profile for hydrogen

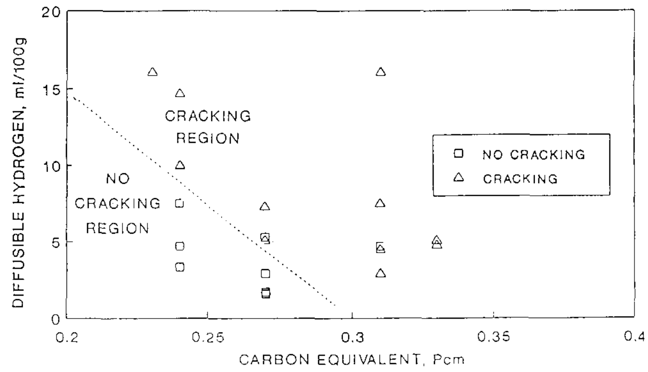

Fig. 3 Diffusible hydrogen content as a function of the carbon equivalent depicting the crack susceptible region (Ref 8)

2.2 *Removal of Hydrogen*

The prevention of hydrogen cracking can be affected by: (a) proper weld preparation, i.e. removal of oil, rust, and paint, (b) use of low hydrogen electrodes without cellulose binders, (c) preheating of the base metal, (d) postheating of the weld metal, and (e) use of austenitic filler materials. Preheat and postheat treatments are commonly used to lower the hydrogen damage susceptibility. As a rule of thumb, preheat is only applied when the carbon equivalent is between 0.40 and 0.65 whereas both preheat and postweld treatments are employed for carbon equivalents over 0.65. The hydrogen cracking tendency as a function of diffusible hydrogen content and the carbon equivalent are shown in Fig. 3 (Ref 8). Two regions are shown in the figure. In one, no cracking occurs (at room temperature). In the other region, typified by higher carbon and hydrogen contents, cracking occurs. The temperature of postweld heat treatment, used to alleviate hydrogen cracking susceptibility, is lower than the temperature used to stress relieve weldments. This thermal experience needs to be sufficient to transport hydrogen away from the weld region and thus reduce the local hydrogen content below the concentration that promotes hydrogen crack growth.

Postweld heat treatment relies primarily on the chemical potential gradient and a sufficient annealing temperature to achieve a large enough mobility factor to transport sufficient hydrogen. The concept evaluated in this paper is the use of the electrical potential gradient to assist in the transport of hydrogen and to offer a new practice of postweld electrotransport to reduce hydrogen cracking susceptibility.

3. Theoretical Approach

3.1 *Electrotransport Theory*

The mass transport phenomenon in condensed phases, such as metals and alloys under the influence of an electrostatic force field, is described as electrotransport. The combined influence of electronic conduction and diffusion make electrotransport dependent on the electronic, vibrational, and spatial aspects of local defects. Electrotransport studies have been made for a wide range of metals, and interesting results have been observed, especially in the case of the electrotransport of hydrogen and its isotope in some metals. The theory of electrotransport is described here. Atomic transport in solids results from various driving forces, as suggested in the generalized flux equation:

$$J = -c\,M\,(\delta\mu/\delta x + \delta T/\delta x + e\,\delta V/\delta x) \qquad \text{(Eq 1)}$$

where J is the flux in number of atoms transported per unit time through a unit cross-sectional area, $\delta\mu/\delta x$ is the chemical potential gradient, $\delta T/\delta x$ is the thermal gradient, $\delta V/\delta x$ is the electrical potential gradient, c is the concentration of diffusing species in number of atoms per unit volume, M is the mobility in transport velocity per unit driving force, and e is the fundamental charge of an electron. All these potential gradients are occurring during the welding process.

Atomic transport due to high electric current density has been well documented (Ref 3). Solute atoms appear to some degree in the solid solution as ions migrating under the influence of an applied field, which may be considered as acted upon by two forces. The first is the field force or electrostatic force, F_{es}, which is given by Z_ieE, where E is the electric field gradient ($\delta V/\delta x$) on the bare ion of valence Z_i (1 for hydrogen). This force is cathode directed for a positive ion and anode directed for a negative charged particle. For a solute atom in a metallic solid solution, such as hydrogen in steel, the particle is most likely to be positive resulting from a positive core and a transfer of an electron or electrons to the conduction band or to an unfilled *d*-level. It has been reported (Ref 9) that hydrogen in iron solid solution transfers its electron to an unfilled iron *d*-level. F_{es} also includes any additional electrostatic shielding effects due to the electrons surrounding the ions (Ref 10).

The second force, F_{wd}, is known as the drag or friction-wind force due to resistance of the migrating atom in the electron current or electron wind and arises from the momentum transfer by the electrons as they collide with the impurity, e.g. hydrogen (Ref 11). This force is anode directed relative to electron current in the metal and is directly related to the current density. This electron-wind mechanism, as the basic force for electrotransport, was proposed by Skaupy (Ref 12). Fiks (Ref 13) and Huntington and Grone (Ref 14) employed a ballistic model, and Bosvieux and Friedel (Ref 15) used a polarization charge model to evaluate the friction-wind force. The friction-wind force is treated as the net momentum lost per second by the electrons to a single defect and is given as nmv_D/t_DN_D, where n and N_D are the electron and defect densities, respectively, m is the electronic mass, v_D is the electron drift velocity, and t_D is the collision time for electron scattering with the defect. Normally in a metal, the size of the electric field, which draws a large current, is very small.

Table 1 Values for effective valence of hydrogen in bcc transition metals at 298 K

Metal	Effective valence, Z^*, of hydrogen
Nb	+1.3 to +2.0
Mo	+0.2 to +1.1
Ta	−0.05 to +0.20
V	+1.3 to +1.5
Fe	+0.23 to +0.27

From Ref 3

The resulting force, F_i, is given by:

$$F_i = F_{es} - F_{wd} = Z_ieE - F_{wd} \qquad \text{(Eq 2)}$$

The friction-wind force, F_{wd}, is given by δ_iE, where δ_i is the friction coefficient for specie, i. Substituting and rearranging Eq 2 results:

$$F_i = e\,E\,(Z_i - \delta/e) = e\,E\,Z^* \qquad \text{(Eq 3)}$$

where Z^* is known as the effective valence and is given by $Z^* = Z_i - \delta_{el}/e$. δ_{el} is the friction coefficient for an electron. Reported values of effective valences for hydrogen in bcc transition metals are shown in Table 1.

The velocity of transport, v_i, of solutes is directly proportional to the applied electric force F_i, mathematically shown as:

$$v_i = B_i \cdot F_i \qquad \text{(Eq 4)}$$

where the proportionality constant B_i, known as absolute mobility, is a measure of the conductance to atomistic transport in the lattice. At steady state, the external driving force is also linearly related to the applied electric field, E, and is given by Eq 3. The absolute mobility and diffusional transport are related by the Nernst-Einstein relation:

$$D_i = B_ikT\,(1 + d\ln\gamma/d\ln c) \qquad \text{(Eq 5)}$$

where k is the Boltzmann constant, T is the absolute temperature, γ is the activity coefficient, and c is the solute concentration. In the case of an ideal solution, the activity coefficient is constant and independent of concentration. The above equations yield:

$$Z^* = \frac{U_i \cdot k \cdot T}{D_i \cdot e} \qquad \text{(Eq 6)}$$

where U_i is the electronic mobility given as the transport velocity per unit applied electric field. Equation 6 requires that the force on the solute be a linear function of the applied electric field. Therefore, Z^* is not the ionic charge on an interstitial

Table 2 Interstitial electromigration and their direction

Solvent metal	Anode-directed solutes	Cathode-directed solutes
αFe	...	H,D,C,N
γFe	N	H,C,B
Ni	...	H,D,C
Co	...	C
V	...	C,N,O
βTi	O	C
W	...	C
Ta	...	C,H
Mo	...	H

From Ref 3

atom. It is the number of fundamental charge units that the migrating solute would need in order that its motion in the electric field be guided by the electrostatic force alone. Equation 6 can be used to determine the migrating velocity of hydrogen, v_H, for a given temperature, T, applied electric field, E, and the diffusion coefficient of hydrogen, D_H.

3.2 *Electrotransport of Hydrogen*

Using the equations developed above, the absolute mobility of hydrogen, B_H, resulting from the electric field and the resulting electric current can be defined by:

$$B_H = v_H/F_i = D_H/fkT \qquad \text{(Eq 7)}$$

where f is the lattice correlation coefficient. Then the velocity becomes:

$$v_H = (D_H/fkT)\, e\, E\, Z^* \qquad \text{(Eq 8)}$$

Equation 8 is the special form for hydrogen electrotransport obtained by rearranging the general Eq 6 described earlier. The electronic mobility, U_H, (transport velocity per unit electric field) for hydrogen becomes:

$$U_H = \frac{\partial v_H}{\partial E} = \frac{D_H}{f \cdot k \cdot T} \cdot e\, Z^* \qquad \text{(Eq 9)}$$

The hydrogen flux can be determined by the following expression knowing the electronic mobility and concentration profile of hydrogen in the weldment for any given applied electric field, E:

$$J_H = N_H\, U_H\, \mathrm{E} - D_H\, (\delta N_H/\delta x) \qquad \text{(Eq 10)}$$

where N_H is the concentration of hydrogen in atoms/cm^3. In case of the movement of hydrogen from the weldment to lower concentration sites in steel, the two fluxes, diffusional and electrical, are additive in Eq 10.

Many metals and alloys are capable of taking large amounts of hydrogen into solid solution even at room temperature, and hydrogen is very mobile over a reasonable temperature range.

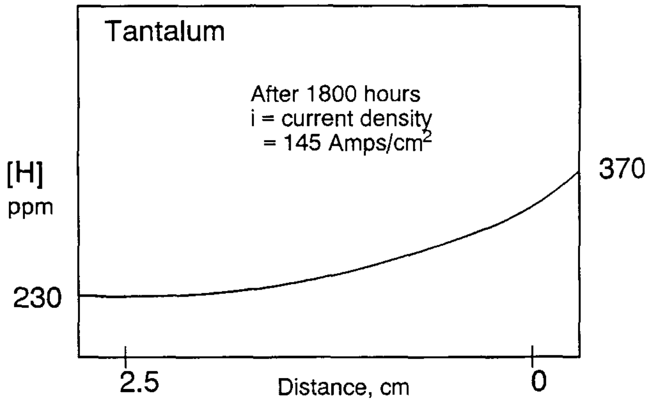

Fig. 4 Electrotransport of hydrogen in tantalum (Ref 17)

Westlake and Miller (Ref 16) described the terminal solubility of hydrogen as a function of temperature. Table 2 shows the metal and alloy systems and the directionality of electrotransport for various solutes.

Mishra and Sivertsen (Ref 17) measured the effective valence of hydrogen and deuterium in tantalum. It was concluded that the effective valence, Z^*, is concentration dependent at low solute concentrations, i.e. <6 at.% H and <10 at.% D. Also, a reversal in the direction of electrotransport was observed for <1.38 at.% H, i.e. H moved from the cathode to the anode. However, the evidence of change in electrotransport direction for deuterium transport is not conclusive. This observation is pertinent to this paper because a reversal phenomenon is not desired when hydrogen is being removed from the weldments. Thus, the critical concentration for electrotransport direction reversal, if any, for hydrogen in steel must be known. The effective valence was found to be independent of the microstructure (Ref 17). Based on the results of Mishra and Sivertsen (Ref 17), Fig. 4 shows that a current density of 145 amps/cm^2 at room temperature can lower the concentration of hydrogen from 300 ppm to 230 ppm in 1800 h over a distance of 1 in. (25 mm). Other researchers (Ref 18-21) also found strong concentration dependence of hydrogen and its isotope in tantalum. Vanadium and niobium (Ref 22) show a weak dependence of Z^* on the solute concentration.

Oriani and Gonzalez (Ref 23) showed an isotope effect and a positive effective valence in favor of deuterium over hydrogen in α-iron and nickel. Einzinger and Huntington (Ref 24) showed a motion of hydrogen towards the anode in silver. Silver has a positive Hall coefficient. Effective valence for hydrogen in molybdenum as a function of temperature was determined by Pietrzak and Rozenfeld (Ref 25). Various other solid and liquid alloy systems were investigated showing the strong possibility of applying electromigration to prevent hydrogen damage in high-strength steel weldments. The phenomenon of electrotransport and its effect on the reliability of IC interconnects was discussed by Thompson and Lloyd (Ref 26) because these circuits carry enormously high current densities causing electrotransport of interstitials in the metal used for circuit manufacturing.

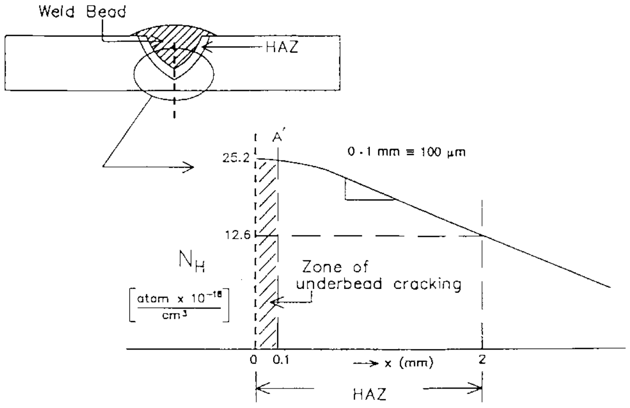

Fig. 5 Hydrogen concentration profile as a function of distance from the weld bead

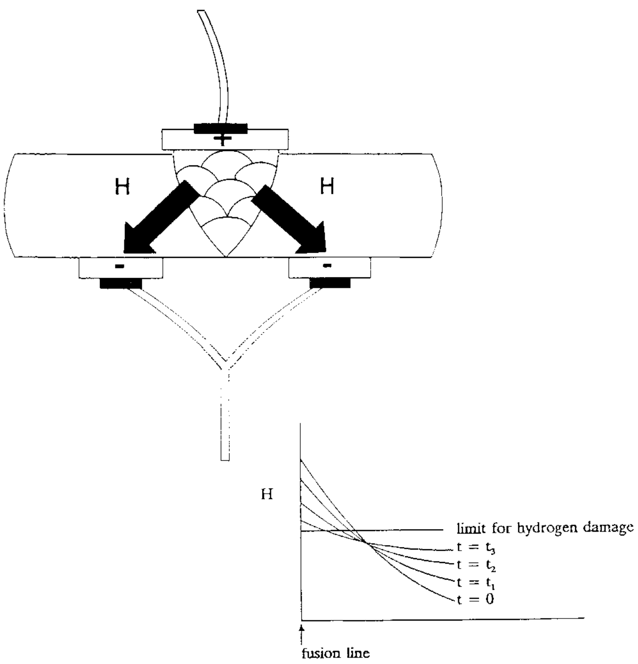

Fig. 6 Schematic diagram of an experimental set-up proposed for the electrotransport of hydrogen in steel to alleviate hydrogen cracking susceptibility

4. Theoretical Analysis

4.1 *Application*

The effectiveness of the electrotransport treatment to alleviate hydrogen will depend on three major factors: the temperature, current density, and time of application. Using a temperature close to room temperature eliminates heating up a large area of heavy or thick sections of steel. Thus electrotransport treatment requires large circuit densities to reduce the necessary application time.

Electrotransport treatment does not require excessively long time because the effort is to reduce a very localized concentration of hydrogen in the heat affected zone along and under the weld fusion line. The necessary transport distance is expected to be <100 μm (Fig. 5).

The application of electrotransport to reduce local hydrogen concentrations proceeds as follows:

1. Welding clamps are attached to the weldment to allow for the passage of high direct-current density across the heat affected zone.
2. Polarity is an important consideration with the negative electrode being positioned in the base metal beyond the HAZ and the positive electrode positioned on the weld deposit (Fig. 6).
3. Special care must be taken to make proper electrode connection so that there is no hydrogen charging at the cathode.
4. Application will occur for a specific length of time.
5. Degaussing may be necessary following the application of the high direct-current density.

Figure 6 is a schematic diagram of the set-up required to conduct the electrotransport of hydrogen from the underbead area into the steel.

4.2 *Calculations*

Examples of the time of application for a specific current density are given in the following two cases.

The rate of hydrogen electrotransport was calculated for moving hydrogen on the basis of a practical scenario presented in Fig. 5. A concentration profile is shown in Fig. 5. A peak hydrogen concentration of 5 ppm (25.2×10^{18} at./cm^3) was assumed at the underbead position, which is known to cause cracking (Ref 27) and which drops to half its value (2.5 ppm) at the HAZ-base metal interface, i.e. ~0.2 cm distance away, providing the initial steepest gradient (63×10^{18} at./cm^4) for hydrogen diffusional transport. An average slope of 31.5×10^{18} at./cm^4 (half of the steepest gradient) was assumed as the gradient under which hydrogen will diffuse, i.e. $\delta N_H/\delta x$ in Eq 10. Also, an average concentration of hydrogen of 24.9×10^{18} at./cm^3 over a distance of 10^{-2} cm away from the underbead, i.e. N_H in Eq 10, was used for these electrotransport calculations with the assumptions of a directed electric field and a linear velocity.

The two fluxes in Eq 10, which are additive in this case, the electrotransport flux, and the diffusional transport flux were separately calculated and are shown in Table 3. A volume element of 0.01 cm^3 (1 cm^2 CS × 100 μm thick one austenite grain size) was used to determine the quantity of hydrogen that has to be removed. The determination of time for the removal of hydrogen to make the weldment resistant to hydrogen damage was made using these fluxes.

The rate was estimated at two temperatures, 298 and 373 K. Use of higher temperature allows a faster rate of hydrogen removal due to an enhanced coefficient of diffusion. The diffusion coefficient of hydrogen in steel is based on expressions developed by Terasaki et al. (Ref 28). The higher temperature electrotransport can be facilitated just after the welding step to take advantage of the heat.

The diffusional and electrotransport fluxes, J_D and J_E, respectively, were calculated using the following values of the required parameters (Eq 8 and 10) and are shown in Table 3:

Boltzmann constant, $k = 1.38 \times 10^{-23}$ J/at.–K	
Fundamental charge, $e = 1.602 \times 10^{-19}$ coulomb	
Calorie = 4.2 J	
Distance of A' from anode: 100 μm	
Calculation of electric field, E in V/cm:	
Cross-sectional area, A	1.0 cm^2
Interpolar distance	4 cm
Applied current	10 000 amps
Steel resistivity	10^{-5} Ω-cm
Steel resistance	4×10^{-5} Ω
Applied voltage	0.40 V
Electric field	0.10 V/cm

Number of hydrogen atoms, $\#_H$, in a steel volume element of 10^{-2} cm^3 (1 cm^2 CS × 100 μm thick) is given by:

$$\#_H = N_H \times N_o \times V_{st} \times \rho_{st}/At._{st} \quad \text{(Eq 11)}$$

where N_o is the Avogadro number and V_{st}, ρ_{st}, and $At._{st}$ are the volume, density, and atomic weight of steel. Substitution of appropriate values for the parameters in Eq 11 yields the number of hydrogen atoms; $\#_H = 24.9 \times 10^{16}$ atoms.

The I^2R heat generated at these temperatures is found to be 0.95 kcal/s. A room temperature treatment is suggested because the net temperature increase of the workpiece (heat generated by resistance heating at 0.95 kcal/s minus heat dissipated by the workpiece) will help reduce the treatment time, yet will not heat the piece excessively to alter its microstructure. The calculated U_H/D_H values at the temperatures being considered for hydrogen removal, i.e. 25 to 100 °C, are in the range of 10.5 to 11.0, which is suitable for the removal of interstitials (Ref 3).

Table 3 Calculation of electrotransport and diffusion time

	At 298 K	At 373 K
Effective valence, Z^*	0.27	0.35
Diffusion coefficient, D_H, cm^2/s	1.3×10^{-6}	8.6×10^{-6}
Mobility, $U_H = D_H \cdot e \cdot Z^* / kT$, cm^2/s · V	1.37×10^{-5}	9.38×10^{-5}
Electrotransport velocity, $v_H = U_H \times E$, cm/s	1.37×10^{-6}	9.38×10^{-6}
Electrotransport flux, $J_E = N_H \cdot v_H$, atoms/cm^2 · s	3.41×10^{13}	2.34×10^{14}
Diffusional flux, $J_D = D_H \cdot (\delta N_H/\delta x)$, atoms/cm^2 · s	4.10×10^{13}	2.71×10^{14}
Time of electrotransport, $t_E = \#_H/A \cdot J_E$, s	7300	1065
Time of diffusional transport, $t_D = \#_H/A \cdot J_D$, s	6075	920

The estimated times for transport of hydrogen (Table 4) assume a complete removal either by electrotransport or diffusion independently. However, one does not need to remove it completely to make the structure safe and resistant to hydrogen damage; therefore, the time could be reduced further in a practical situation. Furthermore, a certain fraction of the hydrogen will be trapped. Thus only the free hydrogen will have to be moved suggesting further lowering in time for electrotransport. The estimated times are, therefore, conservative estimates and predict an upper limit. The actual behavior needs only a fraction of this time, which can be experimentally measured.

This paper analyzes and describes the electrotransport technique for lowering the concentration of hydrogen from underbead region in a weld. The practice will not require any special instrumentation because coupling the needed numbers of standard welding power supply allows for capability of delivering ten kiloamperes. The electrodes can be hooked to the workpiece, as depicted in Fig. 6, at a desired interpolar distance, and the current can be passed for a predetermined length of time. Such a system can be organized to work on-line. This novel application of the electrotransport technology can alleviate hydrogen related problems associated with high-strength steel structurals. Experimental verification needs to consider the primary concerns in electrotransport of hydrogen, which are (a) the competitive interaction of hydrogen with other solute atoms, (b) hydrogen transfer out of cathode surface, and (c) the hydrogen electrotransport in wrought compared with cast material.

5. Conclusion

This analytical work shows that electrotransport is a viable technique for the removal of diffusible hydrogen from the high-strength steel weldments to enhance the resistance of steel to hydrogen damage. The technique can be applied at room temperature to transport hydrogen. An underbead crack susceptible zone, containing 5 ppm of hydrogen, can be made resistant to hydrogen damage by adopting the method of electrotransport using an electric field of 0.10 V/cm (current density of 10 000 amp/cm^2). A treatment time of 30 min, under these conditions, is recommended to lower the hydrogen content of the susceptible zone by 50%.

Acknowledgment

The authors acknowledge and appreciate the support of U.S. Army Research Office and the Division of Materials Science, U.S. Department of Energy, under contract No. DE-AC05-840R21400 with Martin Marietta Energy Systems, Inc.

Table 4 Percent of hydrogen removed as a function of time

t, min	Electrotransported hydrogen, atoms	Diffusion transported hydrogen, atoms	Total removed hydrogen, atoms	Percent of total H
1	2.05×10^{15}	2.46×10^{15}	4.51×10^{15}	1.8
10	2.05×10^{16}	2.46×10^{16}	4.51×10^{16}	18
20	4.1×10^{16}	4.92×10^{16}	9.02×10^{16}	36
30	6.15×10^{16}	7.38×10^{16}	1.35×10^{17}	54

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