ELSEVIER

Journal of Nuclear Materials 307–311 (2002) 1498–1501

journal of nuclear materials

www.elsevier.com/locate/jnucmat

Hydrogen permeability over the joint weld of the steel parts of fusion reactor with magnet confinement of plasma

V.V. Fedorov [a], E.V. Dyomina [b,*], T.M. Zasadny [a], L.I. Ivanov [b], M.D. Prusakova [b], N.A. Vinogradova [b], A.M. Zabelin [c]

[a] *Physico-Mechanical Institute, pr. Nauky, 5, 290601 Lvov, Ukraine*
[b] *Baikov Institute of Metallurgy and Materials Science, RAS, Leninsky pr. 49, 117334 Moscow, Russia*
[c] *Institute of Laser and Information Technologies, Svyatoozerskaya, 1, 140700 Shatura, Russia*

Abstract

Hydrogen and its isotopes diffusion and permeability over the laser joint weld of low-activation 10Cr9WVA ferritic steels have been studied. Welding of steel sheets were produced with the help of Russian gas laser TL-5M type ($l = 10.6$ mm, $P = 2.5$ kW) in He atmosphere with the rate of 66 mm/s. Hydrogen diffusion over the joint welds was detected by the conventional method of electrical resistance measurement. By this way, the kinetics of resistance changes during hydrogenation of specimens engraved from weld metal, neighboring zone of thermal effect as well as basic metal have been determined. Coefficients of hydrogen diffusion were measured in the temperature range from 773 to 1073 K. So, for 10Cr9WVA steel at 873 K it was established that the hydrogen diffusion coefficient in the weld metal is approximately 10 times higher than in the basic metal, and three times higher than that in the zone of thermal effect. Hydrogen permeability over the joint weld specimens was measured by the Dines–Barrer method on the volummetric setup. It was established that the hydrogen flux over the laser joint weld is significantly (up to two orders) more than that over the basic metal. Using the data on the hydrogen permeability and diffusion coefficient, the hydrogen solubility in the weld metal was estimated, which is several ten times higher than that in the basic metal of the steel investigated. As a result, it was concluded that welding the steel parts of the first wall of thermonuclear reactors with magnet confinement of plasma is undesirable due to possible tritium leaking into the environment. A possible way of decreasing the joint welds hydrogen permeability, including application of protective impermeable for hydrogen coatings, is considered.

---

1. Introduction

Austenitic stainless steels are used for manufacture of components of reactors and tubes for heat transfer in modern energy devices. Main difficulties of the application of such steels connected with radiation induced swelling, are activation under irradiation and high gas permeability, which result in hydrogen isotopes loss into environmental atmosphere. Most of the tritium loss is connected with weld joint of reactor steels [1]. In accordance with the demand that tritium loss must be not higher then 1 Ku per day we have calculated ecologically the safe value of the hydrogen permeability coefficient $P_{cr} = 2.4 \times 10^{-12}$ mol/(g s Pa$^{1/2}$). The comparison of the value with hydrogen permeability of reactor steels in the range of ITER working temperature (673–973 K) [2–4] shows that the hydrogen flux through these steels must be reduced to 6–200 times lower. Commonly it is achieved by alloying or surface modification using cover coatings and films. However, formerly the hydrogen permeability of welding joint (excluding room temperature range and electrolytic hydrogenation [5–7]) was not studied. Therefore, we have investigated the parameters of the hydrogen diffusion in the steels with weld joint and try to find the change of diffusivity and

---

* Corresponding author. Tel.: +7-095 135 8680 4540; fax: +7-095 135 4540 8811.

E-mail address: elenadyom@mail.ru (E.V. Dyomina).

0022-3115/02/$ - see front matter © 2002 Published by Elsevier Science B.V.
PII: S0022-3115(02)01289-8

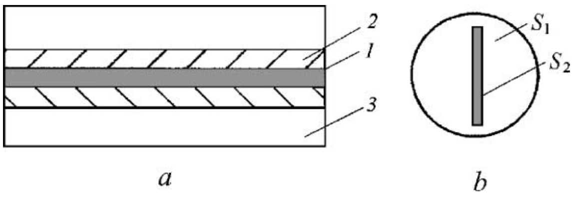

Fig. 1. The scheme of the selection of specimens for conductivity measurements (a) and hydrogen permeability (b). 1 – welding seam, 2 – ZTE, 3 – origin specimen, S_1 and S_2 – square of base specimen and welding seam, respectively.

solubility of hydrogen in the zone of thermal effects (ZTE) and in welding seams in comparison with bulk metal (Fig. 1(a)) in the temperature range 773–1073 K. Main found features are illustrated for example on low activation ferritic/martensitic steel 10Cr9WVA (Fe – base, C – 0.11%, Cr – 9.2%, W – 1.0%, V – 0.3%, Mn – 0.65%, Si – 0.3%, Ta – 0.1%, S – 0.007%, P – 0.02%). Low activation ferritic/martensitic. steels are prospective structural materials for making equipment for the thermonuclear reactor [8] and it is impotent to take into account peculiar interactions of these steels with hydrogen [9,10].

2. Experimental procedure

Diffusion parameters of hydrogen were determined by the so-called penetration technique on special volummetric device using the automatic McLeod manometer. Permeability P and diffusivity D of hydrogen were determined on the base of measurement of the rate of the change of hydrogen flow. Then using the correlation $L = P/D$ the solubility L of hydrogen was calculated. It should be noted that it is impossible to determine the hydrogen diffusivity in the welding seam directly by the penetration technique because we cannot separate the time of gas flow though the welding seam and the bulk metal (Fig. 1(b)). That is why we used the electroconductivity technique. We measured the kinetics of the change of the conductivity during the hydrogenation process at constant temperature (Fig. 2, curve 1) and calculated D on the base on two methods using the formulae [8]:

$$D = \frac{V_R(t_0)}{\Delta R(t_0)} \frac{S^2}{5.78},$$

$$D = \frac{l_1^2 l_2^2 l_3^2}{\pi(l_2^2 l_3^2 + l_1^2 l_3^2 + l_1^2 l_2^2)} tg\alpha.$$

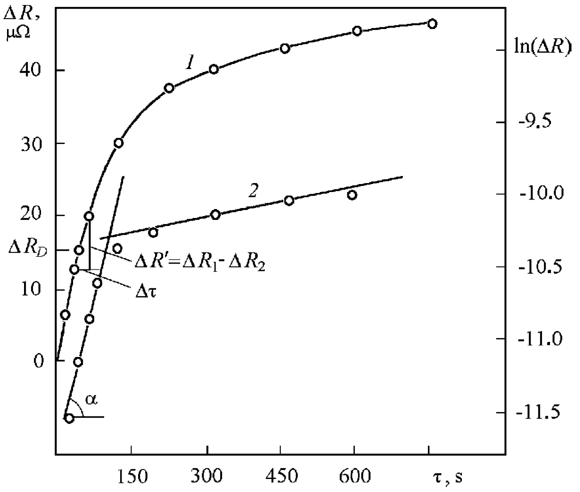

Fig. 2. Origin (1) and logarithmic (2) kinetic dependence of the change of conductivity of steel 10Cr9WVA during hydrogenation at $T = 873$ K. The rate of the change of the conductivity was determined by the formula $V_R = \Delta R'/\Delta\tau$.

Here V_R is the rate of conductivity change, ΔR is the absolute change of conductivity, S is the square of cross section of a specimen, l_1, l_2, l_3 are dimension sizes of a specimen, a is the inclination angle of the kinetic dependence of conductivity change in logarithmic coordinates (Fig. 2, curve 2).

Welding joints are prepared by laser weld in helium atmosphere using the TL-5M laser device in continuous regime with 2.5 kW power and 66 mm/s velocity of weld. Specimens for conductivity measurements were cut in accordance with scheme on the Fig. 1(a).

3. Results and discussion

Obtained values of D for the temperature range 673 – 1073 K are described by the following dependence:

Bulk metal:

$$D(\mathrm{m^2/s}) = 1.4 \times 10^{-8} \exp(-9.1\ \mathrm{kJ/mol}/RT).$$

ZTE:

$$D(\mathrm{m^2/s}) = 6.2 \times 10^{-8} \exp(-16.3\ \mathrm{kJ/mol}/RT).$$

Welding seam:

$$D(\mathrm{m^2/s}) = 1.2 \times 10^{-7} \exp(-27.1\ \mathrm{kJ/mol}/RT).$$

Absolute values of the diffusivity D for 873 K presented in Table 1 allow us to compare the influence of the structural state of the material upon the rate of hydrogen migration. One can see that the hydrogen diffusivity in ZTE is more than three times and in seam about one order of magnitude higher than in bulk metal.

For determination of the hydrogen solubility in welding seam the combined approach was applied. The approach is based on the determination of the hydrogen permeability of welding seam and calculations on the

Table 1
Values of hydrogen diffusivity (D) in 10Cr9WVA steel at 873 K

Specimen	Welding seam	ZTE	Bulk metal	Curve 1 (Fig. 2)	Curve 2 (Fig. 2)
D (m^2/s)	8.7×10^{-8}	3.4×10^{-8}	9.2×10^{-9}	1×10^{-8}	1.6×10^{-8}.

formula $L = P/D$, where D is the hydrogen diffusivity determined by the change of conductivity measurements.

At first, preliminary experiments were carried out and diffusion parameters of hydrogen in initial normalized specimens (0.5 h, 1320 K with cooling in air) with a structure similar to ZTE and normalized specimens after vacuum annealing 600 h at 873 K steel 10Cr9WVA were determined. It was found that the hydrogen permeability of initial (normalized) specimens (Fig. 3, curve 1) up to 1035 K corresponds to an exponential dependence:

$$P(\mathrm{mol}/(\mathrm{g\,s\,Pa}^{1/2})) = 1.6 \times 10^{-8} \times \exp(-51.6\ \mathrm{kJ}/(\mathrm{mol}/RT)).$$

The sharp drop of the hydrogen flux at this temperature and growth of the activation energy of penetration up to 60 kJ/mol indicate that a first-order phase transition take place with a crystal lattice type change (α–γ transition). The same changes are observed for the hydrogen solubility and diffusivity curves; and their equations before the phase transition are:

$$D(\mathrm{m}^2/\mathrm{s}) = 1.8 \times 10^{-8} \exp(-3.4\ \mathrm{kJ}/(\mathrm{mol}/RT)),$$
$$L(\mathrm{mol}/(\mathrm{m}^3\mathrm{Pa}^{1/2})) = 0.9 \exp(-9.1\ \mathrm{kJ}/\mathrm{mol}/RT).$$

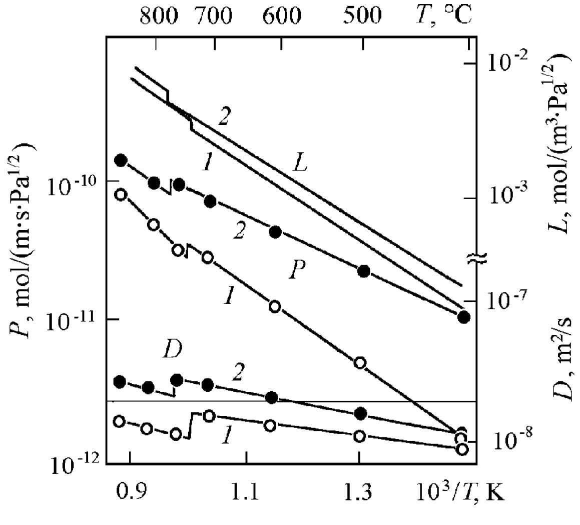

Fig. 3. Temperature dependence of P, D and L for the steel 10Cr9WVA with welding seam. 1 – specimen after normalizing (0.5 h, 1320 K), 2 – specimen with welding seam.

Annealing of specimens in vacuum results in an increase of both hydrogen flux P and critical temperature of phase transition from 1035 to 1055 K.

Direct measurements of hydrogen diffusion parameters in specimens with welding seam (Fig. 3) have shown that in this case both permeability and diffusivity of hydrogen increased. Values of D for bulk metal obtained at 873 K by the penetration technique (curve 1) and by conductivity measurements are very close one to another, but the value D for the specimen with welding seam (curve 2) is highly different from data in Table 1. This value is connected with the superposition of contributions into diffusion from hydrogen flux through both welding seam and bulk metal.

So as sizes of origin specimens and specimens with welding seam are equal so the change of hydrogen flux in the second case depends upon the relation of squares of bulk metal and the seam ratio S_1/S_2, which was of 16 in the experiment. Taking into account the hydrogen permeability of bulk metal at 873 K and an addition of hydrogen flux we have calculated the hydrogen permeability of welding seam P^*, which proved to be two order of magnitude higher than bulk metal one.

$$P = 1.12 \times 10^{-11}\ \mathrm{mol}/(\mathrm{g\,s\,Pa}^{1/2}),$$
$$P^* = 1.07 \times 10^{-9}\ \mathrm{mol}/(\mathrm{g\,s\,Pa}^{1/2}).$$

Dividing the obtained value P^* by hydrogen diffusivity D obtained by conductivity measurements (Table 1) we got the hydrogen solubility L^* in welding seam which is about 90 times more than one in bulk metal

$$L = 1.41 \times 10^{-3}\ \mathrm{mol}/(\mathrm{m}^3\,\mathrm{Pa}^{1/2}),$$
$$L^* = 0.123\ \mathrm{mol}/(\mathrm{m}^3\,\mathrm{Pa}^{1/2}).$$

It should be noted that the hydrogen permeability both of the bulk metal and especially of the welding seam is highly more than the ecology safe coefficient $P_{\mathrm{cr}} = 2.4 \times 10^{-12}\ \mathrm{mol}/(\mathrm{g\,s\,Pa}^{1/2})$, therefore, when using welding joints of steel 10Cr9WVA, an additional modification of the surface in the welding zone is quite necessary.

4. Conclusion

1. A new combined approach has been supposed to investigation of the solubility of hydrogen in welding joints based on the determination of the hydrogen permeability by the penetration technique, the dif-

fusivity by conductivity measurements and the calculation of the solubility in accordance with the relationship $L = P/D$.
2. For the first time the high-temperature solubility of hydrogen and the hydrogen permeability of welding seam have been determined. It has been shown that at 873 K these parameters are about to orders of magnitude higher than those of bulk metal.

References

[1] A.S. Nikiforov, Z.V. Ershova, N.K. Vinogradova, et al., Structure Materials for Fusion Reactors, Moscow, Nauka, p. 28.

[2] V.V. Fyodorov, E.V. Dyomina, V.I. Pokhmursky, et al., Fiz. -Khim. Obrabotki Materialov (Phys.-Chem. Mater. Treatment) 6 (1995) 88 (in Russian).

[3] V. Fedorov, V. Pokhmursky, E. Dyomina, et al., J. Fusion Technol. 28 (1995) 1153.

[4] E.V. Dyomina, V.V. Fedorov, L.I. Ivanov, et al., J. Fusion Eng. Des. 51&52 (2000) 93.

[5] V.D. Tarlinsky, I.G. Bleherova, T.K. Sergeeva, Autom. Weld. 11 (1980) 21 (in Russian).

[6] I.K. Pohodnja, A.P. Palzevich, Autom. Weld. 1 (1980) 37 (in Russian).

[7] V.V. Fyodorov, O.P. Sokolovsy, V.I. Pokhmursky, Using of the Conductivity Method for Definition of the Diffusion Coefficient in the Metals, Lvov, 1987, 34 pp. (preprint 121, Phys.-Mekh. Inst. AS Ukraine, in Russian).

[8] R.L. Klueh, in: Fusion materials, Semiannual Progress report for period ending, 31 December 1998, Oak Ridge National Laboratory, DOE/ER-0513/25, p. 111.

[9] E. Serra, G. Benamati, J. Mater. Sci. Technol. 14 (1998) 573.

[10] F. Schliefer, C. Liu, P. Jung, J. Nucl. Mater. 283–287 (2000) 540.