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HYDROGEN TRAPPING, DIFFUSION, AND RECOMBINATION IN AUSTENITIC STAINLESS STEELS

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Abstract

Trapping, diffusion, and recombination of hydrogen in austenitic stainless steels are reviewed. It is suggested that since all of these processes are strongly interdependent and since the measured recombination rates are found to vary four orders of magnitude at any temperature, the data analysis techniques used to date are insufficient. A two-region diffusion model with surface recombination is proposed in which the surface layer is characterized by a smaller diffusion coefficient than the bulk.

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1. Introduction

The use of the hydrogen isotopes deuterium and tritium as fuel for magnetically confined fusion reactors presents important areas of concern, including plasma fueling, recycling, hydrogen inventory, changes in structural integrity due to embrittlement, and loss of fuel by permeation through the vacuum vessel. All of these areas are strongly affected by surface recombination, diffusion, and trapping. These three effects are reviewed for austenitic stainless steels. Previous reviews [1,2] have addressed the more general area of hydrogen recycling. Gaps in the data base are noted.

The effects of hydrogen surface recombination, diffusion, and trapping are strong functions of temperature and are strongly interdependent. Experiments to measure one of these quantities independent of the other two are essentially nonexistent for a wide range of fluence, flux, concentration, and temperature. In addition, ion-beam-enhanced diffusion and detrapping can play significant roles in fusion reactors.

Present-day plasma confinement experiments are characterized by short discharge times, low wall temperatures, low wall fluxes, and small-area limiters (relative to the first wall area) that receive both high particle and high heat fluxes with a concomitant increase in surface temperature. In future confinement experiments and prototype fusion reactors, the discharge time will be extended, the wall temperature and the fluxes will be substantially increased, and limiters will essentially be eliminated. Advanced devices will probably have some type of impurity control mechanism, such as divertors or pumped limiters. These differences will

significantly change the effects of hydrogen recombination, diffusion and trapping. In existing experiments these effects are non-steady-state, and observed results are dependent on both the wall and the limiter; in future experiments steady-state conditions will be approached, and observed results will be dependent on both the wall and the impurity control system.

Figure 1 illustrates the various processes that can occur during and after the introduction of hydrogen in a solid. Thermal hydrogen can be adsorbed onto the surface with subsequent dissociation and diffusion into the bulk. Energetic hydrogen will be implanted to depths that increase monotonically with increasing energy. Along the particle trajectory, damage is produced and can serve as trapping sites for hydrogen. Other energetic radiation emanating from the plasma (e.g., electrons and neutrons) can produce traps. Implanted fuel and fusion-product particles and electrons will produce damage in the surface region, while neutron damage will be introduced relatively uniformly throughout the bulk. Hydrogen introduced into the first wall will tend to diffuse both into the bulk and to the surface. At the surface it can recombine to form H_2 molecules and desorb.

Stainless steels exposed to oxygen under mild conditions are known to form protective surface layers consisting mostly of Cr_2O_3 with thicknesses in the range of 30-160 Å [3]. These surface layers also contain small quantities of the oxides of iron, nickel, manganese and other alloying elements [3,4]. Surface finish, chemical treatment and operation in a plasma environment can significantly influence the oxide formed. Therefore, it is necessary to consider the solid as consisting of two components, a surface layer and bulk material, each with diffusion and trapping

effects that can differ considerably. Although much can be learned from experiments in the laboratory and in existing devices, much remains to be investigated before it will be possible to reliably predict hydrogen effects in future devices.

In section 2 a formalism for hydrogen retention and release is described. In the succeeding sections trapping, diffusion, and recombination of hydrogen in stainless steel are reviewed. Section 6 is a discussion of work to date, and conclusions are presented in section 7.

2. Formalism

Figure 1 illustrates the processes that can occur in laboratory experiments or in describing hydrogen retention and release in a plasma device or a fusion reactor. The following formalism describes mathematically the various processes and gives the diffusion equation with appropriate boundary conditions to describe hydrogen trapping and release in a material. It is taken in part from Refs. [5] and [6].

The solubility S of hydrogen in a metal exposed to hydrogen gas at pressure p is given by Sievert's law:

$$S = S_0 \sqrt{p} \exp(-E_s/kT) , \quad (1)$$

where S_0 is the pre-exponential factor and E_s is the heat of solution; for stainless steel $S_0 = 7.7 \times 10^{19} \text{ (H/cm}^3 \cdot \sqrt{\text{atm}} \text{)}$ and $E_s = 0.11 \text{ eV}$ [5]. Diffusion in a defect free lattice is characterized by a diffusivity D :

$$D = D_0 \exp(-E_D/kT), \quad (2)$$

where D_0 is the pre-exponential factor and E_D is the migration energy.

A two-region classical diffusion model that includes surface recombination and trapping is described by

$$\frac{\partial C_1}{\partial t} = - \frac{\partial J_1}{\partial x} + G_1 - \frac{\partial C_T}{\partial t}, \quad 0 \leq x < x_1, \quad (3)$$

$$\frac{\partial C_2}{\partial t} = - \frac{\partial J_2}{\partial x} + G_2 - \frac{\partial C_T}{\partial t}, \quad x_1 < x \leq x_2,$$

where C is the mobile hydrogen concentration, J the bulk hydrogen flux, G the hydrogen implant source term, C_T the trapped hydrogen concentration, and x the distance from the surface, with the subscripts 1 and 2 indicating near-surface and bulk. The flux is given by

$$J = -D \frac{\partial C}{\partial x} + \frac{CQ^*}{kT^2} \frac{\partial T}{\partial x} \quad (4)$$

where hydrogen diffuses from a concentration gradient (first term), and by a temperature gradient driving force known as the Soret effect [5]. Q^* is the heat of transport. For negative Q^* , diffusion is to the hotter region, as is the case for stainless steel.

The boundary conditions are as follows. At the surface ($x = 0$),

$$\phi_s = \sigma K_r C_1^2 = -D \frac{\partial C_1}{\partial x} + \frac{C_1 Q^*}{kT^2} \frac{\partial T}{\partial x} \quad (5)$$

(conservation of flux), where ϕ_s is the flux of hydrogen molecules leaving the surface, C_1 is the near-surface mobile hydrogen concentration, K_r is the phenomenological molecular recombination rate constant, and σ is the ratio of surface area to geometric area.

At the interface ($x = x_1$),

$$\frac{C_1}{S_1} = \frac{C_2}{S_2} \quad (6)$$

(chemical potential), and

$$D_1 \frac{\partial C_1}{\partial x} - \frac{C_1 Q^*}{kT^2} \frac{\partial T}{\partial x} = D_2 \frac{\partial C_2}{\partial x} - \frac{C_2 Q^*}{kT^2} \frac{\partial T}{\partial x} \quad (7)$$

(conservation of flux). Not included in these equations are the effects of ion-induced desorption [1] and deviations from Fickian diffusion [7,8].

These general equations have been used with appropriate initial conditions to describe experiments that measure permeation, diffusion, and recombination parameters and to model hydrogen retention and release effects in many devices.

Doyle [9] and Waelbroeck [2] have defined dimensionless transport parameters that can be used in a simplified manner to describe hydrogen transport in materials. In addition, computer codes have been developed to solve the set of coupled differential equations under particular initial and boundary conditions [10-12]. All of these approaches treat only one-region diffusion with surface recombination.

3. Trapping

Hydrogen trapping in stainless steel occurs in radiation damage sites (e.g., vacancies, interstitial loops, etc.), in voids, in highly strained regions around crack tips, in grain boundaries, and at impurities [1].

Trapping sites for hydrogen can be characterized by a trap concentration C_T and a trap binding energy E_B . The experimentally measured detrapping energy E_T is defined as the sum of the migration energy E_D and the trap

binding energy E_B see (fig. 2). The trapping term in Eq. (3) is given by

$$\frac{\partial C_T}{\partial t} = \frac{DC \cdot C_T^e}{\lambda^2} - C_T \nu_0 \exp - (E_b + E_D)/kT \quad (8)$$

where

$$C_T^e = C_T^0 - C_T, \quad (9)$$

and where C and C_T are the concentrations of mobile and trapped hydrogen; C_T^0 and C_T^e are the concentrations of traps and empty traps, respectively [5]. All concentrations are functions of position and time, and the temperature is a function of time. Here λ is the jump distance and ν_0 is the attempt frequency [5]. Measurements indicate that two dominant radiation damage traps exist with binding energies of ≈ 0.1 and 0.3 eV [13]. The effects of trapping have been observed at temperatures below ~ 450 K and for low concentrations; see Sections 4 and 5 for specific examples. These results indicate that for present-day machines trapping in the wall may be important under certain conditions but trapping effects in the limiter are probably unimportant.

A synergistic enhancement of trapping has been observed when stainless steel samples were predamaged by implanting helium [14-16]. An increase in the binding energy of ≈ 0.1 - 0.2 eV was observed. Myers [16] suggests that this increase in binding energy may be due to chemisorption of hydrogen on internal surfaces of helium bubbles. In future fusion reactors, helium created as a fusion product will be implanted into the surface of the first wall, and helium will be created in the bulk of the vacuum vessel by neutron-induced transmutation. Therefore, trapping of hydrogen may be more

important in reactors than in present-day devices although if the wall temperature is greater than 450 K even this type of trapping will have a small effect. Much more experimental work is needed before suitable predictions can be made.

4. Diffusion

Diffusion coefficients are determined experimentally by permeation techniques and by studying the approach to equilibrium from an initial non-equilibrium distribution [17]. In both cases the effects of surface recombination and trapping can significantly affect the results as well as the concentration of the diffusing species. For stainless steel all of these effects have been observed.

A comparison of experimentally measured diffusion coefficients for hydrogen and its isotopes in austenitic stainless steels is given in Table 1. For experiments in which more than one type of austenitic stainless steel was studied, the general conclusion has been that within experimental error all have the same hydrogen diffusion characteristics see, e.g., Refs. [18-22]. The data were chosen because they represent what appears to be reasonably well done experiments; that is, each experiment attempted to account in some manner for surface effects and trapping. In general, measured activation energies for diffusion decrease with increasing time. The data of Tison and Fidelle [22] span the largest temperature range and show what could be a change in the activation energy around 500 K (see Fig. 3). From the data given in Table 1, it appears that an activation energy of about 51 kJ/mole is appropriate in the temperature range from 298 to 500 K while at higher temperatures, 500 to 1200 K, an activation energy of about 59 kJ/mole is more appropriate. The

isotope effect was studied extensively by Quick and Johnson [21]. They confirmed the classical square root of mass scaling relations, $D(H)/D(D)/D(T) = 1/2^{-1/2}/3^{-1/2}$, and that the activation energy was independent of isotope mass. The data of Ref. [21] yield a lower activation energy than the other results. It is difficult to understand why these results are lower, since the experimental technique should have been more than adequate. Many errors occur in the reporting of these data, especially in the exponents [23]. The data presented in Table 1 are the corrected values of Ref. [21].

If trapping does affect the results, it will be more prevalent at lower temperatures. The effective diffusion coefficient will be lower than the intrinsic diffusion coefficient and is given by the relation

$$D_{\text{eff}} = D C_L / [C_L + C_X (1 - \theta_X)] , \quad (10)$$

where C_L is the lattice concentration of hydrogen, C_X is the concentration of hydrogen in trapped sites, and θ_X is the fraction of occupied trapping sites [24]. The high activation energy obtained by Austin and Elleman [19] could be a consequence of this effect. In all other experiments noted in Table 1, hydrogen was introduced by thermal diffusion, while in this experiment tritium was injected into the stainless steel by exposing a coating of ${}^6\text{Li}$ to a neutron flux. The reaction ${}^6\text{Li}(n, \alpha){}^3\text{H}$ produced injection of both alpha particles and tritons with concomitant implantation damage. If trapping by this damage affected the results, then a higher activation energy would be expected.

An experiment that demonstrates a concentration-dependent diffusion coefficient has been performed by Lewis and Farrel [25]. Figure 4 presents

their results for specimens of type 310 stainless steel that were cathodically hydrogenated. This procedure can produce a very high ratio of hydrogen atoms to metal atoms (e.g., as high as 1.0). Concomitant with the procedure to obtain this high concentration, intensive microstructural damage developed in the form of dislocations and cracks and probably influenced the results to some degree. The depth distributions of deuterium are shown for increasing egress times (i.e., time after completion of cathodic hydrogenation). The depth distributions were determined by nuclear reaction techniques and are shown hatched to give an idea of the uncertainty. Fits to the data (solid lines) were obtained using Fick's laws with different diffusion coefficients and assuming diffusion-controlled kinetics as opposed to recombination-controlled kinetics. This assumption appears to be acceptable because the surface concentration decreases rapidly with increasing time. The only free parameter in the fit is D , the diffusion coefficient. For relatively short egress times (i.e., high concentrations of deuterium), a value of $D = 1.4 \times 10^{-16} \text{ m}^2/\text{s}$ was obtained. At the lowest concentrations, trapping effects appear to dominate the diffusion.

Austin, Elleman, and Verghese investigated surface effects on the diffusion of tritium in stainless steel [6]. Their analysis assumed a surface layer diffusion coefficient different from the bulk diffusion coefficient, but they did not consider recombination. They experimentally determined that the surface layer diffusion coefficient was about 2×10^{-3} of the bulk diffusion coefficient in the temperature range from 298 to 475 K (see Fig. 5) and had approximately the same activation energy.

5. Molecular recombination

The molecular recombination process is a combination of hydrogen surface migration, recombination, and finally desorption. The phenomenological rate coefficient defined in Eq.(5) is the rate for the combined processes plus any other surface-related rate effect. For other species, namely carbon and oxygen, it has been argued that the surface migration step is the rate-limiting process and the other two steps are instantaneous by comparison [26]. Equation (5) implies a two-body process, but this is in fact not energetically possible. Estimates of three-body recombination processes involving two hydrogen atoms and a constituent of steel have been made yield to values of K_r some ten orders of magnitude larger than present measured results, leading one to suspect that surface migration or some other effect is the rate-limiting step [27].

Surface migration, recombination, and desorption should be dependent on only the first few monolayers and should be independent of the thickness of any oxide layer as long as its thickness is nonzero and the layer is continuous.

Figure 6 shows a comparison of laboratory measurements, plasma device measurements of σK_r for hydrogen on stainless steel, and theoretical calculations [15, 28-36]. The values span four orders of magnitude at any one temperature; the lowest values were measured for stainless steel that had been electropolished and had thick oxide layers, while the highest values were obtained with sputter-cleaned material and had essentially no oxide layer. In each of the measurements the data were analyzed using a one region diffusion model and surface recombination. For the data of

Waelbroeck [28], a pre-exponential factor and activation energy have been determined and are shown. These data span the largest temperature range of all experiments. The theoretical approach of Baskes [36], denoted by dashed lines in the figure, is given by

$$K_T = \frac{5.25 \times 10^{25} \times \alpha}{S_0^2 T^{1/2}} \exp(E_g - E_D/kT) \text{ (K}^{-1/2} \cdot \text{atm}^{-1} \cdot \text{cm}^{-2} \cdot \text{s}^{-1}\text{)}, \quad (11)$$

where all terms except α are defined in section 2; α is the molecular sticking coefficient. This approach leads to an activation energy of about 0.47 eV (47 kJ/mole). Recent experimental data have not been included in Fig. 6 but show the same effects and variances [37,38].

The most extensive measurements for both ion-sputtered and electropolished surfaces have been obtained by Myers (see Fig. 7) [16]. He derived recombination coefficients for these two conditions; they had very different activation energies E_R . For an ion-sputtered surface, $E_R = 0.34$ eV, and for an electropolished surface, $E_R = 0.81$ eV.

6. Discussion

Recombination measurements made to date have assumed that the recombining flux is proportional to C^2 , as presented in Eq. (5), with a phenomenological molecular recombination rate constant K_T . If diffusion in the surface layer is a factor, it has been implicitly included in Eq. (5). For many experimental arrangements either recombination limited or surface-diffusion-limited kinetics will fit the observed experimental results (see, e.g., Ref. [32]). Since the recombination results have been observed to vary over many orders of magnitude and to have large differences in activation energies it appears that most of the previous data should be analyzed

using the two-region diffusion model with recombination. The "sputter-cleaned," oxide-free results, which give the highest values, should represent the most accurate measurements of the recombination rate for clean stainless steel, while lower results probably include the effects of surface diffusion through the oxide and also a change in the recombination rate with changing surface composition. Waelbroeck has presented a possible mechanism for enhancing surface recombination on exposure to energetic ions [2]. Effects other than those suggested here may also be present.

From a theoretical perspective, Baskes [36] has determined the activation energy for recombination to be $(E_D - E_g)$, which for stainless steel is about 0.47 eV, while Myers [16] has measured an activation energy for a sputter-cleaned, oxide-free surface of 0.34 eV. For an electropolished surface, "thick oxide layer," Myers has measured an activation energy of 0.81 eV, while Austin [6] measured an activation energy for diffusion of 0.65 eV for a surface oxide layer. These comparisons, while not exact, are indicative that surface oxide layers can be a significant factor in analyzing reemission data. In addition, Chang [27] has observed an apparent increase in K_T as the thickness of the oxide layer is reduced.

7. Conclusions

A review of the literature suggests the following:

- Trapping characteristics of hydrogen in austenitic stainless steels are reasonably well understood, and trap concentrations and activation energies are known sufficiently accurately for use with most models.

- The diffusion parameters of hydrogen in undamaged bulk austenitic stainless steels, including the exponential factor and the activation energy, are rather well known. All types of austenitic stainless steels appear to behave in essentially the same way.
- The classical square root of mass scaling of diffusion for H, D, and T has been confirmed to sufficient accuracy for use in modelling fusion reactors.
- There is insufficient basic understanding of surface recombination and diffusion in surface oxide layers to permit accurate predictions of the reemission in future fusion reactors.
- More experimentation is needed to elucidate the effects of oxide surface layers on diffusion through surface layers and surface recombination.

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FIGURE CAPTIONS

FIG. 1. Illustration of the various processes involved in the interaction of hydrogen and solids.

FIG. 2. Schematic of the energy levels used in the formulation of diffusion. E_s is the heat of solution, E_D is the migration energy, E_B is the trap binding energy, and E_T is the detrapping energy.

FIG. 3. Arrhenius plots of the undamaged bulk diffusion coefficient for different types of stainless steels. γ SS represents austenitic stainless steel, types 304L and 316L; α SS represents martensitic stainless steel, type 430; and $\alpha + \gamma$ SS, type 325. Data are taken from Ref. [22]. Activation energy is in units of kJ/mole.

FIG. 4. Plot of deuterium concentration versus depth for various egress times. The hatched lines show the measured concentration and their widths represent the uncertainties. The solid lines represent results of the diffusion model with various assumed diffusion coefficients. The dashed line represents the initial distribution for an ingress time of 6000 s. The experiment was conducted at 298 K. Data are taken from Ref. [25].

FIG. 5. Arrhenius plots of the bulk and surface layer diffusion coefficients for tritium in type 304 stainless steel. Data are taken from Ref. [6]. Activation energy is in units of kJ/mole.

FIG. 6. Arrhenius plots of measured and theoretical values of the phenomenological recombination coefficient K_r . σ is the surface roughness factor. References are to Waelbroeck [28], Braun [29], Ali-Khan [30], Myers [15], Kerst [31], Wilson [32], Wienhold [33], Baskes [34], and Langley [35]. Theoretical estimates from Ref. [36] are shown as dashed lines. The factor α is the molecular sticking coefficient. The solid line is a fit to the data of Waelbroeck. Activation energy is in units of kJ/mole.

FIG. 7. Arrhenius plots of the measured phenomenological recombination coefficient, σK_r , for an ion-sputtered surface and an electropolished surface.

**COMPARISON OF MEASURED HYDROGEN DIFFUSION COEFFICIENTS
IN AUSTENITIC STAINLESS STEELS**

TYPE OF STEEL	ISOTOPE	TEMPERATURE RANGE (K)	E_D (kJ/mol)	D_0 ($10^{-6} \text{m}^2/\text{s}$)	$D(H, 500 \text{ K})$ ($10^{-12} \text{m}^2/\text{s}$)	METHOD OF MEASUREMENT ^a	REFERENCE
304	T	373-473	57	0.72	2.3	D	CHANEY & POWELL (1970)
304, 316	T	298-495	61	1.1	1.5	D	AUSTIN & ELLEMAN (1972)
304N, 304L 309S, 310 21-6-9, A216	D	400-714	54	0.34	1.3	P	LOUTHAN & DERRICK (1973)
310	H, D	472-779	49	0.52	4.0	P	QUICK & JOHNSON (1979)
304, 316	D, T	298-500	51.5	0.24	1.1	P	TISON & FIDELLE (1981)
		500-1173	59.5	1.7			

^aP = PERMATION; D = DEPTH PROFILE DETERMINATION. $D = D_0 e^{-E_D/RT}$; D_0 IS ADJUSTED TO REPRESENT VALUE FOR H.

SCHEMATIC OF HYDROGEN EFFECTS IN SOLIDS

ORNL-DWG 84-2504 FED

T (K)

1000

500

333