Diffusive Behaviors of Deuterium in Nickel under Irradiation of Helium-3

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Diffusion, permeation and trapping of deuterium in nickel exposed to a deuterium plasma were studied experimentally at temperatures of 455 to 568 K. One side of a nickel membrane was exposed to the plasma and the plasma-driven permeation was monitored. A beam of $^3$He irradiated the same side simultaneously to observe deuterium concentrations near the surface by means of the nuclear reaction analysis. The beam also produced traps and the observed concentration increased to be nearly saturated with the dose of $^3$He, while all deuteriums were dissolved at low doses. The trap concentrations were in the orders of $10^{-4}$ to $10^{-3}$ atom fraction. They tended to increase with decreasing temperatures. The traps were not bubbles but would be associated with radiation induced defects. The traps would cause a large amount of hydrogen isotopes retained in plasma-facing walls even at elevated temperatures.

KEYWORDS: nuclear reaction analysis, diffusion, deuterium, plasma-driven permeation, nickel, trapping, inventory, radiation damage, temperature dependence, plasma-facing wall, helium 3, beams, diffusion equation, hydrogen isotopes

I. INTRODUCTION

The diffusive behaviors of hydrogen isotopes in plasma-facing walls in fusion reactors are directly related to the leakage into coolant and the inventory in the walls. Especially, influences on these processes caused by hydrogen trapping by radiation induced defects are of great concern, since the walls are exposed to high energy particles such as 14 MeV neutrons and helium ions from D-T reactions.

In order to study on the hydrogen trapping in solids, a concentration of hydrogen must be known. The nuclear reaction analysis (NRA) with the reaction $D(^3$He, p)$^4$He is suitable to observed the deuterium concentration, and there have been many experimental works on deuterium trapping in metals using the NRA (1)–(6). In their works, the temperatures of samples were not high. Otherwise most deuterium, which was implanted preliminary, would be released from the sample. However, operating temperatures of the plasma-facing walls will be several hundred degrees centigrade(7), and experimental data on high temperatures are needed.

In the present work, deuterium concentrations in nickel being exposed to deuterium plasma were in-situ observed using the NRA. By charging deuterium to nickel continuously during the NRA, the diffusive behaviors of deuterium such as diffusion, permeation and trapping were observed at elevated temperatures. The beam of $^3$He was used for both observing deuterium and producing traps in nickel.

II. EQUATIONS

Equations which include the processes of diffusion, permeation, recombination, trapping and detrapping are derived here. A diffusion equation with a single trap was first given by McNabb & Foster(8). It is written in a one-dimensional form by

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where $C_s$ and $C_t$ are the hydrogen concentrations in solution sites and traps, respectively, and $D$ represents a diffusion coefficient.

The boundary condition on the upstream side is

$$F = k_u C_s^2(0, t) - D \frac{\partial C_s}{\partial x} |_{x=0}. \tag{2}$$

Here, $F$ is the penetrating flux of hydrogen from plasma into metals on the upstream side. The first term of the right side represents the release rate of hydrogen. It was assumed to be proportional to the square of the dissolved hydrogen concentration just beneath the surface $C_s(0, t)$. The proportionality constant was defined as the recombination coefficient $k_u$. Similarly, the boundary condition on the downstream side is

$$0 = k_d C_t^2(l, t) + D \frac{\partial C_t}{\partial x} |_{x=l}. \tag{3}$$

where $l$ is the thickness of the metal membrane and $k_d$ the recombination coefficient on the downstream side.

The term $\frac{\partial C_t}{\partial t}$ in Eq. (1) represents the time-dependent change of the concentration of trapped hydrogen. In the present work, one kind of trap associated with point defects was assumed. The trap of bubbles will be discussed in Sec. IV-3. Equations to describe the term have been given by Wilson & Baskes, Myers et al., and Doyle & Brice. Their equations are essentially the same but slightly different in details. So a new equation was derived by modifying their equations, which covered all of them as shown later.

The following assumptions were made for hydrogen diffusion and trapping:

- Hydrogen is located as atom in either a solution site or a trapping site. Both the sites are interstitial.
- A hydrogen atom diffuses by jumping from one site to the nearest neighbor sites. The jump length is the nearest neighbor distance $a$.
- When a hydrogen atom goes into or escapes from the trapping site by a jump, the jump length is equal to $a$.
- One trapping site can trap one hydrogen atom.
- The enthalpy of hydrogen in the trap is lower by $E_t$ than that in solution. The energy $E_t$ is defined as the trapping energy.
- Neither the trapping site nor the combined hydrogen-trap pair diffuses.
- The density $C_o$ of the trapping site is much lower than that of the solution site $C_a$.
- The concentration $C_s$ of hydrogen in solution is much lower than $C_a$.
- The trap density $C_o$ is independent of time.

The assumption of the trapping site being interstitial may be added some proofs. Myers et al. observed the lattice location of deuterium in iron using the ion channeling technique. At 200 K, deuterium in the trap associated with a monovacancy was displaced from the octahedral site by a small distance toward a nearest neighbor host site. Besenbacher et al. have shown by calculation that deuterium trapped to vacancies in nickel was displaced from the vacancy. Hence, the trapping site was considered to be the interstitial site rather than the lattice site.

In order to derive the equation with the above assumptions, a small volume at the depth $x$ is considered, in which the traps and hydrogen atoms distribute uniformly. The increase $C^+$ in the concentration of trapped hydrogen $C_t(x, t)$ in a short time $\Delta t$ is the product of the following four terms; a jumping rate $Y_o \Delta t$ for a dissolved hydrogen to neighbors in $\Delta t$, the probability $C_o(x)/[C_o(x)+C_a]$ that the neighbor is a trapping site, the probability $(C_o(x)-C_t(x, t))/C_o(x)$ that the trapping site is empty and the concentration $C_s(x, t)$ of dissolved hydrogen, where $Y$ is the number of the nearest neighbor sites, $\omega$ the jumping rate for a dissolved hydrogen in a unit time. Considering the above condition of $C_a \gg C_o$, $C^+$ is written by

$$C^+ = Y_o C_s(x, t) |C_o(x)-C_t(x, t)| \Delta t/C_a. \tag{4}$$

Similarly, the decrease $C^-$ in the concentration of trapped hydrogen is the product of the following four terms; a jumping rate
Yo’Dt for a trapped hydrogen to neighbors in Dt, the probability \( \frac{C_a}{C(x)+C_a} \) that the neighbor is a solution site, the probability \( \frac{C_a-C(x, t)}{C_a} \) that the solution site is empty, and the concentration \( C_t(x, t) \) of hydrogen in the traps, where \( \omega' \) is the jumping rate for a trapped hydrogen in a unit time. The above conditions of \( C_a \geq C_o \) and \( C_a \geq C_i \) give

\[
C^- = \frac{C_a}{C(x)+C_a}\omega' C_t(x, t) \Delta t. \tag{5}
\]

Using Eqs. (4) and (5), the time dependence of \( C_i \) is given by

\[
\frac{\partial C_i(x, t)}{\partial t} = Y[\omega C_s(x, t)\{C_d(x) - C_t(x, t)\} - \omega' C_a C_t(x, t)] / C_a. \tag{6}
\]

The density \( C_a \) of the solution site is expressed by

\[
C_a = hN, \tag{7}
\]

where \( N \) is the atomic density of the host metal and \( h \) the number of the solution sites per host atom. The diffusion coefficient \( D \) of hydrogen in dilute solution\(^{12}\) is expressed by using the jumping length (nearest neighbor distance) \( \alpha \);

\[
D = \alpha^2 Y\omega / 6. \tag{8}
\]

A parameter is introduced, which is defined by \( f = \omega' / \omega \). This is equal to \( \exp(\Delta G / kT) \), where \( k \) is the Boltzmann constant, \( T \) the absolute temperature of the metal and \( \Delta G \) the difference of the free energies between the trapped and dissolved hydrogens. The difference in the enthalpies between them is \(-E_t \) (\( E_t \): the trapping energy) under our assumptions. The difference in the entropies is written by \( \Delta S \). Consequently, \( f \) is given by the form

\[
f = \mu \exp(-E_t / kT), \tag{9}
\]

where \( \mu \) is \( \exp(\Delta S / k) \) and independent of temperature \( T \). The parameter \( \mu \) has the same meanings as used in Doyle’s equation\(^{10}\).

Combined with Eqs. (7)~(9), Eq. (6) is rewritten as

\[
\frac{\partial C_i(x, t)}{\partial t} = \frac{6D}{hNa^2} \left[ C_i(x, t)\{C_d(x) - C_t(x, t)\} \right.
\]

\[
- \left. hNfC_t(x, t) \right] / C_a. \tag{10}
\]

This is the general expression for the concentration change of trapped hydrogen with time. The term \( 6/hNa^2 \) in Eq. (10) depends on the structure of metals as listed in Table 1.

<table>
<thead>
<tr>
<th>Table 1 Values of 6/hNa^2 represented by lattice constant a</th>
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<td>Site</td>
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Equation (10) will be compared with the others equations. Doyle’s equation\(^{10}\) is expressed as

\[
\frac{\partial C_i(x, t)}{\partial t} = 4\pi R_T D \left[ C_i(x, t)\{C_d(x) - C_t(x, t)\} \right.
\]

\[
- \left. hNfC_t(x, t) \right], \tag{11}
\]

where \( R_T \) is the trap radius. This equation is the same as Eq. (10) when the value of \( R_T \) is taken as \( 3a/4\pi \) for fcc metals and \( a/\pi \) for bcc metals. Myers’ equation\(^{6}\) coincides with Doyle’s equation when the parameter \( \mu \) in Eq. (9) is unity. Wilson’s equation\(^{5}\) is written as

\[
\frac{\partial C_t(x, t)}{\partial t} = \frac{D}{Na^2} \left[ C_t(x, t)\{C_d(x) - C_t(x, t)\} \right.
\]

\[
- \left. 6\nu \alpha^3 \right] / D_{0}f \frac{\partial C_t(x, t)}{\partial t} / D_{0}f, \tag{12}
\]

where \( D = D_0 \exp(-Q_0 / kT) \).

Here, \( Q_0 \) is the migration energy, \( D_0 \) the pre-exponential and \( \nu \) the attempt frequency for detrapping. Assuming that \( D_0 = \nu_0 \alpha^2 / 6\mu \) by analogy with Eq. (7), Eq. (12) is the same as Eq. (10) in the case where hydrogen disolves in the tetrahedral site for bcc metals.

In the present work, hydrogen was assumed to solute in an octahedral site in nickel\(^{3}\). From Eq. (10) and Table 1, we obtain

\[
\frac{\partial C_i(x, t)}{\partial t} = 3aD \left[ C_i(x, t)\{C_d(x) - C_t(x, t)\} \right.
\]

\[
- \left. NfC_t(x, t) \right], \tag{13}
\]

where \( a \) is the lattice constant. This equation...
was used to analyze our experimental data.

The solutions of Eqs. (1) and (13) at the steady state are

\[ C_i(x) = C_{i\text{su}} + \frac{(C_{ss} - C_{i\text{su}})x}{l}, \]  
\[ C_i(x) = C_{i\text{d}}(x)C_i(x)/\{C_{i\text{d}}(x) + N_f\}, \]

where \( C_{i\text{su}} = C_i(0) \) and \( C_{i\text{d}} = C_i(l) \). The permeation rate \( J(t) \) is given by

\[ J(t) = -D(\partial C_i/\partial x)_{x=0}. \]

The boundary conditions of Eqs. (2) and (3) at the steady state are

\[ F = k_a C_{i\text{su}} + J_s, \]
\[ 0 = -k_a C_{i\text{d}} - J_s, \]

which give the values of \( C_{i\text{su}} \) and \( C_{i\text{d}} \). The parameter \( J_s \) is the permeation rate of hydrog

\[ J_s = DC_{i\text{su}}/l. \]

The permeation rate at the steady state is proportional to the dissolved hydrogen concentration just beneath the upstream surface. The concentration \( C_i(x) \) at the steady state does not depend on \( C_i(x) \) (see Eq. (14)). This means the permeation rate of hydrogen is independent of the trapped concentration \( C_i \). So the term of "steady state" is used in the case where the permeation rate becomes a saturated value. At the "steady state", however, the trapped concentration \( C_i(x) \) was not always constant in our experiment, since the trap density \( C_o(x) \) increased slowly with the dose of helium ions as shown later. Even in this case, the "steady state" was considered to be achieved because the change of \( C_o(x) \) with time was very slow.

In the case of the non-steady state, the coupled differential equations Eqs. (1) and (13) cannot be solved analytically. A numerical calculating code using the Gauss-Seidel method was developed. The code yields the hydrogen concentrations of \( C_i(x, t) \) and \( C_i(x, t) \), and the permeation rate.

\[ \text{III. RESULTS} \]

1. Experimental

A 0.1 mm thick nickel membrane mechanically polished was used for the sample. It separated two vacuum chambers, i.e. the upstream and the downstream chambers, which were evacuated by turbo molecular pumps. After the sample was heated up and kept at an intended temperature, deuterium gas was fed into a discharge tube in the upstream chamber and radio frequency (rf) was applied to it. The gas was weakly ionized to become a rf plasma and energetic deuterium particles such as \( \text{D}^+ \) and \( \text{D}^0 \) were injected into the sample. This caused a dramatic increase in deuterium permeation rate—which was known as the plasma—driven permeation (PDP).

The analyzing beam of 1.3 MeV \(^3\text{He} \) irra-

\[ \text{diated the sample at an angle of 45° to the surface normal on the same side as exposed to the plasma while the discharge was continuing. A typical beam current was 200 nA (7 \times 10^{16} \text{ m}^{-2} \cdot \text{s}^{-1}). Energy spectra of protons produced by the nuclear reaction D}^{(3}\text{He, p})^{4}\text{He were detected by a solid-state detector and deconvoluted to a deuterium depth-profile up to the depth of 1 \mu m. Both the permeation rate to the downstream side and the deuterium concentration near the upstream side were observed at the same time.} \]

The observed deuterium concentration on the surface was rather high as reported previously\(^{(12)} \). It extended to 0.2 \mu m depth because of a finite resolution of the detecting system. This high concentration would be attributed to the deuterium adsorption on the surface. As we did not check impurities on the surface, an oxide layer may increase the concentration. In this case, the release rate of deuterium was considered to be still proportional to the square of the bulk concentration just beneath the surface as shown by Wampler et al.\(^{(14)}^{(15)} \)

On the contrary, the deuterium uniformly distributed in the bulk. Since the probing depth of 1 \mu m was much smaller than the sample thickness of 0.1 mm no gradient of
the concentration was seen. As the concentration beyond 0.8 μm widely varied because of the small cross sections of the nuclear reaction, the average value over the depth of 0.2 to 0.8 μm was taken as the observed deuterium concentration \( C_m \) just beneath the upstream surface. This value represented the deuterium inventory near the upstream side and was expressed by

\[
C_m(t) = C_s(0, t) + C_i(0, t).
\]

2. Concentration at Low Doses

In this section, the diffusion coefficient and the recombination coefficient on the upstream side of deuterium in nickel will be obtained from measurements. A sample was held at 523 K and the rf plasma was discharged. After the permeation reached the steady state, the deuterium concentration \( C_m \) was observed by means of the NRA. The permeation rate was subsequently changed by increasing the pressure of the deuterium gas or the rf power, and \( C_m \) was observed again at the new steady state. This procedure was repeated several times. The total dose of \(^3\text{He} \) was \( 4 \times 10^{20} \) m\(^{-2} \). The permeation rate \( J_s \) was plotted against the observed \( C_m \) as shown in Fig. 1. This figure shows a linear relation between \( J_s \) and \( C_m \). It is indicated that the diffusion was the rate-limiting process for the deuterium permeation (see Eq. (20)), if \( C_m \) was equal or proportional to the dissolved \( C_{su} \) just beneath the upstream surface.

In the diffusion-limited regime, the diffusion coefficient could be directly obtained from Eq. (20), since both the values of \( J_s \) and \( C_{su} \) were observed and \( l \) was known. The permeation rate at the steady state and the deuterium concentration were observed between the temperature of 455 and 568 K. The discharge condition was kept constant. A non-irradiated sample was used for each measurement and the \(^3\text{He} \) dose was kept less than \( 1 \times 10^{20} \) m\(^{-2} \) to minimize effects of the traps. The diffusion coefficients, which were obtained from Eq. (20), are shown by open circles in Fig. 2.

![Fig. 1 Linear relation between observed deuterium concentrations just beneath upstream surface and permeation rates at low doses of \(^3\text{He}\).](image)

![Fig. 2 Diffusion coefficients of deuterium in nickel.](image)

Open circles represent experimental results in the present work.

The data in the figure are on a straight line in an Arrhenius diagram, which confirmed the diffusion-limited regime between above temperatures. By the data fitting, the diffusion coefficient \( D \) was expressed by \( 5.75 \times 10^{-7} \exp(-0.41 eV/kT) \) m\(^2\)s\(^{-1}\). This agrees well with those of other works\(^{(16)-(19)}\). It was concluded that there were no traps before the irradiation of \(^3\text{He} \) and \( C_{su} \approx C_m \) at this low dose condition.

Among the parameters in Eq. (17), \( J_s \) and \( C_{su} \) were observed. The recombination coefficient \( k_u \) on the upstream side was estimated when the penetrating flux \( F \) was known. It
was difficult to measure the value of $F$ since both of neutral particles and ions contributed to it. Then, $F$ was determined assuming that the values of $k_u$ came on a straight line in an Arrhenius diagram\(^{(14)(15)}\). As the result, $F$ was $4.3 \times 10^{19} \text{atoms/m}^2\cdot\text{s}$ and $k_u=3.1 \times 10^{-33} \exp(0.24 \frac{\text{eV}}{kT}) \text{m}^4\cdot\text{s}^{-1}$.

In order to confirm the values of $k_u$ and $D$, the time-dependent change of the permeation rate was observed at 523 K after the plasma was abruptly discharged and stopped. The beam of $^3\text{He}$ was not irradiated in this measurement. The evolution of the permeation rate is shown in Fig. 3. Calculated results are also shown in the figure, which was obtained by solving Eq. (1) under the boundary conditions of Eqs. (2) and (3) and substituting Eq. (21). Equation (13) was not used, since no traps would exist. The values of $k_u$ and $D$ were taken from the above results. The recombination coefficient $k_d$ on the downstream does not affect the permeation rate in the diffusion-limited regime, so a value of $3 \times 10^{-28} \text{m}^4\cdot\text{s}^{-1}$ was used to assure the regime. The calculated results in Fig. 3 agreed very well with the experimental curves.

![Fig. 3](image)  
**Fig. 3** Time-dependent changes of the permeation rates after the deuterium plasma was discharged and stopped (a solid line). The calculated results are also shown as open circles.

### 3. Concentration at High Doses

The beam of $^3\text{He}$ was continued to irradiate the sample at the steady-state permeation rate. The temperature and the condition of the plasma were kept constant. As the result, the $C_m$ increased with the dose, while no changes in the permeation rate were monitored. This indicated an increase of the trap density. The evolutions of the trapped deuterium at several temperatures were shown in Fig. 4. They were obtained from Eq. (21), since the dissolved concentrations were estimated from the permeation rates. At higher doses, the concentrations of the trapped deuterium were nearly saturated. This suggested that the disappearance and the production of the traps were in equilibrium.

The concentration $C_{tu}$ of dissolved deuterium was $0.8 \times 10^{23} \text{m}^{-3}$ at 523 K. The observed concentration $C_m$ at the dose of $2.5 \times 10^{21} \text{m}^{-2}$ was $6.8 \times 10^{23} \text{m}^{-3}$. The trapped concentration $C_{tu}$ was then $6.0 \times 10^{23} \text{m}^{-3}$. It was much larger than $C_{tu}$ at high doses. Traps would affect the inventory in the plasma-facing materials, which will be discussed in Sec. IV-2.
4. Trap Density

A sample was held at 523 K and irradiated by $^3$He up to $3 \times 10^{21}$ m$^{-2}$ under the steady state of permeation. The observed concentration became constant at this dose. The discharge and the feed of gas were subsequently stopped but the irradiation continued to obtain depth profiles. The observed concentration $C_m$ slowly decreased with time as shown in Fig. 5.

In the calculation, the trap was assumed to distribute uniformly up to the depth corresponding to the mean projected range of the incident ion. From Eq. (15), the trap density is expressed by

$$C_o = C_{tu}(C_{su} + Nf)/C_{su}.$$  \hspace{1cm} (22)

The values of $C_{su}$ and $C_{tu}$ were determined from the measurement at the steady state, and the values of $f$ could be obtained by substituting $C_o$ into Eq. (22). Hence, the values of $C_o$ was fitted so that the calculated result reproduced the experimental data in Fig. 5. The coupled differential equations Eqs. (1) and (13) were numerically solved with the boundary conditions of Eqs. (2) and (3) to yield $C_m$. The calculated result was also shown in Fig. 5. The trap density $C_o$ was found to be $8.3 \times 10^{25}$ m$^{-3}$ (9.0 x 10$^{-4}$ atom fraction).

Similar experiments were performed at different temperatures between 455 and 568 K. The calculations were also conducted. As the results, the time-dependent changes in the observed concentration were all reproduced by calculation with the fitting parameter $C_o$. It was found that $C_o$ tended to increase with decreasing temperature. For instance, values of $C_o$ were $4.5 \times 10^{24}$ m$^{-3}$ (4.9 x 10$^{-3}$ atom fraction) at 568 K and $1.2 \times 10^{26}$ m$^{-3}$ (1.3 x 10$^{-3}$ atom fraction) at 455 K, respectively. This was a reasonable result considering that the traps disappeared by some thermally activated processes such as the diffusion of defects.

The traps were assumed to distribute uniformly in the calculation. However, the average energy transferred to target atoms by incident ions is generally largest near the projected range of the ion. The concentration of radiation induced defects would be higher in the projected range of $^3$He than near the surface. Hence, the trap density $C_o$ obtained above is a mean value over the...
surface to the projected range. The actual trap density is considered to be lower near the surface and higher near the projected range.

IV. DISCUSSIONS

1. Permeation Rate

An influence of the traps on the permeation rate at the non-steady state was studied. After the measurement of the permeation rate shown in Fig. 3, the plasma was discharged and $^3$He irradiated the sample up to $3 \times 10^{21}$ m$^{-2}$. The steady-state permeation rate was not changed by the $^3$He irradiation. The discharge was subsequently stopped and the time-dependent change of the permeation rate was observed again. It is shown in a dashed line in Fig. 6. The permeation rate observed before the irradiation is also shown as a solid line in the figure. The two experimental curves agreed well with each other within experimental errors. The traps caused little effects on the permeation rate both at the steady state and the non-steady state. The reason is that the traps existed in a shallow region where the incident ion could reached and produce radiation damages.

2. Inventory

The inventory is defined by the sum of the hydrogen concentrations in solution and in traps. In a region just beneath the upstream side, it is the observed deuterium concentration $C_m$ in our experiment. A relation between the inventory $C_m$ and the concentration $C_{su}$ of dissolved deuterium at the steady state is discussed here.

In the experiment, a sample was held at 568 K and continued to be exposed to the deuterium plasma. The beam of $^3$He irradiated the sample simultaneously. After the observed concentration saturated at the dose of $5 \times 10^{21}$ m$^{-2}$, the permeation rates and the deuterium concentrations were observed repeatedly by the procedure described in Sec. III-2. Open circles in Fig. 7 show the plot of $C_m$, i.e. the inventory, against the dis-
solved concentration $C_{su}$ determined from the permeation rate. Both $C_m$ and $C_{su}$ are represented by the atom fraction to nickel.

Traps will distribute uniformly in the plasma-facing materials, which are irradiated by fast neutrons. They will cause a significant increase in the hydrogen inventory $C_m$. The relation between $C_m$ and $C_{su}$ at some values of $C_o$ were simulated using Eq. (23) and are shown in Fig. 7. The curves in the figure show that the ratio of $C_m$ to the dissolved hydrogen concentration $C_{su}$ becomes larger with decreasing $C_{su}$ and increasing the trap density $C_o$. The contribution of traps to the inventory becomes significant when $C_{su}$ is small and $C_o$ is large.

3. Traps

The trapping site was assumed to be associated with radiation induced defects in Chap. II. Hydrogens, however, are also trapped in bubbles, which will be discussed. We consider a case that deuteriums are trapped in bubbles as $D_2$ gas. In the diffusion-limited regime, Sievert’s law gives a linear relation between the square root of the deuterium-gas pressure $P$ in a bubble and the dissolved deuterium concentration $C_{su}$ around it. That is, $C_{tu}=K\sqrt{P}$, where $K$ is the solubility constant. The concentration of trapped deuterium $C_{tu}$ is proportional to $P$ when the gas is ideal. Consequently, $C_{tu}$ is proportional to $C_{su}^2$. Therefore the observed deuterium concentration $C_m$, which is the sum of $C_{su}$ and $C_{tu}$, is expected to deviate from $C_{su}$ with increasing $C_{su}$. On the contrary, the experimental result in Fig. 7 showed the inverse tendency. It was concluded that bubbles, if existed, did not act as traps in our experiment.

Some values of $f$ were already obtained in Sec. III-4. The plot of $f$ on an Arrhenius diagram gave the trapping energy of 0.14 eV. Besenbacher et al.(33)(34) observed three kinds of traps in nickel. They were monovacancy with the trapping energy of 0.24 eV, multiple vacancies with 0.43 eV and helium bubbles with 0.55 eV. Nurskov et al.(24) have derived trapping energies theoretically as 0.17 eV for the interstitial associated trap, 0.43 eV for the multiple vacancies and 0.57 eV for the helium bubble. Yasukawa et al.(25) observed interstitial loops in nickel which trapped deuterium...
at elevated temperatures. Under the irradiation of electrons, the loop density tended to be saturated with time, or electron dose, which was similar to the dose-dependence changes of the trapped deuterium concentration as shown in Fig. 4. From above discussions, the observed trap would be associated with defects such as vacancies and interstitials.

V. CONCLUSIONS

The plasma-driven permeation in the nickel membrane exposed to the deuterium plasma was monitored and, simultaneously, the deuterium concentration beneath the upstream surface was observed at elevated temperatures. One kind of trap associated with point defects was assumed to be produced by the irradiation of $^3$He ions, and a new model was derived to describe the diffusion and the trapping of hydrogens. The followings were concluded in the present work.

1. The dissolved deuterium concentration at the steady state could be determined from the permeation rate and the diffusion coefficient, since the permeation was limited by the diffusion process. The diffusion coefficient was expressed by $5.75 \times 10^{-7} \exp(-0.41 \, \text{eV} / kT) \, \text{m}^2 \cdot \text{s}^{-1}$.

2. At low doses of $^3$He, no traps were observed and all deuteriums were dissolved in the solution sites. At higher doses, the traps increased even at the elevated temperature and approached to the saturated value.

3. The saturated density was $9 \times 10^{-4}$ atom fraction at 523 K. It increased with decreasing temperature.

4. The trapping energy was estimated to be 0.14 eV. The trap was not associated with bubbles but would be with defects such as vacancies and interstitials.

5. The contribution of the trapped hydrogen to the inventory in the plasma-facing materials will be significant when the trap density is large and the concentration of the dissolved hydrogen is small.

REFERENCES