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Deuterium pumping experiment with superpermeable Nb membrane in JFT-2M tokamak

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Abstract

A new divertor pumping system with superpermeable membranes has been installed in the JFT-2M tokamak to investigate the applicability of membrane pumping to fusion devices. The membrane pumping is based on suprathreshold (>1 eV) hydrogen permeation through a niobium membrane. Deuterium pumping is observed with divertor plasmas in tokamak environment. The pumping flux through the membrane increases by NBI heating and additional gas puffing and it is found that the pumping flux is directly proportional to the pressure in the divertor chamber where the pumping system is placed. The maximum permeation flux density is 7.3×10^{19} D/m² s for a tokamak discharge with a dense and cold divertor plasma. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

There has been big progress in the last decade towards the demonstration of high performance plasmas in nuclear fusion research. This progress has often been coupled to improved wall materials and better conditioning techniques for impurity control and lower recycling. Wall coating techniques such as carbonization, boronization and beryllium evaporation not only reduce oxygen contamination and metal influx from the wall but also provide reduced hydrogen/deuterium recycling around the wall [1], which is responsible for preventing a confinement degradation. Due to finite wall capacities, however, these techniques are limited to transient pumping and may not be effective for long pulse operation in superconducting machines such as Tore Supra, LHD and ITER. The future direction to-

wards long-pulse or steady-state experiments requires the development of a new type of divertor pumping system.

In this context a ‘superpermeation’ of suprathreshold (>1 eV) hydrogen particles through metallic membranes [2] is of interest. As it was found for thermal hydrogen atoms and accelerated ions [3,4], the permeation probability may be not only very much larger than that for thermal hydrogen molecules but it may be close to unity and not depend on membrane temperature and thickness. The superpermeability originates from an activation barrier for the dissociative absorption of molecular hydrogen (and consequently also a barrier for the desorption of absorbed atoms through their recombination into molecules). In the case of transition metals such a barrier appears due to a non-metal impurity monolayer chemically passivating the metal surface [2,3,5]. Absorption of energetic hydrogen is not influenced by the barrier, but the barrier is effective in hindering absorbed atoms from the thermal re-emission through the recombination into molecules. As a result the most part of absorbed atoms reaches the downstream membrane side

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with a larger probability than that for re-emission back. The downstream surface also ‘reflects’ the absorbed atoms and they cross the membrane many times before desorption. The probabilities of desorption through the up- and downstream sides depend in this case only on the relative height of up- and downstream barriers. If the downstream barrier is lower than the upstream one (favorable membrane asymmetry), virtually the whole incoming flux may pass through the membrane with no dependence on its temperature and thickness. The Group Va metals (V, Nb and Ta) are the most promising for superpermeable membranes due to their lowest (among all metals) level of the bulk potential energy [9] combined with a high surface barrier. This strong reflection condition for absorbed atoms puts the limits on the incident flux density, membrane thickness and on the temperature range at which superpermeability is possible, and it allows us to achieve a high permeation flux density through the membrane.

The superpermeable membranes may be employed for hydrogen pumping in various places in fusion devices. For instance, metal membranes are proposed for pumping of D/T fuel and its separation from He ashes in ITER [5]. Another possible application is to install the membranes just into the divertor region for strong active pumping of suprathreshold hydrogen being present here in order to provide for the high-temperature divertor plasma operation [6]. In applying the membranes to fusion devices, especially in the divertor where the membranes directly face plasmas, there are several subjects which should be investigated: (a) limitation of permeation flux density, (b) reliability of membrane operation in the presence of chemically active gases, (c) sputtering of non-metal film at the inlet surface by suprathreshold hydrogen particles and (d) deposition of non-metal (e.g. B, C) and metal (e.g. Fe, Ti) impurities. Some of them have been already investigated experimentally. A permeation flux density of $3 \times 10^{21} \text{ m}^{-2} \text{ s}^{-1}$, which is comparable to that expected in fusion applications, has been already achieved in the membrane experiment [7,8] while $\sim 10^{23} \text{ m}^{-2} \text{ s}^{-1}$ is a theoretical limit [9]. A stable long-term operation of superpermeable membrane has been demonstrated in the presence of chemically active gases [9] including the interaction with the plasma containing various gaseous impurities [8,10]. Recently, an energy dependence of effect of bombardment by hydrogen particles on Nb membrane was investigated in the plasma where the energy of incident ions was controlled by membrane biasing [11]. A stable plasma driven superpermeation persisted up to the ion energy of 70 eV without special efforts to maintain the surface film. Investigations as to the effects of impurities originating from sputtering of plasma facing materials are in progress now. It is found that the stable superpermeation through Nb membrane may exist at a high flux ($10^{18} \text{ m}^{-2} \text{ s}^{-1}$) and high deposited

dose ($>10^{22} \text{ m}^{-2}$) of stainless steel components [12]. A high enough membrane temperature is found to be a key factor for both the maintenance of superpermeation regime under the metal impurity flux and in situ restoring of the membrane.

In addition to that, it is very important to apply a membrane pumping system to a real fusion device and to investigate the permeation flux density through the membrane which is located in the divertor region. Therefore we designed a prototype of a membrane pumping system to evacuate deuterium particles in the JFT-2M tokamak divertor and investigate the outcoming flux of deuterium atoms produced by the divertor plasma. In this paper, we describe the experimental arrangement including the setup of the membrane pumping system (Section 2). In Section 3, we present the results on first application of membrane pumping to a fusion device. Finally, we discuss the applicability of such membrane pumping systems to fusion devices.

2. Experimental arrangement

2.1. Membrane pumping system

A small pumping system with superpermeable membranes was installed into the divertor region of JFT-2M tokamak as shown in Fig. 1. The JFT-2M is a medium size tokamak (major radius $R = 1.31 \text{ m}$, minor radius $a < 0.35 \text{ m}$, elongation $\kappa < 1.7$, magnetic field $B_t < 2.2 \text{ T}$). Details of this machine were given elsewhere in conjunction with a review of confinement and fueling studies [13]. Recently, the divertor configuration was modified from an open to a closed one in order to

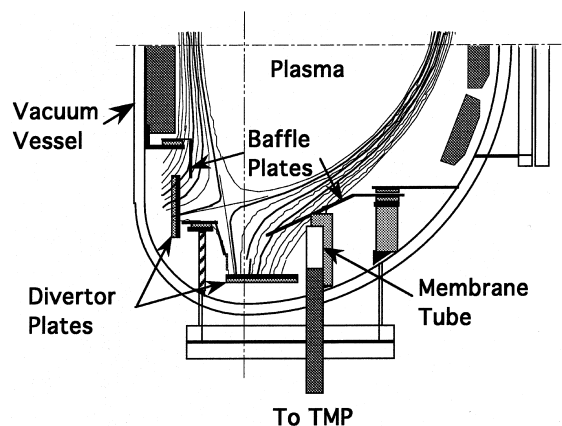


Fig. 1. Schematic view of the JFT-2M closed divertor showing the location of membrane tube. The magnetic field lines are indicated in the scrape-off layer (SOL).

improve particle control capability of the divertor and to demonstrate a possibility of dense and cold divertor with high confinement plasmas [14,15]. The scrape-off plasmas flow toward the divertor plate along magnetic field lines and interact with the target material (i.e. the divertor plate made of stainless steel) through various physical and chemical processes (sputtering, erosion, recycling, etc.). The recycling process increases the neutral hydrogen density in the divertor region and a large number of hydrogen atoms are produced by the divertor plasma through atomic and molecular processes. The incident flux of energetic hydrogen particles to the membranes, which are located at the position of 15 cm from the center of divertor plasma, is provided by these hydrogen atoms (mainly Frank–Condon atoms).

The structure of the membrane pumping module is shown in Fig. 2. This module has two Nb tubular membranes with the diameter of 1.5 cm, the length of 9.4 cm and the thickness of 0.02 cm. The two membrane tubes were arranged in the toroidal direction with the distance of 4.2 cm. The inner surface of the membrane was processed by the bombardment of small metal globes like sand to enlarge the effective surface area for release of hydrogen atoms as molecules through recombination. The basic membrane performance will be described later in Section 3.1. The membrane tubes were

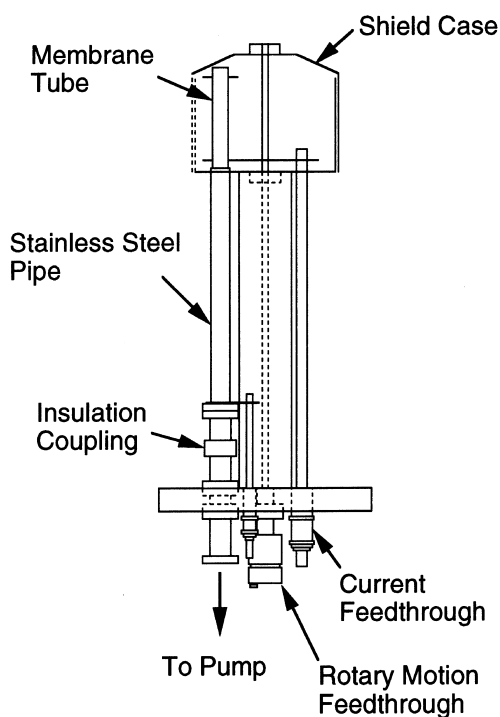


Fig. 2. Schematic view of a prototype membrane pumping system.

connected to a pumping system (turbo-molecular pump with the pumping speed of 500 l/s) through stainless steel pipes and the hydrogen particles permeated through the membranes were evacuated. The tubular membranes were electrically isolated with a ceramic from the plasma chamber and resistively heated by an alternating current through the membranes. The membrane temperature could be raised up to 1200°C with an ohmic current of 250 A. In order to reduce the partial pressure of gas impurities in the downstream volume, the insulated membrane tube components including the connecting stainless steel pipes were baked up to 300°C by ohmic heating with the current through the components. The vacuum chamber on the downstream side was also bakable up to 200°C. As a result the total pressure of impurities was in the order of 10^{-6} Pa in the downstream volume. To make clear that the incident hydrogen flux to the membrane originates from the divertor plasma, the membrane tubes are surrounded with a double cylindrical shield box whose outer cylinder is able to rotate by a rotary motion feedthrough.

The permeation flux through the membrane was measured by using a diaphragm with the known conductance (2.6 l/s for D_2). The pressures on both sides of the diaphragm were measured with ion and Pirani gauges. The pumping down of the volume of membrane tubes was performed through a bypass pumping duct.

2.2. Experimental procedure

After baking the ultra-high vacuum components of the membrane pumping system together with heating of the tokamak vessel, Taylor-type discharge cleaning was carried out in order to remove impurities on the surface of plasma facing components. During the discharge, the shield box was closed to prevent the membranes from facing the hydrogen plasma at low temperature since the membrane absorbs a large amount of hydrogen and hydride phase may be formed at the membrane temperature $T_M < 200^\circ\text{C}$. When the membrane system was not operated, the membranes were also shielded with a stainless steel case and absorbed hydrogen particles were released by heating the membranes at intervals.

In membrane experiments, prior to tokamak discharges, the membranes were heated by ohmic current (150 A) up to high temperature ($\sim 870^\circ\text{C}$) and the membrane temperature was sustained until the rising of the toroidal magnetic field. The circuits of the membrane as well as the thermocouples was cut off during tokamak operation on account of the Lorenz force due to the interaction between the membrane current and the magnetic field. In this phase the membrane temperature decreased gradually and it was about 650°C in the course of plasma discharge according to our calculation. Within the interval of tokamak operation (7 min), the

membranes were heated up again with the same current to release the hydrogen atoms absorbed during the discharge. This time sequence was repeated in the course of the membrane experiment.

The tokamak machine was mainly operated in an ohmically heated discharge with a lower-single-null (LSN) divertor configuration. The time history of typical tokamak discharge is shown in Fig. 3. The tokamak plasma was initiated with a fueling gas of D_2 and then gradually proceeded to the divertor configuration. When the divertor configuration is established at around 0.4 s, the plasma flows and reaches the divertor plate, and the pressure in the lower divertor chamber (P_{div}) increases due to the localization of recycling. The divertor configuration was maintained almost unchanged for 0.5 s in the second half period as indicated in Fig. 3. The plasma current was $I_p = 200$ kA with a toroidal magnetic field of $B_t = 1.3$ T. The line averaged electron density of the main plasma was around $\bar{n}_e = 2 \times 10^{19} \text{ m}^{-3}$. For divertor plasma, the density was around $5 \times 10^{18} \text{ m}^{-3}$ and the temperature 10–20 eV. In the course of the membrane experiment, there were several discharges additionally heated with neutral beam injection (NBI) with the power of 320 kW. An upper-single-null (USN) divertor configuration with open geometry, where there was no plasma in the closed divertor region, was attempted to compare the membrane permeation flux between two divertor configurations.

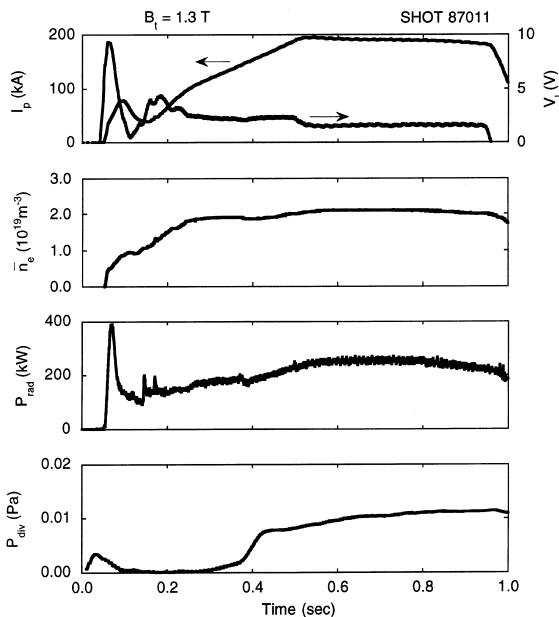


Fig. 3. Time traces of a typical tokamak discharge with a lower-single-null (LSN) divertor configuration. The divertor configuration is established at around $t = 0.4$ s and maintained up to the end of the discharge.

3. Experimental results

3.1. Superpermeation characteristics of membrane

The properties of membrane on superpermeability have been investigated using a Plasma Membrane Test Device (PMTD) [12], where a tubular membrane quite similar to that used in the tokamak experiment was surrounded with a hydrogen plasma (n_e and T_e were 10^{16} m^{-3} and a few eV, correspondingly, gas pressure $p = 10^{-2}$ Torr). The permeation flux was measured as described in Section 2.1. Fig. 4 shows the permeation flux driven by thermal molecules and plasma. The molecular driven permeation (MDP) rate exponentially decreases with decreasing membrane temperature as it is usually observed, while the plasma driven permeation (PDP) does not depend on the metal temperature and this temperature independence is one of most characteristic features of superpermeation (Section 1). In this way, it was confirmed that our tubular membrane was actually superpermeable to the suprathreshold particles (mainly thermal atoms in the given experiment) as that was repeatedly observed earlier with Nb membranes in the beam [16] and plasma [10,12] experiments.

We also investigated a membrane asymmetry (i.e. the ratio of desorption rate constants at both sides) using one more characteristic feature of superpermeation regime: the uniform distribution of absorbed hydrogen

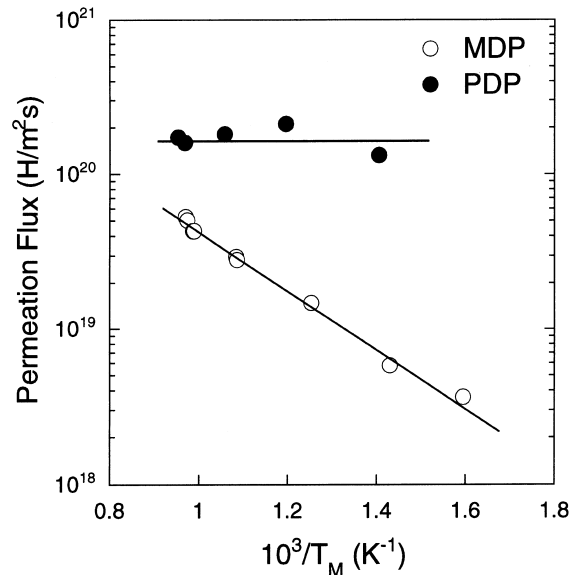


Fig. 4. Hydrogen permeation through Nb membrane in a Plasma Membrane Test Device (PMTD). Permeation to H_2 molecules (molecular driven permeation: MDP) decreases exponentially with decreasing membrane temperature, T_M , while the plasma driven permeation (PDP) is independent of the temperature.

over the membrane thickness [2–4]. In this experiment, after a significant amount of hydrogen was absorbed in the metal from the plasma, we decreased the membrane temperature to confine the absorbed particles and pumped the hydrogen from the plasma chamber. Then the membrane was heated up again and the thermo-desorption fluxes from both membrane surfaces were measured. The desorption flux from the downstream side was twice that from the upstream one. Taking into account the equality of hydrogen concentration at the both membrane sides, we come to the membrane asymmetry of around 2.

3.2. Membrane pumping experiment

The deuterium pumping experiment with superpermeable membranes was started with heating the membranes. Thereafter the tokamak machine was operated with the LSN divertor configuration. Fig. 5 shows the time history of the pressure at the downstream side in the course of the membrane pumping experiments. The decrease of pressure like a spike, which is observed in the intervals, corresponds to the decrease of membrane

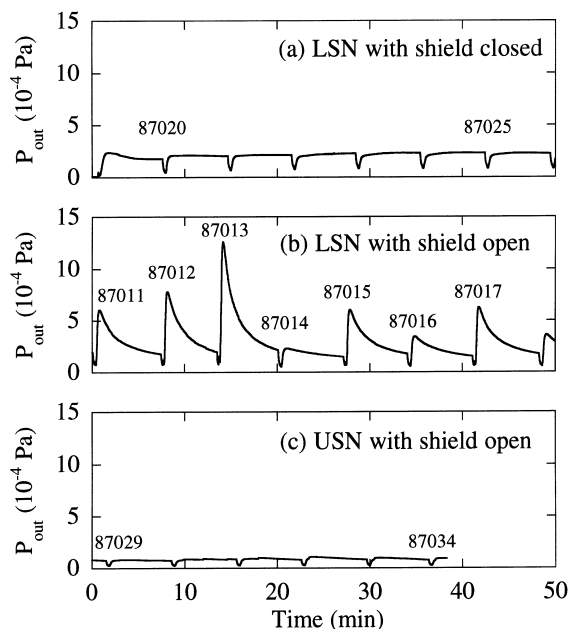


Fig. 5. Time history of the pressure at the downstream side of membrane in the course of membrane pumping experiment: (a) with membrane shielding in LSN; (b) without membrane shielding in LSN for the sequential shots of a density scan experiment (87011–87013), disruption occurred before (87014) and after (87015–87017) the establishment of divertor configuration; (c) without membrane shielding in USN. LSN and USN represent the lower- and upper-single-null divertor configurations, respectively. The pressure increase is observed only when the membrane faces plasma.

temperature caused by the suspension of the membrane heating during tokamak operation (as described in Section 2). The deuterium fueling gas was puffed just before the initiation of plasma discharge. The pressure in the plasma chamber (upstream pressure) was sustained at lower than 10^{-5} Pa during the interval between the plasma discharges, although the pressure increased just after the discharge. Observed evidences for membrane pumping are summarized as follows: The pressure rise at the downstream side is observed only when the shield is open and the membrane faces the divertor plasma (e.g. shots 87011–87013 in Fig. 5(b)). When closing the shield to prevent the membrane from facing the plasma, we observed no increase in the downstream pressure, that is, no permeation flux through the membrane as indicated in Fig. 5(a). On the contrary, when the membrane faced the divertor plasma, the downstream pressure increased on a large scale after reheating the membrane as seen in Fig. 5(b). This pressure increase is attributed to the permeation of deuterium atoms originated from the divertor plasma. In the phase of decreasing the membrane temperature during the tokamak operation, the incident deuterium atoms are absorbed and confined in the metal. With increasing membrane temperature, a major part of the absorbed particles are released at the downstream side. Another evidence for plasma driven permeation was revealed in the plasma disruption that occurred prior to establishing the divertor configuration (shot 87014 in Fig. 5(b)) and the USN divertor discharges (Fig. 5(c)). As expected, no pressure increase was observed in the downstream chamber because there was no plasma in the divertor region near the membrane module. Thus a deuterium pumping with superpermeable membrane has been demonstrated for the first time in fusion devices.

The particle permeation rate (pumping rate) Γ_p was estimated by integrating the gas flow through the constant conductance C_0 ($= 2.6$ l/s) with time:

$$\Gamma_p = \int P_{\text{out}} dt \frac{C_0}{\Delta\tau}, \quad (1)$$

where P_{out} is the pressure at the downstream side and $\Delta\tau$ the duration of plasma discharge in the divertor configuration. In our calculation, the effective discharge duration is supposed to be 0.5 s ($\Delta\tau = 0.5$) as seen in Fig. 3. The pumping rate was reduced by shortening of discharge duration due to plasma disruption (e.g. shots 87015–87017 in Fig. 5(b)). For full duration discharges, it was also dependent upon the type of plasma discharges (ohmically heated, with NBI, with additional gas puffing). The maximum pumping rate of 2.8×10^{17} D/s was obtained in the discharge with a strong gas puffing into the divertor chamber. Fig. 6 shows the dependence of the pumping rate on the divertor pressure measured by a Penning gauge. One can

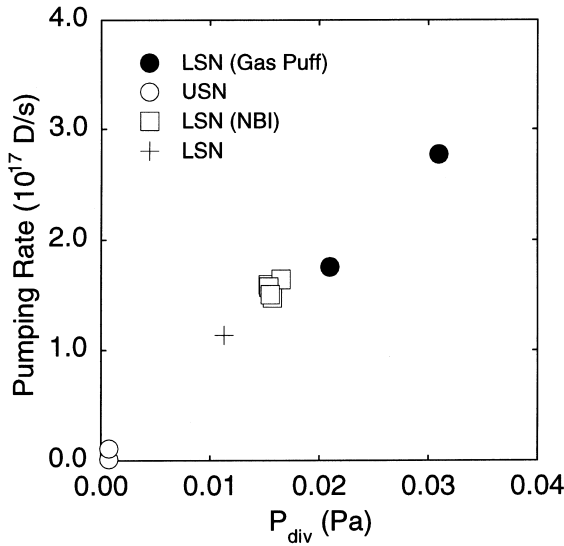


Fig. 6. Dependence of pumping rate on divertor pressure. The pumping rate is proportional to the divertor chamber pressure and depends on the magnetic configuration, additional heating (NBI) and gas puffing.

see that the pumping rate increases in proportion to the divertor pressure, which is closely related to the divertor particle flux. In the USN divertor discharges, the pressure in the lower divertor chamber is much less than that for the LSN divertor discharges because there is no plasma flow from the main plasma to the divertor plate. As mentioned before, we can observe no permeation flux in this configuration. For discharges with NBI heating, both main and divertor plasma densities increase by 20–30% and consequently the divertor pressure goes up due to the increase of divertor particle flux. Then one can expect that the increase of plasma density in the divertor region results in the increase of suprathermal atoms, which are responsible for the membrane permeation. Another important discharge with a strong gas puffing was carried out for the purpose of obtaining a dense and cold divertor plasma, which is effective for reducing the heat load of divertor plate during NBI heating. In these discharges, as reported in Ref. [15], the plasma density in the divertor region increases by two or three times, although the electron temperature decreases with increasing gas puffing. This divertor plasma serves as an efficient generator of suprathermal deuterium (mainly atoms) and the permeation flux increases proportionally to the divertor pressure. This proportional dependence is greatly advantageous in applying the membrane pumping to the closed divertor in which neutral particles may be compressed by orders of magnitude in future machines.

One should note that the pressure dependence of the permeation flux includes important information on the

recycling process in the divertor region. Since the membrane permeation flux corresponds to the atomic hydrogen flux, it is suggested that the atomic concentration in the divertor region is proportional to the gas pressure in the divertor chamber. The gas pressure increases with the influx of plasma particles to the divertor plate (divertor particle flux) due to the recombination of atoms on the walls of closed divertor chamber. In the discharge with additional gas puffing, though there is little change in the main plasma density, the divertor plasma represents a large change in the density and temperature. A major part of admittance gas contributes to the production of dense and cold divertor plasmas and consequently it results in high recycling plasmas localized in the divertor region.

4. Discussion

Superpermeable membranes were available for exhausting hydrogen particles in the divertor region. Here we discuss the incident particle flux from divertor plasma and the capability of membrane pumping in order to design a large membrane pumping system for fusion devices.

At first, we focus on atomic source in the divertor region. In order to discuss the permeation probability through the membrane, we must consider the reflection of incident deuterium atoms and the release rate of the absorbed ones at both sides of membrane. Assuming a plane source of thermal deuterium atoms with the density n_0 and the temperature T_0 in the divertor region, the permeation flux density Γ_D through the membrane can be estimated as

$$\Gamma_D = n_0 \left(\frac{kT_0}{2\pi m} \right)^{1/2} f_g (1 - f_R) f_s, \quad (2)$$

where m is the mass of deuterium atom. The geometrical factor f_g with respect to the acceptance solid angle for the plane source assumption is about 0.3. The reflection coefficient f_R may be supposed to be 0.4–0.6 for low energy particles ($E = 2$ –5 eV) from the recent data in the experiment where the deuterium reflection coefficient was measured with a compound ion beam (ArD^+) and an Nb target covered with the impurity oxygen [17]. The membrane asymmetry factor $f_s = A_s / (1 + A_s)$, A_s : membrane asymmetry) can be calculated to be 0.67 from the experimental result ($A_s \sim 2$) on the desorption rate at both sides of membrane as described in Section 3.1. From our measurements, the permeation flux density through the membrane can be estimated to be 7.3×10^{19} D/m² s for the discharge with a maximum divertor pressure of 0.03 Pa, assuming that the effective membrane area equals half of the surface area for each tubular membrane ($S_{\text{eff}} = 38$ cm²). Thus, for this

discharge, we can find that the atomic deuterium density in the divertor region is $0.9\text{--}2.1 \times 10^{17}$ D/m³. This atomic density is the same order of magnitude as that evaluated by computer simulations where a two dimensional fluid code coupled with a Monte Carlo method for neutral gas behavior was applied to the closed divertor configuration [18]. Consequently, it is found that our membrane is in the superpermeation regime and the outflux of deuterium atoms at the divertor plasma surface is estimated to be $5.6\text{--}8.4 \times 10^{20}$ D/m² s for the discharge with a strong gas puffing.

It is interesting to compare the neutral flux from the divertor plasma with those from main plasmas measured in several tokamaks. In TEXTOR, a permeation probe with a thin iron membrane was employed to measure the radial atomic hydrogen flux [19]. The typical hydrogen flux was 1.4×10^{19} D/m² s for an ohmically-heated limiter discharge with $\bar{n}_e = 2.5 \times 10^{19}$ m⁻³. The other measurements were carried out using a time-of-flight (TOF) method. Neutral particle fluxes onto the walls of ASDEX were obtained for both limiter and divertor discharges with the same density of $\bar{n}_e = 3.0 \times 10^{19}$ m⁻³ [20]. The neutral fluxes (7.1×10^{19} D/m² s) for the limiter discharge were much larger than those (1.9×10^{18} D/m² s) for the divertor one. This is related to the fact that the detector was very close to an intense localized gas source, namely, the limiter. Another information on the neutral flux was obtained in the PLT tokamak [21], where the density dependence of the flux and the toroidal distribution were investigated for ohmically-heated limiter discharges. The maximum flux of 4.5×10^{19} D/m² s was obtained with $\bar{n}_e = 1.0 \times 10^{19}$ m⁻³ and it was also found that the neutral particles were localized near the limiter. Here, one should note that the neutral flux measured in the closed divertor region is much larger than that in the main plasma region by orders of magnitude. This indicates that the closed divertor is essential in applying the membrane pumping to fusion devices.

In our experiment, it is suggested that the capability of membrane pumping depends on the divertor pressure. In the next generation tokamak as ITER, a major part of neutral particles are greatly compressed into the divertor region and one can expect that the divertor pressure increases much higher than 1 Pa. In this case, the membrane permeation flux density can be estimated to be more than $\sim 2 \times 10^{21}$ atoms/m² s from the linear pressure dependence of the permeation flux. This higher permeation flux density has been already achieved in our experiment [7] and the theoretical limit conditioned by a maximum permissible hydrogen concentration in bulk metal is expected to be $\sim 10^{23}$ atoms/m² s [9]. Therefore, if a large effective membrane area can be acquired in the divertor region, an efficient exhausting capability could be obtained by membrane pumping.

On the other hand, we have performed the membrane pumping experiment in the short-pulse machine by using

a transient pumping (i.e. absorbing deuterium atoms in the bulk of metal and desorbing at the downstream side after plasma discharges). However, in order to employ the membrane pumping system in long-pulse or superconducting machines, it would be required to develop a new membrane heating method in a steady magnetic field for steady state operation of membrane pumping. Technological development are under investigation, such as resistive heating with a high frequency alternating current and membrane heating through the eddy current due to a high frequency electromagnetic wave introduced into membrane tubes.

5. Conclusion

A prototype membrane pumping system with superpermeable Nb tubular membranes has been installed into divertor region in the JFT-2M tokamak and the pumping capability has been investigated. A deuterium pumping through the membrane has been demonstrated in tokamak environment for the first time. The pumping flux increases in proportion to the divertor pressure and the maximum permeation flux density amounts to 7.3×10^{19} D/m² s with the divertor pressure of 0.03 Pa. Application of membrane pumping to future large machines, where the divertor pressure may be greatly increased by the compression of neutral particles into the divertor, seems to be rather promising. Further investigation concerning metal membrane heating method would be required for steady state operation of membrane pumping system in long-pulse machines.

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