

Demonstration of the feasibility of the D/T-mixture selective pumping in fusion reactors using superpermeable membranes

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The niobium superpermeable membrane (SPM) in the shape of thin-walled tube with an area of 600 m^2 was coupled with incandescent tantalum ribbons purposed for thermal dissociation of H_2 molecules. The SPM had resistive heating, which made it possible to carry out its high-temperature treatment. This was used to in-situ sulfidate the SPM input surface through H_2S pyrolysis in order to form an effective energy barrier against the recombinative re-emission of absorbed hydrogen atoms and thereby drastically improve the SPM performance. Permeation experiment was carried out in the H_2 pressure range relevant to divertor operation (H_2 $6 \cdot 10^{-4} - 1 \cdot 10^{-2}$ Torr). A high efficiency of atomic hydrogen utilization was demonstrated: the SPM pumping speed was 480 l/s whereas the speed of H_2 molecule dissociation with tantalum ribbons did not exceed 880 l/s. The SPM pumping speed remained as high as 440 l/s when compression ratio reached 400. The permeation flux density achieved $\sim 10^{21} \text{ H}_2 / (\text{m}^2 \cdot \text{s})$ at the total SPM throughput of $\sim 6 \cdot 10^{19} \text{ H}_2 / \text{s}$. The hydrogen pumped out and pre-compressed by the SPM was eventually compressed to normal pressure directly by the foreline pump without any intermediate steps.

Keywords: superpermeable membrane, plasma fusion reactor, D/T-mixture, selective pumping, atomic hydrogen.

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Since the He concentration in the operating plasma of nuclear fusion reactors (NFR) should not exceed 5%, the gas mixture containing $\sim 95\%$ D/T should be continuously evacuated from the operating plasma through a divertor and purged of He. The solution to this problem for post-ITER machines is to selectively pump out most of the D/T mixture in the vicinity of the divertor and return it to the working plasma by the shortest path, bypassing the tritium plant [1–3]. It is proposed to accomplish this by using metal membranes that are super-permeable to superthermal hydrogen particles whose energy is of the order of 1 eV or more [1–4]. Such superthermal particles are, in particular, thermal hydrogen atoms possessing an excess of chemical energy (2.2 eV) [1,3,5,6]. Super-permeable membrane (SPM) allow super-thermal hydrogen particles to pass through with a probability comparable to one per surface collision, automatically compress permeating hydrogen, and are completely impermeable to other gases, including He [5].

It is proposed [1–3] to install the SPM in combination with a superthermal hydrogen generator along the walls of the pumping duct of the NFR in close proximity to the divertor (Fig. 1). Either a cold hydrogen plasma [1–4,7–9] or a incandescent metal surface (atomizer) [1,3,6] is considered as the superthermal hydrogen generator. To prove the feasibility of such a concept, modelling experiments of technically relevant scale [1–3] are required, demonstrating achievement of the required pumped hydrogen flux density at the $10^{16} - 10^{17}$ molecules/($\text{cm}^2 \cdot \text{s}$) scale [1–4] and spe-

cific pumping speed at the $11 / (\text{cm}^2 \cdot \text{s})$ scale in the pressure range $10^{-4} - 10^{-2}$ Torr [1–3], as well as compression of pumped hydrogen to normal pressure [1,2].

We believe that the achievement of the above parameters should first be demonstrated with pure protium (H). Based on previous data [6], we do not expect that isotopic effects (D, T) and the presence of He can significantly alter the results obtained.

The experiment was carried out on the SUPMEM-1 setup (Fig. 2), modelling the scheme presented in Fig. 1. The

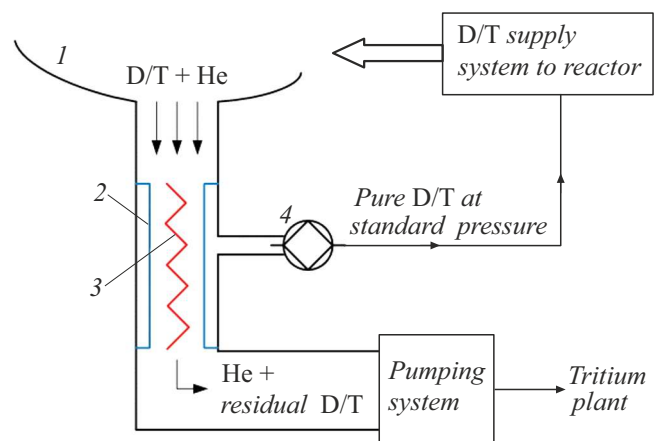


Figure 1. Conceptual scheme of selective pumping of the D/T mixture from the NFR using SPM. 1 — NFR, 2 — SPM, 3 — atomizer, 4 — foreline pump.

vacuum chamber of SUPMEM-1 simulates the pumping duct of the NFR shown in Fig. 1. Along the walls of the vacuum chamber, there is a cylindrical (in the form of a thin-walled tube) SPM interfaced with the atomizer as schematically shown in Fig. 1. The cylindrical SPM is made of niobium, which along with other group five metals (V, Ta) is the most suitable material for SPM [5]. The membrane area is 0.06 m^2 , the thickness is 0.1 mm . SPM can be heated to any temperature (up to melting) due to ohmic (direct current) heating. The atomizer is two ohmically heated free hanging tantalum ribbons, their total area is 80 cm^2 .

The SPM separates two vacuum chambers (Fig. 2): an inlet chamber, where the flux H_2 enters and where the atomizer is located, and an outlet chamber, where the hydrogen passing through the SPM is released. The turbomolecular pump (TMP) present in the output volume pumping system (Fig. 2) is switched off in normal operation, and thus the output side of the SPM according to the conceptual scheme (Fig. 1) is pumped directly by the foreline pump (without any intermediate stages like mercury pump [2]). The inlet volume is continuously pumped by the TMP, which models the pumping system of He and residual D/T (Fig. 1).

In order to ensure the required parameters, it is necessary to create a sufficiently high and dense energy barrier on the SPM entrance surface, which inhibits the associative desorption of absorbed atoms and dissociative absorption of gas hydrogen molecules, but does not prevent the free absorption of H atoms in the gas phase [5]. For this purpose, it is possible to *in situ* modify the chemical state of SPM surfaces. This possibility is realized due to 1) ohmic heating of the SPM, which allows to vary the temperature of the SPM up to the melting point, while leaving the water-cooled walls of the vacuum chamber cold; 2) controlled change of the environment in which the SPM is located.

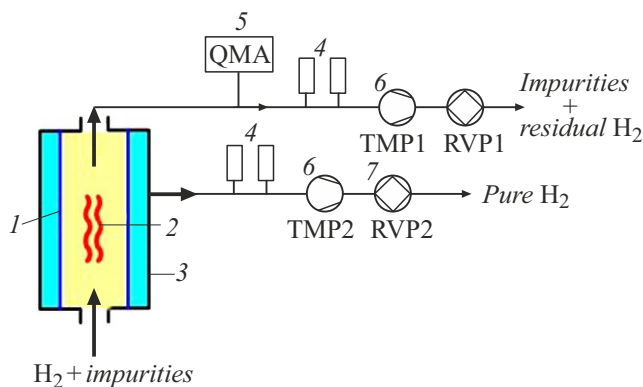


Figure 2. Schematic diagram of SUPMEM-1 unit. 1 — niobium ohmically heated SPM, 2 — tantalum ribbons (atomizer), 3 — water-cooled vacuum chamber, 4 — pressure gauges, 5 — quadrupole gas analyzer, 6 — turbomolecular pumps, 7 — foreline pumps. The input and output volumes are highlighted in yellow and blue, respectively. A color version of the figure is provided in the online version of the paper.

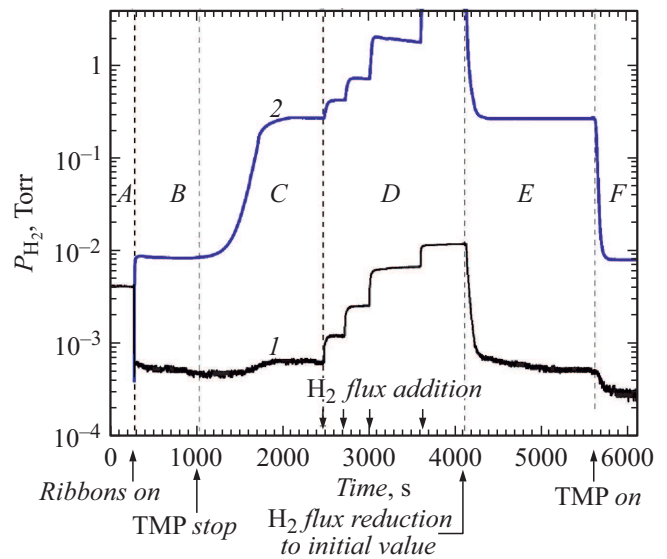


Figure 3. The observed pattern of pressure changes at the inlet (1) and outlet (2) of the SPM. In the interval B–F, the SPM temperature is $\sim 970 \text{ K}$ (the result of heating by radiation from the atomizer).

In this experiment, the SPM was heated in ultrahigh vacuum at $1250\text{--}1280 \text{ K}$ for $\sim 3 \text{ h}$ before starting, then sulfur was deposited on the input surface in an amount corresponding to ~ 15 monolayers. S deposition was performed by pyrolysis of H_2S at a SPM temperature of 1000 K and monitored using a quadrupole gas analyzer (Fig. 2). The choice of S as the element responsible for the barrier is based on the known ideas about sulfur as an extremely strong catalytic poison inhibiting recombinative (in the form of H_2) reemission of H atoms absorbed in the metal and thus contributing to the superpermeability regime [5].

The experimental results are demonstrated in Fig. 3. In the initial state (interval A), the atomizer is switched off, the inlet volume is evacuated by the TMP at a rate of $701/\text{s}$ and a flux of hydrogen enters, which creates a pressure $4 \cdot 10^{-3} \text{ Torr}$ (measured by a baratron). The outlet volume is pumped out by the TMP at a rate of $301/\text{s}$, and there is a deep vacuum.

Further (interval B), the tantalum ribbons of the atomizer are heated to $2075\text{--}2085 \text{ K}$, H_2 dissociation occurs and the formed H atoms permeate through the SPM. This causes the pressure in the inlet volume to drop to $4.9 \cdot 10^{-4} \text{ Torr}$ and the pressure in the outlet volume to rise to $7.9 \cdot 10^{-3} \text{ Torr}$ (measured by the Pirani gauge). This implies that the SPM pumping rate is $4801/\text{s}$ ($0.81/(\text{cm}^2 \cdot \text{s})$) with a compression ratio of 16. Assuming that the dissociation probability of the molecule H_2 on the surface of tantalum ribbons is 0.25 (the maximum value for atomically pure surface [6]), the volumetric dissociation rate of H_2 by 80 cm^2 atomizer ribbons does not exceed $8801/\text{s}$. Thus, despite the inevitable recombination losses at the ends of the cylindrical SPM, more than half ($4801/\text{s}$) of the dissociated hydrogen passes through it. This means that H atoms are effectively absorbed

through the Sulfur-coated surface and that, due to the thermal re-emission barrier created by the S film, most of them permeate through the SPM.

Further (interval *C*), the output TMP is switched off and the SPM output is directly pumped out by a foreline pump (rotary type with oil trap). This is possible due to the fact that the continuous flow of hydrogen in viscous mode blocks the access of any gases to the SPM from the side of the foreline pump and the outlet surface of the SPM is in a favorable environment of ultrapure hydrogen. While the output pressure increases sharply and the compression ratio reaches ~ 390 , the inlet pressure increases only slightly and the pumping speed of the SPM with S film-coated inlet surface remains high (440 l/s). At this time the SPM pumps H₂ flux ~ 0.3 sccs (standard cubic centimeters per second) at a pressure of $6.4 \cdot 10^{-4}$ Torr.

Further (interval *D*), the hydrogen inlet flux increases stepwise, and at time $t = 3100$ s the flux pumped by the SPM is ~ 2.1 sccs or $\sim 5.7 \cdot 10^{19}$ H₂/s. For the SPM area of 0.06 m², this corresponds to a flux density of $0.94 \cdot 10^{17}$ H₂/(cm² · s) at a pressure of $6.4 \cdot 10^{-3}$ Torr. Thus, the SPM with a S-coated inlet surface provides a flux density of the required scale over the relevant pressure range. When the flux is further increased ($t = 3700$ s), the pumping of the SPM continues and the permeate flux density further increases, but the output pressure is beyond the correctness of the Pirani gauge measurements.

Then (interval *E*), the hydrogen flux into the inlet volume is reduced to the initial level, and then (interval *F*) the output TMP is switched on. The output and input pressures return to the previously observed values (intervals *C* and *B*), which demonstrates stable operation of the SPM:

1) under conditions when its input side, covered by the monolayer film of S, interacts with atomic hydrogen (we assume that at the pyrolysis temperature H₂S sulfur deposited on the niobium surface dissolves in the metal volume and only the extremely chemically and thermally resistant monolayer S remains on the surface [5]);

2) when the output side of the SPM is pumped out by the foreline pump, which provides the cleanliness required for SPM operation by a stream of ultrapure hydrogen flowing through the SPM and blocking any gases from the foreline pump side.

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Conflict of interest

The authors declare that they have no conflict of interest.

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