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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

SELF-PUMPING IMPURITY CONTROL BY IN-SITU METAL DEPOSITION

by

J. N. Brooks and R. F. Mattas

Fusion Power Program

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ABSTRACT

A system for in-situ removal of helium by trapping in freshly deposited metal surface layers of a limiter or divertor has been studied. The system would trap helium on a limiter front surface, or a divertor plate, at low plasma edge temperatures, or in a limiter slot region, at high edge temperatures. Fresh material, introduced to the plasma and/or scrape-off zone, would be added at a rate of about five times the alpha production rate. The material would be reprocessed periodically, e.g. once a year. Possible materials are nickel, vanadium, niobium, and tantalum. Advantages of a self-pumping system are the absence of vacuum ducts and pumps, and the minimization of tritium processing and inventory.

1. INTRODUCTION

High energy particles (deuterium, tritium, and helium) impinging on material surfaces are known to become trapped within the material up to saturation levels which depend on the particle species and energy, the type of material, and the material temperature. Passive helium removal using this approach was first examined for a short pulse experimental reactor,^{1,2} but saturation limits the effective pulse length for power reactors. By continually plating the surface, however, it may be able to achieve long burn times (>6 mo) with a relatively simple system. A conceptual view of this approach is shown in Fig. 1. A key requirement for this application is for the material to trap helium much better than hydrogen. It has been demonstrated experimentally that nickel preferentially traps helium,³ and that several other materials (iron, vanadium, niobium, and tantalum) are believed to be capable of preferential trapping. The selective trapping in certain metals is a result of

Fig. 1. Conceptual view of self-pumping system.

the negligible solubility of helium compared with the relatively high solubility of hydrogen in the lattice. The injected helium will diffuse through the lattice until it reaches a nearby trapping site where it can come out of solid solution. Hydrogen, on the other hand, remains in solid solution until it diffuses to the surface and escapes. Thus, the material surface can act as a selective helium pump. However, helium trapping occurs only up to a saturation level, typically 10^{17} - 10^{18} /cm², after which it is released at the same rate it is injected. In order to be used as a pump in a tokamak reactor, a fixed trapping surface would have to be regenerated after every pulse by such means as thermal spiking. By continuously plating the surface, however, much longer burn pulses, suitable for a commercial power reactor, may be feasible.

The system envisioned depends on the plasma edge temperature. At low temperatures ($\lesssim 50$ eV) where self-sputtering is not a problem, the trapping surface could be the front surface of a limiter with no leading edges, or a divertor collector plate. At higher edge temperatures, trapping might be accomplished in a limiter slot region, with low-Z material being used for the front face and trapping material used for the slots.

The sections that follow address the requirements, feasibility and major plasma engineering, and materials concerns for an impurity control system using surface trapping of incoming helium ions. Parameters from the INTOR reactor design are used as a convenient reference in the analysis.

2. MATERIALS SELECTION

There are several materials requirements for selective helium pumping:

- (1) high hydrogen solubility;
- (2) high hydrogen diffusivity;
- (3) absence of hydride formation;
- (4) high thermal conductivity;
- (5) adequate operating temperature window;
- (6) high probability of helium trapping ($\gtrsim 0.25$);
- (7) high saturation trapping level; and
- (8) self-sputtering coefficient < 1 .

Only a few materials can meet these requirements. Low-Z materials, where the self-sputtering coefficients are always less than unity (e.g., beryllium and carbon) will tend to trap hydrogen as well as helium.³⁻⁶ Metals that will selectively trap helium, such as iron or nickel, do not have adequate thermal conductivity to accommodate high heat fluxes.⁷ Other metals, such as titanium or zirconium, can be eliminated because of hydride formation at low hydrogen concentration levels.^{5,8} Metals that appear capable of meeting the above requirements are some of the transition metals such as vanadium, niobium, and tantalum, and possibly tungsten, and molybdenum. The first three metals are known to form hydrides, but they also have high hydrogen solubilities.⁹ These metals are considered to be high-Z materials, and therefore they can only be used at low particle energies due to self-sputtering limitations.^{10,11}

The materials properties of interest are:

- (1) hydrogen solubility;
- (2) ion trapping/reflection;
- (3) saturation trapping levels;
- (4) hydrogen diffusion and release mechanism;
- (5) material surface damage;
- (6) self-sputtering damage;
- (7) helium release versus temperature;
- (8) material vapor pressure; and
- (9) ion penetration depth.

In this report, vanadium (for helium trapping behavior) and tungsten (for erosion/redeposition behavior) have been investigated in somewhat more detail to determine the feasibility of the self-pumping system. The other transition metals are believed to operate in a similar fashion.

3. FEASIBILITY

In order to test the feasibility of selective helium pumping two specific designs were examined: (1) a divertor plate at low edge temperatures; and (2) a limiter/slot system at higher temperatures. Parameters for the first case

are shown in Table 1. The total number of helium ions to be pumped is $2.2 \times 10^{20} \text{ s}^{-1}$. The self-pumping system must be capable of pumping this amount of helium over an extended period of time, typically 6 mo of steady-state operation or its equivalent (e.g. 1 yr at 50% duty factor). The basic concept use here is to use the metallic neutralizer plate as the removal mechanism.

TABLE 1
Parameters for Self-Pumping System Using a Divertor

Parameter	Value
Concept	Self-pumped divertor
Plasma temperature at separatrix, eV	50
Helium production rate, s^{-1}	2.2×10^{20}
Helium trapping rate, s^{-1}	2.2×10^{20}
Area of plate, m^2	32
Metal (vanadium, etc.) current to plate, s^{-1}	1.1×10^{21}
Energy of helium ions striking plate (at separatrix), eV	450
Energy of D-T ions striking plate (at separatrix), eV	300
Energy of redeposited metal ions striking plate (at separatrix), eV	≤ 700

Particles entering the slot region impinge on a neutralizer plate where a significant fraction of the helium particles are trapped. The escaping D-T particles are allowed to flow back out of the divertor and re-enter the plasma scrape-off region. The surface of the neutralizer plate is continuously regenerated by an incoming flow of metal atoms that deposit at a rate that is sufficient to prevent saturation of helium trapping. Thus, over time, the neutralizer plate increases in thickness. Metal could be added to the plasma, e.g., by pellets or to the divertor channel region, e.g., by vaporization of metal rods, as will be discussed later.

The rate at which metal needs to be supplied to the surface depends upon the saturation levels for the helium. As shown in Table 2, saturation levels for hydrogen in several materials range from 16 to 100% of the host atoms.⁶ It is believed that trapping levels for helium are also in this range.³ For this study it has been assumed that the saturation trapping fraction for

TABLE 2

Saturation Levels for Hydrogen Trapped in Various Materials

Material	1.5-keV Deuterium			Saturation Concentration	
	Range (Å)	Straggling (Å)	Saturation (10^{17} D/cm ²)	(10^{22} D/cm ³)	H/Host (%)
Carbon	247	101	2.50	6.0	53
Silicon	335	161	1.91	2.5	50
Boron	216	95	3.40	5.9	45
B ₄ C	288	117	3.57	6.9	57
TiC	183	91	1.22	2.5	26
VB ₂	189	92	0.96	1.9	16
TiB ₂	208	102	1.00	1.8	16
316 stainless steel ^a	100	117	3.00	8.5	100

^a-120°C.

helium in vanadium is 20%. Therefore $\sim 10^{21}$ vanadium atoms/s are required to trap the helium. For FED/INTOR conditions the energies of the incoming deuterium, tritium, and helium particles to the neutralizer plate are estimated to be $\lesssim 300$ eV (including the sheath potential). At these energies, the penetration depth is predicted to be only ~ 40 Å,⁴ and the reflection coefficient is estimated to be ~ 0.3 for particles incident normally to the surface.¹²

Self-sputtering of the metallic surface is a critical issue. Self-sputtering coefficients will, in general, be greater than unity for material added to the plasma and impinging on the surface (because of the high charge state and equilibration with the plasma flow velocity) but what is more important is the self-sputtering coefficient of the redeposited-sputtered material. If the latter coefficient is less than unity a stable surface can result,¹⁰ with the only growth being due to the intentional addition of material. Plasma contamination is minimal in this regime. Based on present estimates of charge state, sheath potentials and sputtering coefficients,¹¹ a plasma temperature of $\lesssim 50$ eV is low enough to keep redeposited energies $\lesssim 700$ eV. This appears to be the general range required for less than unity sputtering coefficients. However, these are fairly crude estimates which need experimental verification.

A REDEP code¹¹ simulation of the self-pumped divertor surface was performed and results are shown in Fig. 2. Tungsten was used as the coating material and added at the rate listed in Table 1. The erosion/redeposition behavior of tungsten is believed to be representative of the other high-Z metals. All plasma parameters used are scaled from the INTOR design.¹⁰ The separatrix is at point 14.5 and all points are linearly spaced. As shown, at the end of 6 mo (of continuous operation), the surface has grown by a maximum of ~ 0.6 cm. The effect of this growth on the surface heat flux is negligible. The total volume of material used during a 6-mo period is ~ 0.25 m³. Thus, the amount of material required for pumping is acceptable to achieve a lifetime of >1 yr at 50% availability, for the impurity control system. For all other things being equal, a limiter system would be expected to give similar results as the divertor. At low edge temperatures the limiter would not need leading edges — only a front face.

Fig. 2. Heat flux and surface growth for a self-pumped divertor using tungsten.

An example of a limiter slot system that might be used (at higher plasma edge temperatures) is shown in Table 3. Beryllium, or other low-Z material,

TABLE 3

Parameters for Self-Pumping System Using Limiter Slot Trapping

Parameter	Value
Concept	Double-edged limiter with two slots
Front face material	Beryllium
Slot material	Vanadium, etc.
Plasma edge temperature, eV	150
Temperature at slot, eV	50
Helium production rate, s^{-2}	2.2×10^{20}
Helium current to limiter, s^{-2}	2.2×10^{21}
Helium entering slots, %	10
Total neutralizer plate areas, m^2	~ 10
Metal (vanadium, etc.) current to plate, s^{-1}	1.1×10^{21}
Energy of redeposited metal ions striking plate (maximum), eV	$\lesssim 700$
Heat flux to plate, MW/m^2	$\lesssim 1$
Plate operating temperature, $^{\circ}C$	$\gtrsim 150$

is used for the front face to minimize erosion from self-sputtering. Vanadium, etc., is used for the slot regions to trap helium. While most vanadium would be confined to the slot region, some beryllium would be transferred from the front face to the slot region. The input of the transferred beryllium on helium pumping needs further examination. The average rate of buildup, over $10 m^2$ of neutralizer plate area to pump $\sim 2 \times 10^{20}$ helium per second is estimated to be $1.4 \times 10^{-9} \mu m/s$ which translates to 4.8 cm/yr (continuous operation) for INTOR.

The divertor and limiter slot system may differ from the limiter-front-face-only system in the physics of hydrogen release, charge-exchange, flow velocities, etc., and this issue needs further assessment.

Another important consideration is the method to be used for providing the necessary material to the trapping surface. For a system using the limiter front face for trapping, metal injection into the plasma, e.g. by pellets

or puffing may be the simplest technique. The required amount of metal is small compared to the D-T flux. For the divertor system or limiter slot system potentially the simplest technique for achieving the necessary material flux would be to allow the incoming D-T flux to heat up metal rods or bars, located in the scrape-off or slot regions, to the point where metal is vaporized from the surface. The vaporized atoms would then be swept into the neutralizer plate with the D-T particle flow. For example, Fig. 3 shows the vaporization rate for vanadium as a function of temperature, and Fig. 4 shows the area of vanadium required to supply a current of $10^{21}/s$. It can be seen that at temperatures >1950 K, the area required is <1 m² or only 10% or less of the neutralizer plate area. It is believed that these temperatures can be achieved simply by inserting vanadium rods into the slot D-T flux. For example, the equilibrium temperature of vanadium receiving a heat flux of 0.5 MW/m² that loses the heat by radiation is given by

$$0.5 \text{ MW/m}^2 = \epsilon \sigma T^4, \quad (1)$$

where ϵ is the emissivity, σ is the Stephan-Boltzman constant, and T is the absolute temperature.¹³ If ϵ is assumed to be 0.4, then the absolute temperature is calculated to be 2166 K ($T_m = 2133$ K). Thus the D-T flux provides enough energy to raise the vanadium temperature and provide an adequate flux of vaporized atoms. The amount of vaporization can be easily controlled by adjusting the height to which the rods are inserted into the slot region.

An important consideration for vanadium is the possibility of a high retained D-T concentration in the surface layer that could lead to hydride formation. The equilibrium concentration depends upon several factors, including the hydrogen diffusion rate, the hydrogen recombination rate at the surface, and the hydrogen partial pressure in the slot region. The operating temperatures for the vanadium range from $\sim 150^\circ\text{C}$ to $\sim 600^\circ\text{C}$. The diffusion coefficient, D , for vanadium is given by

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

where $D_0 = 5.2 \times 10^{-4}$ and $E_D = 0.08$ eV.¹⁰ At 150°C and 600°C the diffusion coefficients are 5.8×10^{-5} cm²/s and 1.8×10^{-4} , respectively. The time required to diffuse the penetration depth of 40 Å is approximated by¹⁴

Fig. 3. Vaporization rate of vanadium as a function of temperature.

Fig. 4. Surface area of vanadium required to supply 10^{21} atoms/s from vaporization (MP = 2133 K).

$$t \approx (\bar{x}^2/D) . \quad (3)$$

At 150°C and 600°C the diffusion times are calculated to be 2.7×10^{-9} and 8.9×10^{-10} s, respectively. Thus, diffusion to the surface is extremely rapid and should result in low concentrations as long as other processes are not rate limiting. However, the release of hydrogen could also depend upon the recombination rate at the surface. The surface release flux, ϕ_s , is given by

$$\phi_s = k_r C^2 , \quad (4)$$

where k_r is the molecular recombination constant and C is the bulk hydrogen concentration.¹⁵ The actual values for k_r are highly uncertain and are expected to depend upon the surface geometry, the impurity (oxygen, nitrogen, and carbon) coverage on the surface, and the influence of the impinging ions and electrons on desorption. Predictions for k_r have been made for several materials including vanadium as shown in Fig. 5.¹⁵ At 150°C, $k_r \approx 10^{-23}$ cm⁴/s, and at 600°C, $k_r \approx 10^{-19}$ cm⁴/s. The average D-T flux for the FED/INTOR conditions is 1.25×10^{18} /s cm². At equilibrium where the release rate equals the injection rate, the D-T concentrations at 150°C and 600°C are calculated to be 3.5×10^{20} /cm³ and 3.5×10^{18} /cm³, respectively. Again the hydrogen concentrations are predicted to be low. The solid solution concentration in equilibrium with the D-T partial pressure is also expected to be low. The hydrogen concentration in vanadium at 0.15 Pa (10^{-3} torr) along with the hydrogen solubility are shown in Fig. 5.⁹ At 150°C and 600°C the predicted hydrogen concentrations are ~0.3 at % and $\sim 7 \times 10^{-3}$ at %, respectively. All values for hydrogen concentration are well below the concentrations needed for hydride formation (see Fig. 6).

A final consideration for the feasibility of a self-pumping system is the operating temperature range for the vanadium. It is expected that the lower limit would be set by hydrogen retention and hydriding. As shown above, at the lowest expected temperature of ~150°C, the predicted hydrogen levels are well below the hydriding levels. The upper temperature limit is expected to be set by the point at which the helium is released from the trapping sites. The release of helium is usually associated surface deformation, in the form of blisters or flakes.¹⁶ Thus the point at which such surface damage occurs gives a rough approximation to the time of helium release. A collection of

Fig. 5. Calculated molecular recombination rate constants (k_r) for various metals, assuming a molecular sticking coefficient (α) of unity (Ref. 13).

Fig. 6. Equilibrium hydrogen concentration at 0.13 Pa hydrogen pressure and hydrogen solid solubility in refractory metals.

surface damage data as a function of temperature is shown in Fig. 7.¹⁶ The helium concentration level at which deformation occurs decreases with temperature. In order to maintain a trapping level concentration of ~20%, the temperature would be limited to ~0.35 T_m which for vanadium is 475°C. A reasonable upper limit for temperature is therefore 450-500°C.

Fig. 7. Summary of the critical helium concentration for surface deformation and helium release, as a function of the fraction of the absolute melting temperature (T/T_M).

In summary, the primary materials requirements for a self-pumping impurity control system are satisfied for vanadium. Other materials, including nickel, niobium, and tantalum, could also be suitable for such applications.

4. UNRESOLVED ISSUES

There are several unresolved materials issues, as shown below:

- (1) properties of deposited vanadium containing trapped helium;
- (2) effect of radiation on trapping of hydrogen within the lattice;
- (3) tritium permeation to the coolant;
- (4) influence of surface characteristics and particle bombardment on hydrogen release characteristics.

The important properties for determining the behavior of the deposited vanadium are the thermophysical and mechanical properties. If these properties are severely degraded, the allowable thickness of the deposited material will be reduced, and the lifetime will be shortened. Radiation damage to the lattice could result in additional trapping sites for hydrogen which could raise the retained hydrogen to unacceptably high levels. Tritium permeation to the water coolant is primarily a safety concern. Should significant levels of tritium reach the coolant, then a large water cleanup system would be required. Finally, D-T release from the vanadium surface could be dependent on the surface characteristics and poorly characterized at this time.

Among the unresolved plasma related issues are the following:

- (1) effect on the plasma of high hydrogen recycle rate via low energy atoms — as opposed to pellet refueling of ~10% of the D-T outflux for a "conventional" pumping system; and
- (2) edge temperatures, flow velocities, and charge states of impurities, in the scrape-off zone and/or the divertor channel, and limiter slot regions.

5. CONCLUSIONS

Although much further work, both experimental and analytical, is needed, a self-pumping helium removal system appears to be promising. Such a system would eliminate all vacuum ducts and pumps (except for a small system for start-up purposes). Tritium recycle and inventory would be minimized. At low temperatures a very simple limiter concept (without leading edges) could be used, or a simplified divertor system.

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