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Boronization in Future Devices -Protecting Layer against Tritium and Energetic Neutrals-

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Boronization in future devices*
– Protecting layer against tritium and energetic neutrals –

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Abstract

A thin boron film is attractive as a deuterium/tritium free wall, and as a protecting layer against impact of energetic charge-exchange neutrals in future fusion devices with a long-time operation. New experimental evidences are given for desorption of hydrogen isotopes at relatively low temperature. All hydrogen atoms in a boron-coated layer are reemitted to the plasma side below 400 ℃ without their penetration into substrate of stainless steel. A problem is maintainability of a thin boron layer during a long-time operation. Boron atoms are hardly removed by pumping because its hydrides are easily disintegrated and redeposited. Gross immigration of boron atoms inside the vessel is a concern. A condition required for avoiding the immigration is discussed.

Key words: boronization, long-time machines, tritium inventory, tritium permeation, protecting layer, damages by energetic neutrals, redeposition

1. Introduction

It has been widely recognized that boronization [1] plays an important role in present fusion devices. However, its roles and availability are not seriously discussed for future devices with a long-time steady state operation. Oxygen and hydrogen gettering function would be lost within a short time due to saturation of these atoms at the top surface of the boron film. It has not yet been conclusive what plays a key role to achieve good confinement scheme after boronization. This paper gives a brief review on the roles of the boronization in present and future devices. The availability and problems in future are also discussed.

2. Possible roles of boron films

2.1. Roles in present machines

Roles of a flesh, thin boron film in the present machines are (i) suppression of oxygen contamination to core plasmas [1], (ii) reduction in hydrogen recycling compared with carbon walls [2] and (iii) suppression of contamination by wall materials [3].

In TEXTOR [1] and Tore Supra [4], oxygen reduction after boronization was clearly seen and it resulted in widening of the operational space in Hugill diagram, namely in higher density limit.

In D III-D [2], impurity reduction was seen after boronization. The levels of O, C, Ni were reduced significantly. It was reported that wall fueling rate in the initial phase of NBI heating was also reduced by a factor of 2 after boronization. A good confinement scheme named "VH-mode" was found for the first time with this situation.

In JT-60U [5], recycling flux was measured as a function of line-averaged electron density. It was found to be reduced after boronization. At the same time, correlation between the recycling flux and confinement improvement was investigated. Enhancement factor of confinement decreased as recycling increased. From these results, it is suggested that reduction in recycling after boronization could be cause of the better confinement.

In Alcator C-Mod [3], boronization was applied to a metallic wall. It is a different situation from other machines such as TEXTOR, Tore-Supra, D III-D or JT-60U, where large area of the plasma facing wall was covered by carbon. Confinement improvement was observed after boronization in C-Mod, too. No clear change in recycling was found but significant reduction in radiation was observed after boronization. Electron temperature at the edge was higher with boronization than that without boronization. It was suggested that reduction in metallic impurity such as Mo caused higher edge temperature, which could trigger the better confinement.

In ASDEX-Upgrade, boronization was applied through a period of tungsten divertor experiments in 1996 [6]. It is said that boronization was necessary

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to realize smooth start up of the experiment and to have a sufficiently wide operation space.

Through these experiences in several tokamaks, it can be said that boronization brings us better performances of the core plasmas in most cases. However, it cannot be generally concluded at present which of (i) - (iii) is the key for better performance of the core plasmas.

The roles of (i) and (ii) are regarded as a utilization of a wide area of the first wall for pumping oxygen or hydrogen. These are useless in a long time discharge because the surface will be saturated with them within a few hundreds or a few thousand seconds [7]. These two roles should be taken over by active pumping of limiter/divertor in future devices. The third role would be useful if the thin film could be maintained for a long time without addition of boron compound during operation. If metallic impurities are really responsible for reduction in plasma performance, and if a maintenance way of the boron film is established, the role (iii) could be promising in future long time machines. A proposal of boronization combined with high Z target plates was given with this consideration in Ref. [8].

2.2. Roles in future long-time machines

In the previous subsection, the roles (iii) is suggested to work possibly in the future machines. A possibility of another role has been pointed out [9], that is, (iv) the thin film would help to reduce tritium inventory and permeation in DT machines. It is based on an experimental fact that hydrogen is completely removed from the B film at a surface temperature below 400 ℃ [9], which is not expected for a carbon wall. In a long-time discharge, it would be not difficult to maintain the temperature of plasma-facing surfaces as high as 300 to 400 ℃ by utilizing heat flux from the plasma. An expected temperature difference between the top surface and water-cooling channels can be less than 300 ℃, which gives a design of the first wall without difficulty of mechanical problems due to a large temperature gradient. If the boronized surface is maintained at 300 - 400 ℃, most of tritium is released from the surface which results in low T inventory. Energetic T atoms from a plasma are blocked by the thin B-film and the tritium is re-emitted to the plasma side at this temperature. This point is discussed more in Sec. 3 based on experimental result.

This B-film protects the wall from micro-damages by energetic particle impact, too. A concern about this type of damages is pointed out by N. Yoshida in ICFRM-8 [10] and in this conference [11,12]. In a long time operation, fluence of energetic charge-exchange neutral becomes enormous. During one-day operation, it could be $10^{25}/m^2$ for hydrogen isotope and 10^{21} - $10^{22}/m^2$ for helium at the first wall, that is, plasma facing part of blanket. These energetic particles give damages as dislocations, loops, blisters at the top surface of the plasma facing components. These damage could result in hardening and mechanical failure of the surface, increase in tritium inventory, and other unfavorable events. The damage production is serious especially with helium impact. Threshold energy of the damage production is found to be below 250 eV, and annealing temperature to be higher than 1500 ℃ [11]. It should be seriously considered how to avoid this kind of damages. A thin boron film is expected to work as a protecting layer against the energetic charge-exchanged particles. This is its fifth possible role (v), which is favorable in a future long-time machine.

A question is whether a thin boron film can be maintained for a long time or not ? This point is discussed in Sec. 4.

3. Experimental results in SUT

Basic properties of boron films have been investigated in the SUT (SUrface modification Teststand) device in NIFS [7 - 8, 13 - 15]. The SUT device has a cylindrical liner inside a vacuum vessel, which can be heated up to 600 ℃ with molybdenum heaters backside of the liner. Total area of the inner surface of the liner is 7000 cm^2. In this experiment, a liner of stainless steel was used. A boron film is coated inside this liner through a glow discharge with helium/diborane mixture gases. Total pressure is 2.6 Pa, discharge current 0.2 A, and voltage 400 V. The coating process is conducted at the liner temperature below 70 ℃. Typical thickness of the film is 200 nm measured by a quartz oscillator with an assumption that mass density of the film is 1.5 g/cm^3.

As discussed in the previous section, one of the interests is hydrogen isotope behavior in the boron film. Deposited boron film includes hydrogen originated from diborane. Most of this hydrogen was desorbed by raising the liner temperature up to 500 ℃ (the first thermal desorption procedure TD1). Then a hydrogen glow discharge was started with the hydrogen depleted boron wall (hydrogen discharge procedure HD1). During the discharge, hydrogen atoms were absorbed to the boron film, which could be indicated by a drop in hydrogen pressure during the glow discharge measured by a residual gas analyzer. Total amount of absorbed hydrogen was 1 - 2 x $10^{17}/cm^2$. Second thermal desorption (TD2) was carried out. Total number of the desorbed hydrogen is roughly the same as absorbed one during HD1. Figure 1

shows a typical hydrogen pressure as a function of the liner temperature during TD2. Changing rate of the temperature was 10 ℃ /min. This indicates that most of the hydrogen in the boron film can be desorbed below 400 ℃. A number of experiments have been repeated with a variation of hydrogen discharge time, with insertion of helium glow discharges etc. The reemitted hydrogen amount is always comparable to the one expected from preceded hydrogen absorption discharge. Thus it can be concluded that most of hydrogen isotopes can be reemitted from a boron film below 400 ℃.

The results above described was obtained for pure boron films produced in a good vacuum condition. Oxygen contamination on the boron film was less than 5 atomic percent. In order to have more realistic condition, hydrogen desorption was investigated for fully oxidized boron film, too, where surface oxygen concentration was around 40 % [15]. Total oxygen uptake was reduced for the oxygen contaminated boron film. compared with the pure films. The temperature of maximum desorption was not changed with oxygen contamination, namely, at below 400 ℃.

Depth profile of hydrogen inside the boron film was investigated with ERD (Elastic Recoil Detection) measurement [8, 16]. The boron film was coated on a small stainless steel test piece on a manipulator in SUT. It was transferred to an ion beam facility in Nagoya University. A helium ion beam of 1.5 MeV was utilized in this ERD analysis. Figure 2 shows results of the ERD measurement for a boron film coated on a stainless steel substrate. It is seen that, after the first thermal desorption TD1, residual hydrogen atoms are distributed uniformly inside the boron layer. Its concentration is relatively small. After hydrogen glow discharge HD1, much higher concentration of hydrogen atoms is seen on the top of the film. Due to poor spatial resolution, apparent profile shown in the figure is broader than real one. According to a numerical analyses, the implanted hydrogen atoms are stayed within 10 nm in depth from the top surface of the film [8].

The results in this figure indicate that the residual hydrogen atoms are seen only inside the boron layer, namely within the thickness of 200 nm. There is no hydrogen penetrating into the stainless steel substrate even after the thermal desorption procedure of TD1.

Thermal desorption of hydrogen has been already investigated for boron-coated small samples. Residual hydrogen inside a boron film was measured with infrared absorption [17]. A standard TDS was applied to a boron film [18]. Results in these experiments indicate that the main peak of hydrogen absorption lies around 400 ℃. The present experiments has confirmed this in a large scale of the boron-coated area with 7000 cm^2. The result with ERD gives an experimental evidence that hydrogen atoms do not stay in the substrate, which has been not clear in the previous experiments in refs.[17] and [18].

A question is why hydrogen cannot penetrate into the substrate. One possible answer is that the film is porous, with which volume recombination in the bulk is the dominant process of the thermal desorption like graphite. Hydrogen molecules are desorbed and move in the direction of either surface side or substrate side through an open pore. The molecules to the surface side can escape the film, but those to the backside cannot penetrate to the substrate due to low permeability of molecule. In order to check this idea, microstructures were examined with a transmission electron microscope (TEM). A boron-coated stainless steel disc was back-thinned to perforation by ion milling. Figure 3 is a TEM image at a position near an edge of a wedge-shaped boron film. The image shows that the film does not contain any pores. Thus the above-mentioned idea has been found to be unlikely. The insert in Fig. 3 is an electron diffraction pattern extracted from the same position. Absence of diffraction spot in the pattern indicates that the film is amorphous. Relatively high concentration of oxygen and carbon is seen near the boundary with an in-situ Auger analysis [7]. Electron diffraction pattern near the boundary showed large number of extra spots which were neither from the boron film nor the stainless steel substrate. Thus, some chemical compounds such as oxides, carbides or borides seems to be formed around the boundary. The layer with these compounds may possibly work as the barrier for hydrogen isotope penetration. Another possibility is that the permeability into the substrate is determined by difference in chemical potentials between the coated layer and the substrate, or the elements in the substrate. In this case, the permeability may be not always small as stainless steel depending on the substrate material. It is worth to note in this context that a thermal desorption experiment was once carried out for a boron film coated on a graphite liner in SUT. Hydrogen behavior in the TD1, HD1, TD2 procedures was quite the same as that with the stainless steel liner. This suggests that hydrogen did not penetrate carbon substrate below 400 ℃ , too. Detail investigation and analyses is interesting and necessary to obtain more conclusive understanding on this subject.

If you combined the ERD results with a fact that all the implanted hydrogen atoms during HD1 are reemitted in the thermal desorption TD2 as described in the first half of this section, you can get an idea of boron film as a deuterium/tritium free

layer. The concept of the boron-coated first wall is given in the next section.

4. Concept of the first wall with a thin boron film

It is essential to keep temperature of the boron film as high as 300 – 400 ℃ in order to get hydrogen free surface. It can be realized as Fig. 4. Water-cooling channels are welded directly to the vacuum vessel. Panels are set on the channels and covered all inner surfaces of the vacuum vessel, which block all fluxes from a plasma, that is, energetic particles and radiation cannot reach the vacuum vessel directly. Thin boron layers are coated on the top of the panels. During a long time operation, heat flux from the plasma keeps the top surface temperature high enough to desorb hydrogen atom if appropriate thermal insulation is set between the panels and the cooling channels. Wide variety of materials can be used as the panels. For instance, stainless steel panels can be used in non-DT machines. Temperature gradient between top surfaces and cooling channels is less than 300 ℃, which gives no serious thermo-mechanical problems to the first wall system. It would be not the case if carbon layers are utilized. In order to get hydrogen free surface, the top surface must be kept as high as 700 – 800 ℃, which gives more difficulty to the design of the first wall system. This type of the first wall with the boron film is actually proposed to Large Helical System (LHD). Cooling channels have already welded to the vacuum vessel of LHD. Panels of stainless steel will be installed near future. One of the problem of superconducting-coil machine is that a helium glow discharge cannot be applied during a break of successive two pulsed high-power discharges. The helium discharge has been necessary in D III-D with a boronized wall in order to access a good confinement VHF mode [2]. In LHD, a medium power ECH discharge is planned to be utilized during the break in a pulse train or steady-state scheme. This would give heat load to the panel and raise the top-surface temperature, which could help to obtain a hydrogen-depleted wall at the start of the next pulsed high-power shot. Thus the concept in Fig. 4 allows us to have a low recycling wall in superconducting-coil machines.

In DT reactors, the thin boron film could be applied as a protecting layer of plasma-facing part of blankets. Blankets are operated with a temperature higher than several hundreds degree C because coolant temperature must be high enough to generate electric power. Then the temperature of the plasma facing surfaces could be maintained with heat flow not only from the plasma side but also from the blanket side. The protecting layer could help to reduce tritium inventory in the first wall and to decouple the tritium flow between plasma side and blanket side. The layer prevents energetic charge-exchange neutrals hitting the blanket surface.

5. Maintainability of boron layer

Different from carbon-hydrides such as methane, boron-hydrides are very fragile, easily dissociated, re-deposited inside the vessel, and do not reach pumping ducts. Then the total number of boron atoms does not decrease in the vessel even during a long time discharge. In SUT, a hydrogen discharge as long as 10 hours has been applied. No indication has been found for the boron film being lost during this long time discharge. Carbon film was removed with hydrogen discharge through a form of methane because methane is relatively stable. This was confirmed with an experiment in TEXTOR with a carbonized wall [19]. Even with methane-seeded discharge, pressure drop of methane at the start of discharge is as small as 20 % of the initial pressure. In contrast to methane, diborane pressure drop is more than 90 % of its initial pressure during a boronization discharge in SUT as shown in Fig.5. A similar result is reported in D III-D [2]. These results indicate that boron atoms are not removed from the vessel through the pump duct.

A concern in a real fusion device is gross immigration of boron atoms from erosion-dominated area to deposition-dominated area. One of the asymmetries is seen between divertor region and wall region other than divertor. In a steady state, total boron flux Γ_{Binw} sputtered from the wall and Γ_{Bind} from divertor is balanced with a sum of losses Γ_{Boutw} across the magnetic field to the wall and Γ_{Boutd} along the field. This can be established for a given plasma density with an appropriate boron density at the separatrix. This condition is described as the following equation;

$$(\Gamma_{Binw} - \Gamma_{Boutw}) S_W + (\Gamma_{Bind} - \Gamma_{Boutd}) S_d = 0 \qquad (1),$$

where S_W is apparent area of the first wall and S_d that of the cross sectional area of the divertor entrance, respectively. It does not necessary mean that each term of the left hand side of Eq. (1) equals to zero. Then, in general, gross immigration of boron atoms occurs from wall region to divertor region or vice versa. There is one more free parameter, that is, the distance δ between last closed surface and first wall. The flux Γ_{Boutw} is proportional to $\exp(-\delta/\lambda)$, where λ is decay length in SOL. On the other hand, Γ_{Binw} is independent of d because it is mainly determined by charge

exchange neutral flux. Then the first term would be zero if the distance d could be chosen appropriately. The fluxes of Γ_{Binw} and Γ_{Boutw} are dependent on operational parameter such as averaged plasma density, and they are not linearly correlated each other. Then a unique operation parameter could give a condition in which the gross immigration is zero for a fixed d. In an experimental device, a wide operation area is necessary. Then gross immigration in each operation resume is not avoided. In a reactor, a limited number of operation schemes would be selected, which enables a choice of the distance for immigration to be minimum.

Discussion in the previous paragraph is based on the assumption that deposition process is dominated by boron ions moving across a magnetic field. Roughly a half of dissociated fragments moves toward plasma direction, which suggests that this process plays a certain role. But another half can directly reach the first wall and redeposited at a location close to that at which it is emitted. Another view could be obtained by taking into account cross sections for dissociation of boron hydrides. For methane, dissociation cross sections have been published and the total dissociation cross section is 1 - 4 x 10^{-16} cm^2 [20, 21]. There is no publication about the cross section for boron hydrides. The result in Fig. 5 suggests the cross section for boron hydrides could be much higher than that for methane. If you assume the cross section as high as 1x 10^{-15} cm^2, mean free path at the electron density of 1 x 10^{12} cm^{-3} is a few millimeter. This is very small compared to the scale of the reactor. Then boron atoms hardly move from one place to another during discharge.

Thus the gross immigration is expected to be zero, or at least minimized in a future machine like a reactor. It might be necessary to make an additional in-situ coating for erosion dominated part of the wall during a break for maintenance. Utilization of bulk boronized graphite would be another choice for a erosion dominated region. In any case, the additional boron amount could be minimized with an appropriate design based on a data which should be accumulated from now on.

Outer striking points of a single null divertor is one of the erosion-dominated regions. In Alcator C-Mod, poloidal distribution of boron was investigated by a post-mortem analysis by an ion beam method. It was found that the boron layer survives at all the investigated location except the outer striking point [22]. The boron layer at locations strongly interacting with plasmas may be completely removed within a short time. But its area is limited. Actually the improved performance in C-Mod was seen even with molybdenum contamination from this limited area. Protection of the eroded surfaces is another issue to be explored.

It should be noted that removability of boron film after boronization was once discussed in Ref.[17]. Positive conclusion for removability was drawn there, which has a negative meaning to the maintainability of the boron film. The conclusions in [17] and the present paper are still preliminary and tentative. A lot of studies will be necessary to reach a final and sound conclusion on this issue. Nevertheless it is worthwhile and interesting to do studies on boronization aiming its application to future long time machines.

6. Conclusions

Boronization is playing favorable role to obtain good performances of core plasma in various tokamaks. Three functions such as oxygen reduction, lowering of hydrogen recycling and decrease in contamination of wall material have been reported. It has not yet been clear which of above three plays the major role for the good performance.

The functions of oxygen and recycling reduction is not available in future long time machines. The third function against the wall material is available when thin boron film can be maintained during long time operations.

A boron film works as a hydrogen-isotope free wall at 300 - 400 ℃, which can be kept by heat flux from plasma or from blanket in long-time machines. It gives a protecting layer against energetic charge exchange neutrals from plasmas. The layer reduces tritium inventory at the wall, tritium permeation inside the first wall, and microdamages at the surface due to the energetic D, T, He atoms' impacts.

It was confirmed experimentally that most of hydrogen isotope atoms are reemitted from a boron film at a temperature below 400 ℃. It was found that hydrogen atoms do not penetrate into the substrate of stainless steel in this temperature region. These results support the idea of the protecting layer.

A thin boron layer could be maintained for a long time with redeposition because boron hydrides are fragile and easily dissociated by electron impact. Gross immigration could be avoided or minimized by an appropriate design in the geometry of the first walls. Detail studies with experiments and numerical analyses are necessary to confirm this point.

Thus the thin boron layer is attractive in long-time machines and reactors. It is worthwhile to have more detail studies on this possibility of the boron film aiming its application to the future machines.

Other materials such as beryllium, lithium,

silicon or some low/medium Z compounds should be investigated, too, from the viewpoint given in this paper, namely a protecting layer against deuterium/tritium and energetic charge-exchanged neutrals.

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Figure captions

Fig. 1. Hydrogen pressure as a function of liner temperature. All the implanted hydrogen atoms in a boron film are reemmitted below 400 C.

Fig. 2. ERD spectra for a boron coated stainless steel. One is obtained after the first thermal desorption procedure TD1, another after exposure to hydrogen glow discharge HD1. Left is plasma side and right substrate side. Hydrogen atoms implanted during discharge are stayed at the top surface of the plasma side. No hydrogen is seen inside the substrate.

Fig.3. The microstructure of the boron film obtained by an transmission electron microscope at a position near an edge of a wedge-shaped boron film. The film is thicker toward top. The microstructure shows that the boron film is not porous. Insert at the bottom-right is its diffraction pattern. Absence of diffraction spot in the pattern indicates that the film is amorphous.

Fig. 4. A first wall concept with a thin boron layer in long -time machines. Water-cooling channels are welded to the vacuum vessel. Panels are set on the channels and covered all inner surfaces of the vacuum vessel. Thin boron layers are coated on the top of the panels.

Fig. 5. Mass spectra with and without glow discharge with diborane. Almost all diborane is disintegrated by plasma.

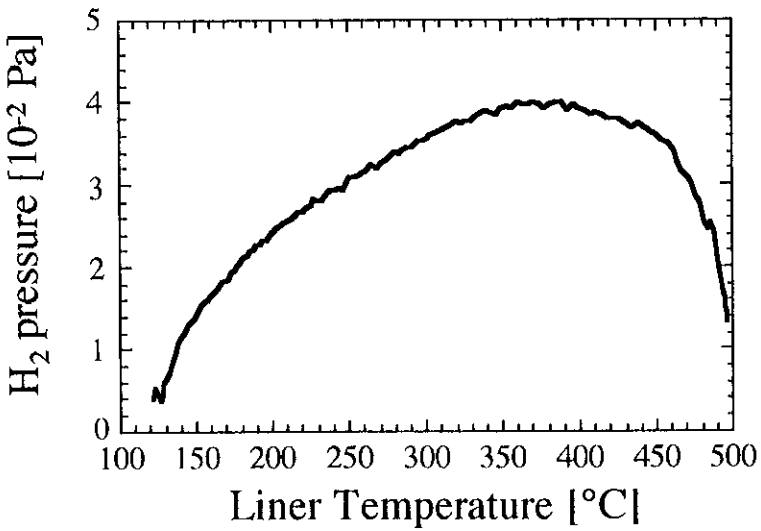
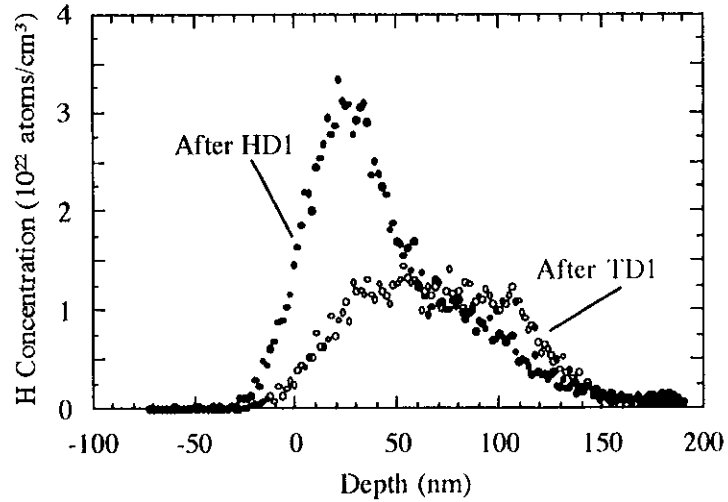


Fig. 1. Hydrogen pressure as a function of liner temperature. All the implanted hydrogen atoms in a boron film are reemmitted below 400 °C.



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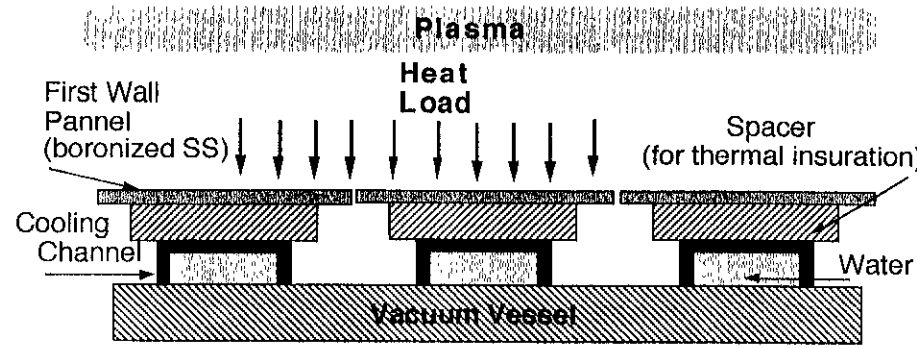

Fig. 4. A first wall concept with a thin boron layer in long time machines. Water-cooling channels are welded to the vacuum vessel. Panels are set on the channels and covered all inner surfaces of the vacuum vessel. Thin boron layers are coated on the top of the panels.

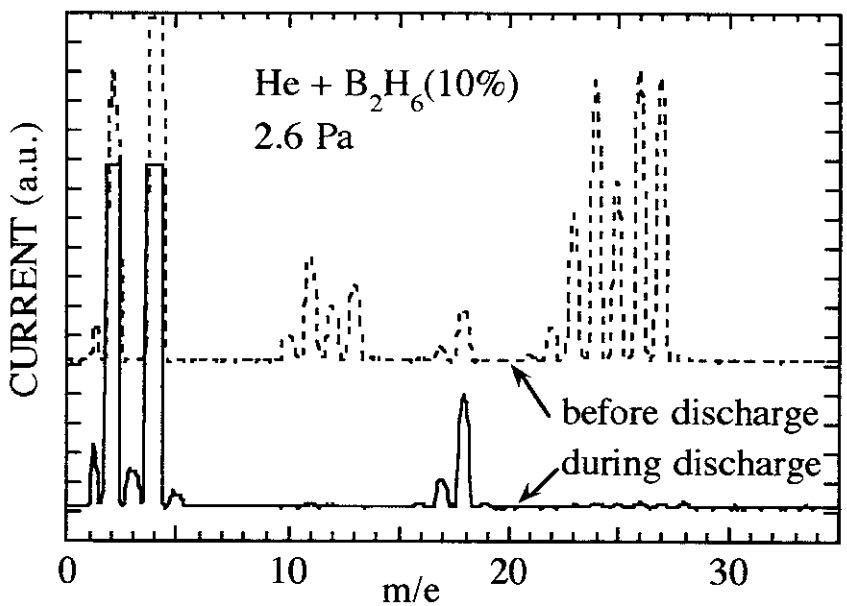

Fig. 5. Mass spectra with and without glow discharge. Almost all diborane is disintegrated by plasma.

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