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Boronization in future devices – protecting layer against tritium and energetic neutrals

N. Noda *, K. Tsuzuki, A. Sagara, N. Inoue, T. Muroga

National Institute for Fusion Science, 322-6 Oroshi-cho, Toki-shi 509-5292, Japan

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 long pulse operation. New experimental evidence is given for desorption of hydrogen isotopes from these films at relatively low temperature. Most hydrogen atoms in a boron-coated layer are re-emitted to the plasma side below 400°C without penetration into the substrate of stainless steel. The maintainability of a thin boron layer during a long pulse operation may be a problem. Boron atoms are hardly removed by pumping because their hydrides are easily disintegrated and redeposited. Gross migration of boron atoms inside the vessel is a concern. A condition required for avoiding the migration is discussed.

Keywords: Boronization; Long pulse machines; Tritium inventory; Permeation; Protecting layer; Damage by energetic neutrals; Redeposition

1. Introduction

It is widely recognized that boronization [1] plays an important role in many present fusion devices. However, its roles and availability are not seriously discussed for future devices with a long pulse steady state operation. The gettering function of oxygen and hydrogen 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 a 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

The roles of a fresh, thin boron film in 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 a widening of the operational space in the Hugill diagram, namely a higher density limit.

In DIII-D [2], impurity reduction was seen after boronization. The levels of O, C and Ni in the plasma were reduced significantly. It was reported that the 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], the recycling flux, measured as a function of line-averaged electron density, was found to be reduced after boronization. At the same time, the correlation between the recycling flux and the confinement improvement was investigated. The confinement enhancement factor decreased as recycling increased. These results indicate that reduction in recycling after boronization could be the cause of 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, DIII-D or JT-

* Corresponding author. Tel.: +81-572 58 2152; fax: 81-572 58 2618; e-mail: noda@lhd.nifs.ac.jp.

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60U, where large area of the plasma facing wall was covered by carbon. A confinement improvement was also observed after boronization in C-Mod. No clear change in recycling was found but a significant reduction in radiation was observed after boronization. The electron temperature at the edge was higher after boronization than before. It is suggested that the reduction in metallic impurities such as Mo caused a higher edge temperature, which triggered the better confinement.

In ASDEX-Upgrade, boronization was applied during the period of the tungsten divertor experiments in 1996 [6]. It is reported that boronization was necessary to enable a smooth start up to the experiment and to have a sufficiently wide operational space.

With this experience in several tokamaks, it appears that boronization gives better performances in most cases of the core plasmas. However, it cannot be generally concluded at present which of (i)–(iii) is the key for better core plasmas performance.

The roles of (i) and (ii) are regarded as the utilization of a wide area of the first wall for pumping oxygen or hydrogen. These are useless in a long discharge because the surface becomes saturated with them within a few hundred or a few thousand seconds [7]. These two roles should be replaced by active pumping of the limiter/divertor in future devices. The third role would be useful if the thin film could be maintained for a long time without the addition of a boron compound during operation. If metallic impurities are really responsible for the reduction in plasma performance, and if a way to maintain the boron film could be established, the role (iii) could be promising in future long pulse 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 pulse machines

In Section 2.1, it is suggested that role (iii) could possibly work in the future machines. Another possible role has been pointed out [9], that is, (iv) the thin film would help to reduce tritium inventory and permeation in DT machines. This is based on the experimental observation that hydrogen is completely removed from a boron film when heated at a surface temperature below 400°C [9], which is not expected for a carbon wall. In a long discharge, it would not be difficult to maintain the temperature of plasma-facing surfaces as high as 300°C–400°C by utilizing heat flux from the plasma. An expected temperature difference between the top surface and water-cooling channels can be less than 300°C, 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°C–400°C, most of tritium is released from the surface which results in low T inventory. Energetic T atoms from the plasma are blocked by the thin B-film and the tritium is re-emitted towards the plasma at this temperature. This point is discussed in more detail in Section 3 based on experimental data.

This kind of B-film also protects the wall from micro-damage by energetic particle impact. Concern about this type of damage is pointed out by Yoshida in ICFRM-8 [10] and in this conference [11,12]. During a long discharge, the fluence of energetic charge-exchange neutrals becomes enormous. During one day operation, it could be $10^{25}/m^2$ for hydrogen isotopes and 10^{21}–$10^{22}/m^2$ for helium at the first wall, that is, plasma facing part of blanket. These energetic particles produce damage such as dislocation, loops and blisters at the top surface of the plasma facing components. This 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. The threshold energy for damage production is below 250 eV, and the annealing temperature higher than 1500°C [11]. It should be seriously considered how to avoid this kind of damage. A thin boron film is expected to work as a protecting layer against the energetic charge-exchange particles. This is its fifth possible role (v), advantageous in a future long pulse machine.

The question is whether a thin boron film can be maintained for a sufficiently long time or not? This point is discussed in Section 5.

3. Experimental results in SUT

Basic properties at the National Institute of Fusion Science, Japan of boron films have been investigated in the SUT (SUrface modification Teststand) device [7,8,13–15]. The SUT device has a cylindrical liner inside a vacuum vessel, which can be heated up to 600°C with molybdenum heaters behind the liner. The total inner surface area 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 using a glow discharge with a helium/diborane gas mixture. 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°C. The typical thickness of the film measured by a quartz oscillator is 200 nm assuming that the film mass density is 1.5 g/cm^3.

As discussed in the previous section, one of the areas of interest is hydrogen isotope behavior in the boron film. The deposited boron film includes hydrogen which originates from the diborane. Most of this hydrogen was desorbed by raising the liner temperature to 500°C (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 by the boron film, measured by a drop in the

hydrogen pressure during the glow discharge using a residual gas analyzer. The total amount of hydrogen absorbed was $1–2 \times 10^{17}/cm^2$. A second thermal desorption (TD2) was then carried out. The total amount of desorbed hydrogen was roughly the same as that absorbed during HD1. Fig. 1 shows the typical hydrogen pressure as a function of the liner temperature during TD2. The temperature was ramped at 10°C/min. This indicates that most of the hydrogen in the boron film can be desorbed below 400°C. A number of experiments were performed varying the hydrogen discharge time. In some cases, a helium glow discharge was inserted between two successive hydrogen discharges in order to investigate the hydrogen desorption by the helium discharge. The re-emitted hydrogen amount was always comparable to that absorbed during the preceding hydrogen absorption discharge. Thus it can be concluded that most of the hydrogen isotopes can be re-emitted from a boron film below 400°C.

The results described above were obtained for pure boron films produced under good vacuum conditions. Oxygen contamination in the boron films was less than 5 at.%. In order to have more realistic conditions, hydrogen desorption was also investigated for fully oxidized boron films, where the surface oxygen concentration was around 40% [15]. Total hydrogen 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°C.

The 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 at Nagoya University. A helium ion beam of 1.5 MeV was utilized in this ERD analysis. Fig. 2 shows the results of the

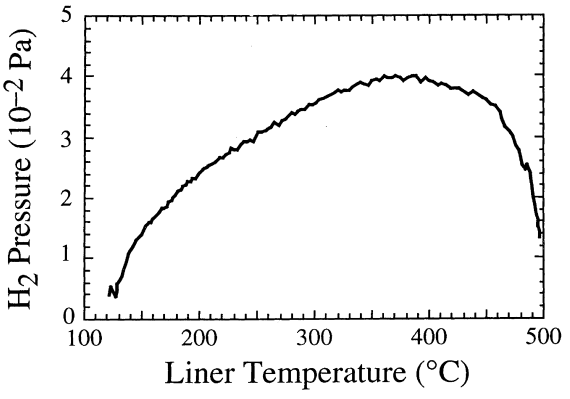

Fig. 1. Hydrogen pressure as a function of liner temperature. All the implanted hydrogen atoms in a boron film are re-emitted below 400°C.

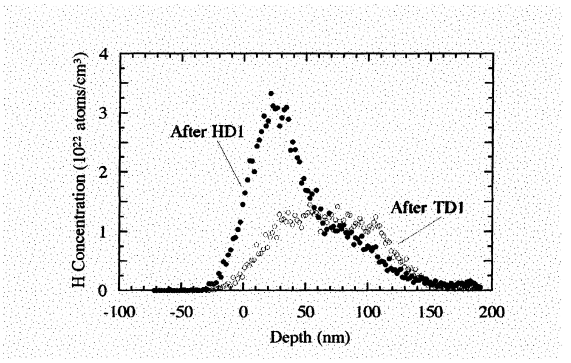

Fig. 2. ERD spectra for boron coated stainless steel. Spectra (a) was obtained after the first thermal desorption procedure TD1, spectra (b) after exposure to hydrogen glow discharge HD1. Left is the plasma side and right, substrate side. Hydrogen atoms implanted during discharge stays at the top surface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

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 with a relatively small concentration. After hydrogen glow discharge HD1, much higher concentration of hydrogen atoms is seen near the surface of the film. Due to the limited spatial resolution, the profile shown in the figure is broader than the real one. According to a numerical analysis, hydrogen atoms implanted are within 10 nm of 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 already been investigated for boron-coated small samples. Residual hydrogen inside a boron film was measured with infrared absorption [17]. A standard TDS (thermal desorption spectroscopy) was applied to a boron film [18]. Results in these experiments indicate that the main peak of hydrogen desorption lies around 400°C. The present experiment has confirmed this on a large scale for a boron-coated area of 7000 cm^2. The result with ERD gives experimental evidence that hydrogen atoms do not migrate into the substrate, which has not been clear in the previous experiments in Refs.[17,18].

One unresolved question is why hydrogen cannot penetrate into the substrate. One possible answer is that the film is porous, where volume recombination in the bulk is the dominant during thermal desorption like graphite. Hydrogen molecules are desorbed and move in the direction of either the surface side or substrate side through an open pore. The molecules which migrate to the surface side can escape the film, but those which

migrate to the backside cannot penetrate in to the substrate due to the low permeability of the molecule. In order to check this idea, micro-structures were examined with a transmission electron microscope (TEM). A boron-coated stainless steel disc was back-thinned to perforation by ion milling. Fig. 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. The absence of a diffraction spot in the pattern indicates that the film is amorphous. Relatively high concentrations of oxygen and carbon are seen near the boundary between the coated layer and the substrate with an in situ Auger analysis [7]. An 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 seem to have formed around the boundary. The layer with these compounds may possibly work as a barrier for hydrogen isotope penetration. Another possibility is that the permeability into the substrate is determined by the difference in chemical potentials between the coated layer and the substrate, or the elements in the substrate. In this case, the permeability may not always be small as for stainless steel but will depend on the substrate material. It is worth noting in this context that a thermal desorption experiment was once carried out for a boron film coated on a graphite liner in SUT. The hydrogen behavior in the TD1, HD1 and TD2 procedures was essentially the same as that with the stainless steel liner. This suggests that hydrogen did also not penetrate into the carbon substrate below 400°C. Further detailed investigation and analysis is interesting and necessary to obtain a more conclusive understanding on this subject.

Combining the ERD results with the fact that all the implanted hydrogen atoms during HD1 are re-emitted in the thermal desorption TD2 as described in the first half of this section, one can get an idea of a boron film as a deuterium/tritium free layer. The concept of the boron-coated first wall is given in Section 5.

Fig. 3. The micro-structure 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 of the picture. The microstructure shows that the boron film is not porous. Insert at the bottom-right is its diffraction pattern. Absence of diffraction spots in the pattern indicates that the film is amorphous.

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

It is essential to keep the temperature of the boron film as high as 300°C–400°C in order to get a hydrogen free surface. This can be realized as shown in Fig. 4. Water-cooling channels are welded directly to the vacuum vessel. Panels are attached to the channels to cover 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 onto the top of the panels. During long pulse operation, heat flux from the plasma keeps the top surface of the panel at a temperature high enough to desorb the hydrogen atom if appropriate thermal insulation is inserted between the panels and the cooling channels. A wide variety of materials can be used as panels. For instance, stainless steel panels can be used in non-DT machines. The temperature gradient between the top panel surfaces and the cooling channels is less than 300°C, which gives no serious thermo-mechanical problems to the first wall system. This would not be the case if carbon layers are utilized. In order to get a hydrogen free surface, the top surface must be kept as high as 700°C–800°C, making it more difficult to design the first wall system. This type of the first wall with the boron film is actually proposed for the Large Helical System (LHD). Cooling channels have already been welded to the vacuum vessel of LHD. Panels of stainless steel will be installed in the near future. One of

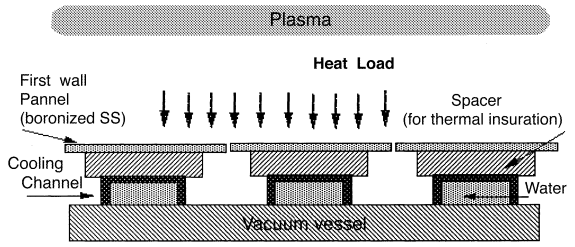

Fig. 4. A first wall concept with a thin boron layer in long pulse machines. Water-cooling channels are welded to the vacuum vessel. Panels are attached to the channels and cover all inner surfaces of the vacuum vessel. Thin boron layers are coated on the top of the panels.

the problems of a superconducting-coil machine is that a conventional helium glow discharge cannot be applied between two successive high-power discharges. The helium discharge has been necessary in D III-D with a boronized wall in order to access a good confinement VH 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 for the plasma-facing parts of the blanket. The blanket must be operated with a temperature higher than several hundred degree C to allow the coolant temperature to 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 the plasma facing surfaces and the blanket. 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 and easily dissociated and tend to be re-deposited inside the vessel, not reaching the pumping ducts. Thus the total number of boron atoms in the vessel does not decrease even during a long discharge. In SUT, a hydrogen discharge as long as 10 h has been applied. No indication has been found for the boron film being lost during this period. A carbon film can be removed using a hydrogen discharge by the formation of methane, as methane is relatively stable. This was confirmed with an experiment in TEXTOR with a carbonized wall [19]. Even with a methane-seeded discharge, the pressure drop of methane at the start of a discharge is as small as 20% of the initial pressure. In contrast to methane, the diborane pressure drop is more than 90% of its initial value 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 migration of boron atoms from erosion-dominated area to deposition-dominated area. One of the asymmetries is seen between the divertor region and other area of the vessel. In steady state, the total boron flux Γ_{Binw} sputtered from the wall and from the divertor Γ_{Bind} is balanced with a

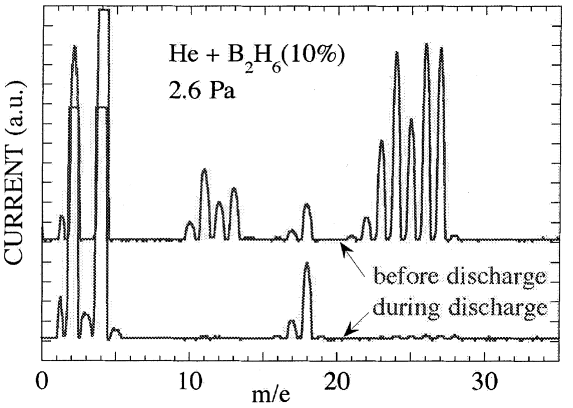

Fig. 5. Mass spectra with and without glow discharge with diborane. Almost all diborane is disintegrated by plasma.

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 by the following equation:

$$(\Gamma_{\mathrm{Binw}} - \Gamma_{\mathrm{Boutw}})S_{\mathrm{w}} + (\Gamma_{\mathrm{Bind}} - \Gamma_{\mathrm{Boutd}})S_{\mathrm{d}} = 0, \quad (1)$$

where S_{w} is the apparent area of the first wall and S_{d} the cross sectional area of the divertor entrance. In general, net migration of boron atoms can occur from the wall region to the divertor region or vice versa. There is one more free parameter, that is, the distance δ between the last closed surface and the first wall. The flux Γ_{Boutw} is proportional to $\exp(-\delta/\lambda)$, where λ is the decay length in the SOL. On the other hand, Γ_{Binw} is independent of δ because it is mainly determined by the charge exchange neutral flux. Thus the first term could be zero if the distance δ was chosen appropriately. The fluxes Γ_{Binw} and Γ_{Boutw} are dependent on operational parameters such as the averaged plasma density, and they are not linearly correlated to each other. Thus a unique operation parameter set could give a condition in which the net migration is zero for a fixed δ. In an experimental device, a wide operation area is necessary. Then gross migration in each operation regime will occur. In a reactor, a limited number of operation schemes would be selected, which enables a choice of the optimum distance for migration.

Discussion in the previous paragraph is based on the assumption that the deposition process is dominated by boron ions moving across a magnetic field. If an isotropic velocity distribution is assumed for the dissociated neutral fragments, roughly half of them moves toward the plasma, suggesting that this process plays a certain role. But another half can directly reach the first wall and be re-deposited 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 1–4×10^{-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 one assumes the cross section as high as 1×10^{-15} cm^2, the mean free path at the electron density of 1×10^{12}cm^{-3} is a few millimeters. This is very small compared to the scale of the reactor. Thus boron atoms hardly move from one place to another during discharge.

Thus the gross migration is expected to be small in a future machine like a reactor. It might be necessary to make an additional in situ coating for erosion dominated parts of the wall during maintenance periods. 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 data which should be accumulated from now on.

Outer striking point of a single null divertor is an erosion-dominated region. In Alcator C-Mod, the 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 locations 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.

The removability of a boron film after boronization is discussed in Ref. [17]. This paper concluded that it was possible to remove a boron film. The conclusions in Ref. [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 pulse machines.

6. Conclusions

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

The functions of oxygen and recycling reduction are not available in future long pulse machines. The third function is available when thin boron film can be maintained during long operations.

A boron film works as a hydrogen-isotope free wall at 300–400°C. This effect can be kept by heat flux from the plasma or from the blanket in long pulse machines. It gives a protecting layer against energetic charge exchange neutrals from the plasmas. The layer reduces tritium inventory at the wall, tritium permeation inside the first wall, and micro-damage at plasma facing surfaces due to the energetic D, T and He atoms' impacts.

It was confirmed experimentally that most of hydrogen isotope atoms are re-emitted from a boron film at a temperature below 400°C and 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 by re-deposition because boron hydrides are fragile and easily dissociated by electron impact. The re-deposition processes are discussed with a simple model aiming to describe some of the major processes for the maintainability, without detailed geometry and plasma operating conditions. Gross migration could be avoided or minimized by an appropriate design in the geometry of the first walls. Detailed studies with experiments and numerical analyses are necessary to confirm this point.

Thus the thin boron layer is attractive in long pulse machines and reactors. It is worthwhile to have more detail studies of the boron films aiming at their application to the future machines.

Other materials such as beryllium, lithium, silicon or some low/medium Z compounds should also be investigated, from the view point given in this paper, namely as a protecting layer against deuterium/tritium and energetic charge-exchange neutrals.

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