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Review of High Z Materials for PSI Applications

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Abstract

Application of carbon based low Z materials to PFM has significantly improved plasma parameters in large tokamaks. There are, however, serious concerns of erosion, neutron damage etc. for application of low Z materials in future D-T burning machine. To apply high Z metals to PFM, there are several issues to be solved; high Z impurity production by sputtering, their accumulation in plasma center, and high radiation loss. Because of these concerns high Z metals are not widely employed nor planned to be used in the present large tokamaks. Since our efforts have been concentrated to optimize the low Z materials, little systematic investigations for high Z materials in tokamak have been done, lacking data base especially those concerning the impacts on plasma core.

In order to employ high Z material as PFM near future, material properties related to impurity production and hydrogen recycling are reviewed and discussed what is important and what shall be done.

[Key words: Divertor, Graphite, Hydrogen recycling, High Z, Impurities, Low Z, Molybdenum, Review, Sputtering, Tungsten]
1. Introduction

In these 20 years, core-plasma parameters have been greatly improved in tokamaks and achieved those comparable to a fusion reactor. It is well known that wall materials have greatly influenced the performance of a magnetically confined plasma in each experimental device. Particularly, adoption of low Z materials, such as carbon, boron and beryllium have led to a great success in the present large tokamaks[1]. These experiences and the achievement have been quite important to make a great contribution to the fusion-research history. Nevertheless it is quite clear that there still remain a lot of questions and works to establish the fusion reactor.

Among all following two are most difficult to be solved in the next step of the fusion research; how to maintain the high temperature plasmas in a steady state, and how long are life times of various structures including the plasma facing components(PFC) against neutron irradiation originated by D-T burning. Up to now, we have been concentrating our efforts to optimize the carbon based low Z materials from a viewpoint to maximize the core plasma parameters. This is of course on the line along the historical requirement for the fusion development.

On the other hand, systematic investigations on high Z materials have been lacking, which results in very poor data base for them, especially those on the impacts to the plasma core. In ITER design, graphite based low Z material is recommended to be used as the divertor plates and first wall protection for the initial operation phase in order to minimize the risk of plasma contamination. At the same time, however, it is envisaged that the development and testing of plasma facing components including high Z materials should be carried out during the physics phase [2]. In SSTR (Steady State Tokamak Reactor)[3], in which engineering feasibility is a top priority, a high Z material, such as Mo, is the primary candidate of the armor tiles because high erosion rate[4] and loss of thermal conductivity[5,6] of the carbon based material enforce the frequent replacement of the armor, which is very hard from engineering aspect[3,7].

Of course recent advances of carbon based material such as C/C composite which has very high thermal conductivity and high thermal shock resistance, is very
promising to reduce surface temperature, but do not help the degradation of thermal conductivity due to neutron irradiation [8,9]. Safety problem of Be and its low melting point can not be avoided, too [10]. Thus we cannot put 100% reliance on low Z materials for a future machine, even though systematic investigations on high Z materials have been lacking, which results in very poor data base for them, especially those on the impacts to the plasma core.

From material aspect, poor ductility and small thermal shock resistance used to be a problem of most of the refractory metals. Recent advances in production and purification techniques of the refractory metals have lowered ductile brittle transition temperature (DBTT) of molybdenum (Mo), for instance, far below room temperature [11,12]. This means that it is now possible to machine Mo at room temperature easily and high Z is becoming within engineering feasibility for PFC.

Thus, facing to the new phase of the research history, we should open our eyes to broader possibilities than to those in these 10 years, including both low Z and high Z materials as PFM. This is the basic idea of this review, in which questions and prospects of the high Z materials are summarized as widely as possible.

Several nice reviews which partly discuss high Z impurity behavior and problems have been published [1,13-15]. A lot of indication and suggestions from these reviews are reflected in the present paper.

2. Experiments on high Z materials in tokamaks

Firstly, let us review what have been the real problem for high Z materials to get a good parameter of the plasma core in fusion devices.

One of the main problem of high Z material is impact of radiation loss on confined plasma. In order to satisfy the burning condition, it has been reported [16] that the concentration of high Z ions at the plasma axis must be below 10^{-4}. It should be noted that this is a calculated prediction and there are not much experimental data which indicate quantitatively the concentration limit required. The central density of impurity ions depends on their generation rate at the surface of plasma facing component, shielding effects by edge plasma, inward flow(degree of accumulation)
inside the plasma core etc. It may be necessary to get a comprehensive understanding on each process to reach a solution. From a practical point of view, the final goal of the studies on high Z impurities is to know the threshold limit value of high Z ion concentration above which D-T burning could not be sustained, and to verify operational conditions to guarantee impurity concentration below this limit.

2-1. Accumulation of high Z impurities in good confinement schemes

One of the severest problems with the high Z impurities is their accumulation in a core plasma in a good confinement scheme. In 1982, a good confinement scheme called "H-mode" was found in ASDEX[17]. This was firstly obtained with titanium divertor plates and stainless steel wall[17,18] This means there is basically no difficulty to access the H-mode with medium Z or high Z wall. However, the accumulation of metal impurities in the plasma occurs during the quiescent H-phase, which leads to the termination of the H-Mode[19]. In Fig. 1 time traces of several plasma parameters are plotted for an H-mode discharge in JFT-2M[20], where most of the wall and divertor plates are covered by titanium (Ti) because of the repetitive application of Ti gettering. One can clearly see continuous increase of the radiation of Ti in the H-mode, which finally results in returning to the L-mode. In JET, it was reported that nickel impurities are confined longer time in the H-mode than in the L-mode (see Fig.2)[21].

The accumulation or longer confinement of impurities has been observed also in other good confinement scheme, such as counter injection of neutral beam (NB) heating in ISX-B[23], pellet fueling, and improved ohmic confinement(IOC)[22]. All these examples suggest that the good energy-confinement scheme is always accompanied by impurity accumulation with longer confinement time, which is the severest constraint for the application of high Z materials to PFC.

It should be, however, stressed that this problem is irrespective to the Z number of impurities. It has not yet been clear how low Z impurities including helium (He) generated by D-T burning influence on sustaining a good confinement scheme. In TEXTOR, it has been shown that He ions are confined much longer in the H-mode than in the L-mode[24]. At present the maximum concentration of He tolerable for continuing D-T burning in a steady state H-mode is not so clear. Thus the fuel
dilution by He and low Z impurity problem are parallel questions to the high Z impurity accumulation in good confinement schemes and should be solved simultaneously.

Many efforts have been paid to get the steady state H-mode. In DIII-D, 10 sec. H-mode has been achieved by careful control of edge localized mode (ELM) instability[25]. In JFT-2M, field ergodization at the edge of the confinement region has prevented the impurity accumulation, giving a quasi-steady state of the H-Mode[26].

Most of these efforts presently carried out are under low Z PFM conditions and a solution with low Z may not directly applicable to the high Z. Still big question has remained unsolved, that is, how plasma confinement would be influenced if PFM was completely changed from low Z to high Z. Wagner et al.[27] clearly showed the difference in the confinement time between SS and B of divertor walls in ASDEX "H-mode" giving better confinement for B. But such comparison is very difficult because the change of the material automatically introduces different impurities and SS often works as a source of low Z impurities such as C and O as discussed in sec3-2. Therefore the comparison must be done well characterized or diagnosed condition.

Thus the questions are summarized as (1) whether it is possible or not to get a steady state in a good confinement scheme with high Z PFM, and if possible, then (2) whether the high Z impurity concentration can be well below the allowable limit in the steady state condition and (3) how plasma parameters would be modified if PFM was completely changed from low Z to high Z.

2-2. Steady state concentration of high Z impurities in ohmic and "L-mode" discharges.

The absolute density or concentration of high Z impurities in the steady state L-mode discharges is a separate question from the impurity accumulation in the good confinement scheme.

It is well known that metal-impurity concentration strongly depends on plasma density, plasma current, neutral beam (NB) power, working gas of discharge and
divertor/limiter configurations. In table 1 are summarized the measured concentrations of metal impurities in the central region of plasmas. Most of the data are for the ohmic discharge and scarce for the NB heated discharge. Figure 3 shows a typical example obtained in DIII-D[28]. Nickel radiation decreases as the density rises and that for the open-divertor discharge is almost one order of magnitude lower than that for the limiter discharge. The density dependence is attributed to the edge temperature[29,30]. It is particularly important to investigate the correlation between measured concentration and edge temperature. However, it is very hard to find a report on simultaneous measurement of these quantities. If it would be allowed to compare edge temperature data by Erents et al.[31], we could put $T_e = 120$ eV for JET case in the table 1. But there is no means to guess the edge temperature for other cases.

One extreme case is included in the table, that is, the lower density discharge in Alcator C[32]. The Mo density is extremely high even with the electron density as high as $7 \times 10^{19}$ m$^{-3}$, of which reason is not clear. In this particular case, electron temperature showed hollow profile because of the strong radiation loss at the plasma center[33].

Such hollow temperature profiles were observed in ORMAK (tungsten (W) limiter)[34], PLT(W and stainless steel (SS) limiter) [35,36] and DITE(Mo limiter)[37]. This has been one of the most important reason for excluding the high Z as PFM. However, one should note that the hollow profile could be avoided by a little stronger gas puffing after Ti getting[36]. It is also noteworthy that the hollow profiles are observed mostly in low density discharges. Figure 4 shows two profiles of electron temperature in PLT with SS limiter[36]. The hollow profile (Fig.4(a)) is without current contraction during start up, while the central-peaked profile (Fig.4(b)) with contraction which could be provide by special preprogramming of gas puffing.

There is no doubt that such a high radiation loss must be avoided in future machine. All example above introduced show that the hollow profiles could be avoided by adopting higher density operations. However, it is again not clear what is the critical condition, or key parameter to avoid such a high concentration of metals.
A role of light impurities is another important issue of metal impurity origin. One of the experience was reported in JFT-2[38], in which Mo radiation decreased one order of magnitude correlating with oxygen reduction due to application of titanium gettering. This suggests that oxygen play a dominant role of the Mo release at the limiter surface. Pospieszczyk et al.[30] have indicated large contribution of C, O and/or metal to the sputtering yields of Cr and Fe, observed in TEXTOR. Since the sputtering yields by the impact of oxygen or carbon is much larger, and the threshold energies lower than those by deuterium, these low Z impurities should have large contribution to the production of the metallic impurities. In ohmic discharge of JET, Ni influx from graphite tile to the plasma has been attributed to sputtering origin, though it is not clear which species among deuterium, carbon and oxygen is the main cause.[39]

Until now diagnostics for PSI have, unfortunately, not been enough and sometimes impurity production due to pyrolytic origin like a gaseous desorption or material sublimation, which can be avoided by reducing areal heat load with a technique such like divertor swing[40], seems to be mixed-up with that due to physical sputtering. The origin of carbon bloom is not finally identified due to either radiation enhanced sputtering or thermal sublimation[41]. Thus the origin of high Z impurity must be more carefully examined. Anyhow reduction of the light impurities is quite important not only because of their direct effect on burning plasmas through dilution, but also on sputtering of high Z impurities.

2-3. Metal impurities during ICRF heating

Although ICRF heating is successfully applied to present tokamaks, impurity contamination has been one of the severe problems of this type of heating. This is partly because an antenna is required to be close to plasma edge in order to get a good coupling of RF power to plasmas. As is well summarized in section 8.2 of ref.[1], there are experimental evidences that a part of RF power directly coupled with scrape off plasmas and e-folding length in particle flux is broaden by RF power with metal or metal contaminated limiters. These are probably reasons for stronger interaction between plasma and antenna/limiters. It is noteworthy that, in contrast to NB heating, metal impurity did actually limit the plasma stability and parameters
even in L-mode. A typical example can be seen in JIPP-T-IIU[42], where adoption of carbon protectors and limiters was not the solution to prevent metal impurity contamination during ICRF heating with 1MW level. In spite of a good coupling of RF power to the plasma electrons, high Te conditions could not be maintained for several tens milli-second and the plasma was cooled down by the strong radiation loss. This problem could be solved only by carbonization[43].

Additional efforts might be needed to apply high Z PFM with ICRF heating scheme compared to other heating techniques.

2-4. Experience in JT-60 with TiC-coated Mo

In the early phase of JT-60, TiC coated molybdenum(TiC/Mo) was adopted as limiters and divertors, and TiC coated inconel 625 as first wall. Mo was selected after various engineering R&D work[44]. The coating was manufactured by CVD and PVD techniques [45] with thickness of 20 mm. In spite of this coating, Mo burst was often observed, especially in limiter operations both with hydrogen discharge and helium one, and divertor operations with helium. This is found to be originated from extraordinary high heat flux concentrated to the edges of several TiC/Mo tiles. This problem was solved by X-point sweeping for divertor discharges, and more than 20 MW of NB power was injected without the Mo burst. Figure 5 shows a typical time behavior of radiation powers with and without X-point swing.[46]

Once the burst was suppressed, one of the advantages of the TiC/Mo wall was that the total impurity concentration was quite low and values of Z\text{eff} are lower than those for carbon wall adopted later in JT-60 as shown in Fig.6 [47,48]. This is because of lower contamination of oxygen and carbon impurities from TiC surfaces, which could be interpreted as a results of stable surface composition with titanium carbide and oxide.

These experiences show that TiC/Mo and/or pure Mo is possibly available in future if excessive heat flow can be avoided and heat flux is removed by an appropriate active cooling. They also suggest that it provides a possibility of smaller contamination with low Z impurities. It should be noted here that
extensive effort was paid to develop this special coating. Some important results are seen in ref.[49-52]

2-5. Self-sputter runaway

It has been pointed out that self sputtering yield of high Z metals normally exceeds unity in energies above several hundreds eV[54,55]. This suggests that if the edge temperature is high enough, an atom, generated at the surface of a limiter or divertor plate, after ionized in the plasma, comes back to its surface and sputters again atoms with a possibility above 1, which results in impurity generation in a avalanche for a while, until it excessively cools down the plasma. Since a certain part of emitted impurity atoms is lost by simply geometric reasons or by shielding effects, this phenomena has not been identified in plasma confinement devices. If this could really occurs, it would be a catastrophic and plasma could not be stable. It is an important question whether and/or at what conditions this phenomenon really occur. Sengoku et al.[53] discussed this phenomena and pointed out that divertor operation possibly remove this problem in JT-60, even in an operation scheme with low density and high edge temperature.

These are a summary of experience and what we could learn from, or what we could not understand from them. In the following section material properties related to impurity production and hydrogen recycling are reviewed in order to employ high Z materials as PFM near future and discussed what is important and what shall be done.

3. Impurity Production and Erosion

3-1. Physical sputtering

Figure 7 shows sputtering yields of Mo and W by various ions[54]. Although higher threshold energy for sputtering of high Z than that for low Z is a large advantage, the self sputtering yield which exceed unity at around 1 keV is a serious concern[55]. In addition, most of outcoming particles from the scrape off plasma are accelerated with the sheath potential and hit the first wall or divertor.
plate with very small angle, enhancing the sputtering yield appreciably[56]. Therefore temperature of scrape off plasma must be well below to keep the self sputtering less than one. However no impurity accumulation by the run-away sputtering has been reported yet. In stead, it should be noted that in the present tokamaks several percent of low Z impurities (carbon and oxygen) are still included which are very likely the source of other medium and high Z impurities. Because the low Z impurities in large tokamaks are highly ionized, they are accelerated up to a few hundreds eV by the sheath potential even the ion energy in the scrape-off layer is below 50eV, giving very high sputtering yield. The effect of sputtering by the low Z impurity is clearly demonstrated by Vojtsyana and Cohen [57] and the high Z is very likely to withstand pure hydrogen plasma. Therefore low Z impurity level as well as the ion energy is important criteria for applying the high Z material.

Nevertheless high Z impurity should be approved in the large tokamaks diagnosed much better than earlier small tokamaks, examining erosion rate, method for reduction, radiation cooling and so on.

3-2. Chemical erosion

Fig. 8 shows chemical stability of various metals in oxidative atmosphere[58,59]. One can clearly see why Be and B, both of which oxides are much more stable compared to H₂O and not reduced by hydrogen atmosphere, work as oxygen-getters in the present tokamaks. Although volatile oxides of Mo and W, MoO₃ and WO₃, are considered to be important impurity sources[61,61], they are not likely produced in hydrogen plasma, because Mo and W are rather noble and can be easily reduced by hydrogen as seen Fig.8. In this respect, chemical sputtering owing to the volatile oxide formation[60,61] may not happen in hydrogen plasma even if it has several % of impurity oxygen. On the contrary, their carbides are much stable than the oxides at elevated temperatures and do not seem to allow the volatilization of pure oxides as already pointed out by Migge[62]. Fresh surface of Mo is reported to have a high gettering action for methane[63].

As discussed later, however, powder metallurgy to produce refractory metals often added a fair amount of gaseous impurities which made outgassing rate of the
metals produced by this technique large[65]. Recent advance in an electron beam melting technique[64] has made possible to produce highly purified metals (electron beam melted Mo : EB-Mo) and the outgassing rate of the purified EB-Mo is found less than half of powder metallurgical one(PM-Mo)[65]. One can clearly see the difference on the surface of the EB-Mo and PM-Mo after NBI heat load test due to impurity gas (see Fig. 15-1(a)) [66]. Unfortunately the EB-Mo was not available for earlier tokamaks at that time and the PM-Mo they employed might have low Z impurities as a gaseous source.

From Fig. 8 one can understand rather complex behavior of stainless steel which often works as a source of impurity oxygen as noted "oxygen recycling"[67], that is, oxygen initially at stainless steel surface with a chemical form of mainly chromium oxide or surface adsorbed species is reduced by impinging hydrogen from plasma forming water molecules[67,68] which immediately dissociate to hydrogen and oxygen, while it comes back to the wall after plasma termination.

As described above chemical sputtering of Mo and W by impurity oxygen in plasma is not likely to occur unless the oxygen amount is fairly large. Nevertheless physical sputtering by oxygen (as well as carbon) is not small compared to that of hydrogen isotopes and this is the most possible source of high Z (not only high Z) impurities in the earlier plasma machines as discussed above. Again we should mention that the amount of oxygen and carbon impurities is one of the most important criteria for PFM application of the high Z materials.

4. Hydrogen Recycling

4-1. Reflection

Figure 9 shows energy distribution of reflected atoms and ions when hydrogen ion of 18keV were impinging to Ta target[69]. One can see that the reflected particles show wide energy distribution from maximum energy given by elastic collision with target atoms to zero energy. Fraction of charged particle is quite dependent on the surfaces cleaness of the target as well as incident energy but usually not exceed several percent of neutrals[70].
Basically the reflection is directed by a collision process and therefore dependent on the mass of the target[71,72] and not on the chemical nature. Because of their heavier mass, reflection coefficients of hydrogen for high Z metals are much larger than those for low Z materials[71,72]. This, in turn, indicates energy reflection by high Z metals is also larger because all reflected particles have certain amount of energy as shown in Fig.9[69]. Figure 10 compares the particle and energy reflection coefficients of carbon and W.[72] It is important to note that the reflection coefficient increases with the decrease of the incident energy and below 100eV most of the impinging energy to W is reflected. Unfortunately, owing to the difficulty of the reflection experiment, experimental data for low energies have not been available until now, giving large uncertainty[71,72]. To confirm the extrapolation from high energy, the experiment is urgently needed. Nonetheless one can see both energy and particle coefficients for W are roughly one order of magnitude larger than those for graphite.

The reflection of high energy electron by high Z material is also larger than that by graphite because it is simply the collision process. Therefore one can expect much reduction of heat load to high Z divertor plates than low Z ones if compared under the same heat and particle load condition. This must be an advantage of the high Z for PFM but not confirmed, for all of the present large machines are working with the carbon based armor and the heat load of the earlier machine was not so high to examine the effect of the heat load.

In addition to these reflected particles, thermalized molecules, which originate from once-implanted ions diffusing back to the surface to recombine, are emitted(reemission)[74]. Different from the reflected particles, the reemission of the thermalized molecules are time dependent. It increases with the increase of implanted fluence from zero at the start to saturation at the steady state where the reflection plus reemission become equivalent to the incident flux and the target is saturated with hydrogen. As depicted from Fig. 10, when the incident energy exceeds a few hundreds eV, the reemission exceeds the reflection and becomes the dominant source for hydrogen recycling, which is discussed in the following section.
4-2. Hydrogen in high Z metals (Reemission, Desorption, Diffusion and Trapping)

Concerning hydrogen behavior[75], high Z metals are divided into two groups, those are, exothermic hydrogen occluders(hydride former) of Nb and Ta, and endothermic hydrogen occluders of Mo and W. Because of their large hydrogen solubility, the former are not considered as PFM but owing to its outstanding thermomechanical properties Nb are considered to be a divertor structure material. [76,77]. The latter group shows a very small hydrogen solubility and hence hydrogen diffusion seems to be influenced by the defect trapping[74,78,79]. These are probably originating from bcc nature like iron in which appreciable trapping effect by defects on hydrogen diffusion below 500K has been observed[80].

Temperature dependence of hydrogen diffusion coefficient and solubility in Mo and W are given in Fig.11 and Fig.12, respectively [74]. One can note very large data scattering in diffusion coefficient of Mo probably owing to the trapping effect. Because most of high Z metals were produced by powder metallurgy( PM), the powder metallurgical Mo (PM-Mo), in nature, included large amount of impurities and defects which work as hydrogen trapping sites. The largest diffusion coefficient given by Katsu et al. [81] is determined with using electron beam melted Mo (EB-Mo). And recent measurement with EB-Mo supports higher values[82].

Although the data for W are not so widely scattered, both were unfortunately rather old and there is a possibility to be influenced by the trapping suggested by the large activation energy as pointed out by Katsu[81]. The experiment for EB-W must be done to confirm the old data. Compared to the diffusion data, agreement of solubility data is very good.

Influence of hydrogen trapping on hydrogen reemission from Mo was first investigated by McCracken et al.[83] and confirmed by Tanabe et al. [78]. The trapping effect in W has been also noted in the ion driven permeation studies by Anderl et al.[79] The trapping effect, however, is not serious at elevated temperatures because not only trapping energy is less than 1 eV[84] but also trapping amount is negligibly small compared to hydride formation. Recombination coefficients so far reported are not enough and no recommendation has been done.
because they are so much influenced by surface state[74,84].

4-3. Hydrogen recycling

Apart from above basic data for hydrogen, it is important to consider global reemission behavior. Figure 13 shows schematic diagram for hydrogen reemission from a solid material[85]. Here only thermalized molecules are taken into account, because the reflection is hardly influenced by implanted fluence or time.

If there were no changes in diffusion, solution and recombination processes in the target material, the reemission decay just after the beam off (1- J(t)) should be the same function to the initial increase of the reemission (J(t)) and all of the retained hydrogen during the beam-on (dynamic retention) should be reemitted (see the solid lines in Fig.13(a)). In actual case, however, the processes are modified by energy deposition by impinging hydrogen as well as irreversible hydrogen trapping or hydride formation. Therefore not all of the hydrogen retained under implantation is reemitted after the beam off (see the dotted lines in Fig.13(a)), retaining certain amount of hydrogen in the target which depend on material itself, impinging energy and target temperature. Changes of recycling and retention with temperature are schematically shown in Fig.13(b). One can see how reemission behavior will be modified when flux and hence temperature are changed.

The retained hydrogen is released by thermal desorption(see Fig.14) and desorption peak temperature of Mo[78] is a little higher than those for SS[86] and Ni [86], indicating larger trapping energy [84]. The total hydrogen retention in Mo, however, is much smaller than that in graphite of which saturated hydrogen concentration is as high as 0.3 in H/C ratio even at 700K[88,89]. This in turn forced the graphite in the present tokamak to be subjected to discharge cleaning or other cleaning technique to assure wall pumping[90,91]. Once cleaned the pumping effect continues to a certain number of shots until the graphite wall is saturated with hydrogen[92]. For long pulse operation where the graphite wall must be saturated, density control of the plasma becomes difficult owing to the huge amount of hydrogen evolution from the graphite. No operational scenario for such high hydrogen recycling regime, where problem comes up from change of output power
and/or thermal fractuation and also plasma termination, has been established yet. Mo and W retain much less amount of hydrogen and a saturation should be attained much lower fluences than the graphite. Above 800K, as depicted from thermal desorption spectra (Fig.14), most of the retained hydrogen is released immediately after the beam off[78]. This indicates that one can expect only limited wall pumping by Mo and W and plasma must be operated at high recycling regime even for a short pulse. However, since the dynamic retention is much smaller, density control may be easier than the graphite.

Hydrogen recycling under high recycling regime is a really important issue for next coming long pulse machine irrespective of wall materials. From this respect, it may not be suitable to compare graphite and W(Mo) as PFM directly, because both reemission and reflection are quite different between them. Instead one can compare Nb or Ta with Mo or W, because hydrogen reemission in the former is similar to graphite as seen in the earlier work [93], while the reflection must not be so different due to similar Z number. Even though Nb and Ta must not be used for the armor in a fusion reactor, such kind of experiment is quite necessary not only for more comprehensive understanding of plasma surface interaction but also for designing future machine.

5. High Heat Load Test and Material Behavior

Under research and development of PFC for ITER, high heat load test of high Z metals has been extensively conducted[94-98]. Whitely et al.[15] gave an excellent review for application of high Z to divertor armor. Since most of the high Z metals are brittle having poor weldability and hence lacking in thermal shock resistance.[11,99,100], many efforts have been done to improve their ductility by alloying and/or purification[11,12,100] and TZM is one of the successful example. Furthermore neutron irradiation increases the ductile brittle transition temperature(DBTT) and earlier data shows neutron irradiation of about 1dpa rose DBTT of Mo over 500K.[101]

Recently single crystalline of Mo and W produced by either sintering[102] or electron beam melting[66] is proposed to be utilized as PFM. The DBTT of the electron beam melted Mo(EB-Mo) is far below room temperature and large block of single crystalline is now available[64]. From the engineering aspect, the
application of the monoblock to PFC with a hole as a cooling channel seems very nice, for blazing or adhesion to the cooling base is not necessary.

High heat load test indicated that the single crystalline EB-Mo showed good performance without appreciable cracks and recrystallization, whereas polycrystalline PM-Mo showed severe recrystallization and intergranular cracks[66] as seen in Fig. 15(b). It should be mentioned that the erosion of PM-Mo by high heat load is four times larger than EB-Mo[103], presumably owing to particle emission from PM-Mo which originates from cracking of boundary of the recrystallized columnar grains as seen in Fig.15(b). Similar cracking is seen in the heavily damaged divertor plates of TiC/Mo in JT-60[104,105]. In recent our high heat load test[66], single crystalline EB-W has also shown nice performance without any cracks owing to recrystallization after once melting[106].

6. Recent Results in RFP(TPE-1RM15) and TRIAM-1M with Mo Limiter

As discussed above, three topics i.e. (1) impurity production and accumulation, (2) hydrogen recycling and (3) material performance under high heat load are main concerns for utilization of high Z metals as PFM. Recently effect of Mo and graphite limiter on plasma performance has been compared in RFP(TPE-1RM15 at Electrotechnical laboratory in Japan). [66,73] Because of the extraordinary high heat load to a limiter of the RFP[66,73,107], surface temperature of the graphite limiter increased over 2000K within ten milli-seconds, which made plasma operation very difficult. When the limiter material was changed from the graphite to Mo the plasma confinement (electron density and temperature and loop voltage) was appreciably improved (examples for loop voltage are given in Fig.16). The improvement of the plasma in the RFP is most probably owing to decrease of the limiter temperature[73,107] with an aid of higher thermal diffusivity. And higher energy reflection coefficient of Mo than that of carbon could play some role.

Nevertheless, when the hot spot temperature of the Mo limiter becomes near the melting point of Mo around 2900K, Mo(I) line emission increased drastically accompanying oxygen and Ha lines as shown in Fig.17. This indicates that increase of Mo(I) is probably due to evaporation of Mo. Similar Mo burst was
observed in JT-60 with TiC/Mo limiter /divertor[103-105], several pieces of which appeared to be severely melted after the opening[104,105]. It is quite clear that the melting must not be allowed in a fusion reactor and the divertor armor must be actively cooled. From engineering aspect, it certainly forces us to make much effort such as active cooling, alinement of tiles and so on but not from PSI aspect.

The effect due to difference in hydrogen recycling between C and Mo in the RFP is not clear, though density control with Mo limiter was much easier than with the graphite limiter[73].

No Mo burst and radiation collapse or sputter runaway have been observed in TRIAM-1M even for 1 hour operation in spite of severe damage and melting of its Mo limiter [108,109]. This is presumably owing to low confinement time and rather low density plasma. Nevertheless one should note that the plasma is successfully controlled with using the Mo limiter without any radiation collapse and difficulty in density control so long as more than 1 hour[108].

These results in addition to the earlier tokamaks, which succeeded to reduce high Z impurity by gas puffing or other techniques as discussed in chap.2, encourage us to use high Z material at least in the L mode. Of course utilization of high Z must be approved in the well confined (H-mode) plasma where the accumulation of impurity is general problem and not specified with high Z.

7. Conclusions and Recommendation

To apply high Z metals to PFM, there are several issues to be solved; high Z impurity production by sputtering, their accumulation in the plasma center, and high radiation loss. Because of these concerns the high Z metals are not widely employed nor planned to be used in the present large tokamaks. However, it should be pointed out that until now no clear evidence has reported on plasma collapses by the sputter runaway both in limiter and in diverted discharges.

High Z impurity accumulation in the earlier tokamaks most likely originates from sputtering by low Z impurities. Until now diagnostics for PSI have, unfortunately, not been enough, and sometimes impurity production due to pyrolytic origin like a gaseous desorption or material sublimation, which can be avoided by reducing areal heat load, seems to be mixed-up with that due to physics origin like sputtering. In addition there are several results showing favour to high Z. In JT-60 TiC/Mo
divertor plates showed good performance with neutral beam heating up to 26MW [47,48]. VH-mode experiments in DIII-D indicate little impurity accumulation in the center [28]. Comparison of graphite and Mo limiter in RFP is also promising.

From material aspect, poor ductility and small thermal shock resistance used to be a problem of most of the refractory metals. Recent advances in production and purification techniques of the refractory metals has lowered ductile brittle transition temperature (DBTT) of Mo, for instance, far below room temperature. This means that it is now possible to machine Mo at room temperature easily and high Z PFC is becoming within engineering feasibility.

We are afraid that the current database in the large machine is too concentrated to low Z and more or less no data is available for high Z. Not only for design of ITER engineering phase but also for any other incoming burning machine we need experience with high Z concerning how to control the impurities as well as hydrogen. As demonstrated in the present review, high Z and low Z materials should behave quite differently to the plasma, in other words, the plasma performance could be changed depending on Z number of PFM. We, therefore, strongly recommend to investigate the effect of PFM comparing Mo (W) and Carbon, or Nb(Ta) and Mo(W). The former pair is different in both hydrogen and collision behaviors, while the latter is quite different in hydrogen behavior but not for collision. Even if Nb and Ta must not be used for the armor in a commercial reactor, such kind of experiment is quite necessary not only for more comprehensive understanding of the plasma surface interaction but also for the designing of future machine.

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Figure captions

Fig. 1. Time behavior of impurity lines during the H-mode of JFT-2M[20].

Fig. 2. Time evolution of the ratio of the nickel intensities to electron density during the H-mode in JET. The nickel is injected by laser blow-off at 11s.[21].

Fig. 3. Nickel-radiation intensities as a function of line-averaged density of DIII-D discharges with and without single-null divertor[28].

Fig. 4. (a) Hollow temperature profile due to W accumulation in the center and (b) Disappearance of the hollow profile with controlled gas puffing[36].

Fig. 5. Radiation loss with and without X-point swing in JT-60[40].

Fig. 6. Comparison of $Z_{\text{eff}}$ in JT-60 discharge with 20MeV NB heating for TiC/Mo and graphite[47].

Fig. 7. Sputtering yield of Mo and W by various ions[54].

Fig. 8. Free Energy of Formation of Various Oxide.[58].

Fig. 9. Hydrogen Reflection from Ta[69].

Fig. 10. Reflection Coefficient of Carbon and Tungsten[72].

Fig. 11. Hydrogen Diffusion Coefficient in Mo and W[74].

Fig. 12. Hydrogen Solubility in Mo and W[74].
Fig. 13. Schematics of reemission behavior [85].

(a) Changes of reemission, recycling and retention with flux and time. Solid lines are for no changes in processes of diffusion, solution and recombination and dotted lines with changes.

(b) Changes of reemission, recycling and retention with temperature and time.

Fig. 14. Thermal desorption of deuterium from Mo carried out immediately after the implantation[78].

Fig. 15. Comparison of high heat load test of single crystalline of EB-Mo, as forged EB-Mo(AF-Mo) and powder metallurgical Mo(PM-Mo)[66].

(a) Surface after NBI test.

(b) Cross section of the RFP limiter.

Fig. 16. Loop voltage vs plasma current in RFP(TPE-1RM15) for different limiter material of graphite, stainless steel and molybdenum[73].

Fig. 17. Changes of plasma parameters of RFP(TPE-1RM15) with Mo limiter. In order to give very high heat load the limiter was inserted deep into the plasma[73].
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Energy distributions of neutral and positively charged hydrogen atoms backscattered from Ta bombarded with 18.5-keV protons. The charged fraction $N^*/N^* + N^o$ is given by dots.

Fig. 9. Hydrogen Reflection from Ta[69]
Arrhenius plot of the measurements of the diffusivity of hydrogen isotopes in molybdenum

$D = 4.0 \times 10^{-4} \cdot \exp[-22.3(kJ/mol)/RT]$  

Arrhenius plot of various measurements of hydrogen diffusivity in tungsten

$D = 4.1 \times 10^{-7} \cdot \exp[-75(kJ/mol)/RT]$  

Fig. 11. Hydrogen Diffusion Coefficient in Mo and W.[74]
Arrhenius plot of the solubility of hydrogen isotopes in molybdenum by various investigators.

Arrhenius plot of various measurements of hydrogen solubility in tungsten

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(a) Changes of Reemission, Recycling & Retention with Flux & Time

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(a) Surface after NBI test
(a) SCEB-Mo

(b) AFEB-Mo

(c) PM-Mo

(b) Cross section of the RFP limiter
Dependence of Vloop on Plasma current.
Comparison of SUS, Graphite and Molybdenum.

TPE-1RM15

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## Table 1. Central density of metal impurities in Ohmic and neutral beam heated discharges in tokamak

<table>
<thead>
<tr>
<th>n, / n, 10^-4</th>
<th>n, 10^14 m^-3</th>
<th>I_p(KA)</th>
<th>P_0, (MW)</th>
<th>gas</th>
<th>config.</th>
<th>material</th>
<th>T_e(wall) (eV)</th>
<th>T_e(w) (eV)</th>
<th>other conditions and remarks</th>
<th>machine</th>
<th>ref</th>
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</thead>
<tbody>
<tr>
<td>Ti: 0.6</td>
<td>2.4</td>
<td>4.0</td>
<td>20(CR)</td>
<td>H₂</td>
<td>div</td>
<td>TiC/No:TiC/No</td>
<td>1.5</td>
<td>3</td>
<td>P_{tot} = 10% n(02)~ 4 x 10^17 m^-3</td>
<td>JT-60</td>
<td>A</td>
</tr>
<tr>
<td>Ti: 40</td>
<td>40</td>
<td>1.0</td>
<td>1.5</td>
<td>11-14(H)</td>
<td>H₂</td>
<td>div</td>
<td>TiC/No:TiC/No</td>
<td>1.5</td>
<td>3</td>
<td></td>
<td>JT-60</td>
</tr>
<tr>
<td>Fe: 2.6</td>
<td>12</td>
<td>4.7</td>
<td>0.3</td>
<td>OH</td>
<td>He lim</td>
<td>Ti: SS</td>
<td>0.3</td>
<td>1</td>
<td></td>
<td>ASDEX</td>
<td>B</td>
</tr>
<tr>
<td>Fe: 4.0</td>
<td>12</td>
<td>3</td>
<td>0.3</td>
<td>OH</td>
<td>He lim</td>
<td>Ti: SS</td>
<td>0.3</td>
<td>1</td>
<td></td>
<td>ASDEX</td>
<td>B</td>
</tr>
<tr>
<td>Fe: 0.9</td>
<td>4.1</td>
<td>4.7</td>
<td>0.3</td>
<td>OH</td>
<td>He div</td>
<td>Ti: SS</td>
<td>0.3</td>
<td>1</td>
<td></td>
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<td>B</td>
</tr>
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<td>Fe: 1.8</td>
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<td>0.3</td>
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<td>He div</td>
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<td>1</td>
<td></td>
<td>ASDEX</td>
<td>B</td>
</tr>
<tr>
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<td>4.7</td>
<td>0.3</td>
<td>OH</td>
<td>D₂ lim</td>
<td>Ti: SS</td>
<td>0.3</td>
<td>1</td>
<td></td>
<td>ASDEX</td>
<td>B</td>
</tr>
<tr>
<td>Fe: 0.5</td>
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<td>3</td>
<td>0.3</td>
<td>OH₂</td>
<td>D₂ lim</td>
<td>Ti: SS</td>
<td>0.3</td>
<td>1</td>
<td></td>
<td>ASDEX</td>
<td>B</td>
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<tr>
<td>W: 93</td>
<td>660</td>
<td>7.0</td>
<td>0.2</td>
<td>OH</td>
<td>D₂ lim</td>
<td>Mo: SS</td>
<td>0.2</td>
<td>1</td>
<td>hollow Te(r)</td>
<td>ALCATOR C</td>
<td>C</td>
</tr>
<tr>
<td>W: 1.0</td>
<td>20</td>
<td>20</td>
<td>0.2</td>
<td>OH</td>
<td>D₂ lim</td>
<td>Mo: SS</td>
<td>0.2</td>
<td>1</td>
<td>hollow Te(r)</td>
<td>ALCATOR C</td>
<td>C</td>
</tr>
<tr>
<td>W: 15-25?</td>
<td>30 - 50?</td>
<td>2</td>
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<td>1</td>
<td>hollow Te(r) ?</td>
<td>PLT</td>
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</tr>
<tr>
<td>Ni: 0.11</td>
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<td>1.9</td>
<td>3</td>
<td>OH</td>
<td>He lim</td>
<td>INCONEL</td>
<td>0.2</td>
<td>1</td>
<td></td>
<td>JET</td>
<td>E</td>
</tr>
<tr>
<td>Ni: 0.08</td>
<td>0.06</td>
<td>1.9</td>
<td>3</td>
<td>OH</td>
<td>D₂ lim</td>
<td>INCONEL</td>
<td>0.2</td>
<td>1</td>
<td></td>
<td>JET</td>
<td>E</td>
</tr>
</tbody>
</table>

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