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Abstract (must not exceed 150 words)

In order to establish a D-T fusion reactor as an energy source, it is not enough to have a DT burning plasma, and economical conversion of fusion energy to electricity and/or heat, a large enough margin of tritium breeding and tritium safety must be simultaneously achieved. In particular, handling of huge amount of tritium needs significant efforts to ensure that the radiation dose of radiological workers and of the public is below the limits specified by the International Commission on Radiological Protection (ICRP). In this paper, after the introduction of tritium as a fuel of DT reactors and as a radioisotope of hydrogen, tritium safety issues in fusion reactor materials, focusing on to tritium retention, permeation and contamination. (118 words)

Keywords:

Tritium, Safety, Economy, Breeding, Retention, Inventory

1. Introduction

In order to establish a D-T fusion reactor as an energy source, economical conversion of fusion energy to electricity and/or heat, having an enough margin in tritium breeding, and insuring tritium safety must be simultaneously achieved. In particular, handling of huge amount of tritium in a reactor system needs a significant effort to ensure that the radiation dose of radiological workers and of the public is below the limits specified by the International Commission on Radiological Protection.

For the safety reasons, tritium in a reactor will be limited to only a few kg orders in weight, with radioactivity up to 10^{17} Bq [1-4]. Since public exposure to tritium is regulated at a level as tiny as a few Bq/cm², tritium must be strictly confined in a reactor system with accountancy of an order of pg (pico-gram). Generally qualitative analysis with the accuracy of more than 3 orders of magnitude is hardly possible. We are facing to lots of safety concerns in the handling of huge amount of tritium as a fuel and to be bred in a blanket.

In addition, tritium resources are very limited. Not only for the safety reason but also for avoiding the shortage of tritium resources, tritium retention in a reactor must be kept as small as possible and tritium breeding should have enough margins to compensate the inventory in all tritium system including the reactor vessel and pumps. In present tokamaks, however, hydrogen retention is significantly large, i.e. more than 10% of fueled hydrogen is continuously piled up in the vacuum vessel, which must not be allowed in a reactor [1,2,5,6].

After the introduction of tritium as a hydrogen radioisotope [7], important issues in interaction of tritium with fusion reactor materials will be summarized considering the handling of large amount of tritium, i.e. confinement, leakage, contamination, permeation, regulations and tritium accountancy. In Japan, a project of strongly related to those subjects “Tritium science and technology for a fusion reactor” supported by MEXT, Grand in Aid for Scientific Research for Priority Area No.476 [8] has been operated and progresses for overcoming such tritium problems in the project is also presented.

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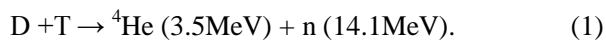
2. Tritium as fuel of DT reactor

For any energy sources, fuel is a concern. In a DT fusion reactor, Deuterium and Tritium are fuels. Since natural water contains 0.016% deuterium, we can extract it from the water mostly by means of electrolysis. While the natural abundance of tritium, which is generated by cosmic rays and also by nuclear

reactions (atomic bombs and nuclear reactors) after the 2nd world war, is very small as show in Fig. 1. Therefore tritium should be artificially produced.

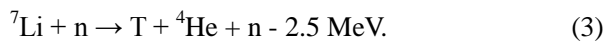
From simple calculations, 56 kg tritium is required per GW year (thermal) of fusion power for a DT reactor. While about 100 g of tritium is produced per year in a standard CANDU fission unit and 20 to 25 kg tritium (mainly in Canada) will be available for operation of ITER. Hence it seems difficult to get sufficient amount of tritium even for a demo reactor. Therefore tritium retained in a fusion reactor should be recovered not only from tritium safety but also tritium economy. Furthermore, tritium must be produced by nuclear reactors of (2) and (3) using ${}^6\text{Li}$ and ${}^7\text{Li}$, respectively.

Once sufficient amount of tritium for a DT reactor is available, the DT reactor produces energy and breeds tritium in a blanket system simultaneously [9] as described below. The energy released by DT reactions is distributed to ${}^4\text{He}$ and neutron,



The energy of ${}^4\text{He}$ is to be used for heating plasma for continuing the DT burning and the energy carried by neutron is transformed to heat in a blanket system for electricity or other use, like hydrogen production.

At the same time, neutron is used to breed tritium as



Although the reaction (2) generates both tritium and energy, tritium is not bred (breeding rate is just 1). If a half of neutrons produced by DT reactions are used for the reaction (2) and the last half for (3), tritium breeding ratio becomes 1.5. However the latter reaction requires energy and reduces the output energy. With using the optimized blanket system, overall tritium breeding ratio in a fusion reactor is expected to be around 1.1 or a little less. In this respect, the tritium inventory increment or retention rate in the reactor $((\text{fueling} - (\text{burning} + \text{recovering}))/\text{fueling})$ must be below 0.1, otherwise tritium for the 2 nd reactor is not generated. At the moment this seems very hard owing to large in-vessel tritium inventory as described later.

3. Tritium, a radioactive hydrogen isotope

Tritium is a radioactive hydrogen isotope decaying to ${}^3\text{He}$ emitting a β -electron and an antineutrino ($\bar{\nu}$) with a half life of 12.323 year [8],

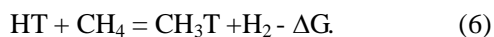


Accordingly the decay rate, 1g of tritium is equivalent to 3.5574×10^{14} Bq. This means that during storage, about ~ 5.5 % is disappearing in a year. Therefore the tritium breeding must have enough margins to compensate this. The energy of emitted β -electrons is widely distributed with the maximum of 18.6 keV and average of 5.7 keV. The integrated decay heat is 324 mW/1gT, which is not very large but could force thermal release of tritium from heavily tritium loaded materials.

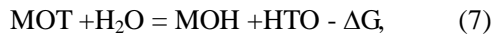
Because of its low energy, the effect of β -electrons on living things is also very week. Hence tritium had been used under very mild regulation or sometimes no regulation. And usually no heavy shielding is required to handle tritium, though cross-contamination must be concerned. Tritium is easily detected by β -electron counting with the detection limit and/or accuracy of several Bq/cm² on solid surfaces and around 0.1 Bq/cm³ in water. However, the β -electron counting is limited to below 10⁹ Bq or mg order of T. For much larger amount of tritium, mass and/or pressure measurements, the same way to measure other hydrogen isotopes are employed. The measurement of decay heat allows calorimetry but its accuracy is only 10⁻² to 10⁻³. All present tritium measurements except the β -counting give only 3 to 4 digit and any loss of tritium less than 0.1% is hardly possible to detect. Since public exposure to tritium is regulated at a level as tiny as a few Bq/cm², tritium must be strictly confined in handling systems. Since, as described above, 56 kg tritium (2×10^{19} Bq) is required per GW year, it is hardly possible to care about such small amount in tritium system. Hence tritium accountancy, or evaluation of inventory in tritium systems, permeation and leakage, is very hard to attain.

Tritium escaping from the handling system by permeation and contamination can be detected by the β -counting method. However, energy of β -electron is so small to escape from materials more than few μm and in air, only 6mm. Thus tritium retained in bulk of solid materials can not be detected and movable tritium in the materials, mostly in metals, is problematic for safety.

Tritium can easily replace the ubiquitous lighter hydrogen isotopes like protium (H) / deuterium (D) in water and hydrocarbons in air,



In particular, any materials surfaces adsorb water molecules and enhance isotopical replacement,



producing hazardous tritiated water.

In addition, the β -electrons could cause and/or enhance undesirable chemical reactions (radio-chemical reactions) in living things appearing as radiation hazard.

Figure 2 is a good example indicating how tritium is transferred by cross-contamination caused by above reactions [10]. Gloves as essential equipment in a tritium handling system are always contaminated and tritium on the glove surface is immediately transferred to non-contaminated materials. However, tritium exposure of skin is not so dangerous owing to thin penetration of the β -electron, while tritium taken into a body usually in the form of HTO or OT is very hazardous. In this respect, gaseous form, HT, is much safer than HTO and OT. It is well known that drinking beer is very effective to remove tritium from the body, and actually beer bottles are prepared at the exit of some tritium laboratories.

It is interesting to note that the β -decay of tritium accompanies the emission of an antineutrino. This means that the precise measurements of edge energy (maximum energy of emitted β electron) would give neutrino mass, which is a positive use of tritium decay [11].

[LF]

4. Tritium fuel cycle

Figure 3 shows a fusion reactor system with a blanket system generating power and breeding tritium simultaneously, and accompanying problems. In the fusion reactor, the amount of tritium to be handled is $\sim 10^{17}$ Bq under accountancy (or under regulation of) of a few tens Bq. The form of tritium handled includes ice pellets (~ 20 K) and gas at RT (300K) (both for fueling), energetic neutrals and ions (for neutral and ion beam heating) and plasmas, having temperatures ranging from $10^1 \sim 10^9$ K.

Therefore the physics and chemistry of the interactions of tritium with materials used in the reactor or tritium confinement systems are very complex [12]. The high level of radioactivity of tritium generates additional problems due to β -electron emission and/or radiation heat, such as excited state chemistry and non-equilibrium thermodynamics. Furthermore defect formation by electron excitation and He production result in various damages in materials. This, in turn, influences basic process of hydrogen-materials interactions such as adsorption, solution, diffusion and permeation in materials, which

are directly related to tritium confinement.

A speciality of the fuel cycle is very poor burning efficiency. i.e. only a few % of input tritium burns and the majority is recovered (pumped out from the reactor vessel) to be recycled. In addition, several % is likely continuously retained in the reactor vessel to become huge in-vessel tritium inventory which is hard to be recovered. Since the recovered tritium includes H, D, T, He and other impurities, it should be refined with isotope separation and then recycled.

The reactor is surrounded by blanket systems to realize power generation and tritium production simultaneously. Different from the fission reactor, in which most of the released energy by nuclear reactions is deposited in a fuel pin with a diameter of only around 10 mm, 14 MeV neutron energy must be transformed to heat for generation of electricity in the large volume of the blanket systems.

Tritium resource is very limited and must be effectively bred in blanket simultaneously with power generation. Tritium breeding in the blanket should have enough margins to compensate the inventory in all tritium systems including reactor vessel and tritium decay. To enhance the recovering rate of T produced in the blanket, addition of water vapor (H_2O) is well known to be effective, but T diluted in H_2O is very hard or need lots of energy to be recovered, which increases fusion energy cost.

In addition, tritium produced in the blanket easily permeates into coolant. The permeated tritium readily reacts with surface contaminants to produce hazardous tritiated water and/or hydrocarbons according to the reactions (7) and (8). In particular, ferrite, a low activation structure candidate material, has very high tritium permeability and needs permeation barrier with the permeation reduction of 5-6 orders of magnitude. For a water cooling system, permeated tritium from the plasma facing surface or blanket to the coolant water easily produces HTO, resulting diluted tritiated water from which tritium recovery is very cost consuming.

Although Lithium (Li) can work as coolant and tritium breeder simultaneously [13], tritium extraction from Li is very difficult. Hence a Lithium Lead (LiPb) eutectic alloy for which T recovery is rather easy, could be promising coolant [14]. Simultaneous generation of electricity and keeping tritium safety still need significant R&D efforts and both must be optimized.

A tritium recycling system, which uses mostly established techniques, can be built for ITER or even for a reactor without very high hurdles and, hence, Tritium Plant is not likely on a critical schedule path towards First Plasma in ITER [1-3]. However, handling of huge amount of tritium in ITER would give

problems to be solved. They are mostly relating tritium behavior in plasma, huge inventory in vacuum vessel and its accountancy, controlled fuelling to keep DT burning, possible permeation and leakage leading to contamination of remote handling system, and so on. Most of those tritium problems are directly related to the safety of operators and/or professionals. But public safety does not seem to become significant problems because the emission to outside of the reactor site can be easily kept below the safety limit as already described.

5. Contamination

In order to handle heavily contaminated materials in the glove box, as already noted in Fig. 2, one should be careful on multi-step or multistage cross-contaminations and multiple use of the box would be required. The cross-contamination is also a serious concern for remote-handling system. i.e. once remote hands touch contaminated components in tokamak, like a divertor cassette, the hands are immediately contaminated and the contamination transferred to other components handled

Such surface contamination on solid surfaces is mostly due to easy oxidation of tritium to tritiated water or isotope exchange between normal water to the tritiated water. Furthermore, most of material surface absorbs water molecule or react to form OH species.

Accordingly, as shown in Fig. 4, surface tritium concentration is not necessarily to be the same as that in the bulk [15]. In the figure, the surface tritium levels are compared with various metals in which tritiated hydrogen were dissolved in hydrogen gas atmosphere at 873K. One can note the surface tritium concentrations are quite different with the solubility. That is because, the surface of most of metals is oxidized with different level (or different oxide thickness) and the amount of retained OT is higher for more oxidized surface. When some of the surface tritium was smeared out, tritium was supplied from the bulk by diffusion and the surface tritium level recovered easily. Of course if bulk tritium concentration was very high, the surface segregated tritium would be hindered. In any way, this makes decontamination of tritium dissolved in the materials very difficult.

6. Interaction of energetic hydrogen and materials (plasma materials interactions), and in-vessel tritium retention

Figure 5 shows schematics of interactions of hydrogen plasma and materials. Different from gaseous

hydrogen, hydrogen from plasma impinging to surface penetrates directly into certain depth except a few tens % reflected. Thus implanted hydrogen diffuses back to the surface and released after recombining to H_2 . The remaining diffuses into the bulk to be distributed to solution sites or trapping sites and some are permeated to the rear surface. It is important to note the incident flux is extremely high and little experiments have been done under such high fluxes. Therefore we have to extrapolate experimental results to higher flux regions. In any way, most of hydrogen returns in plasma. Nevertheless, hydrogen is accumulated in the plasma facing materials by diffusion. The energetic particle incidence induces sputtering erosion of the surface. And the eroded materials are transported to somewhere in particular plasma shadowed area. In the redeposited layers, hydrogen is incorporated to increase tritium retention (inventory). For plasma facing carbon materials used in most of current tokamaks, their chemical sputtering to make methane results in significant erosion and redeposition [16], both of which are the main reason to avoiding carbon in ITER.

The current estimation of the in-vessel tritium inventory or fuel retention rate is fairly large as shown in [fig. 6 \[17\]](#). ITER shall be stopped every hundreds shots which result in tritium retention of nearly 1 kg (the site limitation of T for the safety) and the retained tritium must be removed for both economical and safety reasons. Most of the retention is caused by hydrogen corporation in redeposited layers at plasma shadowed area and could not be saturated. The removal of tritium at the plasma shadowed area is not easy and the methods for reduction and recovery (removal) of the in-vessel tritium still under development and need large efforts.

The current estimation of the fuel retention fraction is around 5-20%, which is too large to compensate by tritium breeding in the blanket, which would be 1.2 at bests. Considering the tritium decay of 5%/y, the breeding ratio should be maximized and the invessel retention must be minimized, otherwise DT reactors can not be an economical energy source.

Tritium pumped out (exhausted) from the vessel, which includes hazardous inorganic tritium, must be recovered (Cryo-pump would have large inventory) and refined to be recycled. Tritium inventory in any tritium systems should be kept as low as possible. Still tritium permeation from system walls is not unavoidable. In a reactor, its higher operation temperature would results in significant amount of tritium permeation, which should be avoided by dual tubing or permeation barrier. In the former, processing of permeated to outer tube gives another problem and permeation reduction factor and reliability of the

barrier must be improved.

Compared to the incident flux, the permeated flux is very tiny. Nevertheless, tritium permeated to the cooling side and released into a coolant is a serious safety concern. In case of water coolant, the permeated T is easily replaced with H in H₂O, which requires T removal from huge amount of cooling water for the safety.

There is another concern. At the cooling water side, corrosion occurs to produce atomic hydrogen which is easily uptaken into the wall. Although concentration of D and T at the plasma facing surface must be much higher than that of H at the corroded water side, chemical potential difference of H between the plasma facing surface (very low) and the cooling water side (very high) makes H diffusion to the plasma facing surface possible and should dilute the fuel. The amount of H uptake in this manner are not clear, it is total dependent on design of the cooling system, but is not likely small.

7. Isotope effect in solution, diffusion and permeation

Owing to large mass differences among three hydrogen isotopes (H, D, T), various isotopic effects are reported. Since most of the effects are include surface kinetics, differences in hydrogen fluxes to and from solid surface among the isotopes should be taken into account.

Simple molecular kinetics tells that an average velocity of a gas molecule with mass m at temperature T is given by,

$$\bar{v} = \sqrt{\frac{8RT}{\pi m}} \quad (9)$$

where, R the gas constant, T , temperature, and m , mass. Therefore, the velocity differences among three hydrogen isotopes are

$$\bar{v}_H / \bar{v}_D = \sqrt{2} \quad (10)$$

and

$$\bar{v}_H / \bar{v}_T = \sqrt{3} \quad (11)$$

Molecular kinetics also gives incident flux to wall surface under pressure P

$$J = nv = \frac{P}{(2\pi mkT)^{1/2}} \quad (12)$$

Accordingly, incident flux ratios are written as,

$$J_H/J_D = \sqrt{2} \quad \text{and} \quad J_H/J_T = \sqrt{3} \quad (14)$$

Since hydrogen permeation and solution are likely proportionally to incident flux, the isotopic effects in solubility (S) and permeability (Φ) would be proportional to the square root of mass ratio, i.e.

$$S_H/S_D = \sqrt{2} \quad \text{and} \quad S_H/S_T = \sqrt{3} \quad (15)$$

and

$$\Phi_H/\Phi_D = \sqrt{2} \quad \text{and} \quad \Phi_H/\Phi_T = \sqrt{3} \quad (16)$$

In diffusion controlled permeation, permeability is represented by,

$$\Phi = SD \quad (17)$$

However, according to the isotope effects in S and Φ , eq (17) is not possible. i.e. the isotope effect could not be represented such simple equations.

Large mass differences among hydrogen isotopes results in large difference in their quantized rotational and vibrational states. Accordingly significantly large isotope effects are observed in diffusion coefficients at temperature far below RT. Contradictions among Φ , D and S are most likely owing to neglecting of the isotope effects in the rotational and vibrational states of hydrogen isotopes in solids.

The isotope effect is also important for keeping DT burning in plasma. Since the lighter mass results in higher flux, better confinement in plasma, and poorer pumping efficiency, the exhausting flux from the reactor vessel should show significant isotope effects. The isotopic effects in solubility would also influence strongly in fuel retention in the plasma facing materials, which is not well examined. Thus the optimized D-T burning should not be realized by equimolar fuelling of D and T. Furthermore, there is no good way to determine concentration of D and T in burning plasma, control of fueling to keep optimized DT burning must not be easy.

Still, other parameters in the interaction of hydrogen isotopes and materials, like reflection, reemission, diffusion, trapping, and so on, should show isotope effects and extensive studies are needed.

[LF]

5. Summary and Conclusion

Not only for safety but also tritium economy, tritium fuel cycling must be very effective and tritium inventory in each system must be kept as low as possible. Fueling system is so inefficient and only a few % of supplied fuel burn and the rest is pumped out (exhausted). Energetic hydrogen injection result in erosion of wall materials, which in turn, return to somewhere to produce redeposited layers. The layers retain significant amount of tritium and the exfoliation of the redeposited layers would be the source of activated dusts. Effects of β -electron emitted from dissolved tritium with high concentration in materials, in particular, carbon, its compounds and organic materials are not studied well.

The in-vessel tritium retention could be significantly large. For both economical and safety reasons, it must be reduced, or removed (recovered). The methods for reduction and recovering (removing) of in-vessel tritium still under development and need large efforts. .

Tritium pumped out (exhausted) from the vessel, which includes hazardous inorganic tritium, must be recovered (Cryo-pump would have large inventory) and refined to be recycled. Tritium inventory in any tritium systems should be kept as low as possible. Still tritium permeation from system walls is not unavoidable. In a reactor, its higher operation temperature would results in significant permeation, which should be avoided by dual tubing or permeation barrier. In the former, processing of permeated to outer tube gives another problem and permeation reduction factor and reliability of the barrier must be improved.

Tritium resource is very limited and must be effectively bred in blanket simultaneously with power generation. Tritium breeding in the blanket should have an enough margin to compensate the inventory in all tritium systems including a reactor vessel and tritium decay. To enhance the recovering rate of T produced in the blanket, addition of water vapor is well known to be effective, but T diluted in H₂O is very hard or need lots of energy to be recovered, which increases fusion energy cost.

In conclusion, tritium safety is the primary issue to establish fusion as an energy source. In addition, energy conversion to electricity or other forms and T breeding must be simultaneously attained. In order to avoid shortage of T resources, T inventory in all T systems should be minimized. And T breeding should have an enough margin to cover tritium inventory in particular in vessel inventory.

Before actual use of large amount of T, we have still time. All problems above mentioned are strongly related to tritium and materials interactions and extensive studies are awaited

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Acknowledgement

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Figure captions

Fig.1 Comparison of tritium amounts, resources, abundance and regulation

Fig. 2 Example of cross-contamination of tritium; Tritium surface profiles of metals handled in a tritium handling glove box.

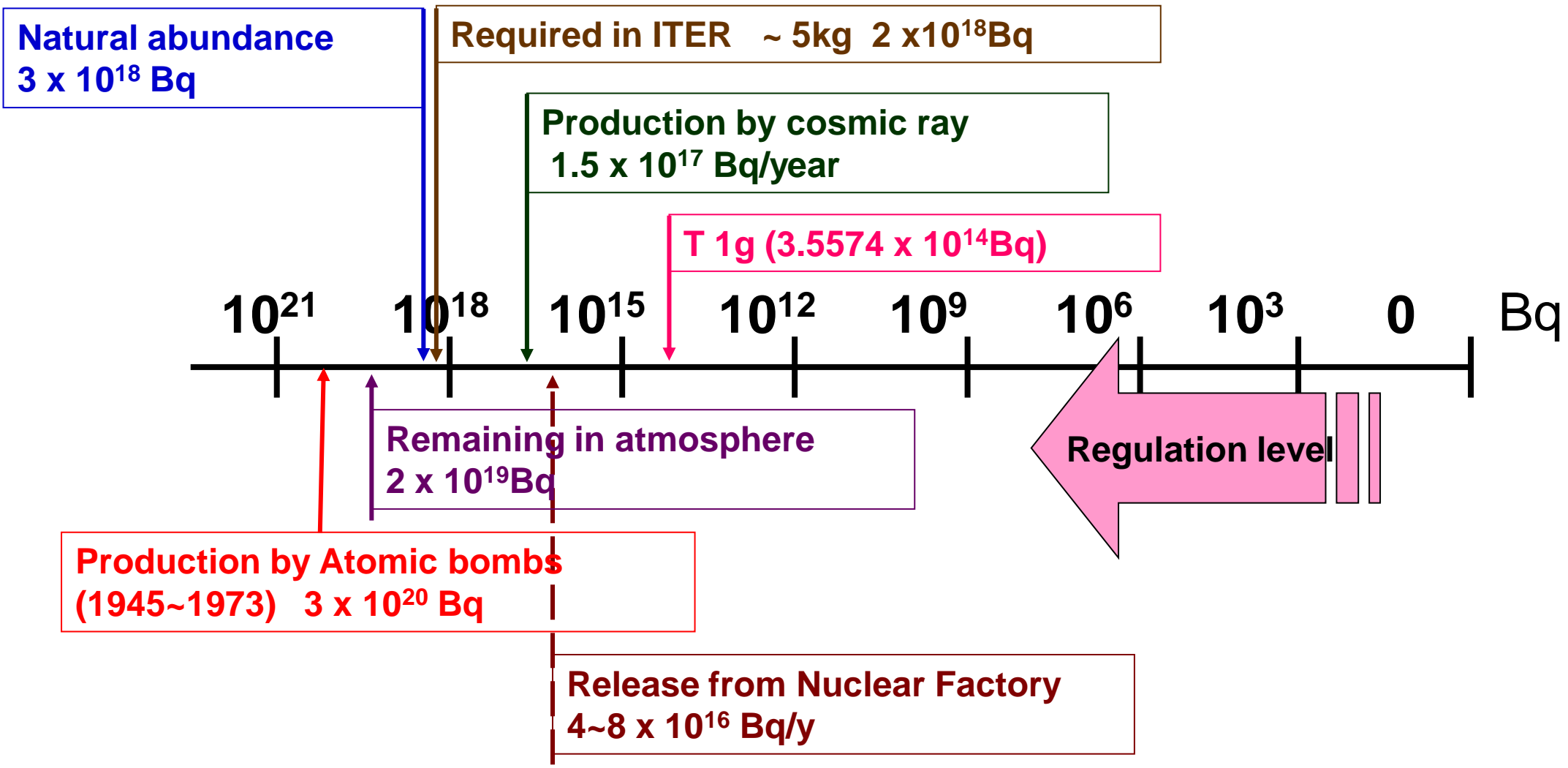
Fig. 3. Fuel cycling systems for a fusion reactor and associated problems in tritium handling

Fig.4 Surface tritium concentration on various metals. Tritium was loaded by gaseous charging.

Hydrogen solubility at 873 K is given below for comparison [15].

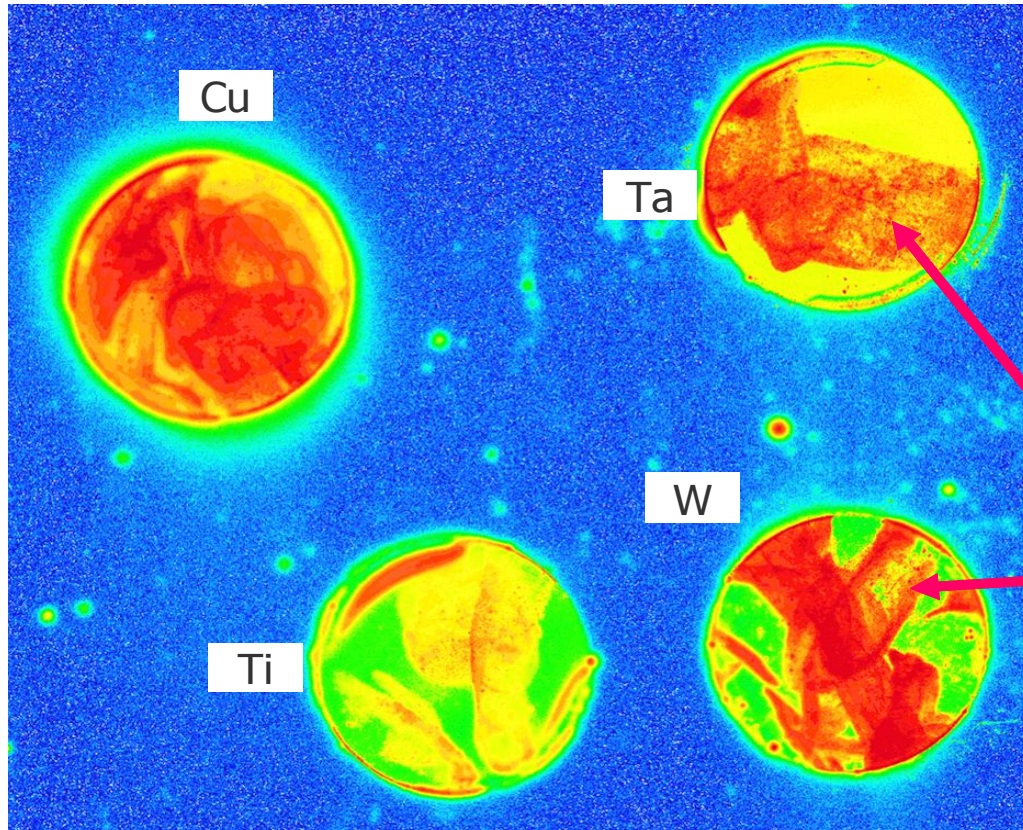
Fig. 5 Schematics of interactions of hydrogen plasma and materials.

Fig. 6 Current estimation of T inventory in ITER by Roth et al. [17]

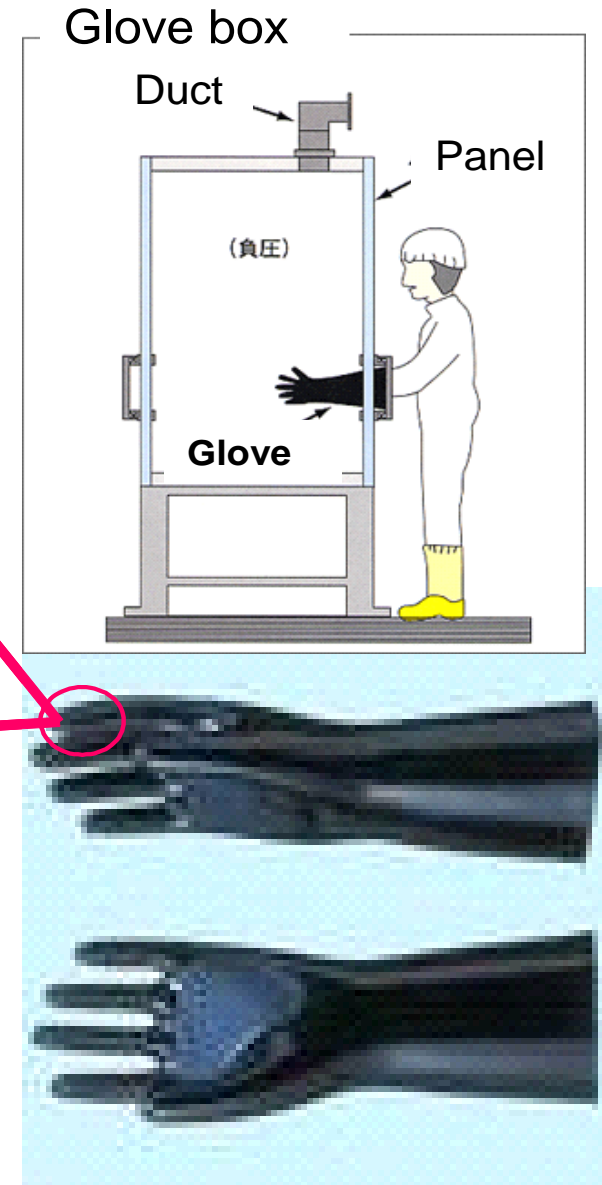


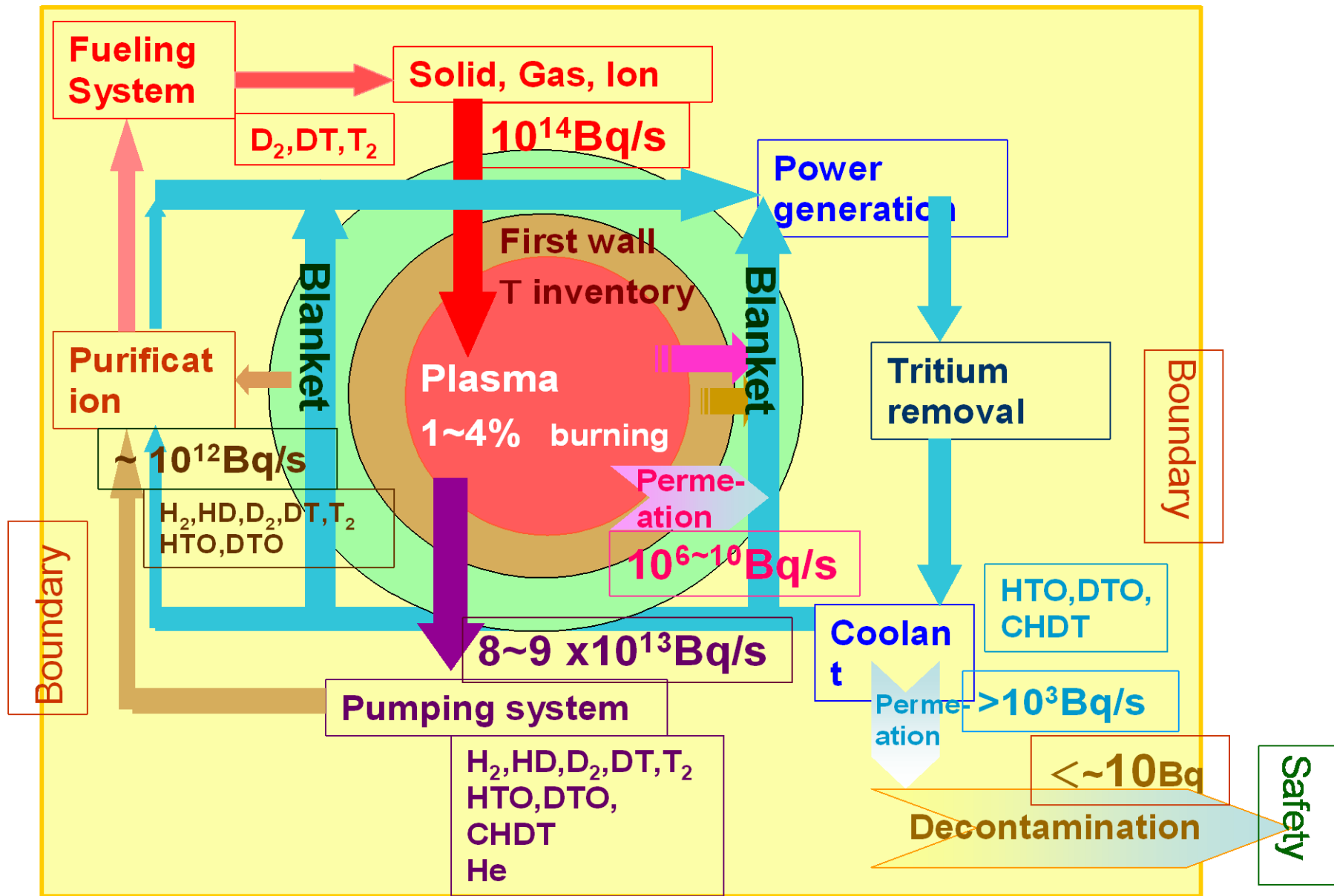
Quantitative analysis	Decay heat	Gravimetric	Disintegration (dps)
		Volumetric (PVT)	

**No single method can cover whole range.
Poor resolution inhibits cross-check**

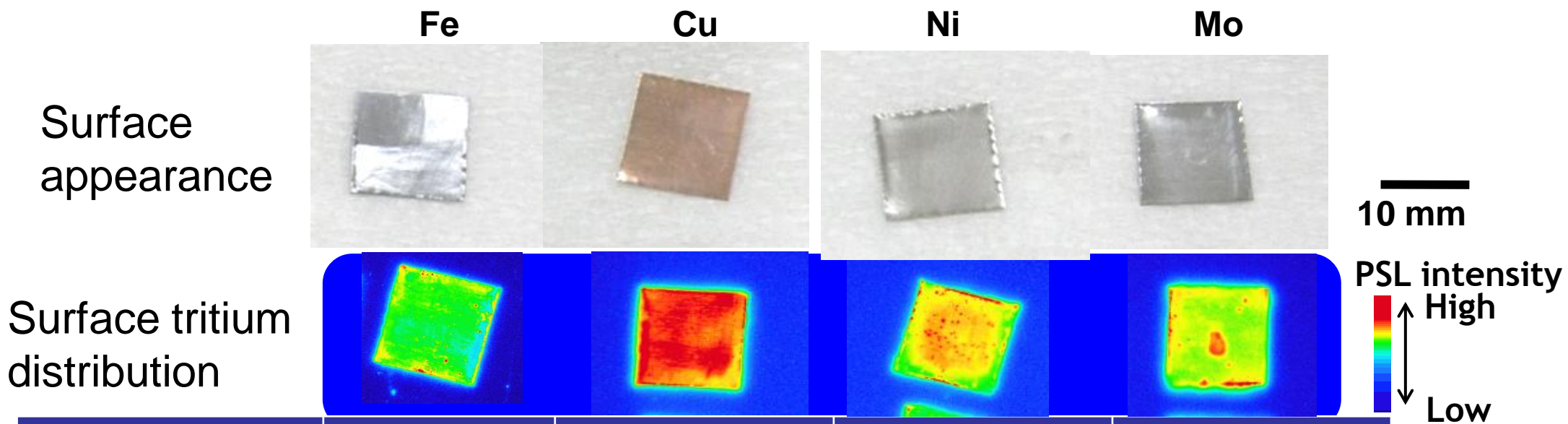


Traces of glove fingers





Tanabe Fig.3 (Single column)



	Fe	Cu	Ni	Mo
PSL intensity (PSL mm ⁻²)	704	2380	1070	811
H solubility at 873K (H/M)	10 ⁻⁵	10 ⁻⁵	10 ⁻³	10 ⁻⁷

Wall inventory is much larger than plasma inventory

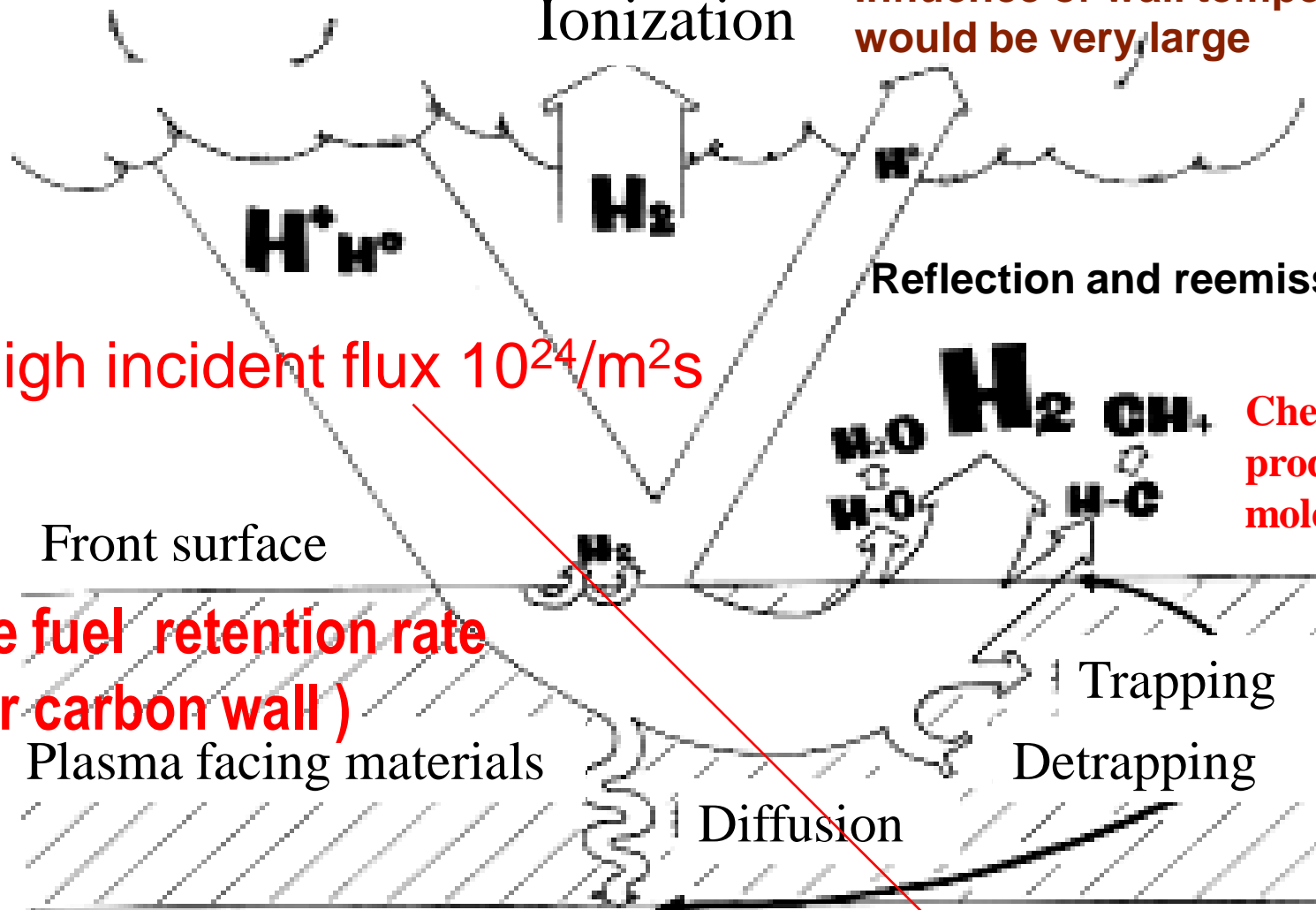
Amount of Hydrogen in wall \gg Hydrogen in plasma

H plasma

Difficulty of D/T control in plasma

Influence of wall temperature increase would be very large

Ionization



Very high incident flux $10^{24}/m^2s$

Reflection and reemission

Chemical reactions produces hazardous molecules and dusts

Front surface

Very large fuel retention rate (~20% for carbon wall)

Plasma facing materials

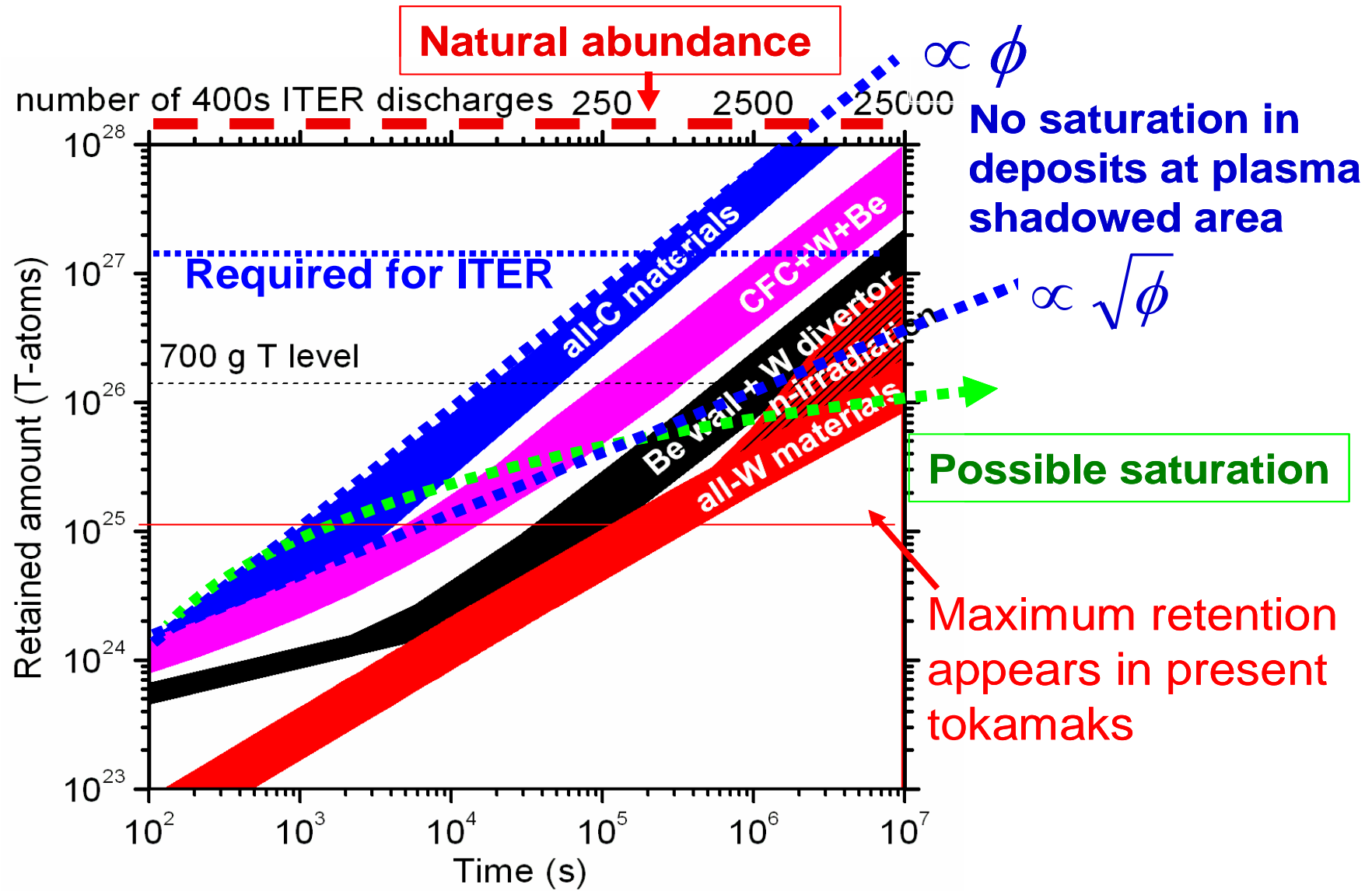
Trapping

Detrapping

Diffusion

Back surface

Permeation result in contamination of cooling water



Tanabe Fig.6 (Single column)