

Chapter 2

Characteristics of Tritium

Tetsuo Tanabe

Abstract For any energy sources, fuel is a concern. In a DT fusion reactor, deuterium (D) and tritium (T) are fuels. Since hydrogen in natural water contains 0.016 % D, we can extract it from the water mostly by means of electrolysis. T, a radioactive hydrogen isotope decaying to ^3He by emission of a β -electron and an antineutrino ($\bar{\nu}$) with a half-life of 12.323 year, is generated by cosmic rays and also by nuclear reactions (atomic bombs and nuclear reactors) after the Second World War. Because of its short lifetime, natural abundance of T is very small. Therefore T, as a fuel of the D-T fusion, must be artificially produced. Although handling and processing of hydrogen is well established in industrial scales, owing to its radioactivity, special care is required for safety in handling T. In this chapter, characteristics of T as a radio isotope of hydrogen are introduced, focusing important properties of T as the fuel of a fusion reactor. Particular focuses are given to handling of large amount of radioactive T, behavior of T in burning plasma, and T breeding in blanket to attain fuel self-sufficiency, all of which we have never experienced.

Keywords Hydrogen • Deuterium • Tritium • Properties • Characteristics • Radioactivity • Resources

2.1 Hydrogen Isotopes

2.1.1 Hydrogen, Deuterium, and Tritium

Hydrogen is the lightest chemical element on the periodic table with chemical symbol H and atomic number 1, and the most abundant element in the universe. There are three isotopes of hydrogen, the most common isotope of hydrogen,

T. Tanabe (✉)

Interdisciplinary Graduate School of Engineering and Sciences,
Kyushu University, 6-1, Kasuga-Koen, Kasuga, Fukuoka 816-8580, Japan
e-mail: tanabe@nucl.kyushu-u.ac.jp

termed protium (name rarely used, symbol ^1H or simply H) constituted of a single proton and an electron, heavy hydrogen termed deuterium (^2H , usually D) involving one neutron into protium and tritiated hydrogen termed tritium (^3H , usually T) involving two neutrons. H and D are stable isotopes, while T is a radioisotope decaying with the half-life of 12.323 year. More heavier hydrogen isotopes can be synthesized, but they are not stable with half-lifetimes of less than a zeptosecond (10^{-21} s) [1].

Main physical properties of H, D, and T are given in Table 2.1 [2, 3]. Because of the large mass differences among them, isotopic differences of those physical properties (isotopic effects) are pronounced much more than isotopic effects of any other elements. At standard temperature and pressure (STP; 0 °C, 101.325 kPa), hydrogen isotopes are a colorless, odorless, tasteless, nonmetallic, highly combustible diatomic gas with the molecular formula of H_2 , D_2 , and T_2 . When three gases are coexisting, total of 6 isotopes, H_2 , HD, HT, D_2 , DT, and T_2 appear. However, the basic exchange between D_2 and T_2 for fusion fuels, for example,



does not take place automatically. Some catalysts or raising temperature accelerates the exchange reaction. The equilibrium constant K of the reaction (2.1) changes with temperature and given by

$$\Delta G = -RT \ln K, \quad \text{or} \quad K = [\text{DT}]^2 / [\text{D}_2] * [\text{T}_2], \quad (2.2)$$

Table 2.1 Comparison of physical properties of H_2 , D_2 , and T_2 gases [3]

	H_2	D_2	T_2
Abundance (in atomic form)	99.985 %	0.016 %	10^{-18}
Density (kg m^{-3}) at STP (0 °C, 101.325 kPa)	0.08988	0.180	0.2705
Heat of vaporization (kJ mol^{-1})	0.904	1.24	1.39
Boiling point (BP) (K)	20.271	23.67	25.04
Liquid density at BP (kg m^{-3})	70.99	163.83	255.6
Heat of fusion (kJ mol^{-1})	0.117	0.197	0.250
Melting point (MP) (K)	13.99	18.73	20.62
Solid density at M.P. (kg m^{-3})	86.3	196.7	45.35 mol/L
Triple point (TP)	13.804 K, 7.030 kPa	18.69 K, 17.13 kPa	20.62 K, 21.6 kPa
Critical point (CP)	32.976 K, 1.293 MPa	38.26 K, 1.65 MPa	40.44 K, 1.85 MPa
Density at CP (mol/m^3)	15,200	16,700	17,700
Solid heat capacity at TP ($\text{J mol}^{-1} \text{K}^{-1}$)	6.2	11.8	16.1
Bond energy (kJ mol^{-1})	435.9	443.4	446.9

using the molar fraction of each molecule. $K = 0$ at 0 K, and $K = 4$ at infinitely high temperature. In an equimolar mixture of D and T, molar fractions are 0.25, 0.25, and 0.5 for D_2 , T_2 , and DT, respectively. In a fusion reactor system, H_2 always remains in the fuel as an impurity, but equilibrium constants for mixture of three isotopes, H, D, and T, were not measured nor calculated. Because of the significant mass differences, gas flow, in particular, evacuation rates for three isotopes are quite different and could influence fuel recycling in the reactor.

There exist two different spin isomers of hydrogen diatomic molecules that differ by the relative spin of their nuclei [3]. In the H_2 molecule, the spins of the two hydrogen nuclei (protons) couple to form a triplet state known as ortho-hydrogen termed o- H_2 with parallel spins, and a singlet state known as para-hydrogen termed p- H_2 with antiparallel spins.

At STP, hydrogen gas contains about 25 % of p- H_2 and 75 % of o- H_2 (or 1/3 in their ratio), which is referred as the “normal form,” n- H_2 . For D_2 and T_2 , the ratios are 2/1 and 1/3, respectively. The equilibrium ratio changes with temperature. At 0 K, p- H_2 , o- D_2 , and p- T_2 account 100 % in equilibrium. Thermal properties of pure p- H_2 differ from those of n- H_2 . The difference in thermo-physical properties of isomers of T_2 would influence the formation process of ice pellets for fueling.

Since hydrogen readily forms covalent compounds with most nonmetallic elements, most of the hydrogen on the earth exists in molecular forms such as water and/or organic compounds. Hydrogen is mostly produced by steam reforming of natural gas and less often from more energy-intensive hydrogen production methods like the electrolysis of water. Most hydrogen is employed near its production site, with the two largest usages being fossil fuel processing (e.g., hydrocracking) and ammonia production, mostly for the fertilizer market. Hydrogen is mainly used in petroleum and chemical industries. Quite recently, H_2 is used as energy carrier with energy density of 120 MJ/kg to give clean energy, and application of hydrogen fuel cell for cars has started.

Hydrogen is a concern in metallurgy as it can embrittle many metals [4], which makes the design of pipelines and storage tanks complicating. Nevertheless, the mechanism of hydrogen embrittlement is not clarified yet, and hence, hydrogen embrittlement should be taken into account for reactor design. Hydrogen is easily dissolved in and permeate through metals. For utilization of T in fusion, the dissolution and permeation are quite important problems for T inventory and confinement from the aspect of radiation safety of which details are discussed in this book.

2.1.2 Tritium, a Radioactive Hydrogen Isotope

Tritium decays to ^3He with emissions of a β -electron and an antineutrino ($\bar{\nu}$) with a half-life of 12.323 year [3],



According to the decay rate, 1 g of T is equivalent to 3.5574×10^{14} Bq and about 5.5 % is disappearing during its storage in a year by the decay. The energy of the emitted β -electrons is widely distributed with the maximum of 18.6 keV and average of 5.7 keV as shown in Fig. 2.1 [3]. The integrated decay heat, 324 mW/1gT, is not very large, but could influence melting of ice pellets used for fueling, and enhance thermal release of T from heavily T-loaded materials. Small particles containing certain amount of T are charged up by the emission of electrons and show unexpected movement due to electrostatic force [5].

Owing to its low energy, the effect of the β -electrons on human beings is very weak compared to other radioactive materials. Hence, T had been used under very mild regulation or sometimes without regulation. “EXIT” signs for emergency and luminous watches commonly used are driven by T with electroluminescence. Usually, no thick shielding is required to handle T, though direct touching is prohibited.

The detection of T retained on materials surfaces is rather easy with using the β -counting method, which has 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 $\sim 10^9$ Bq or mg order of T. Furthermore, T retained in the bulk of solid materials is quite hard to detect, because the β -electron can penetrate through or escape from the solid materials only a few μm in depth. (Its maximum range in air is 6 mm and less than 1 μm in metals.) Therefore, movable T in the solid, mostly in metals, causes safety problems.

For much larger amount of T, mass and/or pressure measurements, the same way to measure other hydrogen isotopes, are employed (see Fig. 2.2). For highly concentrated T in liquids and solids, its decay heat allows a calorimetric measurement, but its accuracy is only 10^{-2} – 10^{-3} . At present, T measurements except for the

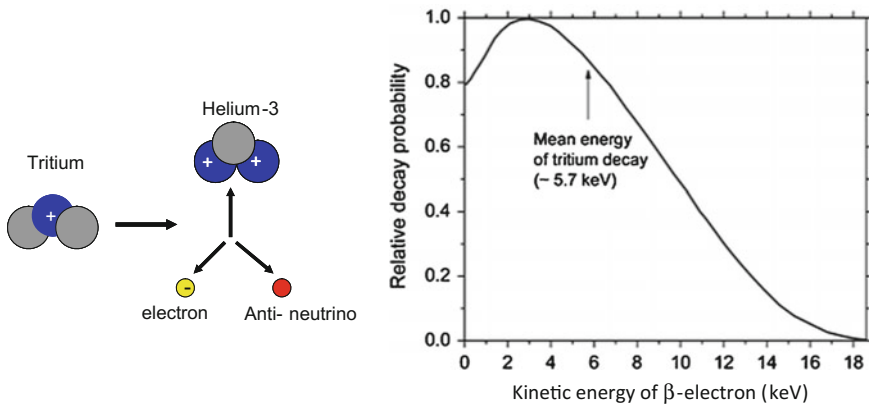


Fig. 2.1 Decay scheme of T and energy distribution of β -electrons emitted at T decay

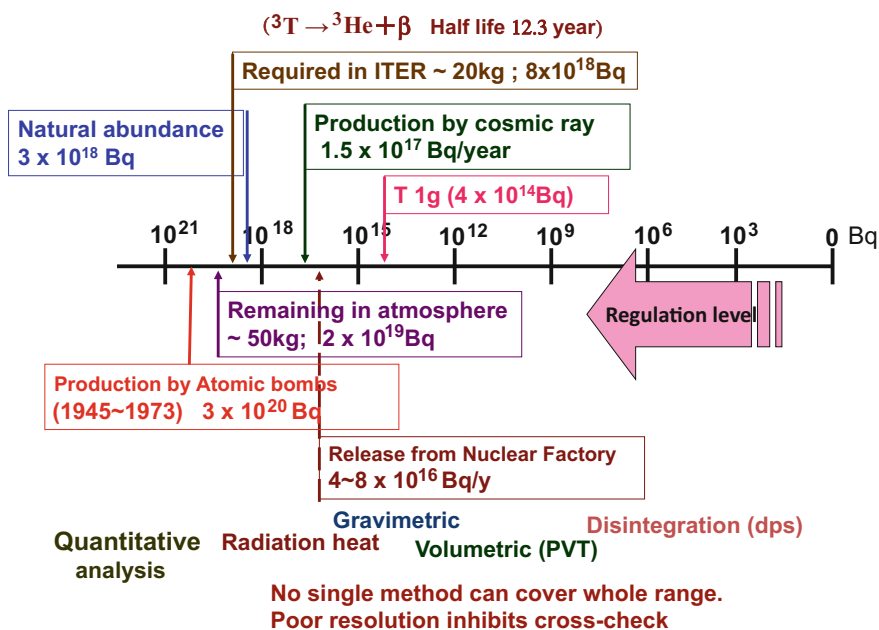


Fig. 2.2 Tritium abundance and measurements [6]

β -counting give the accuracy of only 2–3 digits and any loss of T less than 0.1 % is hardly possible to detect. Therefore, attaining exact T accountancy in a fusion reactor seems quite hard and significant efforts are required to comply with the safety regulation. Details of quantitative analysis of T are given in Chap. 7.

Since public exposure to T is regulated at a level as tiny as a few Bq/cm², T must be strictly confined in T handling systems. But it is quite hard in reality, because T having the same nature as hydrogen easily dissolves in and permeates through materials resulting in easy leakage. T escaping from the handling system by permeation and leakage contaminates materials outside the system. Hydrogen permeation is described in Chap. 9. Furthermore, once T escapes from the system, it is easily replaced with H in ubiquitous water to make HTO and in organic molecules to make OBT (Organic Bound Tritium). Both are more hazardous than HT.

2.2 Tritium Resources

For any energy system, resources of fuel are critical. Since hydrogen in natural water or standard mean ocean water (SMOW) contains 0.016 % deuterium (D) (see Table 1), we can extract D from the water mostly by means of electrolysis. Because

the surface waters of earth contain more than 1 billion tons of D, D can be considered to be a practically infinite fuel resource.

In nature, T is produced by cosmic rays, mostly high-energy neutrons as,



At the same time, T decays with half-life of 12.3 years or around 5 %/year. Consequently, the amount of T is balanced between the production and the decay resulting nearly constant concentration of 10^{-18} in natural hydrogen (T/H) which is referred as tritium unit (TU). Thus, the total abundance of T on the earth is around of several kg ($\sim 3 \times 10^{18}$ Bq) (see Fig. 2.2), existing with chemical form of HTO [7]. Before Partial Test Ban Treaty at October 1963, nuclear weapon tests produced much more T than natural abundance. Afterward, such artificially produced T has been decaying to the present level of ~ 50 kg. Still every nuclear reactor is releasing $\sim 10^{14}$ Bq/year.

Since T is a critically important component for nuclear weapons, it is produced in nuclear fission reactors, for example, in heavy water cooled reactors,



and in a special rod containing ^{10}B or ^6Li in light water cooled reactors,



Reaction (2.6) is used in CANDU (CANada Deuterium Uranium) reactors, and produced T is commercially available for peaceful use, while reaction (2.8) is mostly for T in military use.

The total T inventory for the military use is estimated to be around 600 kg at maximum during the early 1970s. Due to various tests of nuclear weapons, T concentration in environment was about 6000 times higher than the natural abundance at 1963 just before the stop of nuclear weapon test at Partial Test Ban Treaty. Afterward, the natural abundance decayed significantly and nearly comes back to the level of old days when no T was artificially produced. Still commercial light water reactors are releasing T around 1 g/year/ 1 reactor.

According to Ref. [8], the total T inventory or reserve in USA for nuclear weapons was estimated to be 60–100 kg at 1984 and the amount would not change very much afterward or has very likely decreased as evidenced by current shortage of ^3He , of which considerable amount has been recovered from nuclear facilities as a decay product of T.

Now an important question arises, that is, how much T is burned in a fusion reactor? From a simple calculation according to the famous Einstein equation,

$E = \Delta Mc^2$, where E is released energy given by the D-T reaction, ΔM , the mass of D and T burned (disappeared), and c is the velocity of light, 56 kg of T is burned per GW year of fusion power (GWth) for a D-T reactor. (About one-third of the fusion power is converted to electric power represented by GWe.) Currently, around 100 g of T is produced per year in a standard CANDU reactor and 20–25 kg T (mainly in Canada) will be available for operation of ITER. Still the amount of initial inventory in ITER, which is the amount of T simply dissolve in materials used in ITER and immobilized (hard to recover), is quite uncertain, but could be significantly large, as discussed in Chap. 3. This means that at present we have no T reserves for DEMO.

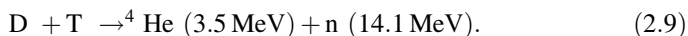
Therefore, any fusion reactor should breed T in its blanket system and the T breeding should have enough margins to compensate T inventory in its tritium systems and the 5 %/year decay (fuel self-efficiency) and reserve for new reactors.

In addition, as discussed in Chap. 3, very poor burning efficiency in a tokamak fusion reactor requires huge amount of total throughput and hence rather large reserve. By using an optimized blanket system, overall T breeding ratio (TBR) in a fusion reactor is expected to be around 1.2 at maximum [9]. In this respect, overall T retention rate in a reactor system including T recovering, refining, isotope separation, and refueling systems must be below 0.1.

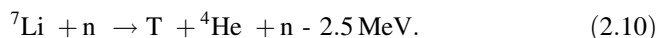
2.3 Tritium as Fuel of D-T Reactor

As described in previous section, it is difficult to get sufficient amount of T even for DEMO. T must be bred by a nuclear reactor or an accelerator with nuclear reactions using Lithium (Li). Since the attainable breeding ratio does not seem to quite high as discussed in Chap. 3, T retained in any T systems in a reactor should be recovered not only for T safety but also for T economy.

As described in Chap. 1, the energy released by D-T reactions is distributed to ^4He and neutron,

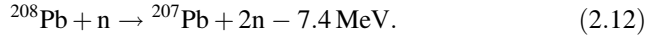


The energy of ^4He is to be used for heating plasma to sustaining the D-T burning and the energy carried by neutron is transformed to heat in a blanket system for electricity or other use, like hydrogen production. At the same time, neutron is used to breed T as



Since, as described in Chap. 1, natural Li includes both ^6Li and ^7Li with their natural abundance ratio of 7.5–92.5, utilization of Li in the blanket seems easily to

give breeding ratio of more than 1. To get enough margin on T breeding, neutron multipliers, such as Be and Pb are planned to use in the blanket,



However, the neutron multipliers (Eqs. 2.10–2.12) require external energy, and their usage in the blanket reduces the output power. By using the optimized blanket system, the overall T breeding ratio in a fusion reactor could be around 1.2 at maximum. In this respect, the overall T recovering rate from the reactor vessel and other T processing systems must be kept as large as possible, above around 0.95; otherwise, T fuel for the 2nd reactor is not constructed. At the moment, this seems very hard owing to large in-vessel T inventory as described later.

2.4 Isotope Effects Among Atoms and Molecules

Various properties of hydrogen isotopes are summarized and tabulated in a book by Souers [3]. In Table 2.1, some properties important for fusion application are summarized.

Because of large mass differences among the three hydrogen isotopes (H, D, and T), isotope effects are most appreciable in properties relating to the velocities of ions, atoms and molecules, and vibration and rotation of atoms and molecules.

Simple molecular kinetics tells that an average velocity $\bar{v}$ of a gas molecule with its mass m at temperature T and pressure P is given by,

$$\bar{v} = \sqrt{\frac{8kT}{\pi m}} \quad (2.13)$$

where k is the Boltzmann constant. Therefore, the velocity differences among the three hydrogen isotopes of H_2 , D_2 , and T_2 are,

$$\bar{v}_{\text{H}}/\bar{v}_{\text{D}} = \sqrt{2} \quad (2.14)$$

$$\bar{v}_{\text{H}}/\bar{v}_{\text{T}} = \sqrt{3} \quad (2.15)$$

and

$$\bar{v}_{\text{D}}/\bar{v}_{\text{T}} = \sqrt{3/2} \quad (2.16)$$

Since incident flux J to the surface of a gas container is given by gas density n times its velocity $\bar{v}$

$$J = n\bar{v}/4 = \frac{P}{(2\pi mkT)^{1/2}} \quad (2.17)$$

Accordingly, incident flux ratios under the same pressure are given as,

$$J_H/J_D = \sqrt{2} \quad (2.18)$$

and

$$J_H/J_T = \sqrt{3} \quad (2.19)$$

These differences appear in reaction rates of any chemical reactions including hydrogen. Isotope separation with using the different fluxes is one of the most useful applications. A turbomolecular pump shows significantly larger pumping speed for D_2 than H_2 . It should be noted that the sensitivity factors of a vacuum ionization gage are also square root mass dependences for all hydrogen isotopes, because the gage measures the molecular fluxes but not the pressure of gases directly. Better confinement time of D than H in plasmas is a well-known and an important isotope effect. All those differences influence the amounts of D and T confined in plasma, and simple fueling with equimolar mixture of D and T does not ensure the most effective D–T burning.

Observed isotope effects between H and D often deviate from such square root mass ratios. For T, little observations have been reported and extrapolation of the isotope effects between H and D to those for D and T might not be correct. Behaviors of D and T fuels are discussed in detail at Chap. 5.

2.4.1 Hydrogen in Plasma (Atoms and Ions)

Different from the kinetic properties given above, differences in electronic properties, ionization, excitation of electrons for atoms and molecules among three isotopes are quite small as given in Table 2.2. This will make distinction of D and T in boundary plasmas difficult as described in the next section. In burning plasma, fuels are fully ionized in core plasma, while neutral atoms and molecules are mixed in boundary plasma. Since heavier ions are confined better in plasma, the mass difference strongly influences confinement times of deuteron (D^+) and triton (T^+) and also evacuation speeds as described above. This requires special care on fueling of D and T in plasma core to maintain high burning efficiency, and hence, determination of D/T ratio in the plasma core is critically important.

Optical emissions from excited atoms and molecules are one of the most important diagnostics to characterize boundary plasmas. Hydrogen in the boundary plasma is usually observed by Balmer series emissions. In particular, α - and β -emissions are used to determine recycling of fuels at plasma-facing surfaces, the

Table 2.2 Balmer series emissions of H, D, and T [3]

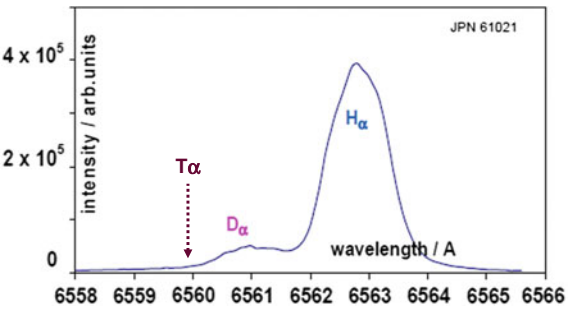
Balmer lines	H (nm)	D (nm)	T (nm)	H–D (nm)	H–T (nm)
α	656.28529	656.10104	656.04166	0.18425	0.24363
β	486.1362	486.00028	485.9563	0.13592	0.1799
γ	434.04946	433.92829	433.88902	0.12117	0.16044
δ	410.1765	410.06191	410.02479	0.11459	0.15171
ϵ	397.0072	396.89992	396.86329	0.10728	0.14391
IP (cm ⁻¹)	109,678.76	109,708.608	109,718.538	-29.844	-39.774
IP (eV)	13.598	13.602	13.603	3.7001 meV	4.9313 meV

ratio of incoming and/or outgoing fluxes. And also temperature of the boundary hydrogen plasma can be determined by the intensity ratios of the Balmer series emissions.

Since electronic structures of three isotopic atoms (H, D, and T) are not much influenced by the differences of atomic masses, the differences in electron energy levels of excited states and ionization energy of each atom are within 3–5 meV and differences in wave lengths of Balmer lines emission are within 0.1–0.2 nm as given in Table 2.2. To distinguish three isotopes in plasma by observation of the Balmer series emission, a spectrometer with very fine resolution is required. Figure 2.3 is an example of the fine-resolution optical measurement in JET [10]. H α and D α were mostly separated. Nevertheless, it seems difficult to quantify concentration of H, D, and T, or any isotope with lower concentration becomes difficult to detect quantitatively.

In burning plasma, because of high temperature, fuel particles of D and T are fully ionized giving no optical emissions except Bremsstrahlung. Therefore, it is quite hard to know concentrations of D and T in the burning core which is indispensable to control D–T burning though fueling ratio of D/T. Since in a D–T fusion reactor, D–D reactions always accompany with D–T reactions, neutrons accordingly produced can be separated with their energies, 2.4 and 14 meV. Hence, measurements of neutron energy distribution in a reactor is expected to be used for a diagnostic to determine, burning rate and D/T ratio in the burning plasma, though it does not seem easy.

Fig. 2.3 Balmer lines emission observed at boundary plasma of D and H mixture in JET (reprinted with permission from [10])



2.4.2 Molecules

As already described, various hydrogen molecules are distinguished. Considering nuclear spins, they are o-H₂, p-H₂, HD, HT, o-D₂, p-D₂, DT, o-T₂, and o-T₂. Since the differences caused by the nuclear spins are only appreciable at very low temperatures, they are not considered in handling of fusion fuels, except for making ice pellets for the fueling. Hence, 6 molecules H₂, D₂, T₂, HD, HT, and DT are generally separated. H is not the fuel but always remains as an impurity and hard to remove any handling systems of D and T. A few percentage of H is expected to be in the exhausted fuels from a reactor vessel.

As described above, translational motion, molecular vibration, and rotation are significantly different among 6 isotope molecules and they influence their thermal properties of their gases, liquids, and solids. The rotational and vibrational levels of hydrogen molecules are quantized and owing to quantum effects, three isotopes show complete different energy levels as shown in Fig. 2.4 [3]. One can note that the ratio of the zero point vibration energies of H and T is just 0.58 equal to their square root mass ratio. Molecular hydrogen spectroscopy in boundary plasma of TEXTOR shows that observed spectra for pure H and pure D discharges agree quite well with the theoretical ones, while the spectral lines for H and D mixtures are overlapping and difficult

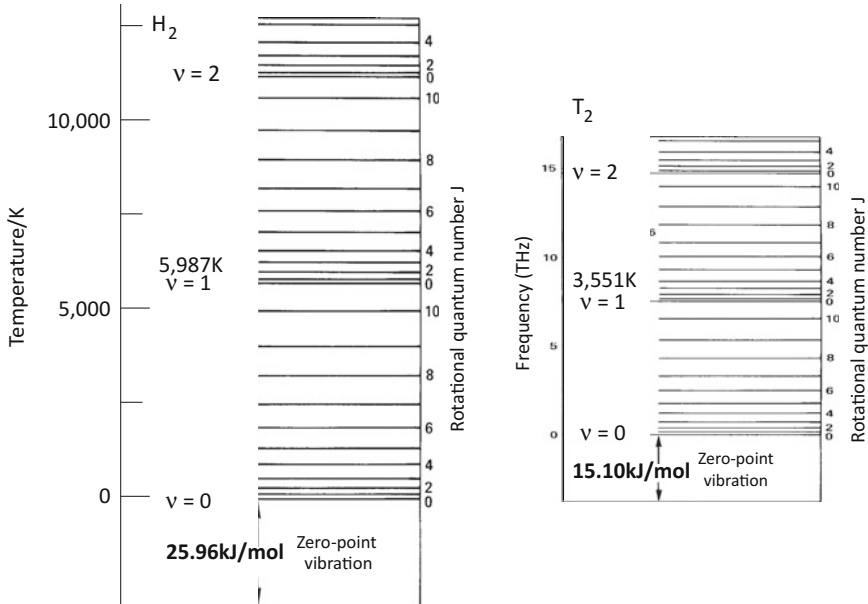


Fig. 2.4 Vibrational and rotational energy levels of isolated H₂ and T₂ (reprinted with permission from [3])

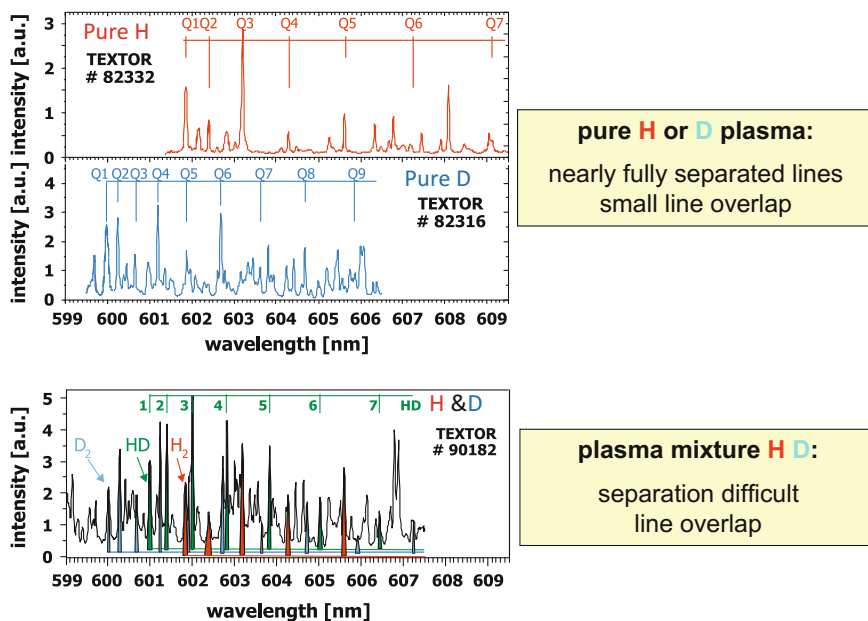


Fig. 2.5 Examples of molecular spectra obtained from plasmas of pure H, pure D, and HD mixture in TEXTOR (reprinted with permission from [11]). For pure H or D, no overlap in dominant lines, while many lines overlap for mixture of H and D to make separation difficult

to separate (see Fig. 2.5 [11]). Hence, molecular spectroscopy seems challenging to be applied for quantitative analysis of H, D, and T in boundary plasmas.

For quantitative analysis of H_2 , HD, HT, D_2 , DT, and T_2 in gases near room temperature, Raman spectroscopy can be applied to distinguish the differences of rotational energy levels of each molecule, which is described in Chap. 12.

The different rotational and vibrational levels give different heat capacity and thermal conductivity of ice pellets of D_2 and T_2 . The heat capacity of solid hydrogen shows quite good square root mass dependence as shown in Fig. 2.6 [12]. As noted above, spin isomers of o- D_2 , p- D_2 , DT, o- T_2 , and o- T_2 give different thermo-physical properties. This directly influences manufacturing the ice pellets, and the decay heat of T could enhance melting behavior and ablation or evaporation flux of D and T in fueling by DT pellets.

Since most of the isotope effects in interaction of hydrogen and materials include surface kinetics, differences in hydrogen fluxes to and from solid surface among three isotopes should be taken into account.

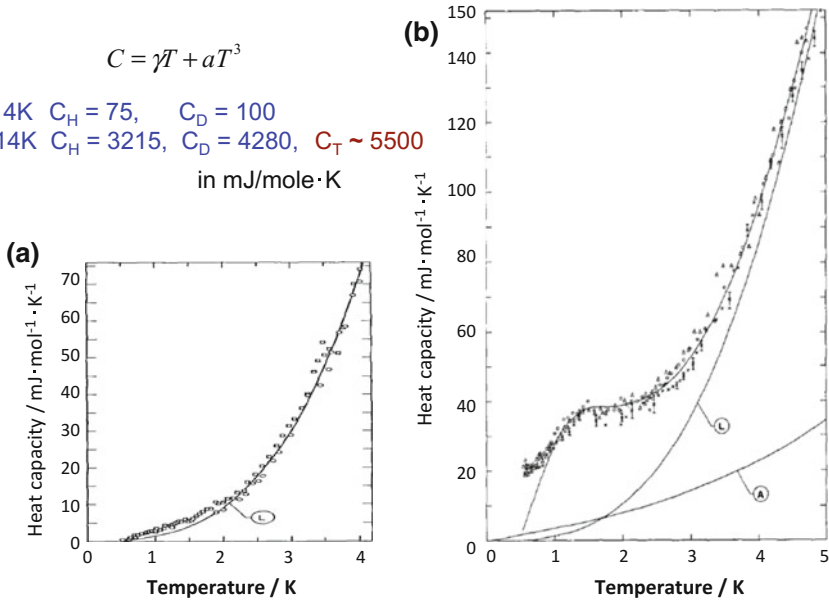


Fig. 2.6 Temperature dependence of heat capacity of H and D (reprinted with permission from [12]). **a** Pure H_2 Solid curve labeled L (proportional T^3) is the lattice contribution. **b** Pure D_2 . Upper curve is theoretical specific heat including lattice contribution given by L and the contribution of empty calorimeter given by A

2.4.3 Hydrogen Properties in Materials

Among various properties of hydrogen in materials, solubility, diffusivity, and permeability are quite important not only for T safety but also for T fuel self-sufficiency. Interaction of hydrogen and materials has been one of the most important subjects in materials science, too.

Compared to other elements, hydrogen is easily dissolved or trapped and migrates in materials. Hydrogen properties in materials have been extensively studied, because of its importance in hydrogen embrittlement which is one of the causes of the materials failure in large structure, bridges, ships, gas tanks, and so on [4].

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

$$S_H/S_D = \sqrt{2} \quad (2.20)$$

and

$$S_H/S_T = \sqrt{3} \quad (2.21)$$

and

$$D_H/D_D = \sqrt{2} \quad (2.22)$$

and

$$D_H/D_T = \sqrt{3} \quad (2.23)$$

Similar isotope effects are often observed in hydrogen permeability Φ as

$$\Phi_H/\Phi_D = \sqrt{2} \quad (2.24)$$

In diffusion-controlled permeation, permeability is represented by,

$$\Phi = SD \quad (2.25)$$

According to the isotope effects in S and D , however, Eq. (2.25) is not possible; that is, the isotope effect could not be represented such simple equations.

Large mass differences among hydrogen isotopes result in large difference in their quantized rotational and vibrational states. Contradictions among Φ , D , and S are most likely owing to neglecting of the isotope effects in the rotational and vibrational states in solids. Actually significantly large isotope effects originating from quantum effects are observed in diffusion coefficients at temperature far below RT as given in Fig. 2.7. The diffusion coefficients do not follow single Arrhenius relationship in their temperature dependence, and the upper shift at lower temperatures is attributed to the quantum tunneling effect. Because of the lightest mass of protium, among the

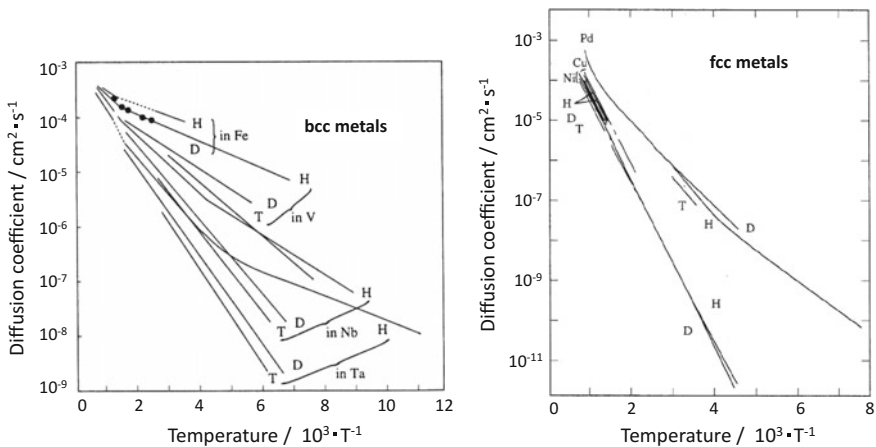


Fig. 2.7 Comparison of diffusion coefficients of H, D, and T in some metals

three isotopes, the quantum effect is the most appreciable. Above the room temperature, the difference becomes less and the differences in not only diffusivity but also solubility and permeability of H, D, and T are within the factor of the square root mass ratio, which is in most case near the experimental error. Therefore, isotope differences in handling of large amount of gases of H, D, and T are not likely over the square root of their mass ratio.

Isotopic effects between H and D are also appreciable in the interactions of hydrogen and materials such as reflection, reemission, diffusion, and trapping. In a fusion reactor, plasma-facing materials are exposed to T plasma or subjected to high-energy T ions and atoms. The isotopic effects among high-energy hydrogen isotopes, in particular between D and T, have not been studied yet. Their difference in confinement time, recycling at the plasma-facing surface, and evacuation could largely influence in burning efficiency.

In case of handling T as a tracer, T sometimes gives anomalous isotope effects mainly caused by isotope exchange with H in ubiquitous water as discussed in the next section.

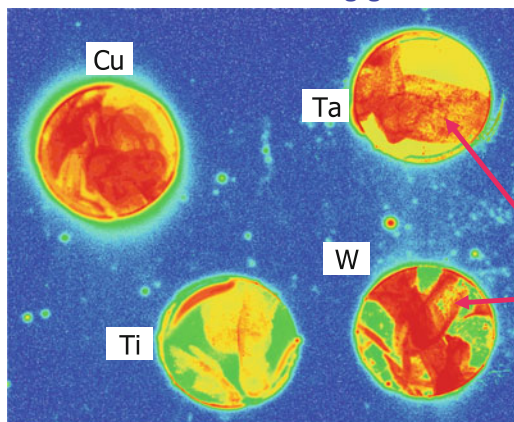
2.5 Contamination by Isotope Exchange Reactions

2.5.1 *Isotopic Replacement on Solid Surface as a Cause of T Contamination*

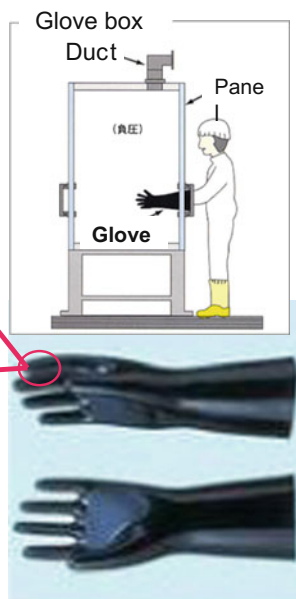
Contamination of materials surfaces by T is one of the most important safety concerns and understanding of the contamination mechanism is quite important to reduce and/or remove the contamination (detritiation). Due to reactive nature of hydrogen, T can be easily transferred from a highly contaminated surface to lower contaminated or noncontaminated surfaces, leading to cross-contamination or multistep contaminations in T handling systems. In ITER, safety handling of a divertor cassette, one of the largest components, is the most serious concern [13]. The cassette retains very high levels of T on their surfaces, which easily transfers to arms or gloves of remote handling systems or gloves boxes. Once the surface of the device/equipment in the systems is contaminated, T can be easily transferred to any materials next to be handled. Figure 2.8 shows an example of cross-contamination remaining fingerprints of T activity on metal plates in a glove box in which heavily contaminated materials were handled [14]. Gloves as essential equipment in a T handling system are always contaminated and T on the glove surface is immediately transferred to noncontaminated materials. The cross-contamination is caused by easily replacement of T with the ubiquitous lighter hydrogen isotopes such as protium (H)/deuterium (D) in water and hydrocarbons in atmosphere. The consequence is multistage contamination and sequential reduction of the contamination by second and third glove-boxes might not so effective.

Easy transfer of T contamination

Metal plates exposed to D plasma in TPL and handled in a T handling glove box



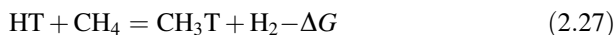
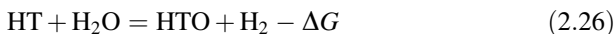
Traces of glove fingers



Possible contamination by permeation

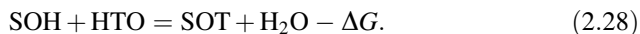
Fig. 2.8 T contamination of metal surfaces caused by handling in a glove box [14]. High T activity appeared as the traces of the gloves

The most important mechanism of the cross-contamination is isotope exchange reactions of T with the ubiquitous lighter hydrogen isotope, protium (H), in water to become HTO and hydrocarbons in atmosphere, as given in following reactions,



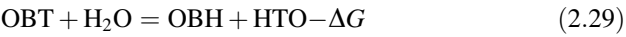
Similar to the reaction (2.27), T involved inorganic molecules, referred as Organic Bound Tritium (OBT) easily remains in a human body and more hazardous than HTO.

Such isotopic replacements easily proceed at any solid surface (S) as



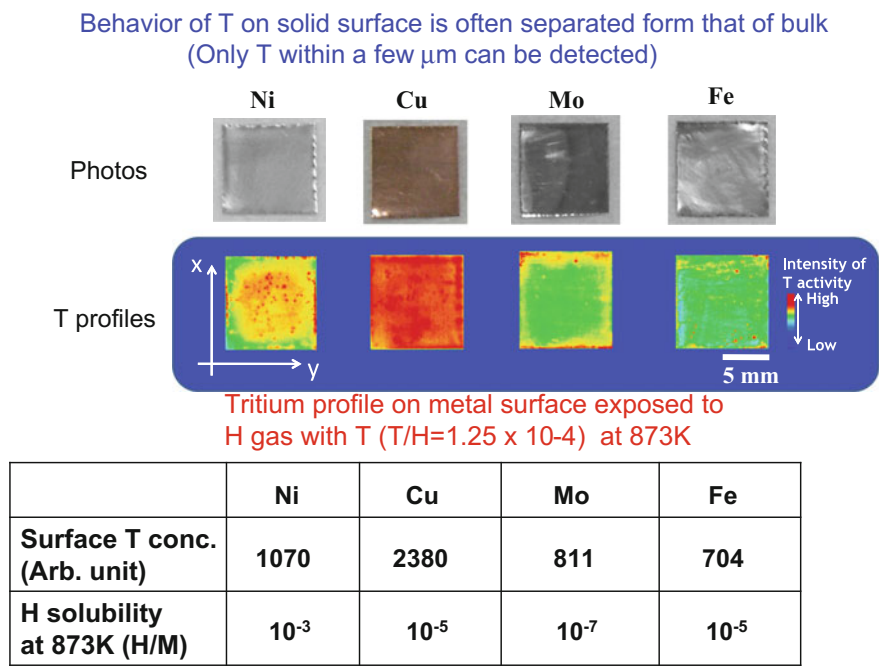
for tritiated water case. This is the main cause of the contamination of the gloves in Fig. 2.8. In addition, T and HTO are very easy to permeate through metals and organic materials, respectively, which also cause the T contamination.

Tritium disintegration at skin is not so important owing to thin penetration depth of the β -electrons, but T can penetrate into a human body by isotopic replacement with light water in tissues to be very hazardous. Once T is going in human body and tissues, a few weeks are needed to be isotopically replaced by H from drinking water as,



On the contrary, dipping in water and/or wiping with wet papers of the T-contaminated surface is very effective for detritiation. For example, drinking beer is very efficient to enhance the replacement of T in a human body. It is also well known that the addition of water vapor is very much effective to recover T in structure and breeding materials and on the surface of any materials, but T diluted in water is very hard or needs lots of energy to be recovered, which increases fusion energy cost.

Owing to those easy surface replacement, surface T concentration is not necessarily to be the same as that in the bulk as shown in Fig. 2.9 [15]. In the figure, the surface T levels are compared for various metals in which tritiated hydrogen was dissolved at 873 K. One can note that the surface T concentrations are quite different with the solubility. That is because, the surface of most of metals was oxidized with different level (or different oxide thickness) and the amount of



retained OT is higher for more oxidized surface. However one should be careful that once some of the surface T was smeared out, T was supplied from the bulk by diffusion and the surface T level recovered easily. Of course if bulk T concentration was very high, the surface-segregated T could be neglected.

2.5.2 Isotopic Replacement with Residual or Impurity H in Materials

Any materials retain hydrogen as an impurity with more than a few tens ppm. Therefore, the isotopic replacement described in the previous section is not limited to the surface. Easy diffusion of hydrogen in the bulk enhances the replacement. In case of H in industrial use, the replacement of H retained in the materials or ubiquitous water with H throughput does not matter. The H throughput into an H processing/handling system is much larger than the amount of H on surface and in bulk of the materials used in the system and tiny loss is not accounted. For T safety, on the other hand, any T retained in the system should be accounted. Furthermore, since absolute amount of T handled in most of the T systems is much less than the amount of H in industrial systems, T retained in the systems does matter. This is quite important in T handling system and T measuring systems and described in Chap. 3 in more detail.

Since the bulk replacement is diffusion-limited process strongly dependent on material's temperature, its replacement rate is generally much slower than that of the absorption and surface replacement and is not appreciable at RT. However, in the fusion reactor vessel probably operated at 700–800 K, the contribution of the isotopic replacement of T with H in bulk would have significant contribution on T retention owing to quite large volume of the vessel. Furthermore, T escaping from the plasma impinges and is retained in a little deep inside of the plasma-facing materials. This enhances the isotopic replacement with H in bulk. The bulk T inventory is discussed in Chaps. 3 and 5.

2.6 Effect of β -Emission on T Behavior

Liquid T₂O is corrosive due to self-radiolysis. The β -particles travel only about 6 μm in water and deposited energy during the travel causes decomposition of water molecules (water radiolysis) to produce gaseous hydrogen (including HT) and redox-active products including highly reactive hydroxyl radicals ($\cdot\text{OH}$ and $\cdot\text{OT}$). This is one of the main causes for T to be hazardous and is discussed in Part III.

Here only direct effect of β -emission on behavior of T fuel is described. In reactions including T, anomaly owing to β -emission and recoil He atoms is sometimes claimed. However, due to the small penetration of β -electron, its direct influence on neighboring T is hardly observed. Instead, the electron colloid with neighboring molecules or atoms, whatever chemical forms, electron excitation of themselves or neighbors could lead some reactions which are not possible thermally like the self-radiolysis. This kind of reaction is highly possible to occur at solid surfaces in water including T or solid surface including T in gas. The reaction caused by the electron excitation could be either reductive or oxidative.

Quite recently, a corrosion test in rather high-dense tritiated water has shown corrosion enhancement [16]. The direct effect of the T decay to ^3He including molecules must be quite seldom unless T concentration is quite high or T is not diluted in H or D. When T concentration in materials is very high, deposited electron energy raises the materials temperature, often referred as decay heat. The decay heat of pure T is 324 mW/g or 67.5×10^{-3} cal/g s, which will cause its temperature rise of 1 K for a few 10 s seconds. The decay heat is not quite large but would influence ice pellet production. The effect of decay heat was observed as enhanced T release from graphite tiles used in T discharges in JET.

If isolated particle retains certain amount of T, electron emission makes the particle charging up and the behavior of the particle becomes controlled by static electric field [17].

2.7 Tritium Issues in Burning Plasma and Self-sufficiency of Fuels

The burning efficiency in a fusion fuel cycle is, unfortunately, very poor; only a few % or less of input tritium in the reactor vessel burns and the majority must be exhausted and recovered to recycle. However, as described in Sect. 2.2, T resources are not enough even for a DEMO reactor. Therefore, T should be artificially produced or bred in a blanket of a fusion reactor. Since T decays about 5 % a year, T breeding in the blanket should have enough margins to compensate not only the T inventory in all T handling systems, but also the disappearance by the decay.

With using an optimized blanket system, overall T breeding ratio in a fusion reactor is expected to be around 1.2 or a little less. In this respect, the in-vessel T retention rate $((\text{fueling} - (\text{burning} + \text{recovering}))/\text{fueling})$ must be below 0.1. Otherwise, T for the second reactor is not generated. At the moment, it seems very hard to make the in-vessel fuel retention rate to be below 0.1, as described later.

Suppose the burning efficiency is ~ 3 %, to burn 56 kg/year of T for 1GWth of fusion power, total throughput becomes ~ 1800 kg/year. Accordingly, fuel retention rate must be below 0.05 % to keep the in-vessel T inventory below 1 kg required for the safety in ITER. It is of course not necessary to reserve 1800 kg of T

at one time. But T inventory in other T systems than the vacuum vessel should be also taken into account.

In the present plasma devices, light hydrogen (H) always remains in the deuterium fueling operation, of which origin was discussed in the previous section. Whatever the sources are, contamination of D and T by H is unavoidable and additional effort to remove H in an isotope separation process is required. For enhancement of radiative cooling of the plasma and also for disruption mitigation, impurity seeding such as Ne and Ar will be employed, which could significantly dilute the fuels. In addition, various chemical forms of hydrocarbons will be formed from carbon plasma-facing materials and impurity carbon in W, if they are used. Thus, D and T fueled into the vacuum vessel are exhausted as a heavily contaminated gas of D and T with H, hydrocarbons and inert gasses (He, Ne, and Ar). Since only D and T must be recycled, the exhaust gas must be recovered, refined, isotopically separated, and refueled. One should remind that this is requested from the self-fuel efficiency but not from the safety.

As noted in Sect. 2.4, behaviors of D and T not only in the plasma but also in materials are significantly different from each other: for instance, confinement times, fueling efficiencies, escaping fluxes from the plasma, retention rates in materials, evacuation rates from VV, and so on. Therefore, the fueling rate is necessarily the same and must be optimized to attain the highest burning efficiency. Because of very poor fueling efficiency of gas puff [18], T is injected mainly by pellets and NBI, while D would be fueled by gas puff and NBI [19]. Such different fueling between D and T does not assure homogeneous mixing of D and T in plasmas. Therefore, it will not be easy to maintain the appropriate D/T ratio in the plasma for continuing the highest burning efficiency. The concentration of D and T in the burning plasma must be separately measured to achieve feedback fueling independently. It is, however, quite difficult to measure the concentration of D and T separately. Separate detection of neutrons with energies of 14 MeV (produced by D-T reactions) and of 2.4 MeV (D-D reactions) is one of the most promising ways, but the signal does not necessarily give spatial distribution of D/T ratio in the plasmas. Different confinement times of D and T would result in their inhomogeneous distribution particularly in their radial distribution.

Furthermore, retention rates of D and T in the plasma-facing wall is not necessarily the same and release rates from the wall may be different as well. This means that if some local thermal load like an ELM hits the wall, thermal release of D and T is different in their amounts and this disturbs the D/T ratio in the plasma. Owing to the huge wall inventory, a small change in the D/T ratio of the wall retention could cause significant change in the D/T ratio of the burning plasma. Thus, we are going to face difficulty in controlling the D/T ratio in the plasma to attain and keep efficient burning. Again, the initial phase of ITER discharges using H and D shall be used intentional control of H/D ratio in plasmas.

2.8 Summary

Hydrogen properties important for fusion application are described. Because of large mass differences, isotope effects among H, D, and T in dynamic behaviors relating atomic and molecular motions are quite large. In particular, differences of confinement in plasma, retention in plasma-facing wall, permeation, and pumping speed between D and T would require independent controls in fueling of D and T to keep D-T burning to be efficient. Unfortunately, however, D/T ratio in the burning is quite hard to observe (determine). The fueling and burning control are one of the most difficult remaining issues to be solved in ITER. Poor T resources also require appropriate fueling, breeding, and full recovering of T to recycle the fuel to attain the fuel self-sufficiency.

For handling of massive amount of T as the fuel, effects of β -electrons emitted at the decay are not concerned, except the 5 % disappearance by the decay. In storage and for safety, the decay could give some impacts through electron excitation by the β -electrons. Self-radiolysis of tritiated water is one of the most important effects, because it is the main cause of hazard of T for human bodies. Isotope exchange of T with H in ubiquitous water molecules, in particular, adsorbed on solid surfaces results in easy cross-contamination. Charging up small particles retaining T with high concentration could be another cause of the contamination.

Nevertheless, the decay heat of 324 mW/1 gT is rather small and would not give significant effect in handling of massive amount of T except at very low temperature for making ice pellets.

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