Hydrogen and its detection in fusion and fission nuclear materials a review

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Abstract

Fusion and fission reactions are profoundly dependent on hydrogen for sustained reactions. Fusion is fueled by the isotopes of hydrogen, and predominantly hydrogen-based moderators slow fission neutrons to propagate chain reactions. Intentional tritium production in fusion and concomitant tritium production in fission reactors introduce challenges. The technology to efficiently extract, harvest, and quantify tritium in and from advanced fission molten salt coolants and fusion molten tritium breeder materials will require more pronounced research and development. Measuring and quantifying hydrogen is necessary in all areas of nuclear materials. Although many characterization techniques cannot directly detect hydrogen, numerous techniques provide the necessary information to understand the behavior of hydrogen in nuclear materials.

Keywords
Hydrogen, fusion, fission, plasma facing components, cladding, moderator, metal hydride, characterization

1. Introduction

Hydrogen plays a central role in nearly every form of power production used by mankind. The hydrocarbons in fossil fuels exist because of the sun's energy and the hydrogen in the biosphere that once vivified ephemeral life. Combustion reactions of hydrocarbons (wood, coal, then fossil fuels) have been the mainstay energy source of humans for millennia. Modern and advanced technologies still rely on hydrogen for electricity production. The power of the sun is derived through proton-proton fusion reactions, which heat the earth and drive electricity generation through solar and wind power. Fuel cells most often rely on the recombination of hydrogen and oxygen to generate an electrical current. Hydrogen also plays a central role in nuclear power. Fission frequently, but not exclusively, utilizes hydrogen bearing materials to provide neutron moderation. And finally, conventional thermonuclear fusion for energy production uses hydrogen as the primary fuel.

While hydrogen is ubiquitous in electricity production, its interaction with materials is complicated and poorly understood. Furthermore, detecting hydrogen in materials can be challenging. This mini-review aims to highlight the behavior of hydrogen in nuclear systems, both fission and fusion, and the available methods for measuring hydrogen in nuclear materials.

2. Hydrogen in fusion

Hydrogen, and especially its isotopes deuterium and tritium, plays a central role in fusion. Deuterium-tritium (DT) fusion has the highest reaction cross section at the lowest center of mass energy (see Figure 1) and is frequently viewed as the most readily achievable form of fusion. Tokamaks compress ionized deuterium and tritium with strong magnetic fields in combination with intense heating to achieve conditions sufficient to fuse the two. This process releases a 14.1 MeV neutron and an energetic (3.5 MeV) helium nucleus. Nearly every concept for advanced fusion will still use some combination hydrogen, whose resulting reactions provide varying advantages. For example, aneutronic fusion with direct energy conversion can be achieved by fusing a proton and boron-11 in the $p(^{11}B,^{4}He)$ + 8.7 MeV reaction, or deuterium helium fusion in the $D(^{3}He, {}^{4}He)p$ reaction. One theoretical non-hydrogenic fusion reaction $^{6}Li(n,nD)^{4}He$ is shown in Figure 1, although no fusion concepts have been proposed under this configuration [1].
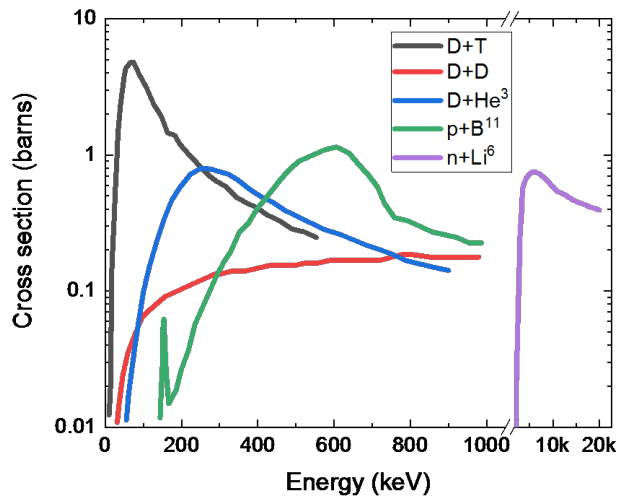
Figure 1. Fusion cross-sections for various fusion fuels as a function of center of mass energy. [2,3] The n(Li6

Managing large quantities of hydrogen (i.e., deuterium and tritium) for fusion research and future reactors is challenging. Ion and energetic neutral DT atoms penetrate and become implanted into plasma facing components. Hydrogen is soluble to some degree in all metals and therefore can be retained in the plasma-facing wall materials or migrate through the wall with a driving force according to concentration and/or temperature gradients. Hydrogen that reemerges out of the plasma facing components back into the plasma is also problematic as hydrogen that accumulates at the edge of the plasma is of lower energy than the core plasma and causes steep energy gradients and plasma instabilities. Substantial efforts have been undertaken to limit this behavior in some fusion devices [4,5]. It is desirable to utilize a plasma-facing material that 1) exhibits low hydrogen retention, 2) has low hydrogen solubility, and 3) low hydrogen permeability. Relative to other metals, tungsten has exemplary performance in these areas.

Tritium is a beta emitting (18.6 keV) radioactive isotope with a half-life of 12.3 years. As a radioactive substance, operational sites that use tritium are regulated for a maximum given inventory. Therefore, it is especially important that fusion reactors minimize the amount of tritium that is retained in structural materials and maximize the amount available for use as fuel. The means for measuring hydrogen in plasma facing components is limited but essential. The two mainstays for post-mortem measurements of hydrogen retention in plasma facing components are thermal desorption spectroscopy (TDS) and nuclear reaction analysis (NRA). Recently, glow discharge optical emission spectroscopy (GD-OES) has been applied to measure deuterium retention with enhanced depth resolution, elemental specificity, and measurement sensitivity compared to TDS and NRA [6,7]. Decades of experiments have been performed to investigate hydrogen retention under a wide range of fusion conditions [8], but further experiments must be performed especially under more reactor-relevant conditions.

Beyond plasma-facing components, hydrogen transport properties, including solubility and permeability, are important in managing fusion safety. Tritium that permeates out of primary confinement systems can result in accidental exposure and release. As mentioned above, tungsten has low hydrogen solubility and permeability. However, structural components and piping systems that contain and transport tritium cannot readily be fabricated from tungsten. More realistically, these systems will likely be fabricated from various types of steel and may include integrated barriers to mitigate permeation. While hydrogen permeates through all metals, there are practical means to reduce hydrogen permeation. One example is to control the

temperature.   Table 1 shows the permeation rates of tungsten, 316L stainless steel, reduced activation ferritic-martensitic (RAFM) steel, and Pd-25Ag [9].   These materials are shown as examples of low permeation (tungsten), realistic structural materials (SS 316L and RAFM), and a high permeability material (Pd-25Ag). At 800ºC, SS 316L and RAFM steels have high permeabilities (closer to Pd-25Ag than W). However, these realistic structural materials are highly thermally activated making permeability very low at room temperature. Therefore, maintaining modest temperatures with the correct material choice can be an effective means for limiting hydrogen permeation in fusion devices. This practice may be applicable for ancillary systems where tritium is stored and processed but be not as feasible for materials in contact with heat transfer fluids, which pose a particular concern for tritium permeation. Lower temperatures reduce tritium permeability, but come with a tradeoff cost of reduced power conversion efficiency.

Table 1.Permeabilities of several materials at given temperatures. Tungsten has exceptionally low permeability and Pd-25Ag has very high permeability. Values taken from Ref [9].

Material	Permeability ($mol\ m^{-1}s^{-1}Pa^{-1/2}$)		
	27˚C	100˚C	800˚C
Tungsten	1.24E-30	5.38E-26	1.46E-13
SS 316L	3.76E-18	5.16E-16	2.77E-10
RAFM steel	2.93E-15	7.43E-14	4.33E-10
Pd-25Ag	4.62E-09	7.53E-09	2.78E-08

Hydrogen permeates through metals in its atomic form, which requires that gaseous hydrogen dissociate from its molecular diatomic form at the metal surface before it begins to diffuse, and then recombine from atomic to molecular form on the downstream surface. If atomic hydrogen comes in contact with a metal in the form of an acid, ionization, or electrolysis, permeation will readily occur because the potentially-rate-limiting molecular dissociation step is skipped [10]. Under certain conditions, such high hydrogen permeation is highly desired and described as follows.

Tritium is not found in any usable abundance in nature; therefore, it is fundamentally essential to breed tritium for fusion power to become a reality. Tritium is bred through nuclear reactions with lithium containing materials [11,12]. Solid breeder materials have included ceramics such as Li_2ZrO_3, Li_8ZrO_6, Li_2TiO_3, $LiAlO_2$, Li_2SiO_3 and Li_4SiO_4, and liquid breeders have mainly included FLiBe and molten PbLi eutectic [11]. ^6Li has a larger cross section, therefore fusion breeder materials will enrich the ^6Li content from its 7.7% natural concentration to >80% to maximize tritium production. Tritium breeders, regardless of solid or liquid form, are contained in a breeder blanket that surrounds the fusion reactor.

One such conceptual implementation of a liquid tritium breeder is to flow molten PbLi through the breeder blanket. As the Li absorbs neutrons, it is converted into tritium, which then must be harvested and processed to be used in gaseous form. The vacuum permeator (VP) [13,14] is one of several proposed methods for harvesting tritium from molten PbLi [15,16]. Experiments are currently being constructed [17] to test the tritium extraction efficiency of various metallic membranes that have very high tritium permeabilities. These materials primarily include Group V metals on the periodic table. The extraction unit of a VP is similar to a heat exchanger where the high thermal conductivity material is replaced by a high permeability material, and where the secondary side of the permeable metal membrane is pumped at vacuum pressures. Depending on the material composition, the permeable membranes can be susceptible to poisoning and may require interfacial engineering to maintain stable extraction efficiencies

over time. Other proposed methods for tritium extraction include the following. A gas-liquid contactor passes PbLi across a high surface-area packed column while an inert sweep gas flows in the opposite direction. Tritium is removed by the sweep gas and carried to another system where the tritium is removed from the sweep gas. A vacuum sieve tray (or vacuum droplet tower) sprays PbLi from a nozzle into an evacuated chamber. High mass transport in the droplet results in very efficient removal from the PbLi. Finally, getter beds can be immersed in the PbLi to preferentially capture tritium. These getters must be removed, processed, and regenerated for cyclical operation. Each of these potential technologies have advantages and disadvantages, which are nicely summarized in [16].

Tritium recovery from solid breeders is more straight forward than for liquid breeders. Tritium generated in solid breeders diffuses out of the ceramic and is captured by a sweep gas. This gas stream is processed for extraction by either a temperature swing absorption system or getter beds [18].

3. Hydrogen in fission

The role of hydrogen in fission reactors is not always as conspicuous as in fusion systems, where hydrogen is prominently the fuel. However, hydrogen plays a marked role in fission moderators, fuel, cladding, and structural materials.

3.1. Fission moderators

Sustained fission is initiated when a fissile material absorbs a neutron, splits into fission products, and releases 3-4 new neutrons. For most fission fuels, the cross section for fission increases as the new neutrons, born at high energies (>1 MeV), slow down to thermal energies (~0.025 eV). This slowing down process, moderation, occurs as neutrons kinetically transfer energy to surrounding atoms through collisions. The transfer of energy per collision from the neutron is maximized when it collides with an atom whose mass is close to its own. Therefore, hydrogen bearing materials are the most efficient moderators. Other factors, such as temperature stability, neutronic performance (scattering and absorption cross sections), radiation tolerance, heat transfer properties must also be considered when choosing a moderator. Water is used around the world as the predominant moderator in reactors. Water is an extremely effective moderator, readily available, and simultaneously serves as the reactor coolant. One method of comparing different moderators is through their slowing down power (product of lethargy and scattering cross section, $P = \xi \Sigma_s$) and moderating ratio (quotient of slowing down power and absorption cross section, $R = P/\Sigma_a$) [19]. High values for both slowing down power and moderating ratios are desirable, and values for some hydride moderators are shown in Table 2.

Table 2. Hydrogen density, slowing down power, and moderating ratio for various hydrogen moderators [20]. Data for D_2O is from [21].

Material	H density (H/cc)	Slowing down power, $P = \xi \Sigma_s$ (cm-1)	Moderating ratio, $R = P/\Sigma_a$
TiH_2	9.1×10^{22}	1.85	6.3
ZrH_2	7.5×10^{22}	1.45	55
LiH	5.8×10^{22}	1.2	3.5

YH$_2$	5.8 x 10^{22}	1.2	25
ThH$_2$	4.9 x 10^{22}	1.0	5.2
H$_2$O	6.6 x 10^{22}	1.35	70
D$_2$O	-	0.175	6000

Heavy water, D$_2$O, is used as a moderator in CANDU reactors [21]. Because deuterium is twice as massive as hydrogen, less energy is transferred per neutron collision thus D$_2$O has a lower slowing down power. However, neutron absorption in deuterium is substantially less, which results in a significantly higher moderating ratio. Neutron absorption is so low that CANDU reactors can operate with natural uranium (0.72% U-235) without any enrichment. Although low from a neutronic standpoint, deuterium in the D$_2$O does absorb some neutrons resulting in the generation of tritium. Consequently, CANDU reactors employ sophisticated methods for detritiating water [22] and have become a predominate supplier of the world's tritium needs [12]. The operational advantage of not enriching the fuel is somewhat offset by the complexity of enriching heavy water and detritiation. Relatedly, most fluoride molten salt coolants (both fission and fusion) contain lithium, such as FLiBe, and produce tritium. The ^{6}Li content, which has a higher absorption cross section than the 92.3% naturally abundant ^{7}Li, is de-enriched to minimize tritium production (where the opposite is true in fusion applications). Nevertheless, small quantities of tritium, comparable to values in CANDU reactors, will be generated and require detritiation [23].

Hydrogen based moderators can also take the form of metal hydrides, where the hydrogen density in these can exceed even that of water or liquid hydrogen (Table 2) [24]. But the most attractive feature of metal hydride moderators is the thermal stability and operating temperatures that can be sustained. Metal hydride moderators have been used in various test reactor programs, including the Aircraft Nuclear Propulsion (ANP) program [25], Systems for Nuclear Auxiliary Power (SNAP) reactor project [26], and the Gas Cooled Reactor Experiment (GCRE) [27]. Currently, metal hydride moderators are of high interest in the development of modern microreactors [28]. Microreactors are designed to be 2-20 MW reactors that are factory fabricated, transportable, and self-regulating [29]. It is planned that they will operate on high assay low enriched uranium (HALEU, 5-20% enrichment). Given the size and mass constraints to be transportable and the high operating temperatures needed to maximize power conversion, the reactor core requires a highly efficient, high-temperature moderator. Yttrium hydride is of particular interest due to having the highest known temperature stability of the metal hydrides [19]. Figure 2 shows the plateau pressures, or minimum hydride dissociation pressure at a given temperature, of various metal hydrides [24]. The relatively low plateau pressure found at high temperatures is what makes yttrium hydride so appealing. The 1950's ANP program performed a tremendous amount of work on yttrium hydride [30], including multiple successful neutron irradiations [31,32], however nearly no physical-property data is available from legacy post irradiation experiments (PIE). Relatively little research has been performed on yttrium hydride moderators over the elapsing decades until recently and consequently they remain at low technology readiness.
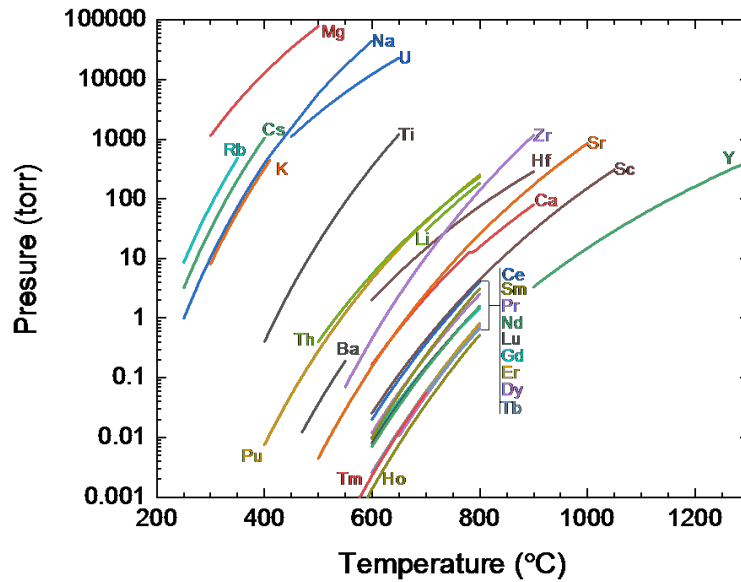
Figure 2. Plateau pressures of the monohydrides and dihydrides of various metals [24].

Yttrium hydride samples were recently irradiated at 600, 700, and 800°C in the Advanced Test Reactor (ATR) [33] and the High Flux Isotope Reactor [34] to quantitatively investigate hydrogen stability following neutron irradiation as well as perform comprehensive PIE to measure thermomechanical and microstructural properties of irradiated YH [35]. Hydrogen concentration in the irradiated samples will be measured using inert gas fusion, mass balance, and qualitatively using x-ray diffraction, each of which are summarized in Table 3.

3.2. Hydrogen in fission fuel and structural materials

Hydrides have been incorporated in some fission fuels - most notably in TRIGA (Training, Research, Isotopes, General Atomics) reactors. TRIGA fuel elements contain a homogenous mixture of uranium and zirconium hydride [36]. The integral union between fuel and moderator results in a prompt negative temperature coefficient that provides inherent safety. Three factors contribute to this effect. 1) Because the fuel and moderator temperatures are uniform, neutrons in the moderated fuel element gain energy as the temperature increases, which decreases the fission cross section, increase the neutron mean free path, and increase the likelihood of absorption in the fuel cladding. 2) Doppler broadening of resonance capture increases with temperature. 3) As the neutron mean free path increases, neutron leakage at the core edge increases [36]. Water in TRIGA reactors functions primarily as the coolant, not the moderator. The SNAP reactor explored both yttrium and zirconium hydride fuels, but ultimately used uranium zirconium hydride fuel due to lower neutron absorption and acceptable temperature ranges for that application [26,37].

3.3. Cladding and structural materials

Fission cladding and structural materials are fabricated from zirconium alloys. Zirconium alloys are generally characterized as very corrosion resistant, having excellent mechanical properties, and, as mentioned above for moderators, having very low neutron absorption.

Although resistant to high temperature corrosion, zirconium reacts slowly with high temperature water to produce a ZrO_2 protective layer and free hydrogen. Over its lifetime in a nuclear reactor, this thin corrosion oxide layer continuously increases to reach thickness up to 100 μm – 17% of the total thickness of the cladding [38]. The native hydrogen concentration in these alloys is on the order of a few weight parts per million (wppm), but increases as high as 600-700 wppm as the zirconium absorbs hydrogen from the corrosion reaction (as opposed to hydrogen from the water) [38]. The mechanical properties of the cladding and structural materials begin to deteriorate as the hydrogen solubility limit is exceeded and hydride platelets precipitate. The corrosion rate of cladding has further been found to increase in the presence of hydrogen [39]. Similarly, although outside the context of cladding materials, hydrogen has also been found to enhance intermetallic diffusion [40]. Enhanced irradiation defect growth has been attributed to the presence of hydrogen [41,42]. Many of these effects may also have impacts to fusion systems but have yet to be explored due to the lack of experiments operating under nuclear conditions. Hydrogen in cladding materials is frequently measured using hot vacuum extraction and inert gas fusion techniques, which are contrasted in Table 3.

4. Emerging hydrogen challenges in fission and fusion reactors

Many challenges relating to hydrogen were discussed in Sections 2 and 3. Containing and selectively removing hydrogen are two significant challenges for fission and fusion nuclear reactors, especially advanced reactors, that are underdeveloped relative to the immediate need.

Hydrogen must be contained in fission microreactor metal hydride moderators. Metal hydrides were discussed in Section 3.1 as attractive moderators due to their high temperature stability. At the high temperatures expected for microreactors (~800˚C), even stable hydrides may gradually lose hydrogen. A more significant concern is the potential for irreversible depletion of hydrogen that could occur if the heat transfer properties of the metal hydride moderator are altered because of swelling, blister formation, delamination, or an off-normal temperature excursion. Hydrogen depletion would result in a decreased moderation worth and lower the fission output. Moderator cladding and hydrogen permeation barriers have been tested to prevent the release of hydrogen from metal hydrides (Chapter 13-4 in [43]). While substantial testing was performed in the 1950-60's for the ANP program, including irradiation testing, those were designed and performed to operate under drastically different conditions (e.g., air working fluid) than what will be used in modern microreactors. In addition, modern advanced fabrication methods can likewise be leveraged to create solutions that were not possible decades ago [44].

Tritium must be removed from fission coolants for safety concerns. The small quantities of tritium produced by lithium-containing fluoride molten salts is comparable to the amount produced in CANDU reactors [45] and must be removed to prevent tritium permeation and migration from the primary coolant into other reactor subsystems. Concerns for tritium management significantly contributed to the decision to discontinue the US molten salt reactor program in the 1960's [46]. Tritium monitoring in heavy-water systems is straight forward using liquid scintillation counting; however, monitoring for tritium in molten salts requires substantial development. Furthermore, detritiation facilities will be required to extract and capture tritium from molten salts. Tritium extraction from molten liquids and gases is quite different from the techniques related to water [47]. Relevant tritium extraction concepts are being explored but require significantly more development [45].

Tritium must be contained in fusion systems. Although tritium is intentionally produced in breeding blankets, it must be contained to the extent possible until it reaches a tritium extraction system. Preventing tritium permeation in thermal systems will require effective permeation

barriers [48]. Similar to a molten salt environment, tritium will need to be monitored in molten PbLi breeders. Some development work has been performed on tritium sensors compatible with molten PbLi, but sensor response times have been demonstrated on the order of days, so further research and new technologies are required [49].

Tritium must be efficiently removed in fusion tritium extraction systems. Utili, et al., summarize the prevailing technologies for extracting tritium from PbLi [16]. The primary candidates include gas-liquid contactors, vacuum permeators, vacuum droplet towers, and getters. Significant work remains on maturing these technologies [50].

5. Techniques for measuring hydrogen retention

Most elements can be detected through a wide variety of means, but hydrogen is unique because of its low atomic number. A number of spectroscopic methods that are standard laboratory instruments for elemental and chemical analysis cannot measure hydrogen directly. For example, the cross section for photoionization in x-ray photoelectron spectroscopy (XPS) is about 5000x smaller than that of the standard carbon 1s XPS reference value [51]. Likewise, auger emission spectroscopy (AES) relies on the Auger process where one electron falls from an outer orbital to an inner electron shell after the latter is ejected. As hydrogen only has one electron, this multi-electron process is not possible. Scanning and transmission electron microscopy (SEM, TEM) are not able to directly detect hydrogen, nor is energy dispersive X-ray spectroscopy (EDS). Some of these techniques allow for indirect detection of hydrogen by measuring the chemical interactions that hydrogen has with other materials. One of many such examples is shown by Taylor, et al., where XPS was used to measure the fundamental chemistry of how deuterium was retained in lithiated graphite plasma-facing components for fusion devices [5].

Despite some of these most common and frequently used techniques having a blind spot for hydrogen, many other techniques are capable of directly measuring and quantifying hydrogen in materials. Excellent reviews have published that contrast a variety of these hydrogen measurement techniques [52-54]. Table 3 provides a list of available techniques used for measuring hydrogen in fusion and fission relevant materials. Techniques used to measure other states of hydrogen are not addressed (e.g., gas chromatography, electrochemical gas sensors, etc.). Invariably, this list cannot be exhaustive of all possible methods used to characterize hydrogen. The table categorizes the techniques by whether they are typically used to measure bulk hydrogen in materials, hydrogen located at the surface/near-surface, or the volumetric distribution of hydrogen. Some of the techniques span multiple categories. For example, laser induced breakdown spectroscopy (LIBS) ablates material at a near-surface and the elemental composition of the resulting plasma plume is measured. By rastering the impinging laser spot, this analysis can provide a two-dimensional map of the surface. Further measurements over the same area can provide a depth profile.

The listed techniques provide information regarding the total hydrogen inventory in a metal, trap energy, temperature stability, transport properties, among others. Calibration is a challenge in many of the measurement techniques. Most calibration methods rely on some standard or certified reference material (CRM), but very few of these certified materials are available in the concentrations, form factors, or isotopes needed for calibration. For example, calibrating a glow discharge optical emission spectroscopy system requires a set of several large diameter thick discs that span multiple concentrations and are ideally the same composition as the material to be interrogated. With the lack of CRMs, some researchers have attempted to produce calibration standards [55,56]. Other measurement techniques rely on theory for quantification. For example,

nuclear reaction analysis (NRA) relies on the nuclear reaction D(^{3}He,p) ^{4}He for detecting deuterium in materials. The energy spectrum of the emitting alpha particles is measured and used to determine the D inventory in the sample [57]. The reaction yield is proportional to the flux of incident ^{3}He ions and the cross section [52].

An order of magnitude sensitivity for the measurement techniques has been listed in Table 3. It should be noted that this is a nominal value and large exceptions could be expected in some scenarios. For the XPS description provided above, a laboratory based monochromatic x-ray source cannot be used to measure hydrogen directly. However, synchrotron x-ray radiation provides sufficient intensity to overcome the otherwise prohibitively low photoionization cross section [58]. The references in Table 3 provide additional information on the technique and/or applications on how the technique has been applied to measuring hydrogen in fusion and fission materials.

Table 3. Available techniques for measuring and quantifying hydrogen in metals.

Technique	Domain	Information learned	Ease of H calibration	Probing method	Detection method	Pros	Cons	Sensitivity	Sample environment	Refs
Thermal desorption spectroscopy	Bulk	Total Q_2 H trapping energy Temperature stability	●	Thermal	QMS	Established technique	Destructive	ppm	High vacuum	[8,59]
Hot vacuum extraction	Bulk	Total Q_2	●	Thermal	pressure	Measure high Q_2	Destructive	ppm	High vacuum	[60,61]
Inert gas fusion	Bulk	Total Q_2	Low conc.: ● High conc.: ◐	Thermal	TCD IR	Established technique	Destructive Saturation at high conc.	ppm	Inert atmosphere	[52]
Sievert's apparatus	Bulk	Diffusivity, solubility Temperature stability	●	Thermal Pressure	Pressure, QMS	Established technique	-	-	High vacuum	[9,62]
Permeation experiment	Bulk	Permeability, diffusivity	●	Thermal Pressure	Pressure, QMS	Established technique	Difficult for hydride forming metals	-	High vacuum	[9,53,63-65]
Mass measurements	Bulk	Total Q_2	●	Mass	Balance	Rapid, easy, inexpensive, NDE	Poor sensitivity with small samples	Improves with sample	STP	[19,24,66]
Thermogravimetric analysis	Bulk	Total Q_2 Temperature stability	●	Thermal, mass	TC, Balance	Established technique	Controlling secondary factors	≥0.1 µg 0.001 K	Inert atmosphere	[54]
X-ray diffraction and scattering	Surface Bulk	Q_2 distribution Crystallography	● By theory	X-ray	Spectroscopy	Established technique, NDE	Synchrotron (availability) for some analyses	-	STP	[66-68]
Sigma pile	Bulk	Total Q_2	●	Neutrons	Neutron detector	NDE	Availability, potential activation	0.035 wt%	STP Radiological	[69]
Positron annihilation spectroscopy	Bulk	Void decoration	○	Positrons	Gamma detector	NDE	Qualitative	Unestablished	STP	[70-77]
Nuclear reaction analysis	Surface	Total H, D Depth profile (0.6-10 µm)	● By theory	MeV ions (^{15}N for H; ^{3}He for D)	Proton detector	No calibration, NDE (mostly)	Availability	~10 ppm	High vacuum	[52,78]
Elastic recoil detection analysis	Surface	Total Q_2 Depth profile (0.6-10 µm)	● By theory	0.3-2 MeV ions (He to Au)	MCP	Easy detection of Q	Availability	0.1 at%	High vacuum	[52,79]
X-ray photoelectron spectroscopy	Surface Mapping	Q_2 chemistry	● By theory	X-ray (keV)	MCP	No calibration, expansive libraries	Cost, no direct H observation	0.1 at%	Ultrahigh vacuum	[5,51,58]
Glow discharge optical emission spectroscopy	Surface	Total Q_2 Depth profile (≤130 µm	◐	Plasma	PMT MS	Rapid No sample prep. nm depth resolution	Calibration for H can be challenging	ppm	STP	[6,52,55,56]
Liquid scintillation counting	Surface	T concentration	●	Radiation scintillation	PMT	Extreme sensitivity	Limited information, Requires tritium	ppq	Liquid	[65]
Laser induced breakdown spectroscopy	Surface Mapping	Elemental composition	○	Laser	OES MS	Min. sample prep Cal.-free method theoretical	Difficult calibration	ppm	STP, vacuum	[52]
Secondary ion mass spectrometry	Surface Mapping	Elemental composition Depth profile (<100 nm)	○	Ions (keV)	QMS, TOF magnetic sector	Directly measure surface H	Slow, expensive	10s ppm	High vacuum	[52]
Ion scattering spectroscopy	Surface Mapping	H absorption site	●	Ions (keV)	MCP	Directly measure surface H	Obscure	Undetermined	High vacuum	[80]
Tritium imaging plate	Surface mapping	2D T_2 distribution	●	Beta radiation	Film	2D distribution Sensitivity	Availability	2 µCi/g	STP	[81,82]
Local electrode atom probe	Volume	Q_2 location	○	high voltage	Ion detector TOF	Easy H detection Volumetric reconstruction	Availability, expensive, slow, quantification	~10 ppm H: Undetermined	High vacuum	[83,84]

Neutron radiography	Volume	Q_2 distribution		Neutrons	Film	Volumetric reconstruction Tomography possible	Availability	~10 wt ppm	Radiological	[33,85,86]
			●							

Acronyms

Q_2: hydrogen isotopes H_2, D_2, T_2
MS: mass spectrometry
QMS: quadrupole mass spectrometer

TOF: time of flight
NDE: non-destructive examination
STP: standard temperature pressure

TCD: thermal conductivity detector
IR: infrared
TC: thermocouple

MCP: multichannel plate
OES: optical emission spectroscopy
ppm: parts per million

Symbols
○: difficult
◐: moderate
●: easy

6. Conclusions

Hydrogen plays a central role in fusion and fission systems. Its isotopes, deuterium and tritium, comprise the fuel in fusion reactions. The challenges due to radioactive tritium are numerous. Tritium implanted into plasma-facing components can become trapped or permeate outside primary systems. Secondary thermal systems that transport tritium must consider losses due to permeation. Multiple concepts have been established and tested for breeding and harvesting tritium, but significant work remains to increase the technology readiness of these technologies for online use in future fusion reactors.

Hydrogen is also fundamental in fission reactors, although in less conspicuous roles than in fusion. Hydrogen bearing materials are the best neutron moderators. Light and heavy-water have been the dominant reactor moderators for decades. High temperature moderators for advanced microreactors will include metal hydrides, which can contain a higher hydrogen density than liquid hydrogen. Beyond hydrogen's functional contributions toward fission power, deleterious effects of hydrogen uptake in cladding and structural materials have been and are an active area of research.

Characterizing hydrogen content in fusion and fission systems has elucidated many of the challenging phenomena relating to hydrogen in materials. Although many conventional analytical characterization techniques have a blind spot toward hydrogen, a wide array of methods are available to analyze most important aspects of hydrogen. Of importance, advancements need to be made on techniques capable of measuring hydrogen in fluoride molten salt coolants and molten tritium breeder materials.

CRediT roles:
Chase N. Taylor: Conceptualization; Data curation; Writing - review & editing.

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