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Hydrogen Permeation in Fusion-relevant Materials

Summary Report of a Consultancy Meeting

IAEA Headquarters, Vienna, Austria

26–27 September 2019

Prepared by

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IAEA Nuclear Data Section

May 2020

IAEA Nuclear Data Section

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Abstract

A Consultancy Meeting was held on 26–27 September 2019 at IAEA Headquarters in Vienna to review data needs for hydrogen permeation in plasma-facing materials and components in fusion devices and to delimit the scope of a possible coordinated research project on that topic. The proceedings and discussions during the meeting are summarized here.

May 2020

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Abbreviations

A+M	atomic and molecular
CM	consultancy meeting
CX	charge-exchange neutral atom
DEMO	demonstration fusion reactor
FPP	fusion power plant
FW	first wall of a fusion device or reactor
GDP	gas-driven permeation
IDP	ion-driven permeation
ITER	international thermonuclear fusion device; next-step fusion device
PDP	plasma-driven permeation
RAFM	reduced activation ferritic/martensitic steel
TBM	test blanket module

1. Introduction

In the magnetic confinement approach to fusion a deuterium-tritium (D-T) plasma at a temperature of around 15 keV (about 150×10^6 K) is trapped in a toroidal magnetic field inside a vacuum vessel. These fusion processes take place in the core of the plasma whereas the significantly cooler outer regions of the D-T plasma (few hundred K) will be constantly interacting with the reactor inner wall component materials. These plasma-facing wall materials must therefore withstand heat and particle loads from the plasma.

It is important to know as much as possible about the behaviour of these wall materials with respect to permeation of hydrogen isotopes from the plasma in order to assess their suitability for containing and isolating the T fuel from the surrounding fusion reactor components. Of particular concern is the trapping and retention of T inside the wall, and the possibility of T diffusing through the material and finding its way into the coolant water of the reactor components, where it would pose a potentially serious environmental hazard. The T inventory plays a crucial role in fusion systems since the dynamical behaviour of T, such as solubility, diffusion, trapping, de-trapping, re-trapping, permeation, etc, in plasma-facing materials determines the in-vessel fuel inventory source term (i.e. T retention) and the ex-vessel fuel release term (i.e. T permeation). These in-vessel and ex-vessel T inventories are critical and being used in the fusion reactor safety assessments for licensing future fusion facilities, such as the fusion demonstration reactor DEMO and the future fusion power plants (FPP). Minimizing the uncertainty associated with T permeation enhances the reliability of safety assessment, and supports licensing of DEMO reactors and FPPs in participating countries.

For investigation and modelling the hydrogen permeation through plasma-facing components, validated hydrogen-material interaction parameters are required. Previous works[1] on these parameters must be revisited and missing data must be addressed in the corresponding interaction categories. Only a full set of qualified parameters allow for fundamental permeation data, which is currently missing for the present plasma-facing components in the first wall (FW) of main chamber and divertor planned for next-step fusion device ITER and for DEMO. These components comprise of various materials, such as tungsten (W), different steels and also beryllium (Be) in the case of ITER, but also of equal importance are elements used as joining materials and cooling pipes. As a crucial example is the divertor design of ITER, which comprises of W mono-blocks having internal copper-chrome-zirconium (CuCrZr) cooling pipe and an interlayer made out of Cu between the W material and the CuCrZr pipe. Further, the ITER port plugs in the main chamber FW recessed areas comprise of stainless steel (SS316L). These ports have been assessed to be the main permeation source in ITER. Moreover, port plugs will be used for testing at least four reference T breeding blanket concepts for DEMO reactor specifications. These so-called test blanket modules (TBM) use reduced activation ferritic/martensitic (RAFM) steel labelled as Eurofer-97 as structural material. Fundamental knowledge of T permeation in TBMs is mandatory for the design of economical operation of DEMO.

Study of hydrogen permeation in the FW plasma-facing materials is further complicated by the elevated temperatures present under normal operation of a reactor and the anticipated damage they will suffer due to irradiation by the flux of energetic 14 MeV neutrons produced in the D-T fusion reactions. Also of relevance is the nature and evolution of the FW material surface, which is altered by interaction with the plasma through erosion and preferential sputtering of certain component atoms. The effect of annealing out of defects during recrystallization of W components under high heat loads may enhance the permeation process at the divertor. The effect of fast ions and charge exchange (CX) neutral atoms escaped from the plasma may have a strong impact to the surface and sub-surface modification of the FW components and their effect to hydrogen permeation needs to be scrutinized. The intense neutron flux from the D-T fusion reactions penetrate the FW surface and interact with the

[1] F. Reiter, K. S. Forcey, G. Gervasini, *A Compilation of Tritium-Material Interaction Parameters in Fusion Reactor Materials*, Joint Research Centre EUR 15217 EN, Commission of the European Communities, Luxembourg1993

atomic elements of the wall creating neutron-induced large-scale lattice damage and transmuting the wall elements. These effects transform the wall component properties, and will have a significant effect to the hydrogen retention and mobility in the wall.

Although the precise mechanism is not understood, it is well-known that trapped hydrogen reduces the ductility of many materials, including steels, a phenomenon known as hydrogen embrittlement or hydrogen-induced cracking. For safety and operational reasons it is also important to understand and mitigate the potential damage that could be caused to key components in a fusion reactor, including the above-mentioned large number of diagnostic ports needed in ITER.

Following advice from the International Fusion Research Council (IFRC) Subcommittee on Atomic and Molecular (A+M) Data the A+M Data Unit is planning to start a Coordinated Research Project (CRP) on fundamental data that are needed to simulate hydrogen permeation and to extrapolate the results to ITER and DEMO plasma conditions. The present report is the output of a two-day Consultancy Meeting on data requirements for hydrogen permeation in the FW plasma-facing materials and components that was held at IAEA to review the state of available data and data needs for permeation studies and to make recommendations about the scope and objectives of the planned CRP. These recommendations will form the basis of a proposal to the IAEA Committee on Coordinated Research Activities (CCRA) to initiate the planned CRP with a start date in Q3 2020.

Section 2 summarizes the presentations at the meeting. Section 3 summarizes the discussions and conclusions. Appendix I provides the list of participants and Appendix II the meeting agenda. Appendix III provides speaker summaries of the presentations. Appendix IV presents the conclusions of the CM in the form of a proposal to CCRA to initiate a CRP on atomic data for vapour shielding. Finally, Appendix V lists recent relevant journal literature from the fusion community related to hydrogen permeation and recent relevant conference contributions at major fusion meetings.

2. Proceedings

Speaker summaries are provided in Appendix III and presentations are available on the A+M Data Unit web pages at https://www-amdis.org/meetings/hydrogen-permeation-cm/.

The first of day of the meeting comprised of presentations of the participants focusing to various aspects of hydrogen permeation in fusion environments. The meeting was opened and participants introduced themselves.

Kalle HEINOLA (IAEA) described the general nature of CRPs and their role in the Agency programme and reviewed the questions to be addressed in the course of this CM.

Gregory DE TEMMERMAN (ITER, France) presented a summary of the methodology and research needs for T permeation in ITER. Because ITER will operate in a pulsed-mode, permeation i.e. the T reaching the coolant is assessed to take place during the baking cycle of the plasma-facing components in the main chamber and in the divertor, and during pulsed operation of the machine. During continuous operation T permeation is not expected to occur. However, providing a good estimate for T permeation in ITER is still unclear due to several unknown parameters and plasma conditions affecting permeation. Permeation modelling has been done with TMAP-7 code, which requires various ITER plasma loading conditions and a validated database of hydrogen diffusion and trapping parameters in ITER-relevant materials, such as W, Be, Cu, CuCrZr, SS316L, RAFM. More advanced modelling taking into account the 3-D features of plasma-facing components is required.

Wolfgang JACOB (Max Planck Institute for Plasma Physics (IPP), Garching, Germany) discussed the data needs for hydrogen permeation in fusion materials and the corresponding experimental activities ongoing at IPP. The permeation modellers require in addition of the plasma information also the solute diffusion coefficients, trap densities, detrapping energies, and surface conditions, but also the conditions at the materials interfaces. An overview of the SIESTA facility and its upgrade was given. SIESTA is used as an ion source for ion-driven permeation (IDP) research at IPP. The IDP activities form the PERMEX-II project, which allows for permeation research with fusion relevant energies (0-10 keV), fluxes (up to 10^{19} $m^{-2}s^{-1}$), and temperatures (up to 1000 K). Recent permeation research focused on gas-driven H permeation and solubility in W and was performed using an UHV oven. The obtained results provided critical corrections to the previous widely used and accepted experimental value for H diffusion in W [2]. Permeation of T in two European DEMO reference breeding modules has been modelled with TMAP-7 and TESSIM codes, and the first results were presented.

Masashi SHIMADA (Idaho National Laboratory (INL), USA) presented recent INL experiments on T permeation in RAFM steel and in W, and in other materials such as nickel (Ni), vanadium (V) and oxide-dispersion strengthened (ODS) steels. The INL's STAR facility is suitable for gas-driven permeation (GDP) experiments up to temperatures ~1200 K. In addition to conventional GDP using D, one can perform gas absorption and GDP using T-gas. This allows measuring real isotopic effects of hydrogen species (according to harmonic transition state theory the diffusivity D has dependence as $D \propto (m)^{-0.5}$, where m is the mass of diffusing particle); molecular T_2 conditions; operations with multi-hydrogen components (H_2, HT, T_2) and with low partial pressures of HT, T_2; experiments with T and the resulted low detection limit allow the use of low temperatures. Further, the STAR facility is compatible for the use of activated samples, which allows T-gas absorption and GDP experiments using neutron-activated samples.

ASHIKAWA Naoko (National Institute for Fusion Science, Japan) discussed on hydrogen isotope permeation in plasma-facing materials towards the Japanese DEMO (J-DEMO) design. Detailed matrix of elements used for J-DEMO plasma-facing components was given. Recent progress on the GDP and plasma-driven permeation (PDP) activities were reviewed. The activities comprise of twin-sample experiments, in which GDP and PDP was performed to various types of W grades with different neutron irradiation conditions and to F82H steel. GDP and PDP provided new data on permeability, effective diffusivity and recombination of hydrogen in W. Hydrogen permeation through co-deposited W was studied with 20 μm thick W thin films on Ni. Effect of high heat fluxes to permeation in W plasma-facing components was simulated experimentally by exposing layered W/Pd/Ti samples to D plasma. Exposure was performed with linear plasma device TPDsheet-U (T_e~4.8 eV, n_e~8×10^{17} m^{-3}, Γ_i ~2×10^{22} m^{-2}s^{-1}) and the samples were analyzed ex-situ with elastic recoil detection analysis (ERDA) revealing permeation throughout the sample.

Dmitry TERENTYEV (SCK•CEN, Belgium) reviewed recent neutron irradiation programmes on W at BR2 reactor. The facility is capable of irradiating fusion-relevant specimens with neutrons by controlling the neutron flux, fluence, and sample temperature. 2017-2019 irradiation programme provided various specimens of different W grades (size and crystallinity) irradiated from 0.1 dpa up to 1 dpa with a temperature range of 673 K to 1473 K. The irradiation programme comprised of mechanical testing of the samples as well as positron annihilation spectroscopy (PAS) analysis for determining open volume defect creation. Obtained data on the microstructure evolution and the tensile properties are of fundamental importance for modelling the materials properties evolution during fusion neutron irradiation. A selection of W samples from the 2017-2019 irradiation campaign can be provided for analyses within this CRP participants. Possibility for a dedicated neutron irradiation programme for this CRP can be discussed.

UEDA Yoshio (Osaka University, Japan) described experiments on hydrogen permeation in W using D-only and mixed C+D, He+D and N+D ion beams. These IDP experiments were done using up to 75 μm thick W sheets for 1 keV ion beams (D_3^+, D_2^+, D^+ with 10^{19}~10^{20} m^{-2}s^{-1}; C~0.1-3.0 %; He ~0.1-2.0 %; N~2%) at temperatures ranging from 500 K to 1000 K. The obtained effective diffusivity with D-only beams was found to be dependent on the W sample pre-treatment history and the corresponding microstructure. A peak in the permeation D flux was seen at 800 K. Using mixed ion beam C+D was found to increase the D permeation. The resulted permeation flux was strongly temperature dependent having a maximum ~700 K, but being decreasingly diffusion limited process at higher temperatures. Addition of He (2%) greatly reduced the D permeation flux, whereas adding N increased permeation up to 900 K. It was concluded, that reactive impurities, such as C and N, tend to increase D permeation via surface or near-surface mixed layers, which could act as diffusion barrier for D recycling back

[2] R. Frauenfelder, J. Vac. Sci. Technol. **6**, 388 (1969)

to surface. Inert gases, like He (Ne, Ar), reduce permeation due to bubble and void formation in the implantation zone.

Anne HOUBEN (Forschungszentrum Jülich (FZJ), Germany) presented FZJ activities on GDP for hydrogen permeation in Eurofer-97 and in ITER-grade SS316L at temperatures 550 K to 900 K. It was found out, that steel surface properties, such as roughness and oxidation, play a critical role on hydrogen permeation – a rough surface on SS316L results in lower diffusion and higher solubility, and oxidation of Eurofer-97 has a lowering effect on permeation. SS316L substrates were exposed to a deuterium plasma with D energies 20 eV and 200 eV and with fluences 10^{25} m^{-2} and 5×10^{25} m^{-2}. GDP measurements were performed afterwards and it was concluded that the needle-like surface modifications created by the plasma exposure did not influence the permeation. GDP permeation experiments with Eurofer-97 having a permeation barrier Y_2O_3 showed only minor influence due to surface roughness, but a strong effect due to oxidation.

3. Discussion and Conclusions

The second day of the meeting was devoted to discussions and conclusions. Participants agreed that a CRP on fundamental data relevant for hydrogen permeation in fusion devices is timely. The discussions focused on the scope and aims of the proposed CRP in the area of development, evaluation and recommendation of permeation data and on the roles within the CRP of physics, trapping, plasma-surface modelling and the corresponding experiments and modelling. Implementation of the data in the preparation of ITER exploitation is one of the major objectives of the global fusion community. Successful ITER operation keeps the long-term nuclear fusion R&D and electricity roadmap on schedule. It is anticipated this CRP will be able to provide an important contribution to mature computational models applied on ITER for the assessment of hydrogen fuel in-vessel and ex-vessel inventories.

It is important to note that the CRP is to be focused on the provision of evaluated permeation data for fusion applications. It cannot address issues such as engineering of the FW plasma-facing components design or the development of the computational methodologies used for the permeation rate assessments.

Topics addressed by the CM, and the conclusions of their discussion are given below.

- Hydrogen isotope permeation in FW plasma-facing materials and components is critical for the fuel cycles of T and for the T decontamination safety issues in ITER and DEMO. It has an effect to the future component design issues and fusion reactor operational scenarios. However, experimental validated data and corresponding permeation modelling using candidate materials for ITER and DEMO is either lacking or is not sufficient.
- Materials relevant to hydrogen permeation are i) divertor: W, CuCrZr, Cu, ii) main chamber: Be (ITER), RAFM steels, SS316L, and other materials considered as part of FW solutions in DEMO and J-DEMO designs.
- Fusion plasma contains various impurity species and their impact on hydrogen permeation of wall materials through surface modifications is very large. Detailed experimental database as well as appropriate modelling on the effect of plasma impurities are still lacking.
- There is a critical requirement of uncertainty minimization associated with
    - hydrogen mass transport properties, such as diffusivity, solubility, recombination coefficient, dissociation coefficient, etc, in unirradiated fusion materials.
    - processes involving different hydrogen species, such as thermalized molecules, hyperthermal CX atoms and fast ions from plasma. The neutral CX particles may modify the surface morphology with high energies and fluxes. Their impinging angle is not the normal incidence, and they can smear the surface or create extensive sub-surface damage. ITER is expecting the FW being irradiated with large fluxes of CX neutrals with a broad spectrum of energies up to keV-range (maximum still around 2-3 eV).

    Also, interactions of mixed hydrogen species can potentially lead to non-linear diffusion effects affecting the permeation.
  - hydrogen isotope effects: experimental mass dependence of hydrogen to permeation processes; databases with T interaction processes largely missing.
  - thermal diffusion (e.g. Soret coefficient).
  - effect of materials surfaces and bulk microstructure, such as lattice structure (BCC, FCC, etc), materials Z-dependence and the effect of recrystallization.
  - geometrical effects in 2-D and 3-D, such as the effect of thermal gradient from the W monoblock surface down to cooling water, and the effect W monoblock gaps.
  - irradiation responses such as neutron irradiation-induced damage and solid transmutations. Especially the thermal evolution of irradiation-induced defects affect the permeation properties.
- Knowledge on the effect of diffusion barriers must be increased. As an example, the above-mentioned plasma impurities get implanted on materials surfaces and may form a reactive layer for hydrogen. This may promote enhanced permeation, or “super”permeation.
- Research on T accumulation and permeation in the joining interfaces i.e. interfaces between different materials of a plasma-facing component, which are bonded with e.g. HIPing, brazing, etc. In the computational simulations, one usually assumes an equilibrium at the interface and uses the continuity of chemical potential, but this assumption might not always be correct. The effect of interface coating on T retention at the interface must be understood with respect to the T retention in the bulk of the material.
- In experimental research, different grades of material tend to provide varying permeation, retention and diffusion properties. Hence it is required to have materials being pre-characterized in addition to the post-characterization. Conditions of intrinsic impurity concentrations and of underlying microstructure must be available in order to understand the effect of e.g. defect evolution and recrystallization of the material to hydrogen permeation.
- Reference materials can be made available for coordinated research within this CRP. Using reference materials allows validation of experiments and uncertainty quantification. Permeation and retention experiments as well as any mechanical testing performed using these materials form a subtask of their own, and the corresponding activities will be separately agreed with the sample providing institute and the corresponding subtask participants. Materials and the providing institutes are
  - neutron-irradiated and un-irradiated W: SCK•CEN, Belgium
  - SS316L: ITER
  - CuCrZr: ITER
  - Ni: INL (benchmarking experiments for permeation with Ni are underway, and samples can be made available for further benchmarking)
- Different experimental methodologies for studying permeation in single material and in layered materials (2 – 3 layers)
  - plasma-driven permeation (PDP)
    - sample exposed to single element plasma or mixed plasmas
    - mixed plasma exposures require understanding of the effects of single element exposures
    - plasma parameters play crucial role: plasma exposures are typically with high fluxes; uncertainty in the loading plasma species (atomic and/or molecular); uncertainty in the amount of plasma impurities; uncertainty in plasma species impact energy (typically a distribution)
    - enhanced surface modification effects may play a role
  - gas-driven permeation (GDP)
    - requires sticking of molecular hydrogen from the gas phase to the sample surface followed by dissociation of molecules to atoms
    - permeation process is initiated once transport of atomic hydrogen over the surface potential barrier to the bulk regions takes place

        - endothermic metal surfaces (typical metallic fusion materials) have high hydrogen solution energy and hence low hydrogen solubility, whereas exothermic metals absorb hydrogen efficiently and have high solubility
        - high gas pressures required for permeation in fusion materials do not mimic fusion conditions, but provide statistically meaningful amount of hydrogen
        - surface modifications are avoided, but GDP may be costly in time due to the lower hydrogen fluxes as compared to PDP
    - ion-driven permeation (IDP)
        - atomistic ion beams used to introduce hydrogen in the sub-surface regions or in the bulk of the studied material
        - effect of mixed ion beams can be used to study non-linear diffusion effects
        - use of low ion beam energies required (energy below the target material's lattice displacement threshold energy) in order to avoid implantation-induced defect creation in the target material
        - use of energetically low ion beams typically results in low exposure fluxes to the sample. IDP experiments may be costly in time due to the lower fluxes as compared to PDP
        - surface modification may take place (sputtering, erosion), but is ion beam impact energy, angle and flux dependent
- Permeation results at fusion-relevant temperatures (few hundred Kelvin and more) are realistically achievable due to the thermodynamic kinetics involved with hydrogen migration in fusion materials. However, experiments at lower temperatures (above or close to room temperature) are typically too time-consuming and hence not feasible
- Highlighted requirements for validated hydrogen permeation data: diffusion, solubility, defect-dependent hydrogen trapping energetics (trapping, de-trapping, re-trapping to impurities, lattice defects, grain boundaries, etc), hydrogen vibrational properties (zero-point energy) for diffusion and trapping, hydrogen isotope mass-dependent effects, recombination coefficient, defect dynamics (mobility, clustering, dissociation), effect of materials lattice and surface properties, effect of surface evolution e.g. in the form of sputtering, effect of diffusion barriers
- Standardized database needed for providing parameters to large-scale permeation modelling with reaction-diffusion methodologies based on rate theories (e.g. TMAP-7, TESSIM, MHIMS, advanced RE, CRDS), and computations based on finite element methodologies (e.g. COMSOL, Abaqus)
    - The CRP can promote code comparison activities, particularly in the area of plasma-material interaction models and multi-scale simulations on retention and permeation.
    - Groups providing experimental data for the validation and comparison of models should be identified. The data needs for hydrogen permeation simulation should be informed as far as possible by interaction with experimentalists.

It was agreed that there is considerable scope for coordinated research within the terms of this CRP, particularly in the areas of benchmarking data and codes against experiment, code comparison, and in the investigation of the diffusivity, solubility, retention and permeation properties of hydrogen isotopes in fusion materials.

The proposal narrative for the CRP under consideration at this meeting is given in Appendix IV.

Appendix I: List of Participants

ASHIKAWA Naoko, National Institute for Fusion Science (NIFS), Toki City, JAPAN.

Gregory DE TEMMERMAN, ITER Organization, Saint Paul-lez-Durance, FRANCE

Anne HOUBEN, Forschungszentrum Jülich (FZJ), Jülich, GERMANY

Wolfgang JACOB, Max-Planck-Institut für Plasmaphysik (IPP), Garching, GERMANY.

SHIMADA Masashi, Idaho National Laboratory, Idaho Falls, USA.

Dmitry TERENTYEV, Studiecentrum voor Kernenergie – Centre d'Étude de l'énergie Nucléaire (SCK•CEN), Mol, BELGIUM.

UEDA Yoshio, Osaka University, JAPAN.

Kalle HEINOLA, IAEA Nuclear Data Section, Division of Physical and Chemical Sciences, P.O. Box 100, A-1400 Vienna, AUSTRIA.

Christian HILL, IAEA Nuclear Data Section, Division of Physical and Chemical Sciences, P.O. Box 100, A-1400 Vienna, AUSTRIA.

Appendix II: Meeting Agenda

Consultancy Meeting on “Hydrogen Permeation in Nuclear Materials.”

Thursday 26 September 2019

09:30 K. Hill: *Welcome and Introduction*

10:00 G. De Temmerman: *Hydrogen Permeation in ITER: estimation and missing data*

10:30 W. Jacob: *Hydrogen Permeation in Fusion Materials: Data needs for fusion and IPP activities*

11:00 *Coffee Break*

11:30 M. Shimada: *D/T permeation behavior in PFC and structural material*

12:00 N. Ashikawa: *Hydrogen Isotope permeation studies on plasma facing materials toward Japanese DEMO design*

12:30 *Lunch*

14:00 D. Terentyev: *Recent neutron irradiation experiments involving baseline and advanced tungsten grades performed at SCK•CEN*

14:30 Y. Ueda: *Hydrogen isotope permeation in tungsten under multiple ion irradiation*

15:00 A. Houben: *Hydrogen Permeation in Fusion Materials: Activities at FZ Jülich*

15:30 *Coffee Break*

16:00 Discussion *(all)*

17:30 End of day one

19:00 Social dinner: *Restaurant König von Ungarn, Schulerstraße 10, 1010 Wien*

Friday 27 September 2019

09:00 Discussion (*all*): *Possible coordinated research activities and data evaluation activities*

11:00 *Coffee Break*

11:30 Discussion (*all*): *CRP outcomes – experiments, databases, code and data comparison exercises; recommendations*

13:00 Lunch

14:00 Discussion (*all*): *Potential CRP participants, draft timetable for the CRP*

15:30 *Coffee Break*

16:00 Final Remarks and Review

16:30 Close of meeting.

Appendix III: Summaries of Presentations

Introduction, meeting objectives

K. Heinola

International Atomic Energy Agency

This presentation briefly summarises the history of the IAEA's Atomic and Molecular Data (AMD) Unit, its activities and place in the organisation. The nature and purpose of Coordinated Research Projects (CRPs) is described and the procedure for initiating one outlined. Typical activities related to this CRP such as requirement for experimental materials pre-characterisation, benchmarking activities, and experimental and computational cross-comparison activities are discussed. Currently active and recently completed CRPs as well as previous CRPs related to Hydrogen Permeation are briefly described.

The questions for the present Consultancy Meeting to consider in the preparation of a proposal for a CRP on Hydrogen Permeation in Fusion-relevant Materials are outlined (see Section 3 of this report), the participants introduced, and the meeting agenda adopted.

Tritium permeation in ITER: methodology and research needs

G. De Temmerman[a], Y. Charles[b], R. Ding[c], J. Mougenot[b], M. Ni[d], D. Matveev[e], K. Schmid[f], H. Zhou[c]
[a]ITER Organization, Cadarache, France
[b]LSPM, Université Paris 13, Laboratoire des Sciences des Procédés et des Matériaux, CNRS, F-93430 Villetaneuse, France
[c]ASIPP, Hefei, China
[d]Sino-French Institute of Nuclear Engi. & Tech.Sun Yat-Sen University
[e]Forschungszentrum Juelich, Germany
[f]IPP Garching, Germany

In the recent years, a significant effort has been made at the ITER Organization to estimate the amount of tritium which could permeate to the cooling water during baking of the in-vessel components. Given that ITER will operate in a pulse mode (albeit with long pulses), permeation during plasma operations is not expected, but tritium could diffuse to the coolant during baking during which the heating is provided from the coolant and when the temperature through the components is more homogeneous. In order to provide physics-based estimates, a clear methodology has been developed:

- Operation scenario is derived from the ITER Research Plan in terms of number of pulses and duration
- Surface conditions are determined from a range of edge plasma simulations which provide incoming particle/heat fluxes which are used to determine temperature gradients through components
- Diffusion/trapping data: because of the large scatter in reported diffusion/recombination coefficients, or even the lack of reliable data for some materials, it was decided to determine a set of consistent parameters using TMAP7 modelling of a set of published experimental data covering a range of parameters such as temperature and loading methods. The obtained values are then compared to those reported in the literature to ensure they are within the range of existing values

In addition, within the ITER Scientist Fellowship on “Fuel retention management”, it has been proposed to establish a database combining existing and valid data for T trapping/diffusion/recombination/solubility as well as to define “standard data” for relevant materials in order to improve the fidelity of permeation estimates done for various devices. It is proposed to apply the method developed for ITER and extend it to materials of interest for ITER and DEMO.

Hydrogen Permeation in Fusion Materials: Data needs for fusion and IPP activities

Wolfgang Jacob

Max-Planck-Institut für Plasmaphysik, Boltzmannstr. 2,
D-85748 Garching, Germany

Planning, Design and finally licensing of any future fusion device will require reliable data for the mobility of hydrogen isotopes in any material that will be in contact with tritium. The most important basic parameters in this respect are diffusivity and solubility. For hydrogen in tungsten, Frauenfelder measured these data in the late 1970ies, but no comparable experimental data exist for deuterium and tritium. Diffusivity and solubility are also the most important parameters to describe permeation through bulk materials. In addition, surface effects, in particular surface recombination, influence permeation as well as interfaces or lattice damage due to the high energy neutrons. Basic data are required for the following materials: Tungsten, RAFM steels, copper alloys (CuCrZr), doped tungsten materials or tungsten alloys, and functionally graded materials to be used in first-wall or divertor components. These parameters will be affected by lattice damage due to neutron irradiation. Furthermore, temperature-gradient-driven diffusion (Soret effect) and the influence of interfaces on diffusion deserve further attention.

IPP Garching started new activities in the field of diffusion and permeation. While most measurements for permeation data are performed by gas-driven permeation experiments where the permeation flux is induced by a high gas pressure, typically of the order of 1 bar on the high pressure side, the species flux in a plasma environment is not only composed of thermal neutral molecules, but also of atoms and ions. Atomic hydrogen atoms may more easily overcome the surface barrier and thus lead to a higher permeation flux than thermal hydrogen molecules. So far, little is known about the permeation probabilities of impinging hydrogen isotopes. On the other hand, it is well known that impinging energetic species are implanted into the near surface region and have, therefore, a much higher permeation probability. IPP is presently commissioning a new device (PERMEX-II) allowing to quantitatively investigating ion-driven permeation through fusion relevant materials. PERMEX-II will be an extension of SIESTA [1]. In SIESTA a high energy (eV to 10 keV) and high intensity ion beam is extracted from an ion source and mass-selected in a magnetic filter field. The mass-separated monoenergetic ion beam impinges on a thin foil at a controlled temperature 300 to 1000 K). The permeation hydrogen flow is measured downstream by a quadrupole mass spectrometer. Start of operation is planned for April 2020.

Diffusivity and solubility of hydrogen in tungsten at high temperature was measured by Frauenfelder in the late 60ies [2]. However, experimental data exist only for protium. IPP has commissioned a new device replicating more or less the set-up used by Frauenfelder. This device was used to measure basic data for protium and deuterium in the temperature range from 1600 to 2600 K [3]. For deuterium diffusion in tungsten activation energy of 0.28±0.06 eV was determined. The activation energy for protium is identical within the uncertainty limits.

[1] R. Arredondo *et al., Rev. Sci. Instrum.* **89** 103501 (2019)
[2] R. Frauenfelder *et al.*, *J. Vac. Sci. Technol.* **6**, 388 (1969)
[3] G. Holzner *et al.*, *Phys. Scr.* **T171**, 014034 (2020)

Deuterium and tritium permeation behavior in PFC and structural materials

M. Shimada

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Tritium permeation plays a crucial role in fusion system since tritium behavior (e.g. diffusion, trapping, permeation etc.) in materials determines in-vessel inventory source term (i.e. tritium retention) and ex-vessel release term (i.e. tritium permeation) in reactor safety assessments for licensing fusion facilities. Comprehensive work on hydrogen isotope behavior in fusion materials is available, but very little database exists for tritium, the radioactive hydrogen isotope, due to the cost and difficulty associated with handling tritium.

INL operates the Safety and Tritium Applied Research (STAR) facility, DOE Hazard Category less than III facility, which is licensed to handle tritium up to 1.6 gram, and developed deuterium and tritium permeation capabilities in fusion materials. This presentation discussed the motivation for needs to measure isotope effects on hydrogen isotope permeation behavior in fusion materials, the development of deuterium and tritium permeation apparatus, and recent results on deuterium and tritium permeability measurements in polycrystalline tungsten, reduced activation ferritic/martensitic steels, nickel, and vanadium. [1-4].

[1] M. Shimada and R.J. Pawelko, "Tritium permeability measurement in hydrogen-tritium system", *Fus. Eng. Des.* 129 (2018) 134-139.
[2] M. Shimada and R.J. Pawelko, "Tritium permeability in polycrystalline tungsten", accepted in *Fus. Eng. Des.* 146 (2019) 1988.-1992.
[3] M. Shimada, et al., "Tritium permeation behavior in reduced activation ferritic/martensitic steel in hydrogen-tritium system", submitted to *J. Nucl. Mater.*
[4] M. Shimada, , "Deuterium and Tritium permeation behavior in plasma-facing and structural material", *presented at Tritium 2019*

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Recent irradiation experiments involving baseline and advanced tungsten materials performed at SCK•CEN

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To reduce uncertainty on the scarce information available on neutron irradiation damage in tungsten and to support the development of advanced tungsten-based alloys, SCK-CEN has recently executed the neutron irradiation programme. The irradiation programme lasted for about 2 years and covered a large range of irradiation temperatures and doses. The primary types of post irradiation experiments (PIE) are mechanical testing and microstructural investigation. Next to the main irradiation programme, SCK-CEN has executed the irradiation programme for internal studies, where a number of samples for advanced characterization such as hardness testing and tritium permeation experiments. The irradiation was performed inside the fuel elements to ensure that the transmutation of tungsten into Re and Os would be minimized and achieve the rates comparable to DEMO/ITER conditions.

As of today, the irradiated materials are dismantled and up to some extent tested. The preliminary results demonstrate that neutron irradiation performed at 600°C up to 0.4-0.6 dpa induces severe embrittlement in baseline tungsten basically reducing the fracture toughness to the level comparable to the lower shelf toughness. The impact of the neutron irradiation at lower temperature (i.e. below 600°C) is expected to be the similar or even more severe. The tensile tests performed on the material irradiated at 600C up to 0.6-0.7 dpa also confirmed that the baseline tungsten (i.e. ITER specification commercially pure grade) exhibits extremely high yield stress (about 1.5 GPa) and nearly zero elongation. TEM was applied to investigate the microstructure and it revealed the presence of both dislocation loops and voids with rather high density (10^{23} m^{-3}). The tests are going on to assess the mechanical properties after irradiation at 800, 1000 and 1200°C.

The samples irradiated in the extra campaign (in similar conditions to the main irradiation programme) are now also dismantled and the activity on the samples is measured. The sample irradiated up to 0.2 dpa exhibit the activity of less than 2 mSv/hour – i.e. can be handled in fume hood. These samples could be used for future CRPs.

1. C. Yin, D. Terentyev *et al.*, *Materials Science and Engineering: A 750 (2019)20-30*
2. C. Yin, D. Terentyev *et al.*, *International Journal of Refractory Metals and Hard Materials, Volume 75, September 2018, Pages 153-162*

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Hydrogen isotope permeation in tungsten under mixed ion irradiation

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Plasma facing surfaces of divertor and blankets are subject to irradiation of fuel ions (D,T) together with various impurity ions from edge plasma such as a fusion ash ion (He), cooling gas ions (N, Ar, Ne), and wall material ions (Be, W, etc.). Concentrations of these impurity ions in plasma are usually small (up to about 10%), but their impact on permeation is very large. In this report, we summarize our work on ion driven permeation in W using mixed ion irradiation with fuel ions (mainly D) and several impurity ions (C, He, Ne, Ar, N).

Mixed ion irradiation was made by an ion beam device with an ion energy of 1 keV and an ion flux of $\sim 10^{20}$ $m^{-2}s^{-1}$. Tungsten specimens with thickness of 30 μm with the purity of ~99.99% were used. Both surfaces were mirror finished. The specimens were annealed at 1573 K for an hour to reduce intrinsic defects. Effective diffusivity of D in the specimens estimated by a time lag method was close to that of Zakharov [1].

Reactive ions (C, N) and inert gas ions (He, Ne, Ar) show different behavior. Reactive impurities (C, N) tend to increase permeation compared with pure D, caused by surface mixed layers, which could work as recombination and/or diffusion barrier. Inert gases (He, Ne, Ar) tend to reduce permeation, caused by surface layer modification originated by bubble formation as well as sputtering. Permeation behavior in all cases strongly depends on temperature.

For C/D mixed irradiation, saturation of permeation against incident flux were observed at temperatures lower than about 700 K. The reason is not well understood, but an analysis based on DFT and thermodynamics [2] indicated that solute D in W over some criticl concentration (temperature dependent) tends to produce D bubbles, which could reduce mobile D concentration in W. This phenomenon could also take place under extremely high flux conditions (up to $\sim 10^{24}$ $m^{-2}s^{-1}$) at divertor. Therefore, it is necessary to make further research by using high density plasma simulators.

For He/D mixed irradiation, reduction of permeation was observed below 1000 K and the reduction becomes more significant with decreasing temperature, which is closely correrated to reduction in retention. At temperatures where this reduction takes place, permeation flux scales to a squar root of incident flux. If we follow Brice-Doyle model[3], surface recombination could be a limiting process of release of implanted D from plasma facing surface, which could be attributed to formation of fast diffusion channls of D_2 by connection of small He bubbles.

1. A. P. Zakharov, et al., Fiz.-Khim. Mekh. **9**, 29 (1973).
2. Lu Sun et al., J. Phys.: Condens. Matter 26 (2014) 395402 (9pp).
3. Brice D.K. and Doyle B.L., J. Nucl. Mater. 120 (1984) 230-244.

Hydrogen Permeation in Fusion Materials Activities at FZ Jülich

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Fuel retention and hydrogen permeation in the first wall of future fusion devices are crucial factors. Due to safety issues and in order to guarantee an economical reactor operation, tritium accumulation into reactor walls and permeation through walls have to be estimated and prevented. Therefore, studies of hydrogen retention and permeation in the first wall materials are performed and tritium permeation barriers (TPB) are developed.

Steels are foreseen as structure materials in fusion devices. Two kinds of steels, the ferritic-martensitic Eurofer97 steel, foreseen for the use in DEMO, and the austenitic 316L(N)-IG steel, implemented in ITER, were investigated. Detailed permeation studies were performed on this steels. Furthermore, the influence of a technical sample surface on the hydrogen permeation was studied [1]. Since the structure material will be exposed to high energy deuterium and tritium particles in ITER and future fusion devices, steel samples were exposed in the linear plasma device PSI-2 to a deuterium plasma. The influence of the exposure on the deuterium retention and permeation was studied in order to estimate the performance in a fusion device [2].

In order to study the influences of interfaces on the permeation behavior, the deuterium permeation through a Cu coated 316L(N)-IG substrate was investigated. By comparing the results with the permeation through pure Cu and 316L(N)-IG samples, the influence of the interface can be calculated and the performance in a component in ITER can be estimated.

Metal oxides were identified as high temperature resistant materials with a low hydrogen permeation. The hydrogen permeation through various materials was measured in the last decades and compared. Due to the strong influence of the microstructure of the coating on the hydrogen permeation, the obtained results can vary widely. Therefore, in order to understand the hydrogen permeation process through the TPB, a detailed phase and microstructure analysis is important.

Y_2O_3 layers prepared by magnetron sputter deposition on steel substrates were developed for TPBs and studied. The stoichiometry, the crystal structure and the microstructure of the deposited and annealed layers were investigated by X-ray diffraction and scanning electron microscopy. The deuterium permeation was studied and compared to a reference measurement of an uncoated substrate in order to determine the permeation reduction factor (PRF). In Y_2O_3, the influence of the microstructure on the hydrogen permeation was investigated in detail. The PRFs of two Y_2O_3 samples with different microstructures differ by two orders of magnitude. Thus, by improving the microstructure of the TPB, the permeation reduction behavior is strongly enhanced [3].

[1] A. Houben *et al.*, *Nuclear Materials and Energy* **19**, 55 (2019)
[2] A. Houben *et al.*, *Physica Scripta*, submitted
[3] J. Engels *et al.*, *International Journal of Hydrogen Energy* **43**, 22976 (2018)

A part of this work has been carried out within the framework of the EUROfusion Consortium and has received funding from the Euratom research and training programme 2014-2018 and 2019-2020 under grant agreement No 633053. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

Appendix IV: Core of Proposal for a CRP on Hydrogen Permeation

The formal proposal to initiate a CRP follows a certain template. What follows are the key narrative sections.

Scope and aims of the CRP

An activity by the A+M Data Unit would be concerned with the production, evaluation and recommendation of data on hydrogen mass transport properties in each fusion material used for T permeation assessment in fusion devices. The relevant FW materials are those foreseen as first wall and divertor plasma-facing materials. The Unit would also coordinate code-code comparisons and benchmark exercises of reaction-diffusion codes as well as experimental permeation systems among the CRP participating parties. Moreover, this CRP can coordinate a joint programme involving specialized institutes to assess the effect of neutron irradiation on hydrogen isotope retention and permeation in the relevant and thoroughly characterized plasma-facing materials so as to enable validation of the computational models by offering a consistent set of experimental data.

The application domain of nuclear fusion motivates priorities among the elemental species, but the applications are not a part of this CRP. The CRP does not have direct objectives to estimate suitability of wall components or materials for fusion applications, or to assess the viability of various plasma scenarios or device designs, which would involve hydrogen permeation.

The most important species for attention in the CRP are those of W (as a divertor target material, but also as a DEMO first wall material), main wall materials Be and iron (steels), hydrogen isotopes (D, T) and intrinsic impurities such as O and C, but also the seeded impurities, such as N, Ne and Ar.

In addition to concrete objectives in the production, evaluation and recommendation of permeation data this CRP will also have an important role in bringing together physicists with expertise in experimental fusion materials research and with plasma-wall interaction modellers that study hydrogen permeation in fusion devices and in controlled laboratory conditions. The CRP must contribute to the transfer of permeation data and associated expertise into the modelling activities.

Relation to other CRPs

The Atomic and Molecular Data Unit has coordinated three CRPs in recent years on plasma-material interaction in fusion devices, each for one specific class of relevant materials.

- CRP F43020 on Data for Erosion and Tritium Retention in Beryllium Plasma-Facing Materials (2012-2016).
- CRP F43021 on Plasma-Wall Interaction with Irradiated Tungsten and Tungsten Alloys in Fusion Devices (2013-2018).
- CRP F43022 on Plasma-wall Interaction with Reduced-activation Steel Surfaces in Fusion Devices (2015-2020).

These three earlier CRPs are concerned with material erosion and with T retention in fusion materials under routine plasma exposure. The information gained from these previous CRPs will be of use when assessing the effects of plasma-surface interactions to long-range and long-term hydrogen permeation in fusion reactor components.

Nuclear Component

The processes considered in this CRP have a key role in nuclear fusion experiments and in future nuclear fusion devices including ITER and DEMO.

CRP Overall Objective

To support fusion energy research in Member States by providing trusted data for processes relevant to hydrogen permeation in fusion materials, and thereby to contribute to the development of fusion energy generation.

Specific Research Objectives

1. To inventorize knowledge of parameters affecting hydrogen permeation in fusion-related materials, including temperature, microstructure and irradiation-induced defects (neutron transmutation products; neutron and ion-induced lattice damage).
2. To increase the knowledge of the isotope effect on hydrogen diffusion
3. To decrease the uncertainties related to diffusion, solubility and trapping parameters affecting permeation
4. To expand the knowledge-base concerning the influence of material microstructure on permeation, including the evolution of microstructure during permeation experiments
5. To assess and, as far as possible quantify, the effect of surface and sub-surface conditions on permeation
6. To perform coordinated experiments and simulations to improve the knowledge-base on hydrogen permeation processes with non-irradiated and irradiated material
7. To compare experimental facilities and techniques in hydrogen permeation

Intended Outcomes

1. Data produced, evaluated and/or recommended in the CRP will be used in fusion plasma-materials interaction modelling and assessments towards the T inventory and permeation in next-step fusion devices, such as ITER and beyond
2. The CRP will reduce the uncertainties in critical parameters used for T transport modelling

Planned Outputs

A meeting report in the INDC (Nuclear Data Section) series will be produced after each Research Coordination Meeting.

Citable, peer-reviewed articles in the academic literature relating to the CRP's activities will be produced by participants in the CRP.

The Knowledge Base and other web pages of the IAEA A+M Data Unit will be kept up-to-date to reflect the work of the CRP.

ALADDIN and other databases of the A+M Data Unit will be augmented with data produced or evaluated in the CRP.

If the data assembled merit it, a searchable online database of critically-assessed parameters relevant to hydrogen permeation in fusion materials, with their provenance and estimated uncertainties will be constructed and hosted at the A+M Data Unit.

A final report summarizing the CRP outputs and making recommendations of best-practice in hydrogen-permeation research and proposing future work.

Schedule of Activities

Q2 2020: Identification of participants, assembly of the CRP.

Q3 2020: First RCM.

Q1 2022: Second RCM.

Q4 2022: Mid-term review of the CRP.

Q4 2023: Third RCM.

2024: Development and publication of CRP final report.

Appendix V: Selected Articles and Reports

This Appendix contains a brief summary of relevant literature.

F. Reiter, K. S. Forcey, G. Gervasini, *A Compilation of Tritium-Material Interaction Parameters in Fusion Reactor Materials*, Joint Research Centre EUR 15217 EN, Commission of the European Communities, Luxembourg 1993

ITER Final Design Report, ITER EDA Documentation Series No. 16, IAEA (1998)

J. Roth, et al., *Tritium inventory in ITER plasma-facing materials and tritium removal procedures*, Plasma Phys. Control. Fusion 50, 103001 (2008).
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J. Roth, et al., *Hydrogen in tungsten as plasma-facing material*, Phys. Scr. T145, 014031 (2011).
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T. Hirai, et al., *ITER tungsten design development and qualification program*, Fus. Eng. Design 88, 1798 (2013).
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D. Stork, et al., *Materials R&D for a timely DEMO: Key findings and recommendations of the EU Roadmap Materials Assessment Group*, Fusion Eng. Des. 89, 1586 (2014).
doi: 10.1016/j.fusengdes.2013.11.007

A. R. Raffray, et al., *The ITER blanket system design challenge*, Nucl. Fusion 54, 033004 (2014).
doi: 10.1088/0029-5515/54/3/033004

Y. Hatano, "*Ch. 9 Permeation and Permation Barrier*" in Tritium: Fuel of Fusion Reactors, Ed. T. Tanabe, Springer, Japan, 2017.
doi: 10.1007/978-4-431-56460-7

K. Noborio, T. Tanabe, "*Ch. 15 Behavior of Tritium Released to the Environment*" in Tritium: Fuel of Fusion Reactors, Ed. T. Tanabe, Springer, Japan, 2017.
doi: 10.1007/978-4-431-56460-7

ITER Organization, *ITER Research Plan within the Staged Approach*, ITER Technical Report ITR-18-003 (2018)

J. H. You, et al., *European divertor target concepts for DEMO: Design rationales and high heat flux performance*, Nucl. Mater. Energy 16, 1 (2019).
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