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D Klimenko et al.

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Definition of the Q-PETE Experiment for Investigation of Hydrogen Isotopes Permeation through the Metal Structures of a DEMO HCPB Breeder Zone

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The Q-PETE (Hydrogen Permeation and Transport Experiment) at KIT is set up to investigate hydrogen isotopes permeation through structural materials with specific relevance to the HCPB (helium cooled pebble bed) DEMO blanket breeder zone. The results are intended to provide validation data for simulation codes. A second objective is the direct determination of material data. In the first stage deuterium permeation through a Eurofer membrane will be studied using a mass spectrometry method.

This paper describes the definition, the design development, the experimental setup and the characterization of the mass spectroscopy method foreseen for the Q-PETE experiment. The permeator setup was designed taking into account results from uncertainty optimization efforts and manufacturing aspects. Based on the decided geometry, engineering analyses concerning thermal responses and tritium transport were performed. The aim of the characterization of the mass spectroscopy method is to provide a proper foundation for the uncertainty analyses, as well as to optimize the technique. The signal to noise ratio was related to relevant spectrometer operation parameters. On this basis measures to reduce the background level for the deuterium concentration by a factor of 5 could be found and implemented to the spectrometer analysis algorithm.

Keywords: tritium transport, permeation, hydrogen isotopes, breeding blanket.

1. Introduction

In the concept for DEMO tritium is required for the foreseen D-T fusion process. One of the top level requirements for DEMO is to demonstrate the tritium self-sufficiency, including a breeding blanket with sufficient tritium breeding ratio (TBR > 1) and a fully featured tritium recovery and fueling process. Tritium and other hydrogen isotopes will therefore be present in DEMO systems, such as the Breeding Blanket. For each of these systems, from the viewpoint of safety, the tritium balance must be studied in respect to: release of tritium to the neighboring systems or the environment in normal operation conditions, the accumulation of tritium in the system or component, the possible release of tritium to neighboring systems or the environment in accident conditions, the accumulated amount of tritium and the tritium mobility in the system or component relevant for decommissioning and disposal.

For according assessments, a number of numerical codes exist and is under development. It was decided to focus this work on the breeding zone of the HCPB blanket and the code on component scale to be developed for this blanket in the frame of Eurofusion program [1]. Also existing codes like TMAP [2] and FUS-TPC [3] can be validated with the planned experiments. The involved physics (exchange of tritium between a gas phase and a solid metal) is furthermore common to the tritium extraction system and the vacuum systems.

2. Experimental setup

2.1 Definition

The Q-PETE experiment simulates the situation in the DEMO HCPB Breeder Zone, where the hydrogen-carrying (H2, HT, T2, possibly H2O) helium purge gas flow is separated by the cooling plates made of Eurofer steel from a coolant helium flow. In this situation, a permeation flux of hydrogen isotopes (of generic hydrogen species “QQ”) from the purge gas side through the steel into the coolant gas will establish. This situation is represented in the experiment by providing three important elements:

1. Retentate side chamber (also called primary side or high pressure side), where a variably controlled hydrogen isotope partial pressure $p_{QQ,1}(t)$ is set;

2. Permeation membrane, which can be made of various materials (most relevant: Eurofer97), in several thicknesses (to access a broad range of experimental parameters) and with defined surface conditions (defined by manufacturing method and other surface modifications like cleaning);

3. Permeate side chamber (also called secondary side or low pressure side), where a quantified purge gas flow is collecting the permeated hydrogen and transports it to a measurement instrument. There a time resolved, quantitative analysis is performed in order to reconstruct the transient QQ permeation flux during the experiment.

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This assembly (1.-3.) is called the permeation setup or permeator. As temperature plays a decisive role in permeation, the setup is foreseen to operate in a wide temperature range of 200 - 600°C at quasi-isothermal conditions all over the membrane.

Important peripheral systems are:

- The retentate side feed gas system. It will supply a (time dependent) flow of argon carrier gas, with a specified hydrogen isotope molar fraction $x_{(QQ,FG)}$, a measured total pressure $p_1(t)$ in the retentate chamber and a molar flow rate $J_{FG}(t)$

- The permeator heater and temperature control system, adjusting and measuring the membrane temperature T_m.

- The guard vessel or vacuum vessel containing the permeator setup.

- The permeate side sweep gas system, providing a controlled flow of a sweep gas with a molar flow rate $\dot{J}_{SG}$, and adjusted total pressure $p_2\ (t)$ in the permeate chamber.

- The permeate side analysis station, measuring the time dependent molar fraction of QQ $X_{QQ,QMS}(t)$.

A simplified schematic of the setup is shown in Fig. 1

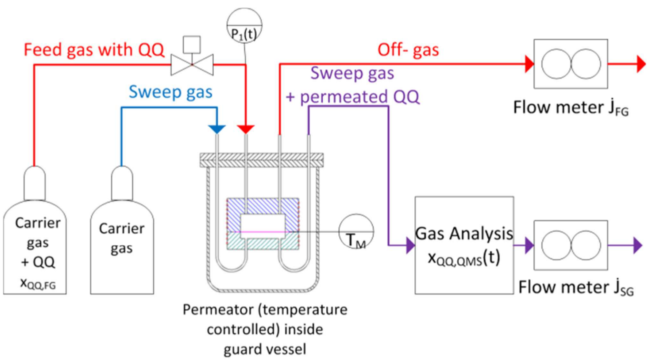

Fig. 1. Functional schematic of the Q-PETE experimental setup

In the first stage, the experiment will be performed in a conventional laboratory (no tritium) and deuterium (D_2) with the molar mass of 4 g/mol will be used as traced hydrogen isotope. As primary gas analysis instrument, an IPI GAM300 quadrupole mass spectrometer (QMS) will be used. The noble gas argon (Ar) is chosen as carrier gas, substituting helium with molar mass 4 g/mol (identical to D_2) to allow for mass spectrometric discrimination from the traced hydrogen isotope. The setup shall enable a good time resolution to reconstruct a good estimate for $J_{(Q,M)}(t)$ and adherent to these specifications the experiment is called Q-PETE/D2.

2.2 Design Development

For the permeator a cylindrical geometry with large diameter/gap width ratio was chosen to facilitate abstraction of this geometry which can be easily manufactured and controlled in the to-be-validated codes. As shown in Fig. 2, the retentate chamber and the permeate chamber are built from thick-walled flange-like structures into which a cavity is machined. The membrane disk made from the material under test (i.e. Eurofer) separates the two cavities. The membrane is supported by two opposing "C"-cross-section (soft) Cu-coated metal sealing gaskets. The two flanges are fixed against each other by screws (not shown). An exchangeable (thickness varying with membrane thickness) spacer ring makes sure that (1) the flanges can be screwed tightly together, (2) the metal gasket receives the prescribed deformation, but (3) the membrane is not clamped/fixated to the flanges (a nominal gap of 0.1mm remains on each side).

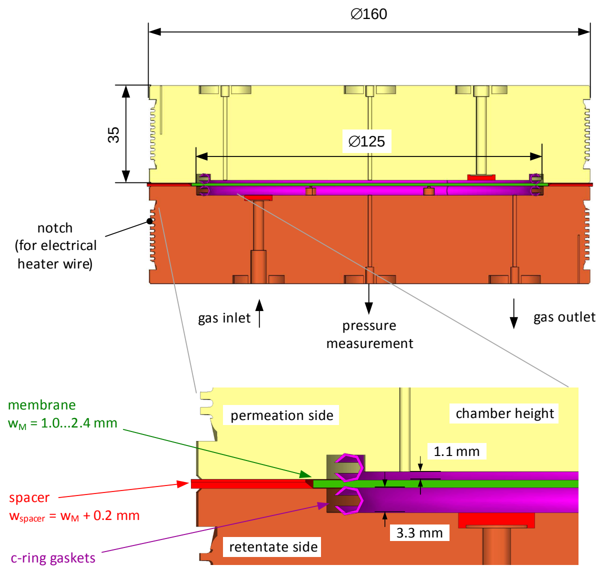

Fig. 2. Permeator composed of retentate side chamber and permeate side chamber separated by the permeation membrane. Top: overall view. Bottom: detail view with gaskets

For thermal insulation, the permeator will be placed inside a guard vacuum vessel as shown in Fig. 3. A concentric assembly of thermal radiation shields will further contribute to the thermal insulation. The support structure on which the permeator is mounted also strives to minimize thermal conduction losses. The massive permeator together with the thermal insulation measures aim at providing isothermal conditions for the membrane during the tests. Heat-up and compensation of steady-state heat losses is provided by electrical heater wires that are wound in spiral notches/groves on the cylindrical outer surface of each of the two flanges of the permeator. Four purge gas pipes, electrical heater power cables and sheathed thermocouples need to penetrate the guard vessel cap. This is done using feed-through Conax glands.

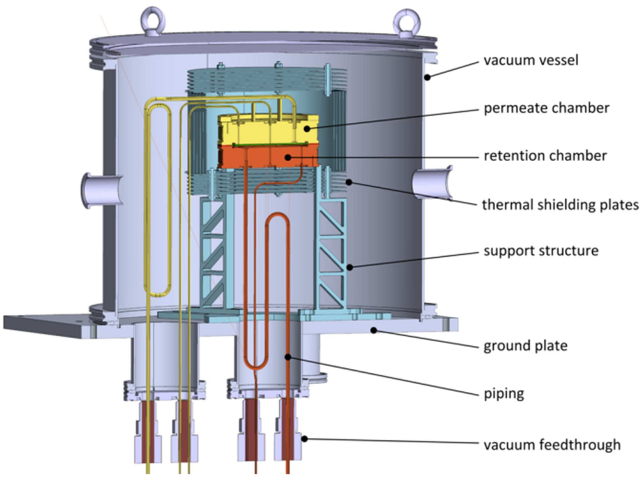

Fig. 3. Guard vessel with permeator inside.

Fig. 4 provides a detailed view of the retentate side flange. The heater wire (Thermocoax type OD=1mm) is clamped into a spiral grove (9 windings), resulting in a wire length of about 4.5m. Six M10 bolts are used to fix the two flanges. 4 spacer knops are machined in the retentate side chamber to support the membrane against bending by overpressure from the permeate side chamber (in the condition when the retentate side chamber is evacuated). Thermocouples are led to the membrane through a central hole.

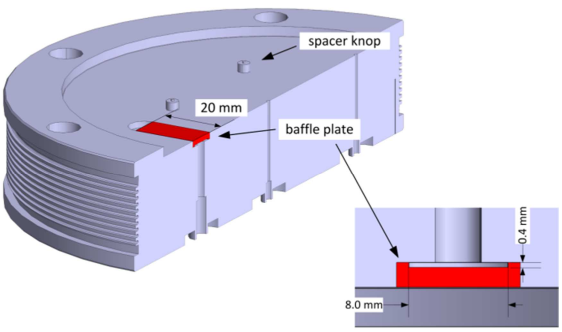

Fig. 4. Cut view of the permeator flange (retentate side).

It is important that the temperature of the incoming gas is close to the membrane temperature to avoid local cooling of the membrane. CFD studies revealed that heating up of the gas passing the drill hole (35mm depth) into the chamber alone is not sufficient. For this reason the feed gas is lead in through a pre-heating duct. Additionally, the contact surface of the incoming gas to the hot chamber is increased by a baffle plate. Figure 4 indicates that the flow cross section along the baffle plate corresponds to a flat duct with a rectangular cross section of 0.4 x 8.0 mm. For a further increase of the contact surface and to reduce the flow speed in the narrow channel, the flow is split at the center of the baffle plate and guided to the chamber through two inlets that are separated by the length of the baffle plate of 40mm.

In a laminar CFD simulation of this channel with according boundary conditions it was shown that the maximum difference between inlet gas temperature and membrane temperature is 13.4K. If the same thermal equilibration is to be reached in a circular channel (drill hole ∅ 1.4 mm), the channel length would have to be at least 160 mm.

3. Characterization of mass spectroscopy method

An IPI GAM300 mass spectrometer will be used as the key analysis instrument in the Q-PETE setup as described above. The capabilities of this spectrometer had been tested against the requirements in a preparative campaign of measurements. The requirements had been estimated by hydrogen transport simulations [4]. Those results provided an expected steady state concentration of 0.5-20ppm of deuterium in argon at temperatures 250-550°C. These estimated data are the basis for the following investigations to the resolution of the mass spectrometer for deuterium in argon. The spectrometer signal for deuterium will appear at m/Z=4.

The process diagram for Q-PETE experiments in Fig. 5 shows how the GAM 300 mass spectrometer is implemented in the permeator setup.

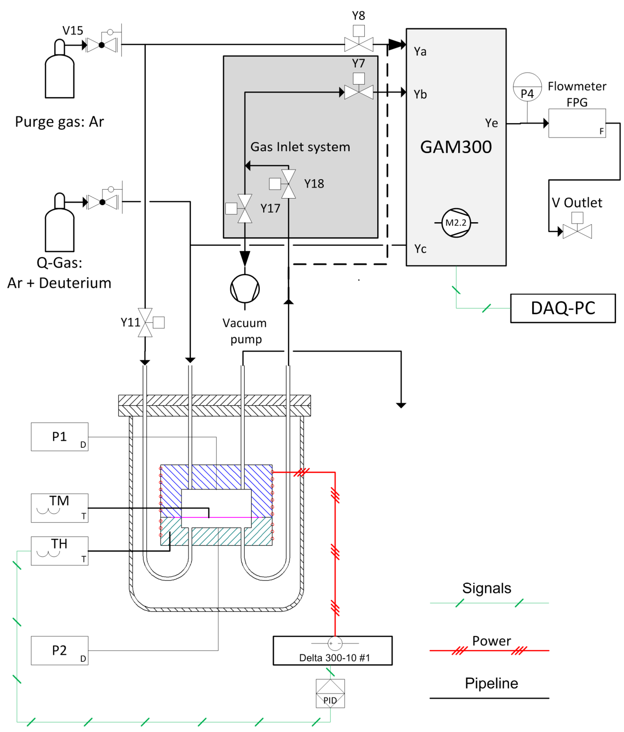

Fig. 5. Process diagram of the PETE-Q setup including the GAM 300 mass spectrometer.

The preparative experiments identified the detection limits (LOQ – limits of quantification) for deuterium in argon as 248 ppb and 141 ppb for SEM (Secondary electron multiplier) voltages 1200 V and 1400 V, respectively. The sensitivity analysis was performed by investigating the signal to noise ratio using several calibration gas mixtures of deuterium in argon.

In order to acquire the transient permeation behavior of D_2, the spectrometer channel integral time or a time a

QMS spent on each measured mass (so called dwell time Δt_{QMS}) should be kept as small as possible. A parameter study of spectrometer resolution vs. dwell time at m/Z=4 of signal and noise behaviors for the gas mixture of 0.91ppm D_2 in argon for a SEM voltage of 1400 V is presented in Fig. 6. Depending on the quantity permeated through the membrane, a reliable acquisition of the deuterium flow is still possible with a dwell time of 0.1 s.

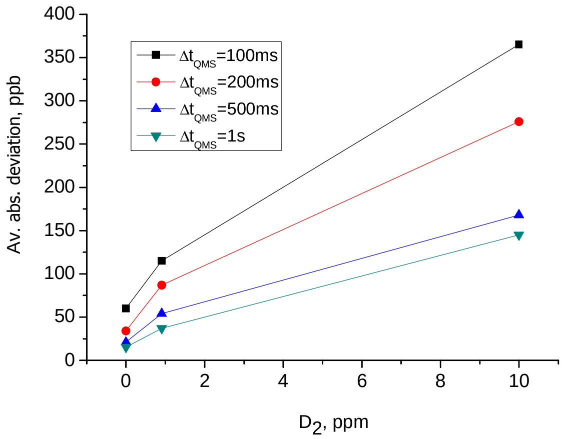

Fig. 6. Average absolute standard deviation at m/Z=4 vs. D_2 concentration. Volume flow: 330-340 sccm. Dwell time: Δt_{QMS}=1s, 500ms, 200ms, 100ms.

Fig. 7 shows the residual background signal at m/Z=4 when pure argon is supplied to the QMS. The signal increases visibly for low volumetric flow, i.e. a m/Z=4 contribution seems to accumulate when no purge is done. Because also components that contain helium (also m/Z=4) share this spectrometer, this background contribution is most probably attributed to helium leaks in the gas inlet system of the QMS.

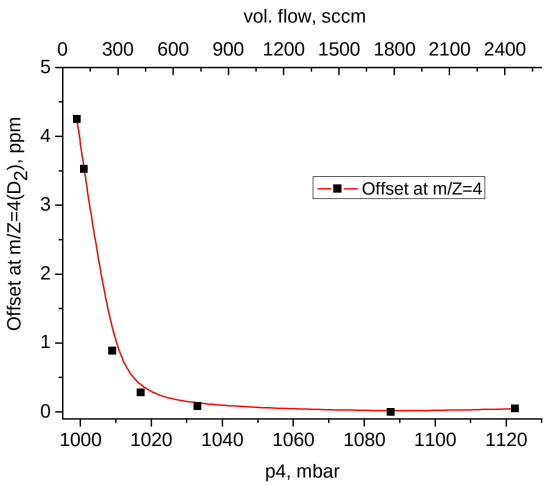

Fig. 7. Signal background at m/Z=4 as a function of pure argon 6.0 volumetric flow (top axis) and inlet pressure p4 (bottom axis) in preparative experiments.

Several measures to reduce the background had been undertaken. First, a needle valve was installed at the spectrometer outlet (= "V Outlet" in fig. 5) to rise the absolute pressure level (p4) in the spectrometer from 1bar up to about 1.5bars. The overpressure should antagonize possible (helium) leaks in the valves of the spectrometer inlet system. Second, for the same reason, the inlet port closest to the analyzer was selected and the evacuation routine and equipment of the inlet system was improved. A turbomolecular pump (HiCube by Pfeiffer Vacuum) or a diaphragm pump could be alternatively connected to the gas inlet system (at valve Y17 in fig. 5) of the QMS to remove possibly leaked in helium from the gas inlet.

The results of the experimental analysis (at a SEM voltage of 1200V) are presented in Fig. 8. The red horizontal line displays the LOQ level, which must not be exceeded by the remaining background signal at m/Z=4. Three curves satisfy this condition. The lowest values of offset was achieved by the configuration with a turbomolecular pump (TP) at p4=1.6 bar (red curve), but the also the configuration with a diaphragm pump (MP) and with p4=1.3 bar and p4=1.6 bar (purple and black curves respectively) still provides appropriate offset values being significantly below the LOQ level.

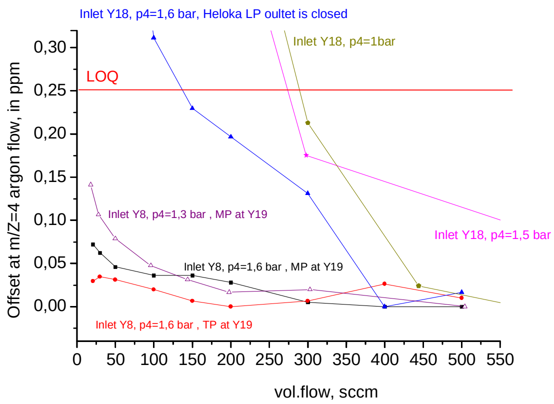

Fig. 8. Signal background at m/Z=4 vs. volume flow for different configurations of the gas inlet of the mass spectrometer (SEM 1200V).

From these finding the foreseen parameters for the Q-PETE experiment are a SEM Voltage of 1400 V and a spectrometer absolute pressure close to p4=1.4bar. The measurements for the offset (i.e. purge with pure argon 6.0) were performed also at these conditions. In order to remove impurities of helium, the gas inlet system was flushed and evacuated with argon several times. After flushing, the unused part of the spectrometer gas inlet was permanently pumped by a turbomolecular pump during these experiments. The results are shown in fig. 9. The background is well suppressed and significantly lower than the corresponding limit of detection LOD (red line on the graph).

Another important issue is the purity of the argon 6.0 gas with regard to helium traces that also contribute to m/Z=4 like deuterium. Suppliers typically do not specify helium impurities separately. It turned out that here large differences (still within the global impurities specifications) exist. For argon 6.0 we found impurities at m/Z = 4 from up to 1 ppm down to values below our detection limit of 43 ppb depending on the supplier. For the same reason we order the carrier gas (argon with several ppm of deuterium) only at the suppliers that also supply argon 6.0 with helium traces below our detection limit.

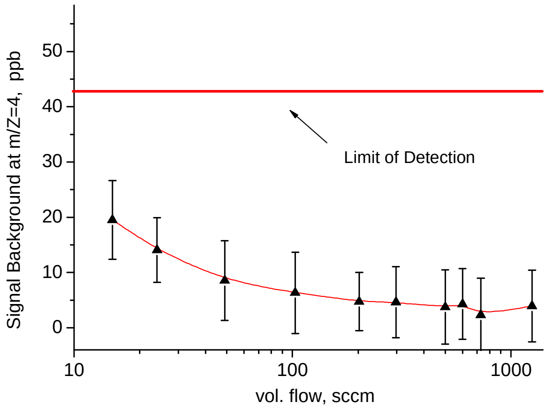

Fig. 9. Signal Background (with average absolute standard deviation for each measuring point) at m/Z=4 vs. volume flow for p4=1.4 bar (SEM 1400 V).

Summary and Conclusions

Details of the permeator setup and the analysis equipment for the Q-PETE hydrogen permeation experiments at KIT are presented. Calculations including CFD analyses provided the expected ranges of data and parameters and indicated that the required boundary conditions for the experiments will be kept by the permeator. A number of measures have been successfully implemented on the experimental setup to improve the background suppression. It was shown in preparative measurements that the mass spectrometer which is foreseen for the evaluation of these experiments can fulfill the expected requirements, too.

Acknowledgments

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