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Development of a hydrogen permeation sensor for future tritium applications

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HIGHLIGHTS

- Designing and testing of a hydrogen permeation sensor.
- Palladium and α-iron have been used as a hydrogen permeation materials in the sensor.
- The experiments performed using both membranes showed that the operation of the sensors in the equilibrium mode required at least several hours to reach the hydrogen equilibrium pressure.

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ABSTRACT

Tritium monitoring in lithium–lead eutectic is of great importance for the performance of liquid blankets in fusion reactors. In addition, tritium measurements will be required in order to proof tritium self-sufficiency in liquid metal breeding systems. On-line hydrogen (isotopes) sensors must be design and tested in order to accomplish these goals.

In this work, an experimental set up was designed in order to test the permeation hydrogen sensors at 500 °C. This experimental set-up allowed working with controlled environments (different hydrogen partial pressures) and the temperature was measured using a thermocouple connected to a temperature controller that regulated an electrical heater.

In a first set of experiments, a hydrogen sensor was constructed using an α-iron capsule as an active hydrogen area. The sensor was mounted and tested in the experimental set up. In a second set of experiments the α-iron capsule was replaced by a welded thin palladium disk in order to minimize the death volume. The experiments performed using both membranes (α-iron and palladium) showed that the operation of the sensors in the equilibrium mode required at least several hours to reach the hydrogen equilibrium pressure.

1. Introduction

One of the questions that the scientific community should overcome in the ITER project is the tritium generation. Lithium–lead eutectic is one of the candidates to be used for tritium generation in the He-Cooled Lithium–Lead Test Blanket Module (HCLL TBM). In order to proof the tritium self-sufficiency in the liquid metal breeding system the on-time and on-board tritium concentration measurement will be required [1,2]. In this framework, the need for a reliable and fast sensor for the measurement of tritium concentration in the liquid breeder system is of high interest.

Metal based membranes have been utilized for hydrogen separation/purification since the early 1960s [3]. These membranes can provide high hydrogen fluxes, resistance to poisoning, and long operational lifetime. In addition, Pd metal membranes are well suited for high-temperature applications. Palladium metal membranes have received much attention for H_2 purification because, in principle, they have perfect selectivity for H_2 over other gas species. Pure Pd membranes, however, are susceptible to H_2-induced embrittlement and to sulfur poisoning.

In order to overcome the drawbacks related with the use of bulk Pd, research has focused on methods of reducing the amount of palladium needed (i.e. Pd-coated substrates or different Pd-alloys), decreasing the effect of hydrogen embrittlement, and/or eliminating palladium as a membrane component (i.e. using other metallic materials). Even though a large number of metals and binary alloys

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have been tested experimentally as metal membranes [4–7], the search for durable and cost effective alloys that show high permeability for H_2 continues to be an active area.

Therefore, tritium permeation through a metal based membrane seems to be an effective principle for a hydrogen sensor. A hydrogen permeation sensor can be seen as small cylinder in which its base is permeable to hydrogen. Then, starting from vacuum the pressure inside the sensor should increase because of hydrogen permeation through its walls until the equilibrium pressure is reached.

The measurement of the hydrogen equilibrium pressure in the sensor or the hydrogen permeation flux through the membrane can be correlated with the hydrogen concentration in the working environment. As a result, this type of sensors can operate in two different modes: the equilibrium mode and the dynamic mode. In dynamic mode, the hydrogen concentration is measured by the rate of its permeation through its walls, however, in the equilibrium mode the pressure inside the vacuum sensor increases until the equilibrium pressure is reached (P_{eq}).

The hydrogen equilibrium pressure measured inside the sensor can be related to the hydrogen concentration in the environment (C_L) by the Sievert's Law for a hydrogen/lithium–lead eutectic system

$$C_H = K_S P_{eq}^{0.5}$$

where K_S is the Sievert's constant. Then, from a theoretical point of view the measurement of the hydrogen concentration relies on the determination of the hydrogen Sievert's constant for the molten alloy in the breeding blanket (lithium–lead). Much information about hydrogen transportation properties in different metals and alloys can be found in literature [8].

The Electrochemical Methods Laboratory at Institut Quimic de Sarria (IQS) has been working in the design and characterization of high temperature gas sensors, mainly electrochemical sensors, for molten alloys since 1998 [9–11]. In this work, a permeation hydrogen sensor based on metal membranes (palladium and α-iron) has been tested. The final goal of this project is to design and develop a high performance and fast response time tritium sensor for molten lithium–lead eutectic.

2. Experimental method

2.1. Design of the sensor

The sensors use two different hydrogen permeation membranes: Pd and α-iron. Palladium was selected because it is high hydrogen permeability ($\phi_{Pd/500\,°C} = 7.31 \times 10^{-8}$ mol/m s Pa$^{1/2}$ [12]) and α-iron ($\phi_{Fe/500\,°C} = 2.56\ 10^{-10}$ mol/m s Pa$^{1/2}$ [13]) was selected as a compatible material with lithium–lead eutectic. Fig. 1 shows a schematic representation of the sensor.

The sensors are based on a design previously published in literature [4,5], and can be seen as a hollow capsule submerged in a controlled environment with a fixed hydrogen partial pressure. The capsule is then connected to a small tube, to a pressure transducer and to a pumping vacuum system.

The sensors consist on the following parts:

Sensor capsule. Two different hydrogen permeation materials have been used in the following configurations:

- A palladium foil (99.95%, thickness 0.25 mm supplied by Goodfellow) was welded to a 316SS body. See Fig. 2a).
- A small cylinder of α-iron (wall thickness 0.5 mm) machined from an iron rod (99.8%, supplied by Goodfellow). It was finally welded to a 316SS body. See Fig. 2b).

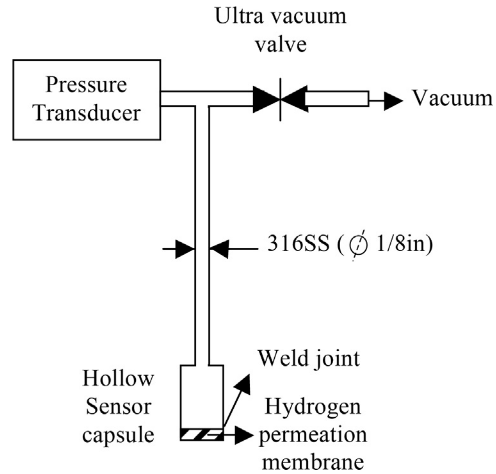

Fig. 1. Sensor design.

- The tube was made of 316SS (diameter 3.175 mm–1/8 in) and the connection between the tube and the vacuum pump and the pressure transducer was done using a tee fitting.
- A vacuum valve (Swagelok® ref. SS-2H) was used to isolate the sensor body from the vacuum pumping system (Alcatel 2010 Pascal Rotary Vane Mechanical Vacuum Pump).
- A pressure transmitter (Sensor-Technik Wiedemann GmbH, ref. STW S04) connected to a computer for data acquisition was used to measure the inner pressure of the sensor.

Due to the geometries of palladium and α-iron membranes employed, the dimensions of the sensors are slightly different. These differences in the dimensions of the sensors cause a different inner volume for each sensor; the palladium sensor has an inner volume of 385 mm^3 whereas in the α-iron sensor is 445 mm^3.

In order to obtain different hydrogen partial pressures in the system high purity argon (99.999%) and a calibration mixture of Ar + 10% H_2 were mixed at different flows. Argon was selected as a non-reactive/inert gas. The gases flow was controlled using a set of mass flow meters (Bronkhorst EL-FLOW®-Select F-201-CV). All gases were supplied by Abello-Linde.

2.2. Experimental set-up

All the tests were performed in an autoclave. Fig. 3 shows a schematic representation of the experimental set-up.

This experimental set-up allowed working with controlled environments. The temperature of system was measured using a thermocouple connected to a temperature controller that regulated an electrical heater. All experiments were performed at 500 °C. The

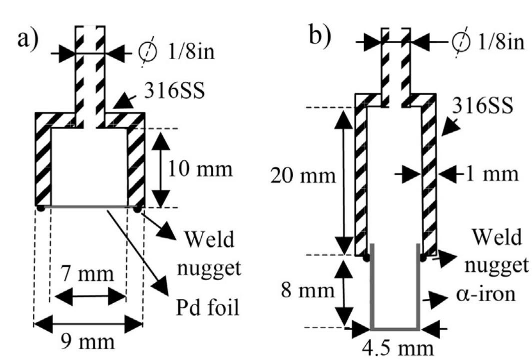

Fig. 2. Sensor capsule (a) using a Pd foil and (b) using α-iron as a hydrogen permeation membrane.