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Hydrogen and deuterium distributed sensing using Chirped-Pulse φOTDR

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ABSTRACT

The detection and quantification of the presence of certain chemical species is of central importance regarding permanent structural health monitoring of key industrial fields and civil infrastructures such as oil extraction boreholes or radioactive waste repositories, where H_2 is released. With this work we propose and test a competitive technique able to measure the concentration of hydrogen and deuterium thanks to their diffusion into the silica glass of a standard optical fiber, already employed for the distributed monitoring of large infrastructures. The proposed technique, based on Chirped-Pulse Phase-sensitive Reflectometry (CP-φOTDR), could represent a novel solution for this problem, thanks to its ability to provide dynamical measurements of refractive index change, with great linearity and sensitivities of 10^{-8} refractive index units, featuring spatial resolutions of a few meters and kilometric sensing ranges.

Keywords: Distributed optical fiber sensors, Rayleigh scattering, chemical sensing, gas diffusion, radioactive waste storage

1. INTRODUCTION

In the last decades, the presence of distributed optical fiber sensors has increased, in particular for those applications related to structural health monitoring and surveillance of big structures. The low intrusion, electromagnetic immunity of the sensing fiber, chemical and mechanical stability of the silica glass or low maintenance cost of the interrogator systems are only some of their advantages. They are able to provide measurements for hundreds or thousands of points along several kilometer long fibers, and for a wide variety of physical quantities. Chemical sensing is only a subset of the target monitoring quantities, and multiple schemes exist that focus on the detection and measurement of gas species like molecular hydrogen. For some facilities or industries, such as oil extraction boreholes or radioactive waste repositories, excessive presence of H_2 may be an indicator or problem itself. In addition to the optical absorption and the enhanced mechanical fatigue of the glass due to the generation of OH, the presence of H_2 in some operating environments can affect some of the measurements of interests (temperature/strain), not only due to the induced signal attenuation (and its consequent effect on sensing range), but also by introducing apparent measurand variations. Luckily, the main detrimental effects can usually be prevented by the application of hermetic coatings (metals/carbon) to the sensing fibers[1].

For these reasons, there is a growing interest from institutions such as Andra in the qualification of distributed gas sensors able to work in harsh conditions. Hydrogen is released in structures such as the French underground repository for high and intermediate level long-lived radioactive wastes (Cigéo project)[2], and much effort has been devoted to identify sensors able to measure its concentration. Some of them benefit from an intermediary transducer such as palladium or platinum[3], but simpler methods are under continuous research to achieve technologies basing on standard telecom-grade fibers.

Due to the permeability of fused silica glass to the diffusion of small molecules such as hydrogen or deuterium, conventional telecom fibers can be exploited for the development of distributed gas sensors. In this work we propose an effective method for detecting/quantifying the presence of these species based on Chirped-Pulse Phase-sensitive Optical Time-Domain Reflectometry (CP-φOTDR).

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2. GAS DIFFUSION IN FUSED SILICA GLASS

When molecules like H_2 or D_2 become present in the optical fiber surroundings, they start diffusing through the fiber glass. The way this happens is determined by the characteristics of the substances involved. A detailed model of the gas kinetics can be found in reference 4. In the case of optical fibers, this model is constrained to the cylindrical symmetry of the optical fiber, where diffusion progresses radially. The solutions to the governing equation describe the evolution in time (t) of the gas concentration at the core of the fiber (where it will be measured) according to, basically, a simple exponential law. This qualitative behavior holds for both the in-gassing and the out-gassing processes.

We will define τ as the characteristic time of the phenomenological exponential variation of the gas concentration in the core (i.e., the non-constant term $\pm exp^{-t/\tau}$). This time parameter strongly depends on the fiber radius and decreases with the experiment temperature (T), but it is not affected by the pressure (P) of the surrounding gas. The saturation value for the gas load, on the other hand, increases with P and decreases with T.

The presence of these molecules in the glass lattice changes its dielectric susceptibility, and there is a linear relation between their concentration or P and the variation of the refractive index of the fiber. This constitutes the basis of the proposed sensing method.

3. EXPERIMENTAL PROCEDURE

The Fiber Under Test (FUT) is a concatenation of three different loose coils (~30 m long each) of Single-Mode Fiber (SMF), including a sample of Corning® SMF-28e®, a second segment of pure-silica fiber manufactured by Fibertronix®, and a third *Hole-Assisted* Carbon-Coated (HACC) fiber sample. The first one is employed as the gas sensing element of the setup. It is 125 µm in diameter with an acrylate primary coating. The others, provided with a carbon coating, are hermetic to the tested analytes. Their purpose is to serve as a distributed temperature sensor, without being affected by the gas, in order to provide a reference to subtract thermal effects in the signal of the permeable fiber. At the output end of the fiber, a few centimeters of coreless fiber were spliced, in order to increase losses and avoid unwanted reflections back to the interrogator.

The prepared FUT was placed inside a sealed high-pressure, stainless steel vessel provided with feedthroughs for fibers of different diameters or other instrumentation, as well as a manometer. The reactor was then filled with hydrogen at a pressure around 150 bars. For the desorption, the hydrogen was substituted by nitrogen (unable to diffuse into the glass), avoiding mechanical effects on the measurement due to a change in the external pressure. A similar procedure was followed with the deuterium, which was kept at 100 bars. The temperature of the whole experiment was around 25ºC (non-controlled, room temperature).

For the interrogation of the fiber, an experimental setup based on CP-φOTDR principles was used. In comparison with a conventional φOTDR sensor, the application of chirped pulses provides a high sensitivity and single-shot, linear readout, allowing to track the dynamic changes of refractive index (Δn). This information is extracted from local delays Δt found when comparing consecutive traces in the regions z where a perturbation has been applied. These induced shifts are proportional to the stimulus applied, through the relation[5]:

$$\Delta t|_z = \frac{\nu_o}{\delta\nu}\,\tau_p\,\frac{\Delta n(z)}{n}, \tag{1}$$

where ν_o and $\delta\nu$ are the spectral center and width of the pulses chirp, and τ_p is their duration. Hence, the measurement of the refractive index change at every fiber point is done by calculating Δt via cross-correlation of each trace region with the corresponding region from a previous trace. As this measurement can be performed from pulse to pulse, the probing frequency is solely limited by the fiber length, in order to prevent the consecutive traces from overlapping. For this reason, the method provides good performance for distributed acoustic sensing. However, by tracking the cumulative local Δt it is possible to carry out longer term measurements, as it is proved with the experiment here presented.

The optical setup used to implement this technique is shown in figure 1-left. The upper branch, devoted to preparing and emitting the pulses to the FUT, starts with an external cavity Laser Diode (LD) source. It operates in Continuous

Wave (CW) mode and its wavelength (1550 nm) is controlled by stabilizing its driving current and temperature (I, T). The signal is chopped into pulses by gating the CW signal with a Semiconductor Optical Amplifier (SOA). This determines the pulse length (6 m for durations of 60 ns). A synchronous ramp signal is superimposed to the LD bias current, producing a chirp that sweeps ~600 MHz during the pulse (providing a sensitivity to Δn of around 10^{-8} for the selected τ_p). Optical amplification of the pulse is done with an Erbium-Doped Fiber Amplifier (EDFA) whose Amplified Stimulated Emission (ASE) is filtered out prior to launching the probe pulses to the FUT.

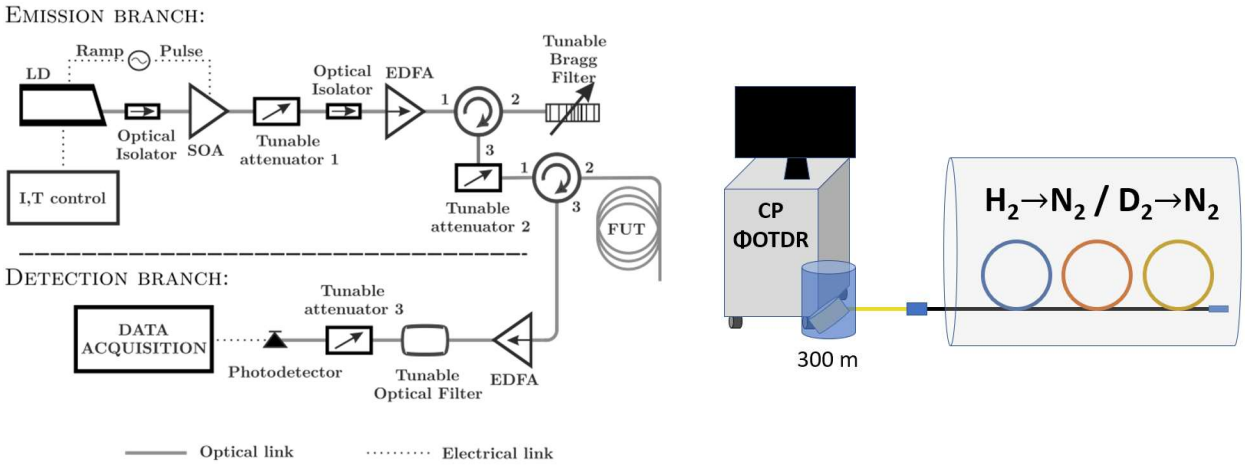

Figure 1. Left: optical setup of the interrogating system; Right: experimental distribution of the sample fibers in the reactor.

To detect the Rayleigh signal coming from the FUT, it is required to apply a second amplification and filtering stage (bottom branch). After this, direct detection is accomplished with a fast p-i-n photodetector that matches the chirp bandwidth. The produced electrical signal is finally digitized and processed by a computer. In this experiment (lasting several days) a technique to mitigate the effect of the laser phase noise was applied[6]. For this, an additional 300 m long fiber segment was placed at the FUT input end (see figure 1-right).

4. EXPERIMENTAL RESULTS AND DISCUSSION

All the points of the FUT were simultaneously monitored along the whole diffusion experiment. The change in relative refractive index (Δn /n) during the in-gassing and out-gassing was registered. The curves corresponding to a representative

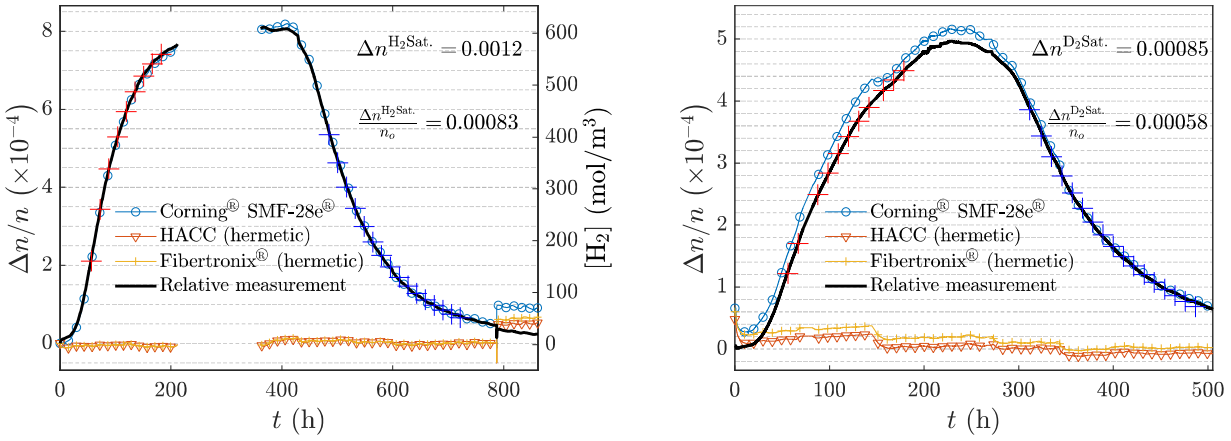

Figure 2. Left: Relative refractive index evolution due to the hydrogen loading as registered at the different fiber samples. The difference between the permeable and one of the hermetic fibers is shown (relative measurement); Right: equivalent deuterium experiment. The red and blue crosses represent exponential fittings of the relative measurement curves.

point of each fiber sample have been plotted in figure 2 for hydrogen (left) and deuterium (right). The figure also includes the difference between the curves measured at the permeable fiber and one of the hermetic samples (relative measurement). This subtracts the room-temperature fluctuations found during the day-night cycles, providing a true readout of the diffusion-induced refractive index change. The right axis showing hydrogen concentration in the silica glass has been calculated according to the coefficients and model provided in references 4 and 7.

The red and blue crosses added represent exponential fittings of the transient relative measurement curves. The parameters obtained from these fittings provide the saturation values of the induced change in the refractive index, that are shown in the plots. The results for hydrogen and deuterium are, respectively: $\Delta n(H_2)_{sat.} = 1.2 \cdot 10^{-3}$ and $\Delta n(D_2)_{sat.} = 8.5 \cdot 10^{-4}$ (considering $n_o = 1.47$). In addition, the characteristic times of the exponentials were extracted: $\tau(H_2) = 3.37$ days for hydrogen and $\tau(D_2) = 4.28$ days for deuterium. These values are consistent with the more massive molecules of D_2 when compared to H_2 (smaller diffusivity of the gas).

5. CONCLUSION

The method proposed in this communication has proven the ability of CP-φOTDR sensors to track and quantify the absorption and desorption of two chemical gas species into a conventional SMF. The experiments were done *in situ* while the fiber temperature was monitored and under a controlled external pressure of the diffusing gas. Considering the performance of the employed technique, this solution could present advantages compared to alternative sensors, potentially increasing the sensitivity to low concentrations and providing a fast response to changes. We continue working on this topic with the aim of achieving a complete model able to map the changes in relative refractive index to gas mole fraction.

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