

# Distributed hydrogen monitoring with phase-sensitive optical time-domain reflectometry

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**Abstract:** We propose a distributed hydrogen sensing system based on  $\phi$ -OTDR and a novel coating configuration with fiber adhesively bonding to palladium foils. An ultra-high sensitive distributed hydrogen sensing with 5 cm spatial resolution is demonstrated. © 2018 The Author(s)

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## 1. Introduction

As a potential clean energy source, hydrogen is used for a variety of applications in the chemical and energy industry. The wide explosion range (4–75% in air), small ignition energy (0.02 mJ), large flame propagation velocity, and difficulty of leak-tight storage, demand hydrogen detection with fast and accurate response [1]. In particular, distributed gas sensing with a long sensing range is highly desired in today's complex industrial facilities, gas delivery systems and civil infrastructure sites.

In the last decades, distributed fiber sensing (DFS) has drawn intense attention regarding its key advantages. For example, DFS is inherently safe in combustible gases, immune to electromagnetic interference, stable at high working temperatures, easy to install, and shows a low-cost per sensing point. These merits make DFS an attractive candidate for distributed hydrogen sensing. Several DFS systems for hydrogen detection have been reported [2–4], however, their performance is limited. In [2], the sensitivity is relatively low since the system is based on the loss of the pulse; in [3,4] the sensing range are very limited because of the large loss of the acoustic wave propagating along the fiber [3] and the coherence of the laser [4]. On the other hand, as one of the most promising DFS techniques, phase-sensitive OTDR ( $\phi$ -OTDR), which exploits the interferences created by coherent Rayleigh backscattering, has demonstrated its capability for long range sensing due to its ultra-high sensitivity, and its low requirement on the coherence length of the laser [5–6]. Rayleigh backscattering is caused by natural thermodynamic microscopic fluctuations of the material refractive index that are “frozen-in” during the transition of the liquid silica to the glassy state. This “fingerprint” scattering pattern keeps stationary over extended years-long periods, unless varying temperature or external strain is applied [7]. By scanning the optical frequency of the interrogating pulses, a stationary random spectrum is obtained at every sensing point along the fiber. Since temperature or strain changes linearly modifies the refractive index of the fiber, the subsequent local phase change is observed as a simple frequency shift of the local random spectrum in successive measurements [8]. To determine the spectral shift between a reference and a live acquisition, a spectral matching comparison is performed for each point along the sensing fiber. Strain or temperature changes will induce a shift in the correlation peak, accordingly. A distributed measurement is formed by compiling the spectral shifts for each segment along the sensing fiber. Via a proper transduction interface this makes  $\phi$ -OTDR a promising technique for distributed hydrogen sensing.

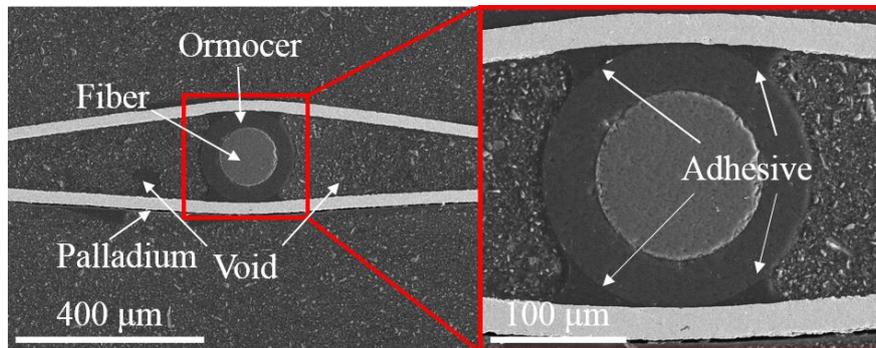
Due to the inert chemical property of silica, optical fiber sensors for hydrogen detection often rely on palladium (Pd) as a transductive sensing element [1]. Atomic hydrogen is dissociated on the Pd surface and absorbed into the lattice. The absorption goes along with a volumetric expansion of the Pd. The expansion, which is correlated to the hydrogen concentration will apply a strain to the fiber, which is then measured by the optical fiber sensor [9]. This process is fully reversible, if any phase change is avoided [10].

In this paper, we propose a distributed hydrogen monitoring system based on  $\phi$ -OTDR and a fiber customized by adhesively bonding thin Pd foils to it. A sensor design has been previously validated for fiber Bragg gratings [11]. In contrast to conventional coating process, Pd foils show a larger cross-sectional area, so that the hydrogen sensitivity is much enhanced. As a result, by measuring the strain change induced by the Pd foils absorption, an ultra-high sensitive distributed hydrogen sensing with 5 cm spatial resolution is demonstrated.

## 2. Sensing fiber design

The customization for hydrogen sensing is based on an Ormocer coated fiber (FBGS, Germany) and segments with adhesively bonded Pd foils. As a conceptual demonstration, a fiber with 4 Pd-foil sensing segments have been prepared. The segments are each 50 mm long and the distance between the segments is ~0.4 m. The segments structuring is made of the fiber sandwiched between two 20  $\mu$ m thick Pd foils (width 2 mm) and bonded using a bisphenol-a epoxy resin. A custom tool, based on vacuum bagging, allows the compression of

the foil-fiber composite while curing the adhesive at 100°C, generating a thin bond line between foil and fiber [11]. An SEM image of the cross-section of the sensor is shown in Fig. 1 (to generate a contrast between adhesive and potting epoxy, the latter was filled with silica nanoparticles). The image reveals a thin bond line on two sides between foil and fiber, and two large voids on the other side of the fiber. This design enables a much larger strain than conventional Pd coated fiber and increase the sensitivity by more than 10 times.

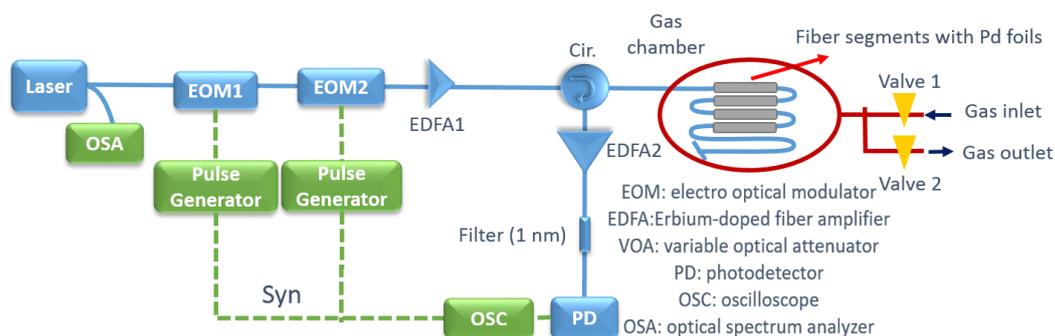


**Fig. 1.** SEM image of the cross-sectional view of the sensor. To increase the visibility of the adhesive, the SEM potting epoxy was filled with silica nanoparticles.

### 3. Experimental setup

A schematic drawing of the experimental setup is shown in Fig. 2. The interrogator in the experiment is based on a direct-modulation  $\phi$ -OTDR with ultra-high spatial resolution. We use a distributed feedback laser with 1 MHz linewidth as a source. Two cascaded electro optical modulators (EOMs) are used to modulate the continuous wave into pulses with a width of 500 ps, which result in a spatial resolution of 5 cm. Then an Erbium-doped fiber amplifier (EDFA) and a variable optical attenuator are used to deliver enough peak power to the pulses while preventing any nonlinear optical effect. The Rayleigh scattering signal is pre-amplified before the photodetector using another EDFA. The amplified spontaneous emission (ASE) noise of the EDFA is filtered out using a 1 nm bandwidth filter. The bandwidth of the photo-receiver is 3 GHz. A sampling rate of 10 Gs/s is set for the oscilloscope to digitize the electrical signal, which is further analysed by the software.

Short optical pulses with high enough extinction ratio are generated using a couple of cascaded EOMs. Since the pulse width is set to 500 ps, corresponding to a signal bandwidth of 2 GHz, a frequency scan step of 200 MHz is fine enough to recover the spectra at the sensing points. In this case, the frequency scanning can be performed by directly tuning the temperature or the current of the laser. In a first stage, a broad frequency range (100 GHz) is scanned by temperature-tuning the laser, as a reference to map large strain changes. Then a relatively small frequency range (20 GHz) is scanned for measurements by laser current tuning to enable a fast scanning (less than 10 seconds).



**Fig. 2.** Experimental setup based on a basic  $\phi$ -OTDR with 5 cm spatial resolution

As a demonstration, the fiber we used is about 5 m long and 4 Pd foil segments are placed in a gas test chamber. To test the hydrogen sensing performance of the sensor, a reference is taken when the chamber is filled with air. Then the chamber is evacuated and filled with gas with different hydrogen concentrations. The expansion of the Pd due to hydrogen absorption induces strain change to the fiber. These changes are interpreted by different frequency shifts in the spectra obtained by the interrogator. To avoid a phase change of the Pd, the used hydrogen concentrations are kept below 1% at room temperature.

#### 4. Results and discussions

To test the monitoring ability of the system, we apply different hydrogen concentrations in the chamber during the 6 hours-long monitoring and the measurements are taken consecutively every minute over the 6 hours. We call each hydrogen concentration change in the chamber an event and 4 events are set during the campaign:

- I. Event 1 at 20th minute: gas injection with 0.30% hydrogen;
- II. Event 2 at 70th minute: gas injection with 0.38% hydrogen;
- III. Event 3 at 85th minute: chamber vacuum, followed by ambient air injection;
- IV. Event 4 at 125th minute: gas injection with 0.60% hydrogen.

First, the frequency shift of the sensing fiber as a function of position along the fiber is shown in Fig. 3. The 6 traces in Fig. 3 correspond to responses taken every 5 minutes immediately after exposing the sensor to the gas with hydrogen concentrations of 0.3% and 0.6%, respectively. Clear peaks can be observed at 4 positions along the fiber, which correspond to the 4 fiber segments wrapped in Pd foils. The frequency shifts increase with time and a higher frequency shift at a given exposure time corresponds to a higher hydrogen concentration. Owing to the large Pd cross-sectional area, and good strain transfer of the sensor design and the sensitivity of the interrogator, the system shows a high sensitivity and observable frequency shifts can be obtained after the first minute of exposure to hydrogen with a concentration as low as 0.3%.

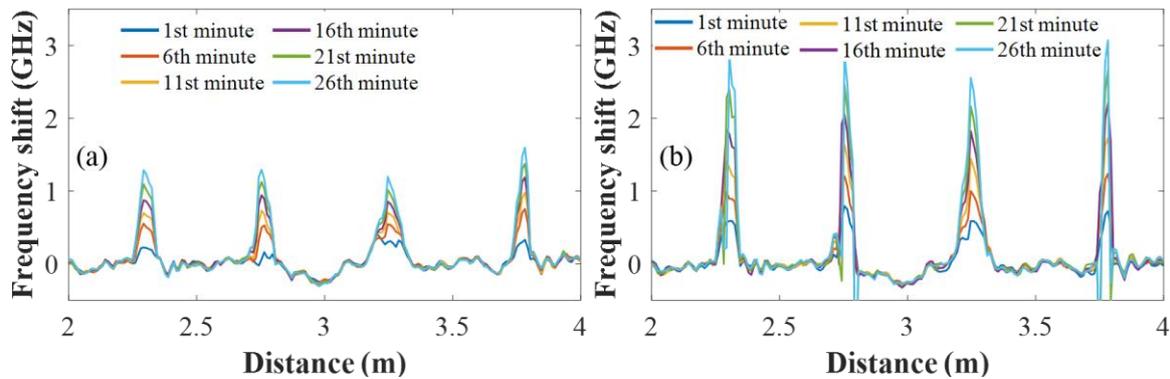


Fig. 3. Frequency shift responses of the sensing fiber every 5 minutes in half an hour

The frequency shift temporal evolution of the 4 sensing segments is illustrated in Fig. 4. All the traces show initial sharp increases of the frequency shift when the hydrogen concentration is raised (Event 1, Event 2 and Event 4 in Fig. 4), even for a small increment from 0.3% to 0.38% (Event 2 in Fig. 4). Details of the trace change of Event 1 and Event 2 are drawn in Fig. 5. On the other hand, the frequency shift decay is clearly observed after the gas with hydrogen is evacuated (Event 3 in Fig. 4) and then a faster increase resumes when the fiber is exposed to 0.6% hydrogen (Event 4 Fig. 4), which makes fully sense for a doubled concentration.

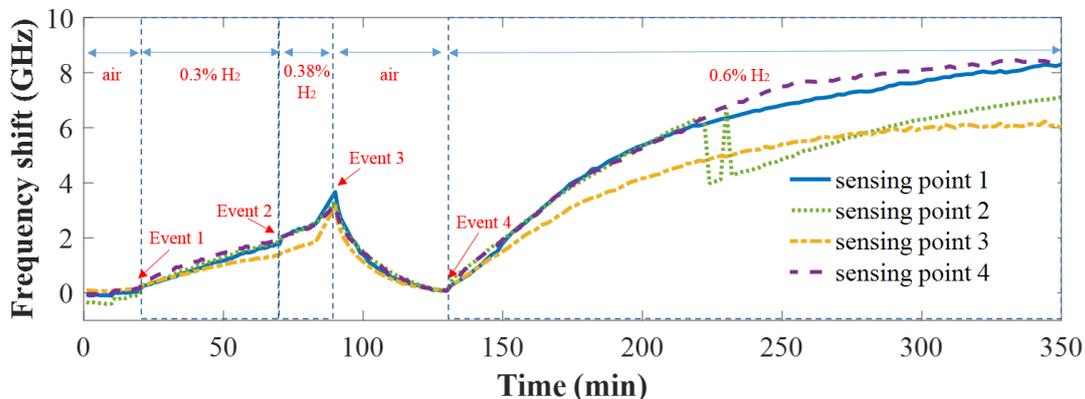


Fig. 4. Frequency shift responses of the 4 sensing segments over 6 hours. The hydrogen concentration-changing events are indicated in the time chart.

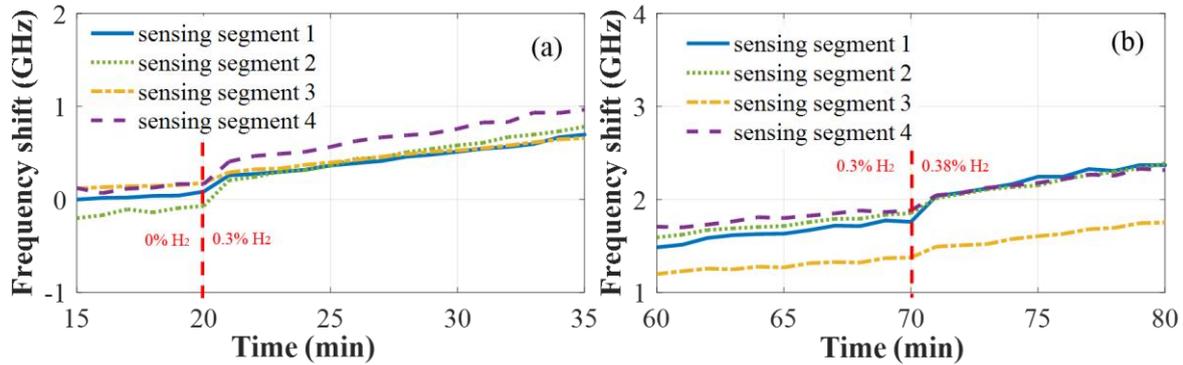


Fig. 5. Frequency shift response when the hydrogen concentration changes from (a) 0% to 0.3%; (b) 0.3% to 0.38%

This essentially demonstrates the ultra-high sensitivity of the system for distributed hydrogen monitoring and its relatively fast response entirely related to the Pd intrinsic reactivity.

It should be noted that the responses of the 4 sensing segments are not identical, which result from the non-uniform strain transfer of the palladium foils to the fiber. This can be calibrated by measuring the temperature or strain sensitivity of the fiber, as addressed in [9]. Besides, there is a sharp drop of frequency shift for sensing segment 2 in 0.6 % after 220 minutes. This is widely unexplained, but a tentative explanation may be a birefringence change in the fiber when subject to an increasing strain, as a result of the asymmetric structure of the sensing segment (as shown in Fig. 1). This leads to a mismatch between the polarization state of the interrogating pulse and the fiber eigen polarization axis and this needs to be confirmed in a future work.

It must be pointed out that the system has a spatial resolution as sharp as 5 cm. In this proof-of-concept implementation, the total length of the sensing fiber is only 5 m; however, the distance range of the sensor can easily reach ~1 km, which offers a potential for an extended sensing coverage.

## 5. Conclusions

In summary, a distributed hydrogen sensing system is proposed for the first time based on  $\phi$ -OTDR and a fiber with adhesively bonded Pd foils. The optimized design of the customized fiber and the high sensitivity of the interrogator allow a fast response to a small change of hydrogen concentration. A concentration 10 times lower than the explosive level of 4% can be detected in less than one minute over potentially up to 20'000 independent sensing points (5 cm spatial resolution over 1 km) along the optical fiber.

## 6. Acknowledgement

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