An All-Fiber Reflective Hydrogen Sensor Based on a Photonic Crystal Fiber In-Line Interferometer

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***Abstract*— A new method based on introducing a photonic crystal fiber (PCF) in-line interferometer for hydrogen sensing is demonstrated. One end-face of the PCF and part of the outer-face of the fiber are plated with a thin palladium film by magnetron sputtering, while the other end is connected to the optical path. Therefore, a set of reflection-type all-fiber hydrogen sensor system is accomplished. Hydrogen concentration from 0% to 5% was detected in the experiment and the related interferometric resonant wavelength-shift was recorded. The maximum wavelength shift is over 1.2 nm. The whole system has an all-fiber optical path without any bulk-optic components, and this design obtains a high level of integration, a high stability, and low production costs.**

***Index Terms*— Photonic crystal fiber in-line interferometer, reflective, wavelength-shift, hydrogen sensor.**

I. Introduction

As ONE kind of industrial raw materials, hydrogen plays an indispensable role in many different fields such as petrochemical, electronics industry, metallurgical industry, food processing, new energy etc. However, hydrogen molecule can easily leak since it is the lightest and smallest molecule. In air, the concentration of hydrogen exceeding 4% will cause an explosion under the emergence of even a spark. Therefore, in the process of transportation, storage and use, real-time monitoring of the environmental hydrogen concentration is a very necessary security work.

Up to now, the study of hydrogen sensors is divided into two main directions: One is electrical hydrogen sensors [1], [2], and the other one is optical hydrogen sensors, especially optical fiber hydrogen sensors [3]–[5]. Compared with electrical sensors, optical fiber sensors are electrically isolated, having no electrical contact. So they neither generate sparks nor rely on heat exchange and have no risk of gas ignition. With the development of optoelectronic technology and micromachining technology, more and more new work such as sensors based on fiber grating [6], [7], evanescent field [8], [9], metallic photonic crystal structures [10] and surface plasmon resonance [11]–[13] are springing up. However, some of them have an all-fiber sensing system but a low sensitivity, while some others own a high sensitivity but introducing bulk-optic components which may introduce instability to optical system.

Manuscript received September 11, 2013; revised November 15, 2013 and November 24, 2013; accepted November 25, 2013. Date of publication November 28, 2013; date of current version February 14, 2014. This work was supported in part by the National 973 Program under Contract 2010CB327803, in part by NSFC under Program 11074117, in part by the National Science Fund for Excellent Young Scientists Fund under Grant 61322503, and in part by the National Science Fund for Distinguished Young Scholars under Grant 61225026. The associate editor coordinating the review of this paper and approving it for publication was Prof. Istvan Barsony.

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Digital Object Identifier 10.1109/JSEN.2013.2293347

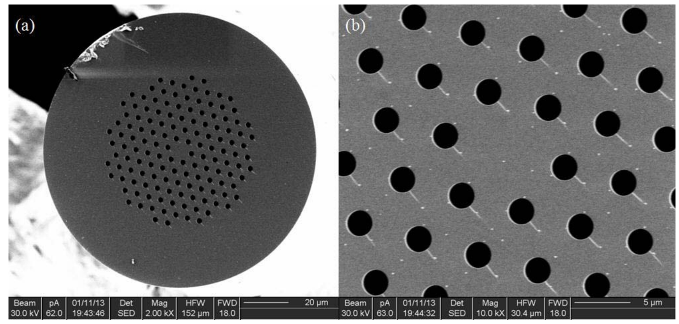

Fig. 1. SEM images of the PCF tip coated with Pd film. (a) Magnification of 2000. (b) Magnification of 10 000.

In this paper, we demonstrate a ultra-compact all-fiber reflective hydrogen sensor based on a palladium (Pd)-deposited PCF in-line interferometer (PCF-IF). The PCF-IF consists of a stub of 125 μm large-mode-area (LMA) PCF spliced with the same size standard single-mode fiber. In the spliced regions, the voids of the PCF are fully collapsed, thus allowing the coupling and recombination of PCF core and cladding modes. The fabrication of the PCF-IF is very simple, only involving cleaving and splicing processes that can be carried out in a standard fiber optics lab. And it is cost-effective, highly stable over time. It has low temperature sensitivity and only needs a small piece of cheap commercial LMA PCF with the standard size ($\sim$1 cm long, 125 μm in diameter). Then Pd is deposited on one end and the outer face of the PCF and constitute an in-line interferometer. When this PCF-IF is exposed to hydrogen, the optical properties of the Pd layer and the interference spectrum of fundamental mode and cladding mode that transmitted in the PCF change. The interaction is detectable with a simple transmission measuring setup. Compared with some typical hydrogen sensors based on fiber Bragg gratings [14]–[16], sensitivity of our device has great improvement. Furthermore, the fabrication procedure is simple and does not need supplementary treatment like etching or tapering. The development of other gas sensors is also feasible if the PCF is deposited with suitable gas-permeable materials [17].

II. The Pd-Deposited PCF-IF

For the construction of the Pd-deposited PCF-IF, we employ a commercial single mode PCF (LMA-8, NKT Photonics) consisting of a solid core surrounded by six rings of air holes.

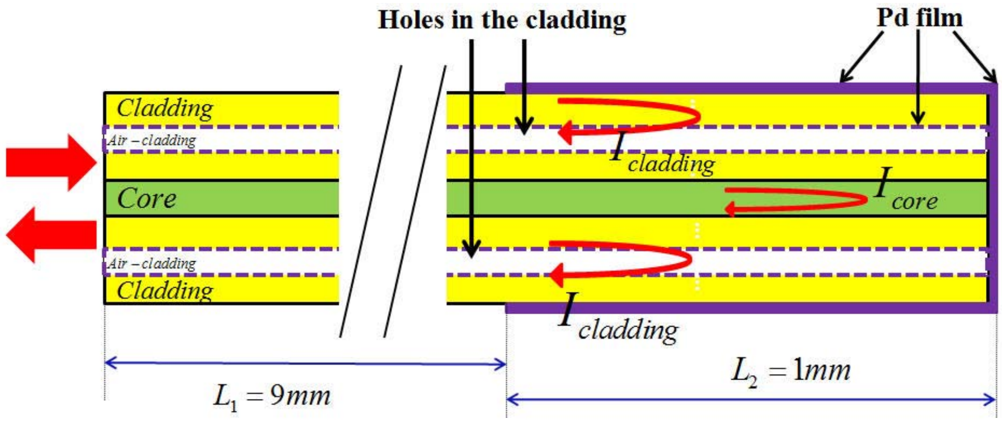

Fig. 2. Schematic of the PCF-IF.

As shown in Fig. 1(a), such a fiber has a core of 8.4 μm in diameter, voids with average diameter of 2.17 μm, and the average separation between the voids is 5.3 μm. The diameter of the PCF is the same as SMF-28 fibers and it is cheaper and easier to be spliced to SMF-28 with higher stability and better repetition, compared with other larger size PCFs. Initially a PCF-IF was fabricated by splicing the end of one SMF-28 fiber to the cleaved end of a 10 mm-long PCF. The voids of the PCF collapsed completely over a short region about several hundred micrometers long. Then the sample was fixed on one metal stage which then was fixed in the coating cavity of the coating machine. At last, a 50-nm-thick layer of Pd was deposited onto one end face, and outer-wall surface of the PCF by magnetron sputtering under operating current and voltage.

Because the PCF is covered with silver-glue when we fix it on the metal stage, Pd film on the outside of the PCF is only 1 mm long. Because of the small hole and unperfected direction, little of Pd is deposited onto the inner-wall surface of the PCF. Fig. 1 shows the scanning electron microscope (SEM) images of the PCF tip coated with Pd film. Fig. 2 shows the schematic of the PCF-IF. The Pd film on the end-face of the PCF acts as a metal mirror.

When the light travels from the SMF to the PCF, the SMF fundamental mode begins to diffract in the collapsed PCF region, and it excites core and cladding modes in the PCF section with different propagation constants. At the end-face of the PCF, both the core and cladding modes are reflected and reach the collapsed region of the PCF. They will thus further diffract and will be recombined. Therefore, the reflection spectrum of our PCF-IF can be expressed as that of a two-mode interferometer:

$$I = I_{core} + I_{cladding} + 2\sqrt{I_{core} I_{cladding}} \cos(\delta + \varphi_0) \quad (1)$$

$$\text{Free spectral range (FSR)} = 2\pi\lambda/\delta \quad (2)$$

where

$$\delta = (2\pi/\lambda)\left[2\left(\int_{L_1}(n_{cl_1} - n_{co})\,dz + \int_{L_2}(n_{cl_2} - n_{co})dz\right)\right] \quad (3)$$

The resonant wavelength

$$\lambda_k = \frac{2}{k}\left[\int_{L_1}(n_{cl_1} - n_{co})dz + \int_{L_2}(n_{cl_2} - n_{co})dz\right] \quad (4)$$

I is the intensity of the interference signal, I_{core} and $I_{cladding}$ are fundamental and cladding modes transmitted in the PCF, respectively. δ is the phase difference of the two modes, n_{cl_1} is the effective index of the cladding mode which is not covered by Pd, n_{cl_2} is the effective index of the cladding mode which is covered by Pd, n_{co} is the effective index of the core mode, λ is the wavelength. The region of the PCF outer surface that covered by Pd is $L_2 \approx 1$ mm long, while the region without Pd is $L_1 \approx 9$ mm long. When the sensing unit is exposed to hydrogen atmosphere, the Pd film both inside and outside the PCF absorbs hydrogen, thus causing its volume to increase and the volume density of free electrons to decrease, and both the real part and imaginary part of the complex electric permittivity of the Pd film are reduced. It results in the change of δ and interference wavelength, which can be used as an efficient sensing value. It is interesting to investigate dominant effect on the spectra. There are several parameters changing simultaneously. The modifications of the Pd complex refractive index lead several variations: the reflectivity of the 50 nm thick film on the optical fiber end face, the refractive index of the core (n_{co}) and cladding (n_{cl}) modes and the cavity length (L_2, via volumetric changes of the outer Pd film, which is ignorable). The reflectivity isn't related to the resonant wavelength shift. Because little Pd was infiltrated within the holes and our calculation also shows that n_{co} changes little with thin Pd film on the inner hole wall, n_{co} can be assumed to be constant with or without Pd. Then the main contribution on the wavelength-shift sensitivity comes from the changes of n_{cl}. As the effective index of the core mode is almost unchanged while the effective index of the cladding mode is decreased, the resonant wavelength of the two signals' interference spectrum shifts toward the short wavelength.

III. Experimental Result and Discussion

Fig. 3 shows the schematic diagram of the experimental setup. A supercontinuum source (SuperK Versa, NKT Photonics) and an optical spectrum analyzer (OSA, Ando AQ6317B) are employed during the whole experiment in order to detect the interferometric behavior of the PCF-IF in real time. The light source emits an optical signal of a wide spectrum. Then the light transmits from a circulator to the PCF-IF

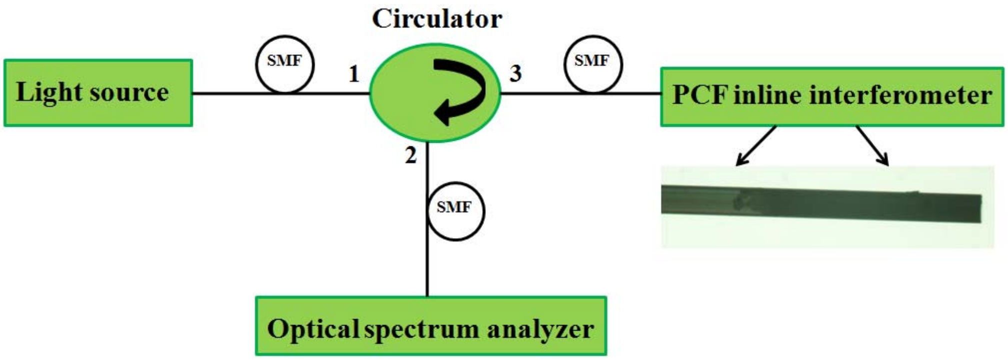

Fig. 3. Schematic of the all-fiber reflective hydrogen sensor based on a PCF-IF. The inset is the PCF-IF.

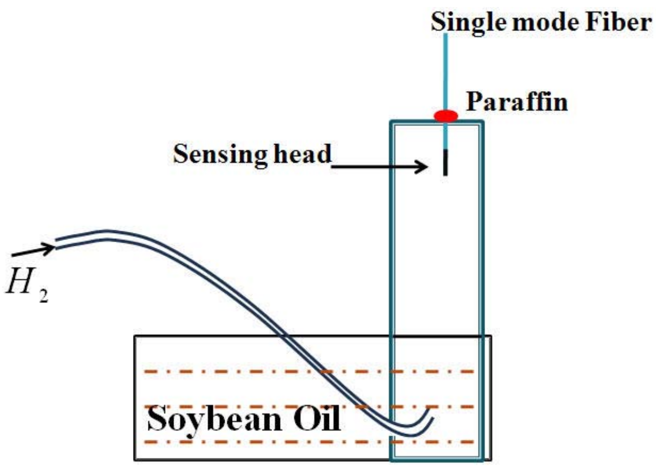

Fig. 4. Diagram of hydrogen concentration controlling device.

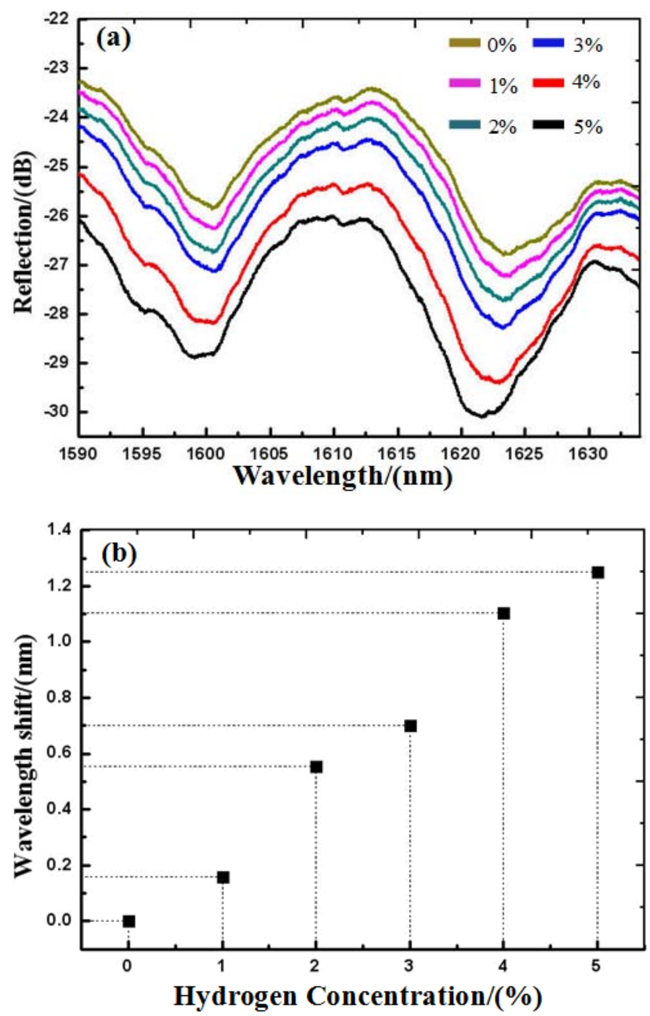

Fig. 5. (a) The reflected spectra for the sensor under different hydrogen concentrations. (b) Wavelength shifts for the system under different hydrogen concentrations.

through SMF. The light is reflected back by the metal mirror on the end-face of the PCF and the circulator returns the reflected light to OSA.

The sensor is tested in a gas chamber made with a section of glass tube which is fixed on the bottom of a crystallizing dish, shown in Fig. 4. The inner diameter and height of the glass tube are 8 cm and 30 cm, respectively. The sensor head is fixed in the tube by paraffin, and the measurements are carried out under room temperature. We fill the crystallizing dish and glass tube with soybean oil which is about 5 cm high. Hydrogen is imported into the gas tube by a plastic hose. And the hydrogen concentration is determined by the change of oil height in the tube. The amount of hydrogen absorbed by the oil is very small, so we ignore this part of the loss of hydrogen. Besides, the oil height in the tube changes in a range of $0 \sim 15$ mm, so the pressure in the tube almost remains unchanged. This method which determines the concentration of hydrogen makes our setup does not allow recovery measurements.

In order to test the influence of the hydrogen on the sensing unit, different concentration of hydrogen were imported into the gas tube. At the same time, the real-time spectra was recorded by OSA. The Pd's thickness of 50 nm provides us a relatively fast response time of about 15 seconds and at the same time achieves a satisfying spectral contrast. Fig. 5(a) presents the reflected spectra of the PCF-IF under different hydrogen concentrations. According to Eq. (2) and (3), the calculated FSR is $\sim$24 nm which is closely consistent with experimental results shown in Fig. 5(a). From the results, it can be seen that with the increase of the hydrogen concentration, the interference spectrum of the fundamental and cladding modes in the PCF moves toward the short wavelength. Furthermore, when the Pd film absorbs hydrogen, its effective index is decreased. So, we can see from Fig. 5(a), the intensity of the reflected light reduces with the increase of the hydrogen concentration. Fig. 5(b) shows wavelength-shifts corresponding to related hydrogen concentrations. The wavelength-shift sensitivity is about 0.25 nm for one percent of hydrogen. As shown in Fig. 5(b), when hydrogen concentration reaches 5%, the resonant wavelength shifts over 1.2 nm. As to some sensors based on fiber Bragg grating, while under the same condition, the sensitivity is only pm magnitude [14]–[16]. Finally, we have to point out that repeatability was not investigated and we measured this sample once in this paper. However, repeatability is very important for real applications. In our setup, it can be done by pulling out the glass tube, cleaning it and measuring its spectra again. In theory as well as reports mentioned in previous literature, we can expect that this device can repeat well. However, because it takes a long time to re-do the measurement, the humidity and temperature will damage the device's repeatability. In particular, the humidity in the tube, coming from the water in the oil, possibly has a great influence on the Pd film.

IV. Conclusion

In conclusion, we propose an all-fiber reflective hydrogen sensor based on a PCF-IF. The whole sensing system is composed of all-fiber elements, without any bulky optical components. This method is very simple and cost-effective, avoiding tapering or etching. The experimental results show that when hydrogen concentration ranges from 0% to 5%, the sensor gives a sensitivity about 0.25 nm for one percent of hydrogen. Under the explosion limit of hydrogen, the wavelength-shift that the system detects is over 1 nm.

Acknowledgment

The authors also acknowledge the support from PAPD and the Fundamental Research Funds for the Central Universities.

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