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Hydrogen Sensor Based on A Palladium-Coated Long-Period Fiber Grating Pair

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We propose a simple hydrogen detection scheme based on a Mach-Zehnder interferometer formed with a pair of palladium-coated long-period fiber gratings (LPGs). Since an LPG pair offered a fine-structured interference fringe in its transmission spectrum, the resolution as a sensor could be appreciably enhanced compared to that of a single LPG. As the palladium layer absorbed hydrogen, the effective refractive indices of the cladding modes were increased so that the interference spectrum was blue-shifted up to 2.3 nm with a wavelength sensitivity of −0.29 nm/min for 4% of hydrogen concentration.

Keywords : Fiber gratings, Fiber sensors, Hydrogen sensors, Palladium

OCIS codes : (060.0060) Fiber optics and optical communications; (060.2310) Fiber optics; (060.2370) Fiber optics sensors

I. INTRODUCTION

For several decades, as is well known, hydrogen has attracted great attention as an alternative energy source to our limited fossil fuels [1]. Since hydrogen can be obtained from electrolysis of water and produces water after burning, it has the advantage of unlimited availability without environmental pollution [2]. However, the quick diffusion and sudden explosion of hydrogen in air limit its use as the energy carrier [3]. In order to overcome these obstacles, many trials have been made to detect hydrogen leakage in advance, leading to the development of various hydrogen sensors.

Among many types of hydrogen sensing methods, the one based on fiber-optics has superior characteristics such as safety from sparking, immunity to ambient electromagnetic interference, and capability of long distance detection [4]. Most of the optical hydrogen sensors utilize the special optical fibers that are coated with palladium. Palladium can absorb hydrogen up to 900 times of its own volume at room temperature and atmospheric pressure [5]. In previous works, a micro-mirror formed at the end of a fiber with a palladium film, a multimode fiber having a palladium coating over an exposed core, a palladium-coated tapered fiber, and a fiber Bragg grating (FBG) covered with a palladium layer have been demonstrated [6-9]. Thickness of palladium plays an important role in deciding the sensitivity and the response time of the palladium-coated fibers. Thicker palladium has a good sensitivity but, unfortunately, at the same time the response time becomes longer. Therefore, there should be trade-off between the sensitivity and the response time.

The fiber-optic hydrogen sensors mentioned above have low sensitivity because usually the core mode of the fiber is employed as the sensing element. Also, the bothersome processing such as tapering, etching, or FBG inscribing makes the fabrication very complex and brings high fragility. On the other hand, the long-period fiber grating (LPG), which causes coupling between the fundamental core mode and cladding modes [10], can be an attractive alternative to improve the weak points of prior works. It is expected that the use of

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cladding modes coupled by an LPG results in a higher sensitivity to external perturbations than the FBG does [11], because the cladding mode encounters the environmental change directly. Furthermore, the fabrication process is relatively simple because any other treatments like etching or tapering are not necessary. When an LPG pair is employed, a Mach-Zehnder interferometer is easily formed, which can enhance the sensor resolution appreciably. The transmission spectrum of the LPG pair has a fine structure due to interference, so that reading the spectral change is much easier and more precise than that of a conventional single LPG which has a relatively blunt loss peak in an interesting wavelength range [12].

In this paper, a highly sensitive hydrogen detection sensor is presented. The proposed sensor is composed of two identical LPGs that are fabricated by UV laser (λ=248 nm) exposure on the core of a single mode fiber with a 500 μm grating period and a 50 mm of center-to-center distance between two gratings. Each grating length is 20 mm, so that the whole sensing length coated with palladium is 70 mm. Thin (50 nm) palladium is uniformly deposited over the cladding of the fiber. While exposing the fiber to 4% concentration hydrogen, which is known as the level leading to an explosive condition, we measure the wavelength change in the transmission spectrum with respect to time and confirm the feasibility as a hydrogen sensor.

II. PRINCIPLE

An LPG induces optical power coupling between the core mode and several cladding modes at the resonant wavelengths satisfying the phase matching condition given by

$$\lambda_{res} = (n_{eff}^{co} - n_{eff}^{cl(m)})\Lambda \tag{1}$$

where λ_{res} is the resonant wavelength, Λ is the grating period, and n_{eff}^{co} and $n_{eff}^{d(m)}$ are the wavelength-dependent effective indices of the core mode and the m-th order cladding mode, respectively [13]. In the equation, the effective index of the m-th order cladding mode $n_{eff}^{d(m)}$ is the main parameter because its evanescence field is easily affected by the refractive index of its surroundings. Accordingly, when palladium exists at out of the cladding surface, the change in the refractive index of palladium can be perceived by the shift in the resonant wavelength of eq. (1).

Figure 1 is the schematic of the light propagation in a long period fiber grating pair (LPGP). When the core mode beam encounters the first LPG, some of its optical power is coupled to the cladding mode at a specific resonant wavelength given by eq. (1). Then, the second

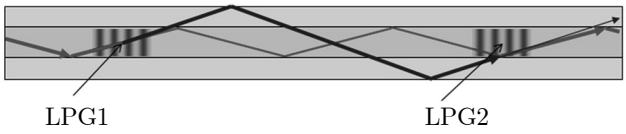

FIG. 1. The schematic of the optical paths of the beam in an LPG pair. A part of the core mode beam is coupled to a cladding mode beam by the LPG1, and then recoupled to the core mode by the LPG2.

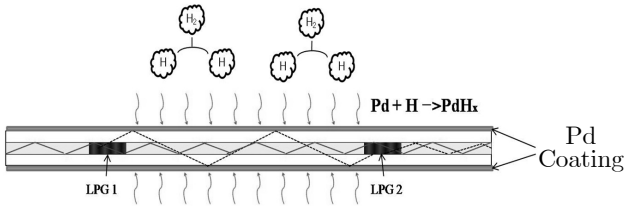

FIG. 2. The schematic of the proposed hydrogen sensor. Hydrogen interacts with the cladding mode of the LPGP through the palladium coated on the cladding surface.

LPG recouples a part of the cladding mode back to the core mode. Therefore, after passing the second LPG, the original core mode and the recoupled core mode interfere with each other, so that the transmission spectrum develops an interference fringe pattern in it. The transmission of the LPGP is represented as

$$T_{pair} = T_{single}^2 + R_{single}^2 - 2\alpha T_{single} R_{single} \cos\psi \tag{2}$$

where α is the rate of the cladding mode loss by any means, T_{single} and R_{single} are the transmission and reflection coefficients of a single LPG, respectively [14]. The phase term, which is related with the path length difference between the original core mode and the recoupled core mode, is approximately expressed by

$$\psi \cong \frac{2\pi}{\lambda}(n_{eff}^{co} - n_{eff}^{cl(m)})L \tag{3}$$

where L is the distance between the gratings [14]. The equation says that the phase of the interference spectrum is affected by the $n_{eff}^{d(m)}$. Thus, we can detect hydrogen gas by measuring the wavelength shift in the interference spectrum of the LPGP.

The schematic of the propose hydrogen detection sensor is shown in Fig.2. When palladium is exposed to hydrogen gas, a hydrogen molecule is divided into two hydrogen atoms ($H_2 \leftrightarrow 2H$) at the palladium surface with a very efficient dissociation rate [8]. As the palladium layer absorbs the hydrogen, its volume expands because hydrogen absorption converts palladium to palladium hydride (PdHx), while causing a low density of free electrons within palladium [15]. The palladium hydride layer has different optical properties compared with those of the hydrogen-free palladium layer. Both the

real and imaginary parts of its complex permittivity are reduced [4]. It results in the change in the boundary condition between the cladding surface and the palladium layer as well as in the effective index difference between the core and the cladding modes within the LPGP [16]. Eventually, the spectral shift in the transmission spectrum is measured as the indicator of hydrogen.

III. EXPERIMENTAL RESULTS

Figure 3 exhibits the LPGP spectra measured before and after palladium coating at a wavelength range from 1300 nm to 1650 nm. It was observed that the spectral peaks were shifted to the longer wavelength direction and their strengths were weakened. The thin layer of palladium has a complex refractive index of roughly 3.35 + i8.06 as reported in Ref. 17. It is also known that both the real and imaginary parts of its complex permittivity are reduced with hydrogen [4]. In our experiment, the fourth order cladding mode located near 1570 nm was used to monitor the hydrogen gas.

The proposed sensor was fabricated and inserted into a gas chamber as shown with Fig. 4. Flowing 4% concentration hydrogen gas mixed with 96% nitrogen gas into the chamber, the transmission spectrum of the interferometer was successively measured with a resolution of 0.05 nm and shown with Fig. 5. For the spectrum measurement, a broadband LED source having a 1550 nm center wavelength and −30 dBm peak power was used. With the hydrogen gas, the resonant wavelength of peak 1 in the figure, initially formed at 1570.7 nm, was shifted to the shorter wavelength direction. About -2.3 nm of the resonant wavelength shift was observed within 10 minutes. As shown in Fig. 6, the interference peak was almost linearly shifted for the first 6 minutes and then saturated after roughly 8 minutes. The rate of the wavelength shift was measured as −0.29 nm/min for the first 8 minutes.

Figure 7 shows the transmission depth change in peak 1 of Fig. 5, which was measured with respect to the hydrogen exposure time. The depth of the peak started deepening after 3 minutes and after 9 minutes the deepening was almost saturated. The total variation

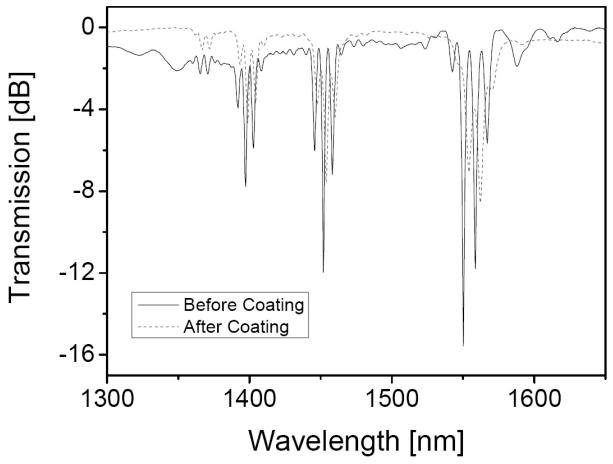

FIG. 3. Transmission spectra of an LPGP before and after palladium coating.

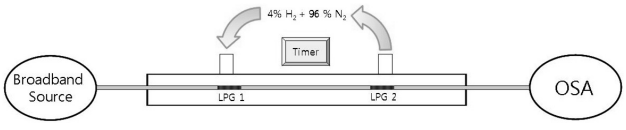

FIG. 4. The experimental setup for the proposed hydrogen sensing. The palladium coated LPGP is inserted into a gas chamber, and 4% concentration hydrogen gas mixed with 96% nitrogen gas is flowed into the chamber.

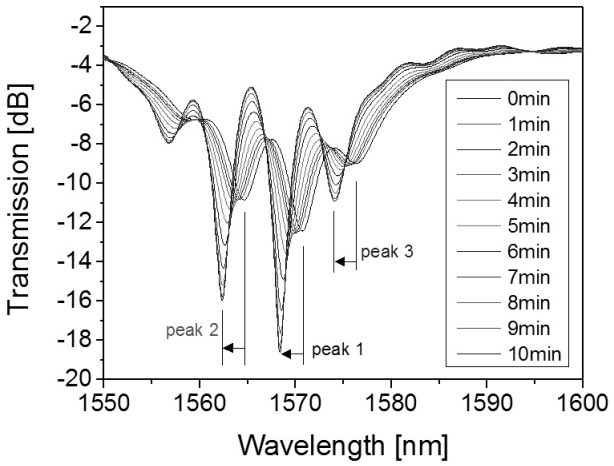

FIG. 5. The interference fringe affected by the hydrogen. The successive measurements were made with respect to time for 4 % hydrogen environment. Each interference peak was blue shifted, moving to the shorter wavelength direction, and deepened in its strength.

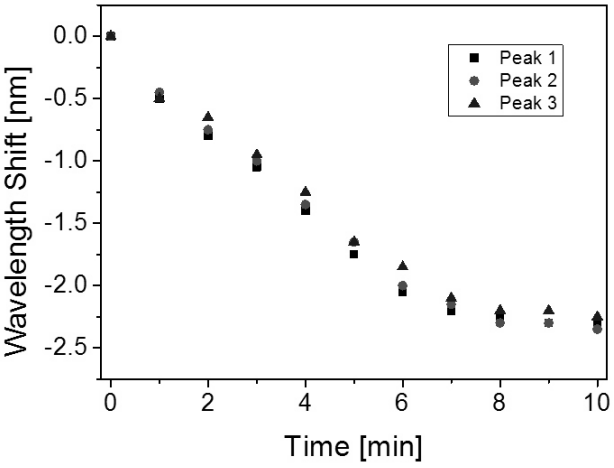

FIG. 6. The wavelength shifts of three interference fringe peaks in Fig. 5, measured with respect to time.

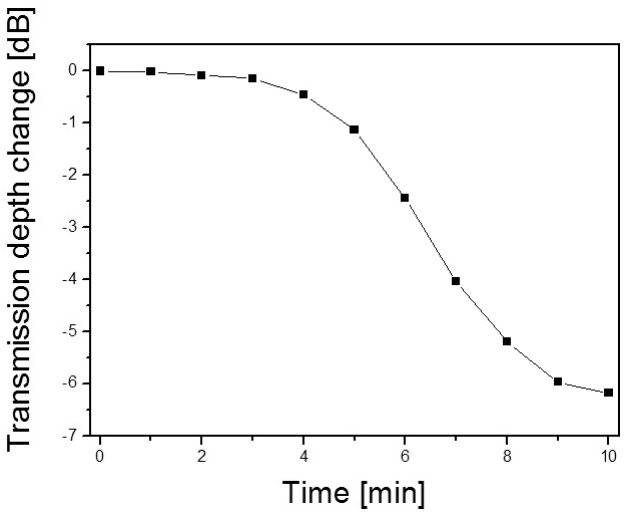

FIG. 7. The transmission depth change of the interference peak measured with respect to time.

in the transmission depth was approximately 6 dB. For getting the exact reason why the transmission depth was so significantly changed, it is necessary to investigate the optical properties of palladium and palladium hydride in more detail. However, one thing we can say is that the formation of palladium hydride affects the coupling strength of LPGP as can be seen with Fig. 3; it weakens the coupling strength as well as shifting the resonant wavelengths. Usually, in an LPGP, the transmission spectrum has the maximum interference fringe contrast when both LPGs are the same and have 3 dB coupling strengths [18]. Therefore, when both LPGs have the same but slightly larger than 3 dB coupling strengths, thus slightly over-coupled, the fringe contrast of the interference pattern can be increased as palladium absorbs hydrogen molecules. However, as mentioned, more detailedinvestigation is necessary to understand all the underlying phenomena.

IV. DISCUSSION AND RESULTS

By experiment, we have verified the possibility of the proposed hydrogen sensor. The sensor had a simple and firm structure with a satisfactory sensitivity compared to previous works. The sensitivity was quite attractive, but the response time was relatively long for actual situations. According to the result of Ref. 2, the normalized response time for the same 4% hydrogen but with an 8 nm coating thickness at room temperature was 90 seconds. In our case, we have used a 50 nm thick palladium coating. Therefore, it seems reasonable that our sensor has a longer response time due to the palladium thickness. As mentioned, a thin coating can improves the response time, however, it deteriorates the sensitivity. Usually, hydrogen sensors require the detection time as quick as possible to ensure safety from gas explosion. Hence, it is necessary to optimize the sensor parameters through additional analysis and experiments.

First of all, in order to find the proper sensitivity with the short response time, many experiments should be carried out under various palladium thickness states. Second, the experimental confirmation is required to make sure that the sensor works at lower hydrogen concentration along conditions with an appreciable sensitivity. Third, it is expected that a supplementary design considering exterior parameters such as temperature and strain might improve the performance of the sensor. Even though we are seeking a way that can improve both parameters, there should be compromise between the sensitivity and the response time. The experiment with various palladium thicknesses is under preparation. A full scale stability test was not done in this work, however, we observed that after stopping the hydrogen flow, the spectrum returned to the original position and shape within 10 minutes.

In short, the hydrogen detection sensor based on a fiber Mach-Zehnder interferometer using a palladium-coated long period grating pair (LPGP) has been demonstrated. When the palladium was exposed to hydrogen, its refractive index was changed; thus, the effective refractive index of the cladding mode coupled by an LPG was changed. The change in the modal index of the cladding mode could be effectively monitored by inserting one more LPG in series and seeing the interference spectrum of the LPGP. With the 4% hydrogen, it was observed that the spectrum moved to the shorter wavelength direction with a wavelength sensitivity of −0.29 nm/min. Total shift of 2.3 nm was observed for the first 8 minutes of the hydrogen exposure and then saturated. At a resonant peak of the LPGP interferometer, it was also observed that the transmission depth was deepened by at most 6 dB in 10 minutes. Additional experiments are under consideration to make up for the relatively long response time of the proposed sensor and to elevate the sensing performance.

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