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Hydrogen Sensor Based on Palladium-Attached Fiber Bragg Grating

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This paper demonstrates the performance of a palladium wire hydrogen sensor based on a fiber Bragg grating as a means of developing a quasi-distributed hydrogen sensor network capable of operating at cryogenic temperatures. The new approach employing a fiber Bragg grating based palladium hydrogen sensor described in this study is advantageous over other traditional hydrogen sensors because of the multiplexing capability of fiber Bragg gratings. The sensitivity of the hydrogen sensor at room temperature is approximately 2.5 times that of the hydrogen sensor at cryogenic temperatures.

I. INTRODUCTION

A number of electronic hydrogen sensors have been demonstrated based on changes in the properties of palladium [1-3]. Palladium is a metal that can absorb and desorb up to 900 times its own volume of hydrogen [4]. Exposure of the palladium to hydrogen results in the formation of the hydride with dependence on the partial pressure of hydrogen. The hydride has larger lattice constant than pure palladium, so that absorption of hydrogen produces deformation in three orthogonal directions. According to the literature [5], the expansion of palladium is reversible when hydrogen is removed from the palladium. The expansion of palladium in the presence of hydrogen has been used with optical fiber technology to develop hydrogen sensors, such as coating palladium on one arm of a Mach-Zehnder interferometer [5], depositing a palladium micromirror at the end of an optical fiber [6], and depositing palladium on an extrinsic Fabry-Perot interferometer [7] because hydrogen sensors utilizing optical fiber technology are advantageous in terms of small size, light weight, and immunity to electromagnetic interference, compared to other traditional hydrogen sensors. In these references, the fibers were coated with palladium using sputtering techniques to produce coatings of a desired thickness in order to maximize the measurand-induced modulation of the guided optical beam in the fiber.

The fiber Bragg gratings have been used for a wide range of applications in both optical communication devices such as dispersion compensators, wavelength selective devices, and fiber lasers, etc. [8] and grating-based sensors for smart structures [9]. The new approach is employed to develop a new hydrogen sensor at lower cost by wiring palladium on the fiber Bragg grating (FBG), having the capability in multiplexing of FBG. This paper summarizes our efforts to use the expansion of palladium with Bragg grating sensors as means of developing a quasi-distributed hydrogen sensor network capable of operating at cryogenic temperatures to monitor the hydrogen leaks from hydrogen fuel tanks in vehicles such as space and high speed civil transport.

II. PRINCIPLE OF SENSOR

Optical fiber Bragg gratings used for the hydrogen sensor are formed by phase mask printing of photosensitive fiber with exposure to ultraviolet light, which modulates a permanent refractive index along the core of the fiber. The sharp reflection resonance causes the Bragg wavelength of a grating, λ_g, which is given by the expression [8]

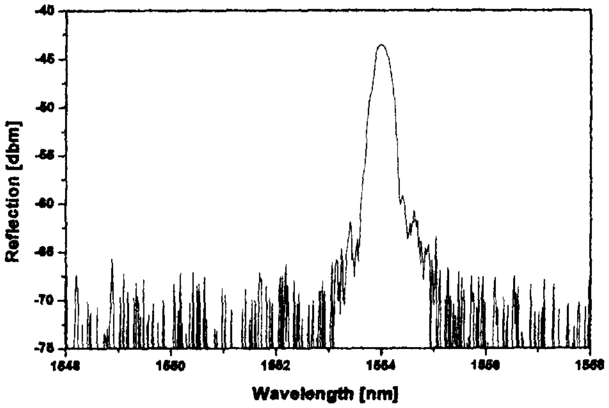

FIG. 1. The Typical Optical Reflection Spectrum of a Bragg Grating.

$$\lambda_g = 2n_e\Lambda \qquad (1)$$

where Λ is the period of the grating and n_e is the effective refractive index of the fiber core. Both Λ and n_e vary according to the ambient temperature and the applied external strain, and as a result the Bragg wavelength, λ_g, shifts linearly. Fig. 1 shows the typical reflection spectrum of the fiber Bragg grating which can be detected in either the reflected or transmitted spectrum. The fractional Bragg wavelength shift induced by strain and temperature can be described by [8]

$$\Delta\lambda/\lambda = (1 - P_e)\epsilon + (\alpha + \xi)\Delta T \qquad (2)$$

where P_e is the effective photoelastic constant, ϵ is the strain, α is the coefficient of thermal expansion for the fiber, ξ is the thermo-optic coefficient and ΔT is the temperature change. The material properties of the silica fiber are shown in Table 1.

The grating exhibits large temperature dependence of approximately 0.01 nm/°C and strain dependence of ~1.15 nm/mϵ at the center peak wavelength, λ_g = 1529~1559 nm. The basic concept of hydrogen detection is to attach palladium wire to the fiber Bragg grating which expands due to absorption of hydrogen into palladium when exposed to hydrogen to be detected. The deformation of palladium stretches the Bragg grating and changes the period of FBG, and as a result the grating peak wavelength shifts linearly. The hydrogen can thus be detected by measuring the shifts in the grating peak wavelength with dependence on the partial pressure and concentration of hydrogen. Fabrication of the hydrogen sensor used in this study is described in the next section.

TABLE 1. The Material Constants of the Silica Fiber [8].

A Glass Fiber	Material Coefficients
Effective Refractive index, n_e	~1.45
Effective Photoelastic Constant, P_e	0.22
Thermal Expansion Coefficient, α	0.55 x 10^{-6}/°C
Thermo-Optic Coefficient, ξ	8.3 x 10^{-6}/°C

III. EXPERIMENTAL ARRANGEMENT

The schematic configurations of the hydrogen sensor based on the Bragg grating and the basic hydrogen detection system used in this study are shown in Figs. 2 and 3. The palladium wire used in the hydrogen detection had a length of 5.0 cm, and originally had a diameter of 1.0 mm. However, this wire was polished into a "D" shape to increase the surface area exposed to the hydrogen while at the same time reducing the volume of the palladium exposed to the hydrogen, and attached to the fiber Bragg grating with crazy glue. Broad band light from an erbium doped fiber amplifier (EDFA) propagates through the fiber coupler, which directs the light to the hydrogen sensor. The second arm of the 2 x 2 coupler is index matched to minimize back reflections. The light reflected from the Bragg grating is directed via the 2 x 2 coupler to an HP multiwavelength meter (Hewlett Packard) or a Ferret II optical scanning card (Research International Inc.) to detect changes in the grating wavelength induced by the deformation of palladium during hydrogen detection. Exposing it to controlled mixtures (by volume) of hydrogen and nitrogen tested the hydrogen sensor. The chamber used in this experiment to mix the gases is a Pyrex flask with three gas ports to allow the gas mixture to flow into and out of the chamber, and to allow the sensor to be inserted. After the sensor was inserted into the chamber, hydrogen concentrations of

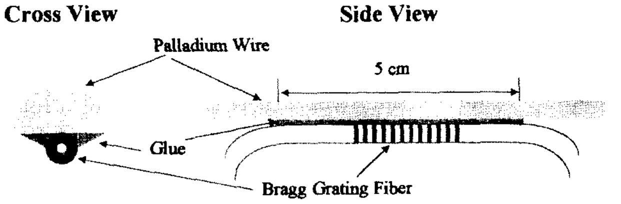

FIG. 2. The Schematic Configuration of Hydrogen Sensor Based on the Fiber Bragg Grating With Palladium.

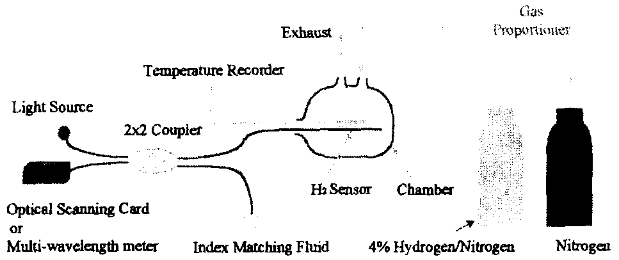

FIG. 3. Schematic Configuration of the System for Testing of the Hydrogen Sensor Using Palladium-Attached Fiber Bragg Grating.

1 %, 2 %, 3 %, and 4 % were produced using a gas proportioner to change the gas mixture volume ratio of hydrogen to nitrogen. A 4 % by volume mixture of hydrogen in nitrogen is the highest hydrogen concentration allowable by safety. The pressure of hydrogen was applied with 50 kpa and 101 kpa in these experiments. To detect wavelength shifts of Bragg gratings, both multiwavelength meter and the optical scanning card were used and compared. The wavelength shifts during hydrogen detection were recorded automatically in the computer and manually.

IV. RESULTS AND DISCUSSION

The first test we conducted was intended to confirm that hydrogen-induced Bragg wavelength shifts could be detected. In this case, a hydrogen sensor and a fiber Bragg grating without hydrogen were exposed to the same gas mixture to confirm that any changes in Bragg wavelength were due to the palladium and not to environmental temperatures. As seen in Fig. 4, the Bragg wavelength associated with the hydrogen sensor increased linearly when the palladium was exposed to 4 % hydrogen with 50 kpa pressure for 110 min at room temperature, while the Bragg wavelength of the grating without palladium showed no measurable wavelength shift. This data confirms that the sensor we design indeed responds to hydrogen. The next series of tests is intended to investigate the cyclic response and the sensitivity of the sensor to various concentrations of hydrogen. Fig. 5 shows the results of cyclic changes of hydrogen concentration. The gas mixture is set to 1 % for 20 minutes, and the hydrogen flow is turned off for 20 minutes. Next, the gas mixture is set to 2 % for 20 minutes and then the hydrogen flow is turned off for 20 minutes. This process is repeated for 3 % and 4 % hydrogen concentrations. This test was repeated six times, and Fig. 5 shows the results of tests 4 and 5. As expected, the Bragg wavelength of the palladium sensor increases when the hydrogen is present. However, higher hydrogen concentrations produce only slightly higher wavelength shifts, and there is no real indication of hydrogen desorption in the 20 minutes spans in which the hydrogen gas flow is turned off. The same test as discussed above was repeated once again. In this case, however, the hydrogen concentration was fixed at 3 % instead of starting at 1 % and increasing to 4 %. The performance of the hydrogen sensor was also shown in this figure with sensitivity of approximately 3 pm/min at 4 % hydrogen concentration and 101 kpa pressure. The results, shown in Fig. 6, are substantially the same as those in Fig. 5. The slopes of wavelength shifts vs. time appear to be the same at the times during in absorption of hydrogen gas. The desorption process was investigated by exposing the sensor to 4 % hydrogen (at 50 kpa and 101 kpa) for a fixed period of time and then purging the gas mixing chamber using pure nitrogen. The HP multiwavelength meter was used for these tests. Of

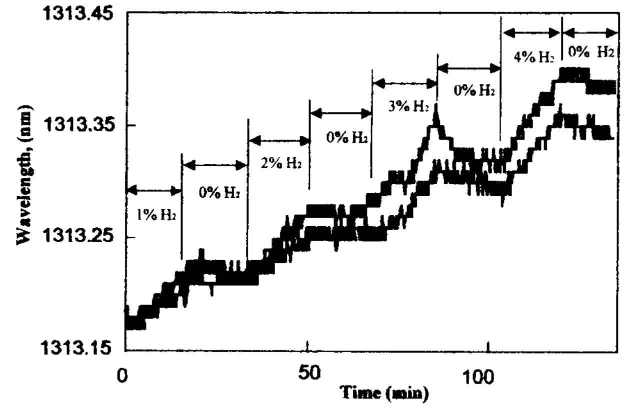

FIG. 5. Hydrogen Absorption During the Hydrogen Concentration Changes at Room Temperature With 1 % to 4 % Hydrogen Concentration and 101 kpa Pressure.

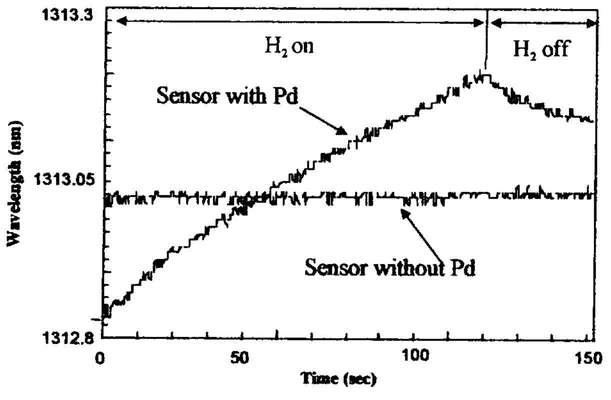

FIG. 4. The Wavelength Shift Measured by the Ferret II Card During the Detection of the Hydrogen (Left Axis: Without the Palladium, Right Axis: With the Palladium).

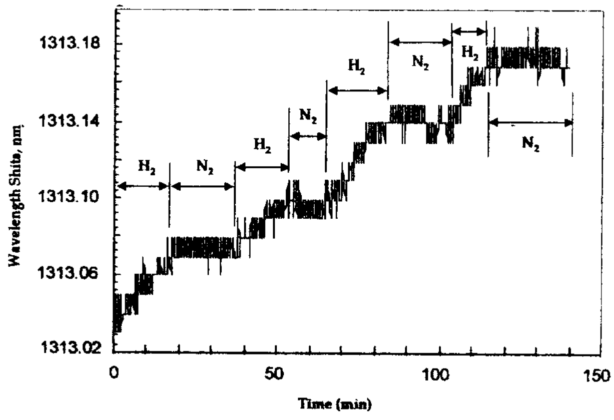

FIG. 6. Hydrogen Absorption During the Hydrogen Concentration Changes at Room Temperature With 3 % Hydrogen Concentration and 101 kpa Pressure.

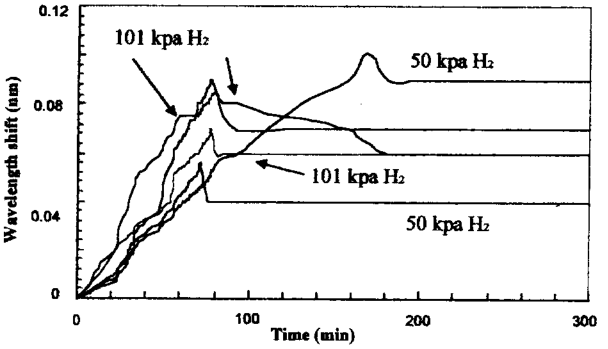

FIG. 7. Hydrogen Absorption and Desorption at the Room Temperature With 3 % Hydrogen Concentration, and 50 kpa and 101 kpa Pressure.

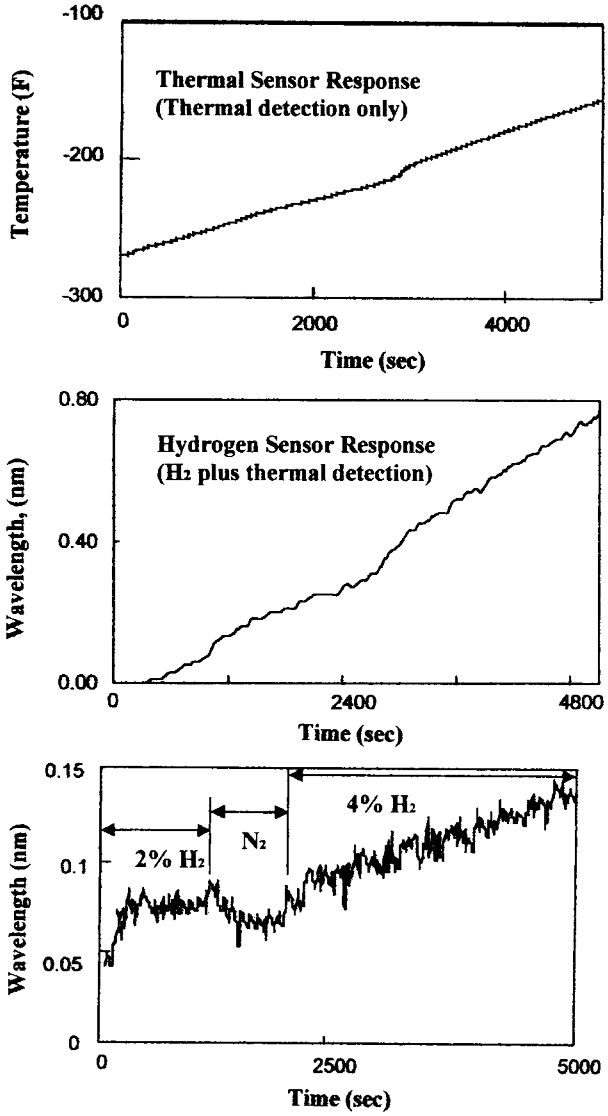

FIG. 8. Temperature Change 8(a) and Hydrogen Absorption (8(b): Before Temperature Compensation, 8(c): After Temperature Compensation) at Cryogenic Temperature With 2 % and 4 % Hydrogen Concentration and 101 kpa Pressure.

the five data sets shown in Fig. 7, only one shows any indication of desorption. We are not entirely certain why this is the case, but we believe the slow desorption response of our sensor is related to its relatively larger volume compared to the coatings for conventional optical fiber sensors used in the literature [5,6]. The final set of tests we performed involved an initial attempt to assess the response of the hydrogen sensor at cryogenic temperatures. A Pyrex flask chamber was immersed in an insulation container filled with liquid nitrogen in order to reduce the temperature of the chamber to cryogenic temperatures to allow the gas mixture to flow into and out of the chamber, and to allow the hydrogen sensor to be inserted. The same hydrogen sensor with palladium as the sensor used at room temperature was utilized at cryogenic temperatures. The light reflected from the hydrogen sensor was directed to the Ferret II optical scanning card of Research International to detect the wavelength change during hydrogen detection at cryogenic temperatures. The results are shown in Fig. 8. Fig. 8 (a) shows the variation of temperature in the chamber with the hydrogen/nitrogen gas mixture during the test, and Fig. 8 (b) shows the Bragg wavelength shift caused by the combination of thermal effects and hydrogen absorption. It is obvious from these two graphs that the thermal effects dominate the Bragg grating response causing in excess of a 1 nm wavelength change as a result of the $\Delta 110$ F temperature change during the course of the experiment. The data in Figs. 4 through 6 indicate that the hydrogen-induced wavelength change should be on the order of 0.05 nm to 0.1 nm. A calibration procedure was used to compensate for the temperature response, and the resulting Bragg wavelength shift was attributable to hydrogen. The data during times which the hydrogen is turned on shows a gradual increase in Bragg wavelength, and the data time duration in which the Hydrogen is turned off appears to be flat, as shown in Fig. 8 (c). The sensitivity of the hydrogen sensor was shown with approximately 1.2 pm/min at 4 % hydrogen concentration and 101 kpa pressure in this figure. This result shows the sensitivity difference of the hydrogen sensor between room temperature and cryogenic temperatures. The sensitivity of the hydrogen sensor at room temperature is approximately 2.5 times that of the hydrogen sensor at cryogenic temperatures. This suggests that there is a temperature dependence of hydrogen absorption of palladium, which should be considered when designing the hydrogen sensors with palladium. Difficulties were experienced during the temperature compensation arising from the fact that the temperature gauge had a slower response time than the Bragg grating. This issue can be resolved by using a colocated grating as temperature sensor.

V. SUMMARY

This study demonstrated the performance of a palladium wire hydrogen sensor based on a fiber Bragg grating. The sensitivity of the hydrogen sensor at room temperature is approximately 2.5 times that of the hydrogen sensor at cryogenic temperatures. This sensor scheme in this paper is able to multiplex many sensors as a means of developing a quasi-distributed hydrogen sensor network capable of operating at both elevated temperatures and cryogenic temperatures and has relevance to monitoring hydrogen leakage from hazardous industrial operations. Further work on sensor configuration will be able to improve the sensitivity and reversibility of hydrogen sensor responses. The simultaneous measurements of strain and temperature using a single sensor will also resolve the temperature compensation for responses caused by changes in temperature during the detection of hydrogen.

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