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Plasmonic Metal-Hybrid Hydrogen Sensor Based on Semiconductor Nanocrystal Micro Ring

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ABSTRACT

We have proposed and demonstrated numerically an ultra-small ($4\times4\mu m^2$*) hydrogen sensor based on micro ring resonator. With a palladium or platinum layer coated on the inner surface of the micro ring resonator, the device is highly sensitive to the low hydrogen concentration variation and the sensitivity is at least one magnitude order larger than the optical fiber-based hydrogen sensor. We have also investigated the tradeoff between the portion coverage of palladium/platinum layer and the sensitivity. The width of the hydrogen sensitive layer is also studied and the minimum feature width is determined to be the length of the ring waveguide evanescent wave. This ultra-small optical hydrogen sensor will be promising to realize highly compact sensor with integration capability for applications on hydrogen fuel economy.*

INTRODUCTION

Because of the superior performance of hydrogen as one of the most promising fuels in the future, it has drawn the attention of the scientific community all over the world for decades. Hydrogen is an invisible, flammable, odorless and highly explosive gas. Therefore, the hydrogen sensor has very different characteristics from the conventional gas detector. Currently, hydrogen sensors are widely utilized in solid oxide fuel cells, hydrogen transportation, hydrogen storage and other applications [1], [2].

In this work, we proposed an on-chip silicon-based optical micro ring resonator hydrogen sensor that could overcome many of the disadvantages of current sensors. It is ultra-small, easy to integrate, and highly sensitive. In addition, hydrogen-sensitive materials as Pd and Pt have good adhesion to silicon ring resonators. This ultra-small optical sensor is environment-friendly and can work in harsh and flammable environments.

Both Palladium (Pd) and platinum (Pt)are suitable for sensing hydrogen because they can react with hydrogen and form Pd/Pt hydride. They have the characteristics of sensitivity, selectivity and specificity to hydrogen, which is another reason for widely using in hydrogen sensors.

We have demonstrated numerically a novel hydrogen sensor based on micro ring resonator with low hydrogen concentration detection capability (0~4%) and ultra-small size (4x4μm^2), as illustrated in Figure 1, with hydrogen sensitive Pd or Pt layer integrated within the inner layer of the micro ring resonator. We have compared the performance of Pd and Pt layers based micro ring resonator sensors. The ring resonator is a wavelength selective photonic device that can act as an optical sensor for gas detection, including hydrogen. We have also investigated the Pd/Pt metal layer coverage within ring resonator, as more Pd/Pt layers will result in more resonance wavelength shifts, also lower the quality factor (Q) due to the loss part of metals. Finally, we studied how the width of the Pd/Pt layer will affect the performance of this type of optical sensor and revealed the relationship between the minimum feature width and the evanescent wave of the ring waveguide mode.

THEORY

The basic structure in our simulation is a silicon based micro ring resonator coated with a thin layer of Pd/Pt on its inner surface. A 3D Finite Different Time Domain (FDTD) technique is used for the simulations.

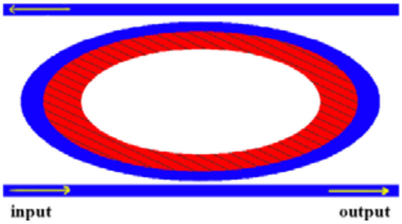

Figure 1. Top view of the hydrogen sensitive materials Pd/Pt layer coated in the ring resonator. The blue part represents Si ring resonator, and the red part (dashed part) represents the hydrogen sensitive material Pd/Pt layer.

The basic model for the device structure is shown in Figure 1. The micro ring resonator with the diameter of 3μm is evanescently coupled with the straight waveguide, which has a width of 200nm and height 200nm to keep the single wave guided mode condition. The gap between the straight waveguide and the micro ring resonator is 100nm. In Figure 1, the layer in red is a layer of hydrogen sensitive material (Pd/Pt) of thickness 400nm, deposited on the inner surface of the ring resonator. The input signal is a sinusoidal pulse, with the wavelength range from 1500nm to 1580nm. The core materials of the micro ring resonator are silicon, a refractive index of 3.45 has been used at wavelength 1550nm, the bottom cladding is SiO_2 with refractive index 1.45.

The resonance condition can be described by the following equation.

$$n_{eff} \cdot L = m \cdot \lambda$$

where L and n_{eff} are the circumstance and effective refractive index of the ring waveguide, λ is the on-resonance wavelength, and m is an integer [3,4].

RESULT AND DISCUSSION

A. Basic model and simulation results

Take palladium as an example, palladium will form palladium hydride if the hydrogen concentration in the surrounding environment increases, and as the refractive index of the palladium layer changes, the n_{eff}value will also change. The refractive index of pure palladium, at a wavelength of 1550nm, is n = 3.164 α = 665790/cm [1], [5], [6], where n and α are the real and imaginary part of the refractive index, respectively. According to the measurement of the refractive index of palladium hydride, both the real part and the imaginary part of the refractive index change when the hydrogen concentration changes from 0% to 4%. For every 1% change in hydrogen concentration, the change of the refractive index of palladium hydride is almost linear to the hydrogen concentration variation.

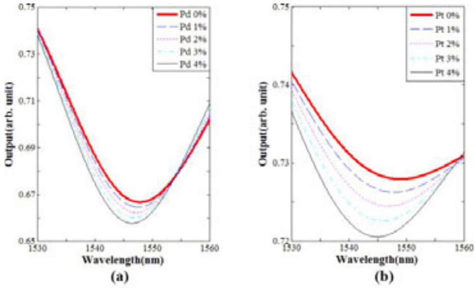

Figure 2. Resonance wavelength shift and relative intensity change of the fully covered ring resonator hydrogen sensor with hydrogen concentration variation from 1-4 % (a) Pd coated structure. (b) Pt coated structure.

For a thin palladium film at an optical wavelength of 1550nm, the real part of the refractive index decreases 0.033 as the hydrogen concentration increases every 1%, at the same time, the imaginary part increases 5432/cm [1], [5], [6]. Therefore, Pd thin film has a refractive index of n = 3.0320 and α = 687518/cm for a hydrogen concentration of 4%. For platinum, the change of n and α are -0.089 and 14674/cm for every 1% increasing in hydrogen concentration.

As the hydrogen concentration is increased from 0% to 4%, the resonance wavelength shift between 1530 and 1560nm for both Pd and Pt coated micro ring resonators. A significant resonance wavelength shift and relative intensity change can be observed. Figure 2a shows the result of the Pd coated structure, with 1% increase of hydrogen concentration, the resonance wavelength has a shift of approximately 0.35nm (i.e. 1547.9nm at 0% hydrogen and 1546.5nm at 4% hydrogen). The relative intensity decreases from 66.66% to 65.77%, i.e. 0.22% for every 1% increase in hydrogen concentration. On the other hand, Figure 2b shows the result of the Pt coated structure, where the resonance wavelength shift and intensity change are about 1.02nm and 0.18% for every 1% increase in hydrogen concentration. Compare to the Pd layer, with the same change in hydrogen concentration, the resonance wavelength shift and intensity change for Pt layer are noticeable larger. From the above results, the micro ring resonator hydrogen sensor can detect very small changes in the hydrogen concentration, for the resonance wavelength change, at least one order of magnitude larger than the fiber-based hydrogen sensor, which is due to the ultra-small size of the micro ring resonator structure.

Figure 3 shows how the resonance wavelength shift / intensity change of Pd and Pt coated layer related to the change of hydrogen concentration. Figure 3a shows that a Pt-coated micro-ring resonator has a resonant wavelength amplitude about three times higher than the Pd coated structure per 1% change in hydrogen concentration. That is due to the real part of the Pt refractive index changes more when the hydrogen concentration changes.

While Figure 3b shows the change in resonance intensity are almost the same for the Pd and Pt coated structures, Pt is even slightly larger. The lower resonance intensity of Pd layer is due to the higher optical loss. Both Pd and Pt have significant imaginary part of refractive index at wavelength around 1550nm, which means these two hydrogen sensitive materials will introduce certain loss in the ring resonator and induce a degradation of the resonance mode quality factor (Q factor). Since the real part of refractive index determines the shift in resonance wavelength, obviously, the tradeoff between the resonance wavelength shift and the Q degradation should be considered. i.e., the induced Q degradation will increase with more resonance wavelength shift at more Pd/Pt coating condition. Thus, we investigated the coating condition of Pd/Pt in the two important parameters, cover proportion of Pd/Pt layer and the width of Pd/Pt layer.

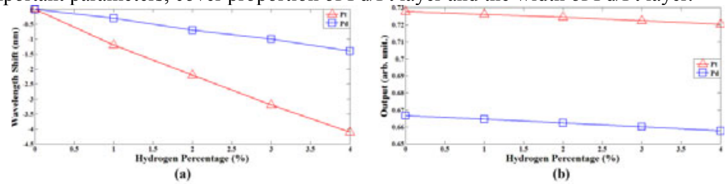

Figure 3. The comparison between Pd and Pt coated structure. (a) Resonance wavelength shift. (b) Relative intensity change.

B. Investigation of the coverage proportion of Pd/Pt layer

Coverage proportion is investigated in this section. Three different structures are used in simulation, i.e. micro ring resonator is covered with 1) one-fourth 2) half, and 3) three-fourths of Pd/Pt layer, and is compared with the above results with fully covered structure. Figure 4 illustrates the micro ring resonator structure with Pd/Pt layer is 1/2 covered.

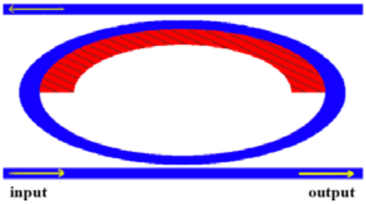

Figure 4. Top view of the ring resonator with partial (1/2) coverage of hydrogen sensitive material Pd/Pt layer.

The simulation results show that, with every 1% increase of hydrogen concentration, the resonance wavelength shift and relative intensity change for Pd is 0.08nm and 0.47% for 1/4 covered structure, 0.155nm and 0.465% for 1/2 covered structure, and 0.30nm and 0.21% for 3/4 covered structure. As described above, the results for fully covered structure is 0.35nm and 0.22%. The corresponding results for Pt based structure are 0.11nm and 0.92% for 1/4 covered structure, 0.31nm and 0.59% for 1/2 covered structure, 0.62nm and 0.30% for 3/4 covered structure, and 1.02nm and 0.18% for fully covered structure.

It can be clearly summarized that higher coverage proportion contributes to larger resonance wavelength shift. As illustrated in Figure 5, take Pt layer as an example, the Q-factor of the resonance mode for more coverage portion only degrades slightly (from nearly 70 for 1/4 covered structure to about 50 for fully covered structure).

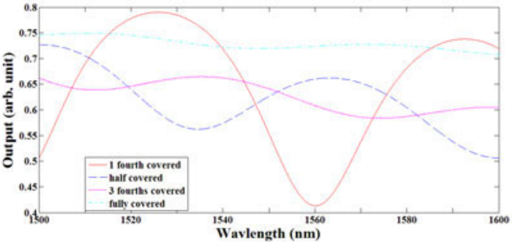

Figure 5. The comparison among different covered proportions for Pt coveredstructure at 4% hydrogen concentration.

However, different coverage proportion causes a significant relative intensity difference between the resonance mode peak and bottom. Note that results for Pt covered structure at 4% hydrogen concentration are used in Figure 5. For fully, 3/4, 1/2, and 1/4 covered structure, the relative intensity difference for Pt structure are 1.7%, 4.5%, 13.5%, and 35.4% respectively. While for Pd layer, the corresponding results are 9.9%, 18.1%, 32.1%, and 54.6%. The pros and cons of the different coverage proportion are obvious from the results Figure 5.

C. Investigation of the width of Pd/Pt layer

The width of Pd/Pt layer, will also affect the resonance wavelength shift, Q-factor and relative intensity difference, is studied in this section. Compared to 400nm Pd/Pt layer width, the 1/2 covered structure with 100 nm Pd/Pt layer width is selected to investigate this parameter, because it suffices to observe how evanescent field of the ring waveguide mode penetrates into the Pd/Pt coated layer and understand how the width of the Pt layer will impact the ring resonator sensor.

Simulation results show that, the resonance wavelength shift and relative intensity change for 100nm width Pd structure are 0.18nm and 0.44% per 1% hydrogen concentration increase. And for Pt, 0.31nm and 0.55%, are observed. These values are almost equal to the corresponding results of 400nm width of Pd/Pt layer structure shown in the above section, reaching a conclusion that width of hydrogen sensitive material Pd/Pt layer has no significant impact on the performance of device, at least when the Pd/Pt layer thickness is more than 100nm.

The electromagnetic (EM) field distribution shown in Figure 6 is used to explain this phenomenon. Simulation with continuous wave at the resonance wavelength (1535nm) applied to 400nm width of Pt layer structures is did, 0% Hydrogen concentration is used as an example in this simulation. Figure 6 shows the EM field intensity of evanescent wave in the Pt layer, the horizontal axis is the distance in nanometer from the interface of Pt layer and Si layer. The EM field intensity has an exponential decay to almost zero at 100nm, which shows that 100nm layer is thick enough to interact with all the evanescent wave, and hence, the simulation results of 400nm Pd/Pt structure has only ignorable difference with the 100nm Pd/Pt structure. In addition, the exponential decay of evanescent wave is the one of the typical characteristics of the surface modes excited by the ring resonator waveguide.

So, in terms of device fabrication, this study should provide us a route to balance the challenges in small Pd/Pt layer width fabrication, while at the same time to ensure enough Pd/Pt layer width to optically interact with the micro ring resonator evanescent field.

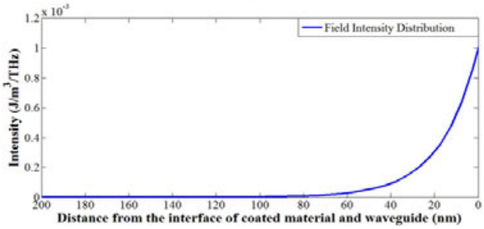

Figure6. Evanescent electromagnetic field intensity distribution in the Pt layer.

In this simulation work, we mainly focus on how the Pd/Pt coating coverage portion play the critical role on the trade-off between the Q factor, and wavelength shift, especially for the optical properties. In real fabricated devices, there are many issues which need to be investigated, such as resonator deformation due to coating deposition stresses, coating uniformity and surface flatness [7], we would like to address them in our future experimental works.

CONCLUSIONS

In summary, we have proposed and numerically demonstrated an ultra-small hydrogen sensor based on the micro ring resonator coated with a hydrogen sensitive palladium/platinum layer. The device is very sensitive to the low hydrogen concentration variation (0 – 4%) with nm resonance wavelength shift, which is at least an order of magnitude higher than the fiber based optical hydrogen sensor. We have also investigated the tradeoff between the portion coverage of palladium/platinum layer and the sensitivity. The width of the hydrogen sensitive layer is also studied and the minimum feature width is determined to be the length of the ring waveguide evanescent wave. The ultra-small size of the hydrogen sensor will have the potential to be applied in integrated optical circuits with large integration capability and portability.

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