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Study of the influence of the agarose hydrogel layer thickness on sensitivity of the coated silica microsphere resonator to humidity

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In this paper, we investigate both theoretically and experimentally the influence of the agarose hydrogel layer thickness on the sensitivity of a proposed relative humidity (RH) sensor based on a silica microsphere resonator coated with agarose hydrogel. The operating principle of the sensor relies on excitation of whispering gallery modes (WGMs) in the coated silica microsphere using the evanescent field of a tapered fiber. A change in the ambient relative humidity is detected by measuring the wavelength shift of the WGMs in the transmission spectrum of the tapered fiber. Using perturbation theory, we analyze the influence of the agarose coating thickness on the sensitivity of the proposed sensor and compare the results of this analysis with experimental findings for different coating layer thicknesses. We demonstrate that an increase in the coating layer thickness initially leads to an increase in the sensitivity to RH and reaches saturation at higher values of the agarose layer thickness. The results of the study are useful for the design and optimization of microsphere sensor parameters to meet a performance specification.

OCIS codes: (140.3945) Microcavities; (140.3948) Microcavity devices; (060.2370) Fiber optics sensors; (140.4780) Optical resonators; (260.6970) Total internal reflection.

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1. INTRODUCTION

Optical microspheres with whispering gallery modes (WGMs) have become the focus of many theoretical and experimental studies, and the basis for a number of reported ultrasensitive sensors of chemical, physical, and biological quantities [1–4,5]. The high Q-factor, very small mode volume, and narrow spectral lines of the WGMs make the microsphere resonators attractive for the design of highly sensitive and accurate sensor systems. In addition, spherical microresonators are attractive because they can be easily fabricated at the tip of an optical fiber, and the WGMs can be excited by evanescent light coupling using a simple setup, for example, based on a tapered fiber. The principle of such a sensor operation relies on measurements of the shift of the resonance wavelength against any variation of the microsphere size or any change in the optical properties of the surrounding medium. Arnold *et al.* reported specific detection of proteins absorbed on the surface of a microsphere [1], and showed theoretically that an atomic thickness can lead to a detectable shift of a given resonance frequency. The reports in Refs. [2,3] described refractometric sensors based on microsphere resonators with a sensitivity of up to 30 nm/RIU (refractive index unit), leading to a detection limit for a refractive index (RI) in the order of 10^{-7} RIU [3]. Subsequently, Ma *et al.* demonstrated temperature and humidity sensing with microsphere resonators [4].

Most of the microsphere sensors reported to date detect the change in the ambient RI because the evanescent "tail" of the WGM penetrates into the surrounding medium. Typically, the surrounding medium and silica microsphere have a large RI contrast, so that the radiation loss is very small, resulting in very high Q-factors [6]. On the other hand, this leads to a limited sensitivity of WGMs to the ambient RI, since the evanescent field is located very close to the microsphere's surface. This means that chemical molecules with poor adsorbability to the microsphere surface are difficult to detect using the sensing principle above. Teraoka *et al.* [7] examined the properties of spherical microresonators coated with high RI materials and demonstrated that the high RI coating layer enhances the sensitivity of the WGM wavelength shift. They also described a perturbation theory for the wavelength shift of WGM resonances in a microsphere coated with a high RI index layer [7]. Later, Lin *et al.* used this approximate model to calculate the thickness-dependent sensitivity of a zeolite-coated microsphere to ammonia gas [5]. They demonstrated that the zeolite coating enhances the typically small microsphere surface-to-mass ratio, and most importantly, acts as an effective concentrator of the analyte molecules, absorbing them within the layer close to the surface.

Recently we proposed and demonstrated an ultrasensitive relative humidity (RH) sensor based on a silica fiber microsphere coated with a thin layer of agarose hydrogel prepared from a 2.25% wt./vol. agarose solution [8]. Agarose is a hygroscopic material which has proved to be highly stable and can be easily applied to the surface of the device by dip coating. Changes in the surrounding RH induce changes in the RI of the agarose coating layer and thus cause a spectral shift in the WGM resonant wavelengths, which with a suitable calibration can be used for the measurement of RH. Experimental results presented in Ref. [8] showed that RH sensitivity of the sensor increases with the increase of the thickness of the agarose coating. On the other hand, increase in the agarose layer thickness also resulted in the decrease of the Q-factor due to higher absorption loss within the coating layer, ultimately limiting the sensor's resolution. This paper aims to develop a deeper understanding of the relationship between the thickness of the agarose layer and sensitivity of the sensor to RH. We analyze the influence of the agarose coating thickness on the RH sensitivity of the proposed sensor using perturbation theory and compare the results of this analysis with experimental findings for different coating layer thicknesses. To the best of our knowledge, this is the first systematic study on this topic, allowing us to determine an optimal agarose layer thickness for the best possible trade-off between sensitivity and absorption loss for a given microsphere diameter.

2. THEORETICAL ANALYSIS

In our analysis, we use the model based on the perturbation theory for the WGM resonances in a coated microsphere developed in Refs. [5,7]. Figure 1 shows a schematic diagram of the microsphere of radius a_0 with a surface coating layer of thickness t considered in the model. The RIs of the microsphere and the agarose coating layer are n_1 and n_2 and the RI of the surrounding medium is n_3.

For simplicity, we consider only fundamental WGM modes ($l = m$ and $n = 1$, where n, l, and m are the radial, azimuthal, and polar mode numbers, respectively) of TE polarization.

First, we considered an uncoated silica microsphere ($n_1 = 1.4682$; $n_2 = n_3 = 1$) with a radius $a_0 = 130$ μm and with a WGM resonance near $\lambda = 1551.12$ nm (matching those in our experiment). Solving the characteristic equation for the fundamental mode given in Ref. [5] for the above parameters gives $l = m = 757$.

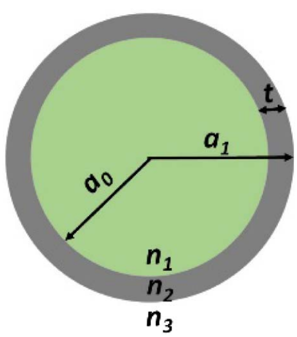

Fig. 1. Schematic diagram of the Agarose-coated microsphere.

Setting the l constant (757), we then introduce the agarose coating in the model by assuming new values for $n_2 = 1.3385$ as the RI of the bulk of the agarose layer and $n_3 = 1.0$ as the surrounding RI. The value of n_2 was measured for the agarose gel experimentally using an Abbe refractometer.

The electric field distribution for the WGM mode [transverse electric (TE) polarization] in a coated microsphere can be calculated as [5]

$$E_{l,n}\begin{cases} A_l\psi_l(n_1k^{(n)}r) & r < a_0 \\ B_l\psi_l(n_2k^{(n)}r) + \chi_l(n_2k^{(n)}r) & a_0 < r < a_1, \\ C_l\chi_l\psi_l(n_3k^{(n)}r) & r > a_1 \end{cases} \quad (1)$$

where $k^{(n)} = 2\pi/\lambda_R$ is the wave vector for nth order radial WGM; $\psi_l(x) = xJ_l(x)$ Ricatti–Bessel and $\chi_l(x) = xN_l(x)$ Ricatti–Neumann functions are calculated from the lth order spherical Bessel J_l and Neumann functions N_l, respectively. A_l, B_l, and C_l are constants determined from the boundary conditions for the given mode number, and the detail calculations were provided by Lin *et al.* [5]. Figure 2 illustrates the electric field distributions for different values of the agarose layer thickness obtained by solving boundary conditions for the mode with $l = 757$ using MATLAB software.

The RI sensitivity for the TE polarization (S_{TE}) of the coated microsphere can be calculated as [5]

$$S_{\mathrm{TE}} = \frac{n_2\lambda_R I_2}{n_1^2 I_1 + n_2^2 I_2 + n_3^2 I_3}, \quad (2)$$

where I_1, I_2, and I_3 are the fractions of the mode's energy distributed within the silica microsphere, agarose coating layer, and the surrounding medium, respectively.

Figure 3 shows the simulated dependence for the RI sensitivity of the fundamental WGM above versus the agarose coating layer thickness. One can see from the graph that the estimated RI sensitivity saturates around the value of 1.5 μm of the coating thickness.

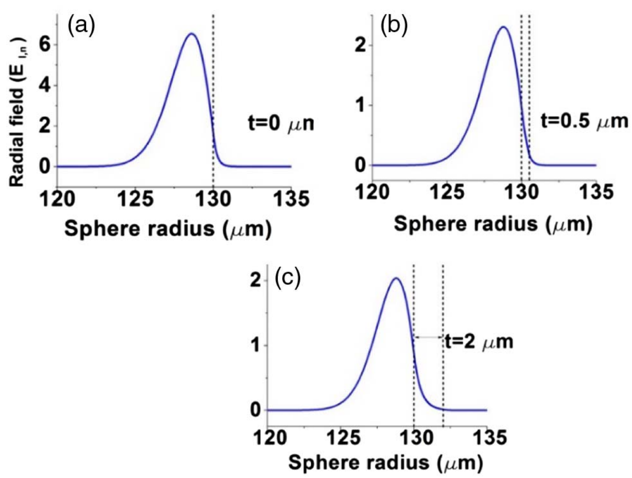

Fig. 2. Radial field distributions for the fundamental WGM with $l = m = 757$ and different agarose coating thickness values: (a) $t = 0$, (b) $t = 0.5$ μm, and (c) $t = 2$ μm.

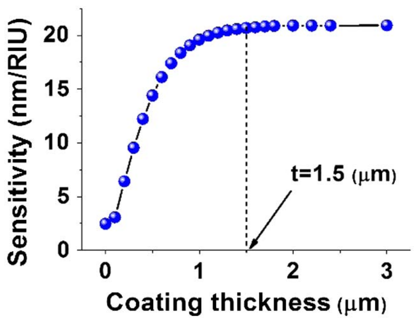

Fig. 3. Simulated dependence for the RI sensitivity of the fundamental WGM above versus the agarose coating layer thickness.

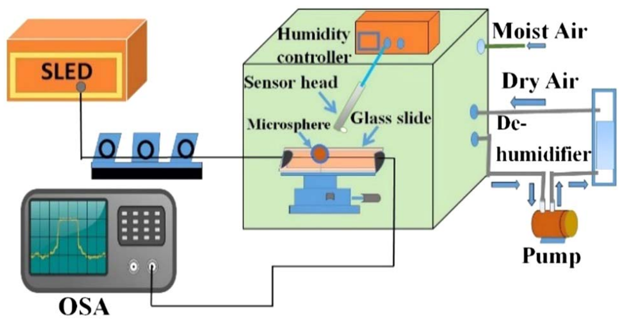

Fig. 4. Experimental setup for RH measurements.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The microsphere for our experiments was fabricated at the tip of a standard SMF28 single-mode fiber by applying a series of electric arc discharges. The sphere diameter in the experiment was measured using an optical microscope as 260 μm. Agarose hydrogel for the coating was fabricated using commercially available agarose powder from Sigma Aldrich (A6013). The hydrogel solution was prepared by adding 2.25% wt./vol. of the agarose powder into deionized (DI) water, followed by stirring at 80°C temperature until the agarose powder completely dissolved in the DI water. The RI of the agarose gel was measured as 1.3385 with the help of an Abbe refractometer. The agarose layer was then applied on the surface of the microsphere by the dip coating method. Coating layers with different thicknesses were realized by repeating the dip coating cycle multiple times (up to six cycles in our experiment). After each coating cycle, the microsphere was kept at room temperature for one day to allow for curing. For light coupling to and from the microsphere, an adiabatic tapered fiber was used with a waist diameter of ~3–4 μm. Such a diameter was chosen to ensure the match between the propagation constant of the propagating mode in the taper with that of the WGM mode of interest [9]. The fiber taper was placed in a close proximity with the microsphere inside a chamber in which both humidity and temperature could be controlled. Light from the broadband superluminescent light source (SLED) operating in the wavelength range 1530–1570 nm was launched into the fiber taper and the corresponding transmission spectrum was observed at the taper output by means of the optical spectrum analyzer (OSA) with a 10 pm wavelength resolution. The light transmitted through the fiber taper was received by a photodetector connected to an oscilloscope, as shown in Fig. 4.

Experimental transmission spectra were recorded and analyzed for the uncoated microsphere and after each of the subsequent six coating cycles in the RH range from 25% to 50% RH. The value of quality factor was estimated from the experimental spectra as $Q = \lambda_R/\Delta\lambda_{\mathrm{FWHM}}$, where λ_R is the resonance wavelength and $\Delta\lambda_{\mathrm{FWHM}}$ is the full width at half-maximum of the resonant lobe, calculated by fitting the resonant dip with the Lorentz function. Figures 5(a) and 5(b) illustrate, as an example, changes in the quality factor of the microsphere due to application of the agarose coating. The Q for the uncoated microsphere estimated as 5.87×10^4 decreases to $\sim 1.61 \times 10^4$ after four cycles of coating due to the increase in absorption loss within the agarose layer.

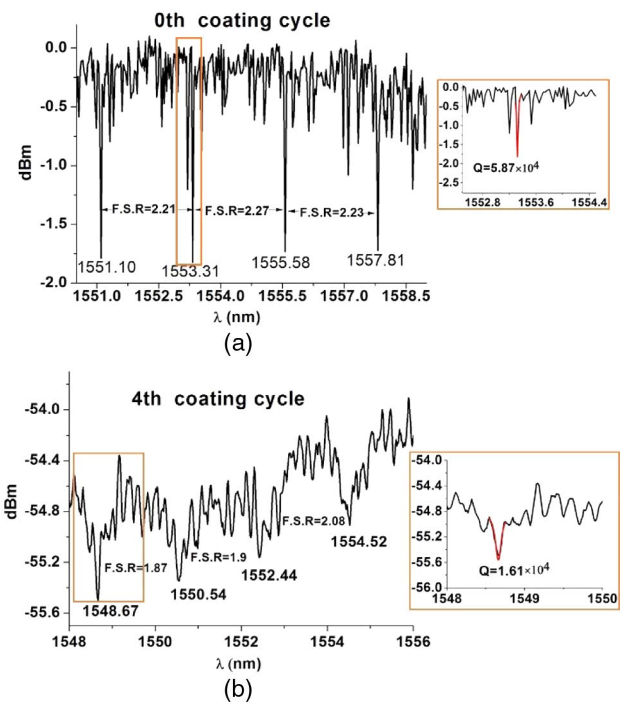

Fig. 5. Transmission spectra with the selected WGM resonances: (a) for uncoated silica microsphere with a 260 μm diameter and (b) for the same microsphere after application of four agarose coating layers. Insets illustrate fitting of the selected resonances and estimated Q-factors.

Figure 6 shows experimentally measured WGM spectra recorded at different humidity levels for the microsphere coated with a single agarose layer. The inset graph illustrates the shift of the selected WGM resonance versus changes in humidity inside the chamber. Any increase in humidity inside the chamber gives rise to the adsorption of water molecules on the surface of the microsphere; water molecules replace the air inside the micro-pores of the coating layer, which in turn increases the effective RI of the agarose layer. Increase in RH from 25% to 50% at a constant room temperature resulted in a red shift in the WGM spectrum, and the value of the spectral shift increased with the increase of the number of agarose coating

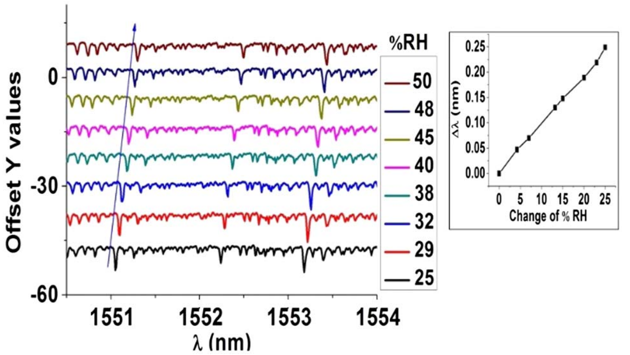

Fig. 6. Experimental WGM spectra for the microsphere coated with a single agarose layer recorded at different humidity levels varying from 25% to 50% RH. Inset graph illustrates the WGMs spectral shift versus changes in RH.

cycles (the coating thickness), as shown in Fig. 7(a). Figure 7(b) summarizes the experimental data related to RH sensitivity and Q-factor values versus the number of coating cycles for all the studied samples.

As expected, the quality factor decreases monotonically with the increase of the coating thickness due to the increased absorption. The measured RH sensitivity, on the other hand, initially grows with the increase of the coating thickness and reaches saturation at a higher coating thickness value. This result agrees well with that predicted by the model (Fig. 3). Correlation between the experimental and theoretical modeling results allows for an approximate estimate of the agarose layer thickness achieved by the technique described above. Since we have estimated the Q-factor of the microsphere after each consecutive coating cycle, it is possible to derive the layer thickness by comparing the resonance wavelength difference for every coating cycle for a certain mode at certain RH and temperature. For this, we consider the effective index of the resonance mode, determined by the refractive indices of both silica and agarose, and the energy fraction of the mode distributed in both parts:

$$\frac{1}{Q} \approx \eta_1 \frac{1}{Q_{\text{uncoated}}} + \eta_2 \frac{1}{Q_{\text{coated}}}, \tag{3}$$

where

$$\eta_1 = \frac{n_1^2 I_1}{n_1^2 I_1 + n_2^2 I_2 + n_3^2 I_3}, \tag{4}$$

$$\eta_2 = \frac{n_2^2 I_1}{n_1^2 I_1 + n_2^2 I_2 + n_3^2 I_3}. \tag{5}$$

Assuming the agarose coating is smooth and defect-free so that intrinsic loss is negligible, the total Q-factor mainly depends on the absorption loss within the agarose layer, and can be expressed as $Q_{\text{coated}} \approx 2\pi n_2/\alpha_2 \lambda_R$, where α_2 is the propagation loss in the agarose coating (the approximate value for the agar material loss coefficient per unit length is 0.5 dB/cm [10]), n_2 is the coating RI, and λ_R is the resonant wavelength. By comparing the results of simulations for the fraction of energy (η_2) within agarose coating with the value of the Q-factor measured experimentally, the coating thickness corresponding to each number of coating cycles is derived and

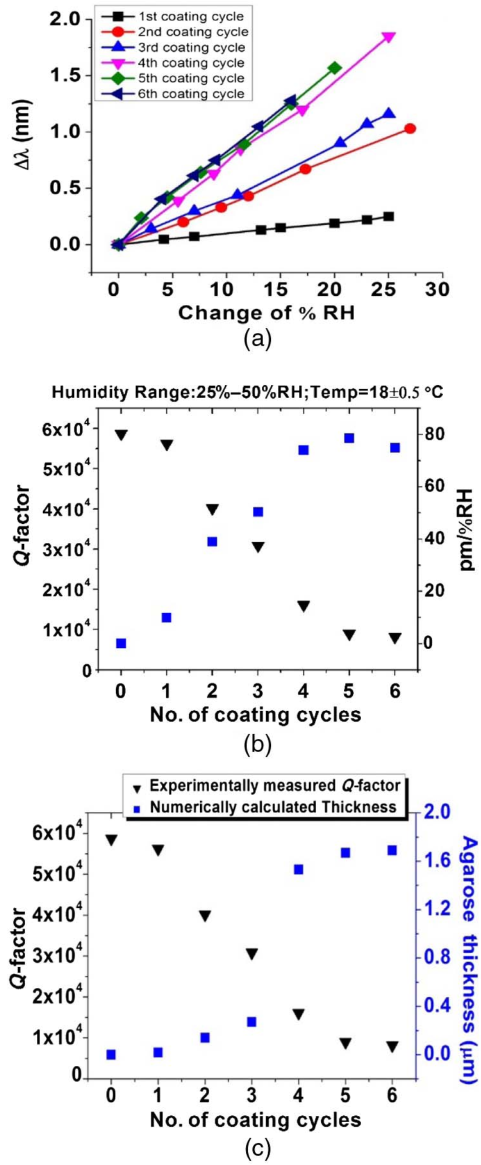

Fig. 7. (a) Measured WGM wavelength shift versus RH changes for different thickness of the agarose coating; (b) Q-factor and RH sensitivity from experimental data; and (c) experimental Q-factor and calculated agarose coating layer thickness as a function of the number of coatings.

plotted in Fig. 7(c), along with the experimental Q-factor values for each number of the coating cycles. With increasing the number of coating cycles of agarose, the absorption loss is also increased significantly, as a result the total Q-factor decreases [5].

4. CONCLUSION

In this paper, we carried out a theoretical analysis of the sensitivity of the proposed sensor based on perturbation theory and compared the results of the simulations with our experimental findings for the coating layers of different thickness. It is concluded that an increase in the coating thickness initially leads to an increase in the RH sensitivity and then reaches saturation at agarose coating layers with greater thickness (>1.6 μm in our experiments). The results of the study are useful for the design and optimization of the microsphere sensor parameters for a specified performance.

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