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Compact humidity sensor based on a multi-layer Agarose hydrogel coated silica microsphere resonator

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ABSTRACT

In this paper we report on a novel approach to implementing a compact humidity sensor that utilizes whispering gallery mode (WGM) phenomena in a silica microsphere coated with Agarose hydrogel. The spectral positions of the WGM resonances for such a sensor depend strongly on the refractive index and thickness of the coating. The WGM's spectral shift occurs due to adsorption/desorption of the water vapor in response to changes in ambient humidity and also due to the corresponding changes of the coating thickness. We experimentally investigated the WGMs spectral shift for a 100 µm diameter silica microsphere coated with Agarose hydrogel over a wide range of relative humidity (RH) values from 30%RH to 70%RH at a constant temperature. Six dip coating cycles of 2.25% wt. /vol. Agarose hydrogel were carried out in sequence with a characterization of the sensor performed for each coating thickness. A resonance shift of 16 nm is achieved in our experiment for the six-layer Agarose hydrogel coating sensor.

Keywords: Optical Resonators; Micro-optical Devices; Fiber Optic Sensors; Micro structured fibers

1. INTRODUCTION

Optical microcavity resonators are versatile building blocks that can provide solutions in a variety of areas, including optical sensing [1] and optical communications [2]. One approach is to adapt traditional Fabry-Perot (FP) resonators, but FP resonators are difficult to scale to micro sizes and are also difficult to integrate. Silica microsphere microresonators are widely used for many applications as they can have quality factors that are several orders of magnitude better than typical surface etched resonators and they also possess as small modal volume, which allows for sensors that required very small amounts of analyte. Light is confined inside the microcavity by total internal reflection. Resonance occurs when light retains the same phase after each cycle of propagation. This is known as a Whispering Gallery Mode (WGM) resonance. Since these microcavities can be shaped by natural surface tension forces during fabrication, the result is a clean, smooth silica surface with low optical loss and negligible scattering. Such microcavity are also inexpensive, simple to fabricate, and are compatible with integrated optics. Optical microcavity resonators are particularly useful for optical sensing given that ultra-high Q factors in the order of ~10^{10} have been demonstrated for a variety of sensing applications [3].
The presence of water vapors in the air is known as humidity. It plays a significant role in agriculture, food industry, clinical medicine, manufacturing, civil engineering, textile, semiconductor industry and many other fields. Humidity measurement in industries is critical because it may affect the quality of the product, for example the shelf-life of foodstuffs. Hence, humidity sensing is very important, especially in the control systems for industrial processes and for human comfort. In recent decades, various humidity sensors have been proposed and developed, which sense relative humidity (RH) in terms of resistance [4,5], capacitance [6], or refractive index (RI) [7]. Fiber-optic humidity sensor has some notable advantages over conventional electronic based humidity sensors, such as miniature dimensions, light weight, a potential for multi-parametric sensing, immunity to electromagnetic interference, water and corrosion resistance and radiation tolerance. The methods involved in the measurement of humidity using fiber optic humidity sensor are based on

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a variety of principles, including absorption [8], evanescent wave [9], interferometric [10], strain [11] and the use of an LPFG (Long Period Fiber Grating) [12]. Due to the ultra-high *Q* factor of optical microresonator, the potential resolution for humidity measurement is better than other fiber optic humidity sensors. For example, for humidity sensing for a long-period fiber-grating device coated with SiO_2 nanoparticles it is estimated the minimum detectable humidity change if of the order of 1% RH whereas for a microsphere coated with same SiO_2 nanoparticles in spherical microresonator the minimum detectable change is estimated be around 0.003%RH [13].

In this paper, we propose a compact spherical micro resonator humidity sensor utilizing WGMs. The microsphere is made of silica fiber and dip coated with several layers of a hygroscopic material known as Agarose. Agarose gel, as a smart material, has proved to be highly stable, not soluble in water and can be handled easily during device fabrication. Humidity changes induce a refractive index (RI) change in coating layer and thus a shift in the resonance wavelength, which with a suitable calibration can be used to measure humidity change. We have observed that the sensitivity to humidity increased after introducing each new layer of Agarose coating to a 100 µm microsphere, over a wide range of humidity levels from 30% to 70% RH. We report a detailed study of the sensor in terms of its sensitivity, repeatability, long term stability and measured accuracy.

2. EXPERIMENTAL INVESTIGATION AND DISCUSSION

The microsphere for our experiments was fabricated at the tip of a standard single mode fiber by discharging a series of electric arcs. The fiber tip was gradually melted by the arcs and assumed spherical shape due to surface tension. The sphere diameter was controlled by the number of arcs induced on the tip of the fiber. Fig.1 shows a microscopic image of a 100 µm diameter microsphere, which is used in our experiment. Agarose is commercially available in a white powder form (Sigma Aldrich, A6013). An Agarose solution was firstly prepared by adding 2.25% wt./vol. of Agarose powder into deionized (DI) water and the solution was constant stirred at 80 ˚C temperature until the Agarose powder completely dissolved the DI water. Next the microsphere was quickly dipped in to and out of the Agarose hydrogel and subsequently was kept at room temperature for one day before use to allow drying to take place. The same procedure was followed for second, third, fourth, fifth and sixth coating cycles on the same microsphere.

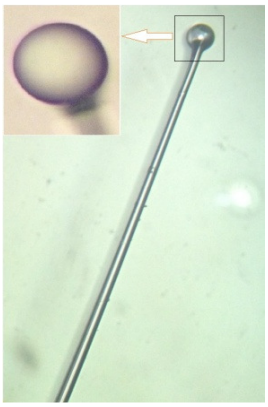

Figure 1. Microscopic photographs of the microsphere having 100 µm diameter used in the experiment

An adiabatically tapered fiber was used to couple light into and out of the microsphere. The tapered fiber was prepared by a micro heater brushing technique [14] from a standard SMF-28^{TM} single mode fiber, having a core and cladding diameter 8.3 µm and 125µm respectively. After tapering to ensure mechanical stability, the tapered fiber was fixed on a microscopic glass slide at a height of approximately 5 mm above the surface of the glass slide using UV curable epoxy. The tapered waist diameter was ~3.3 µm with a length in our experiment of approximately 2 mm. Fig. 2 shows the experimental setup for humidity measurement. The light from a fiber coupled superluminescent diode (SLD) light source (Thorlabs S5FC1005S) was transmitted through a polarizer to the tapered optical fiber. The other end of fiber taper was connected to optical spectrum analyzer.

The coated microsphere was positioned in contact with the tapered fiber inside a controlled humidity chamber (ETS5503) as shown in Fig. 2. The environmental chamber consists of ~0.11 m^3 airtight space with an inlet and an outlet. The upper inlet is made for the entry of the moist air into the chamber and the outlet is used for dragging the moist air to the dehumidifier through a pump. The dehumidifier box contains anhydrous calcium sulfate which absorb

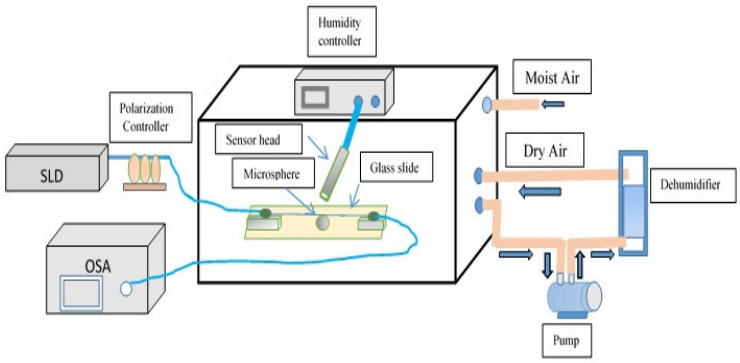

Figure 2. Experimental setup with controlled humidity chamber

moisture from the air. The compressed dry air is pumped in to regulate the RH in the chamber. Humid air was obtained by bubbling dry air into an ultrasonic humidification system (ETS5462) controlling the ratio of humid air and compressed dry air in the chamber gives a fixed RH. The chamber controller system allows for independent setup of both temperature and humidity inside the chamber. The accuracy of the chamber is ±2% RH. Each humidity measurement was recorded five minutes after the RH level reached a set value to allow for the humidity throughout the chamber to stabilize. Light from SLD source is coupled into the coated microsphere by evanescent coupling from the fiber taper.

The WGM spectral positions are recorded at the output end of the taper fiber by detection of the transmitted light intensity using an optical spectrum analyzer (OSA, Advantest Q8384) with a resolution of 0.01 nm. In order to eliminate the effect of temperature variations, the temperature of the humidity chamber was set to a constant at 23 ˚C (close to room temperature) throughout the entire RH measurement cycle. The transmission spectrum achieved from the 100 µm sphere is shown in Fig.3.

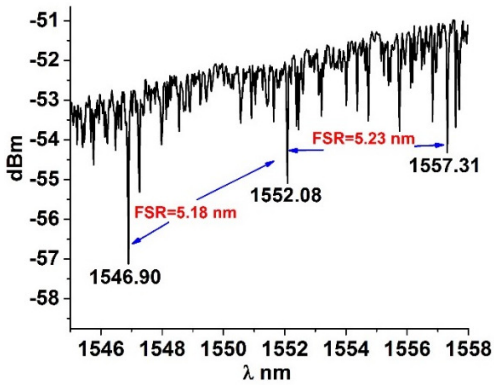

Figure 3. Transmission spectrum recorded with an Agarose coated microsphere with a diameter of 100 µm

The value of the free spectral range (F.S.R) determined from the graph (5.187 nm) can be used to estimate the microsphere diameter or its effective refractive index based on the approximate formula [3]

$$\text{F.S.R} \approx \frac{\lambda_0^2}{\pi D n_s} \qquad (1)$$

where λ_0, n_s and D are the resonant wavelength, the refractive index of the microsphere and the microsphere diameter, respectively. Assuming that the RI of the micro resonator is close to that of silica (n_s = 1.4628), the resulting calculated microsphere diameter is 101 µm at resonance wavelength 1552 nm, which is in a good agreement with the result of the microscopic measurement (Fig. 1). Experimental measurement of humidity inside chamber was carried out with the coated microsphere by observing one of the resonance wavelengths of the micro resonator under the influence of a controlled humidity variation. The sensing mechanism of Agarose gel is based on the swelling behavior of the Agarose gel upon adsorption of water vapors with an increase of ambient humidity [15]. As shown in Fig 4(a), an increase of humidity inside the chamber gives rise to the adsorption of more water vapors on the surface of the microsphere. As a result, the air inside the micro pores of the coating layer is replaced by water molecules due to the hygroscopic nature of the material and capillary forces, which in turn gives rise to an increase in the effective refractive index of the Agarose coating layer [16]. As a result, a redshift in the WGM spectrum is observed with an increase in the humidity from 30 % to 70% at a constant temperature of 23±0.4 ˚C. A commercially available electronic hygrometer sensor head is used to calibrate the humidity measurement inside the chamber. Fig.4 (b) is the linear plot of the change in resonance wavelength vs change in humidity inside the chamber. From the graph Fig 4(b), the sensitivity of the sensor to the humidity was calculated as 22.4 pm/%RH. Similarly, when the humidity decreases inside the chamber, water vapors are desorbed from the coating layer, hence the effective refractive index of the coated layer decreases resulting in a blue shift of the resonance wavelength as shown in the Fig.4 (c).

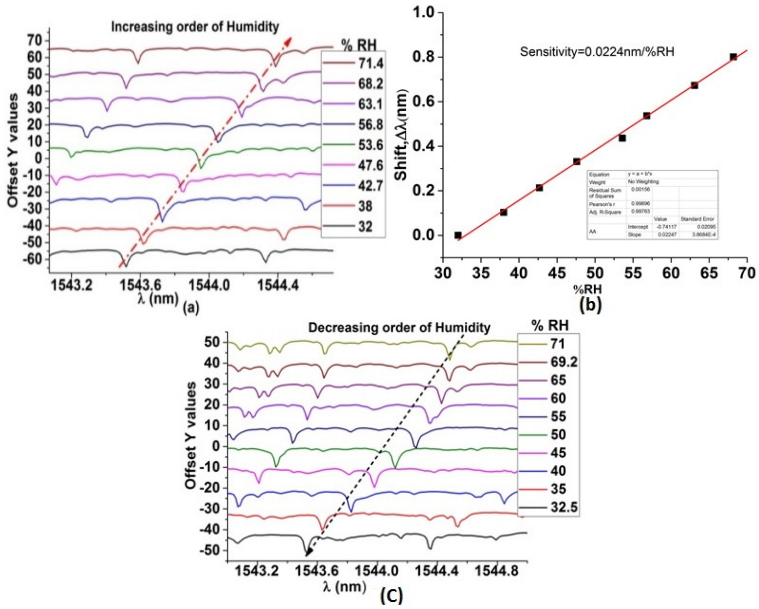

Figure 4. Experimental results for the WGM resonator based on a 100 µm diameter microsphere coated with two cycles of 2.25% Agarose gel coupled with a 3.3 µm tapered fiber in the range of humidity from 30 % to 70% RH at a constant temperature of 23 ^{0}C. (a) WGM spectra at different RH levels at gradually increasing humidity; (b) Linear response for resonance shift wavelength vs a change in humidity; (c) WGM spectra at different RH levels for gradually decreasing humidity.

Since it is known [17] for conventional fiber sensors that an increase in the coating thickness can increase sensitivity, in order to determine if the same is true for the sensitivity of the micro resonator, the microsphere was subjected to 2nd,3rd ,4th ,5th and 6th coating cycles of Agarose hydrogel. After finishing each coating cycle, the microsphere was characterized for its humidity response inside the test chamber. The humidity inside the chamber was varied from 70% RH to 30% RH at a constant temperature. From Fig. 5, it is clear that with an increase in the number of coating cycles of Agarose on the microsphere, the shift in the WGM resonance wavelength also increased accordingly. The maximum shift in the WGM wavelength was found to be 370 pm for the 1st coating cycle of Agarose layer whereas the shift for the 3rd coating cycle was approximately 1.54 nm, which was four times greater than the 1st coating cycle for the same humidity variation. After

the sixth coating cycle, the sensor RH sensitivity was further increased, with a resonance wavelength shift around 16.5 nm and a sensitivity calculated as 518 pm/%RH in the range of 30-70%RH. The exact Agarose coating thickness was unknown in our experiment but it is known that coating thickness influences [17] sensitivity.

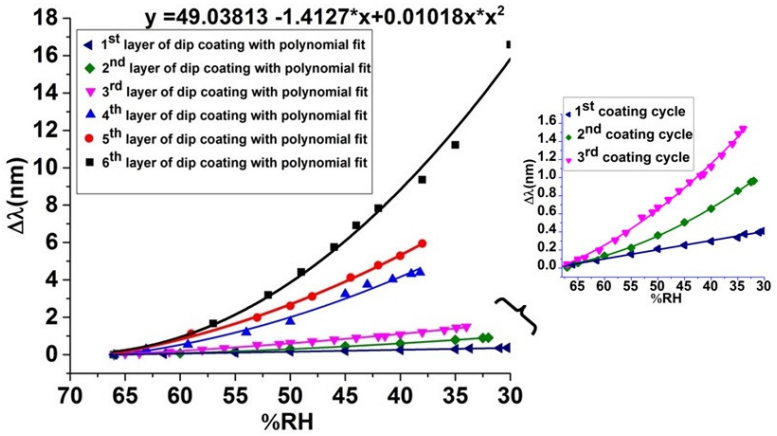

Figure 5. WGM spectral shift versus RH change of 1-6 cycle of 2.25% wt./vol Agarose coated on 100 µm microsphere at 23 ˚C

A physical explanation for why sensitivity increases with the number of coating cycles is that when the Agarose layer thickness increases, a greater proportion of the light energy is contained in the polymer and thus the change in the polymer's RI (n_a) in response to changing RH leads to a higher RH sensitivity for the micro resonator. However, as the thickness of the polymer layer increases, the larger material absorption loss in the Agarose compared to silica should also result in a decrease of the Q factor, which can be understood as follows. For a relatively large silica sphere coated with an Agarose layer which has a smooth surface and assuming an absence of impurities in the Agarose that could increase losses, the quality factor mainly depends on the absorption loss within the resonator material [3], which can be expressed as in Eq.(2).

$$\frac{1}{Q_{in}} = \frac{1}{(Q_{abs})_{silica}} + \frac{1}{(Q_{abs})_{agarose}} \tag{2}$$

where $1/(Q_{abs})_{silica}$ and $1/(Q_{abs})_{agarose}$ denote the material absorption loss within the silica microsphere and the Agarose layer, respectively. The absorption limited Q factor can be calculated as $(Q_{abs})_{silica} = 2\pi n_s/\lambda_R\alpha_s$ and $(Q_{abs})_{agarose} = 2\pi n_a/\lambda_R\alpha_a$, where α_s, n_s and α_a, n_a are the optical attenuation coefficients per unit length and the refractive indices of silica and Agarose layer respectively. From Eq (2), as the Q factor of the coating decreases as it gets thicker, because of the larger material absorption loss in the Agarose compared to silica, the coating Q begins to dominate the overall Q-factor, in effect the total Q factor is mainly determined by the large absorption loss in the polymer and thus for the thicknesses typically created by dip coating, the overall Q can be approximated by $Q = 2\pi n_a/\lambda_R\alpha_a$.

To verify that increasing the number of coating cycles will decrease the Q, we carried out a series of experiments for a silica microsphere with a fixed diameter (100 µm) which was coated with 2.25 wt./vol.% Agarose gel applied multiple times through repeated coating cycles, forming coatings with progressively larger thickness values. The WGM spectra of the coated sphere were recorded after one- ,two- ,three- ,four-, five and six coating cycles respectively using the same setup and the same tapered fiber at a constant temperature (23±0.4˚C) and humidity(66%RH).The spectra were further analyzed to estimate the corresponding Q-factors, by calculating as $Q = \lambda_{res}/\Delta\lambda_{FWHM}$, where λ_{res} is the resonance wavelength and $\Delta\lambda_{FWHM}$ is the FWHM of the resonant lobe calculated by fitting the resonance dip with Lorentz equations illustrated in Fig. 6 (a, b, c). As expected the Q factor decreases as the number of coating cycles increases.

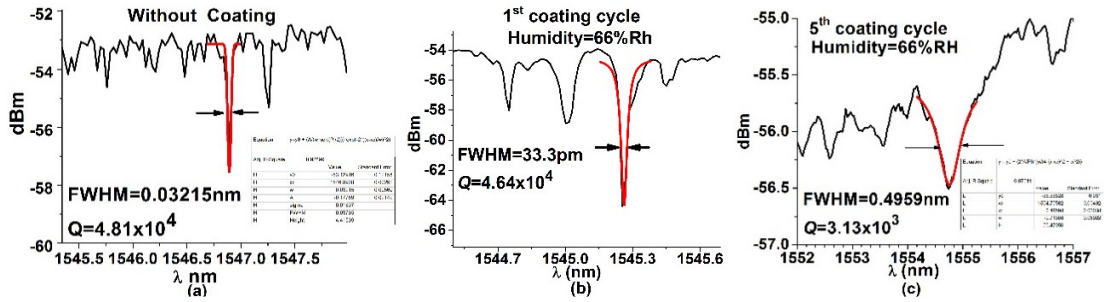

Figure 6. Experimental spectra and Lorentzian fitting for a 100µm diameter microsphere coated With 2.25% Agarose hydrogel: (a) without a coating; (b) after one coating cycle; (c) after 5 coating cycles.

Fig 7 summarises the impact of the number of coating cycles on the Q factor and on the sensitivity. The Q factor gets smaller as the number of cycles increases while the sensitivity increases with the number of coating cycles. The maximum sensitivity in our experiments was observed for the sensor coated six times and was estimated at 518 pm/%RH which is significantly higher than that of 10 pm/%RH for the sensor with a single coating as illustrated in Fig 8.

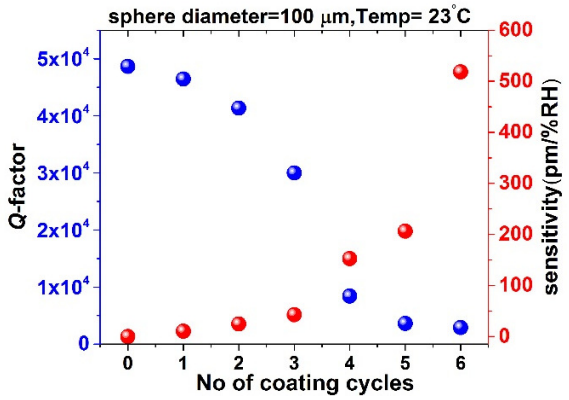

Figure 7. *Q*-factor and estimated RH sensitivity for the 100 µm diameter microsphere with a different number of Agarose coating layers

The detection limit (*DL*) is the critical parameter to quantify the device sensing capability. The detection limit represents the smallest measurable physical parameter change and it can be expressed as [18]:

$$DL = \frac{R}{S} \tag{3}$$

where R is the resolution of the sensor and S is the sensor sensitivity. Large Q-factor values may result in poorer detection limit values, due to the increased temperature sensitivity, while low Q-factors are typically limited by amplitude noise and spectral resolution [18]. Considering all the factors, R can be calculated as,

$$R = 3 \times \sqrt{{\sigma_N}^2 + {\sigma_T}^2 + {\sigma_{SR}}^2} \tag{4}$$

where $\sigma_N, \sigma_T, \sigma_{SR}$ represent the standard deviations associated with amplitude noise, temperature and detector spectral resolution, respectively. We have calculated the the detection limit of 100 µm microsphere coated with a 1st cycle and a 5th cycle of Agarose layer using the same procedure described detailed in a previous paper [18]. The parameters and resulting DLs are compared in the Table 1. This assumes the noise level of the system is 60 dB, the spectral resolution of

our spectrum analyzer is 10±0.03pm and the standard deviation due to temperature stabilization is 10 fm.

Table 1. comparision the detection limit of two different coating cycles of 2.25% wt./vol. Agarose layer onto 100 µm microsphere

	One of coating cycle	Five coating cycles
Q-factor	4.64×10^4	3.13×10^3
RH sensitivity (pm/%RH)	10	206.32
Standard deviation of amplitude noise(σ_N)	0.23358pm	3.4848
Standard deviation of temperature stabilization (σ_T)	0.010pm	0.010pm
Standard deviation of spectral resolution (σ_{SR})	0.2236 pm	0.2236 pm
Total R	0.97052 pm	10.4759 pm
Detection Limit (DL)	9.7×10^{-2} %RH	5.8×10^{-2} %RH

To investigate the stability and repeatablity of the sensor, the humidity response of a 171 µm diameter coated microsphere was measured twice, but with a week long time gap between the measurmement, with the same temperature and same testing parameters without disturbing the setup inside the chamber. The sensor proved stable and repeatable between the two measurements with a small fluctuation as shown in Fig 8.

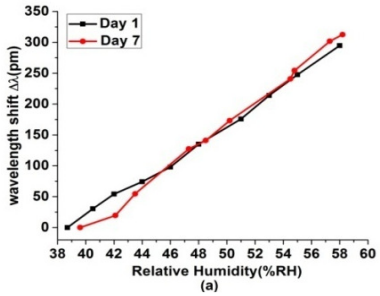

Figure 8. Relative humidity responses of the sensor, measurements recorded seven days apart for a 171 µm diameter microsphere;

The hysteresis characteristics using data from humidification-dehumidification cycle for a first, second and third cycle of 2.25 wt./vol.%Agarose coating on a 100 µm diameter microsphere was studied. The swelling rate of Agarose polymer

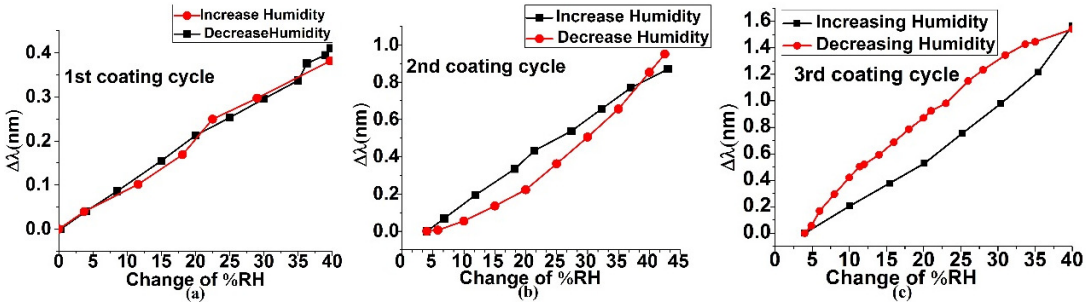

Figure 9. Studies of the sensor hysteresis: humidity response for the 100 µm microspheres coated with 2.25% wt./vol. (a) 1st coating cycle (b) 2nd coating cycle, (c) 3rd coating cycle of Agarose concentration coatings during an RH increase-decrease cycle at 23˚C.

depends on the thickness of the layer. Lower thickness tends to high rate of swelling and de-swelling. As the thickness of Agarose layer increases, it takes more time to desorb water molecules compared to a thinner layer, leading to in increase in hystereis, which is experimentally proven in Fig.9 (a, b & c).

Finally, to investigate the effect of temperature on the RH sensor performance, the humidity in the chamber was set to a constant value of 31% RH. The temperature was then gradually raised from 16 °C to 24°C. The wavelength shift of the WGM spectrum with temperature is shown in Fig. 10. It can be noted that the temperature sensitivity of the sensor is small compared to its RH sensitivity. For a 100 µm diameter sensor coated with 2.25% of Agarose hydrogel, the temperature sensitivity it is estimated as ~11.3 pm/°C in the temperature range of 16 -24°C.

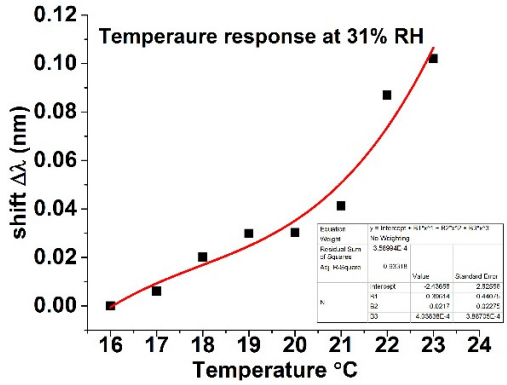

Figure 10. Resonance wavelength shift versus temperature for a 100 µm microsphere coating of one cycle of 2.25% wt./vol. Agarose solution at 31% RH

3. CONCLUSION

A compact relative humidity sensor based on a whispering gallery mode microresonator has been proposed and experimentally demonstrated. WGMs are excited in the silica microsphere dip-coated with an Agarose gel, evanescently coupled to a tapered fiber. A change in the refractive index of the Agarose coating arising due to changes in the surrounding relative humidity, which leads to a spectral shift of the WGM resonances which can be related to the RH value after a suitable sensor calibration. The RH sensitivity of the proposed sensor can be enhanced by applying more coating layers of the Agarose hydrogel. Studies of the humidity response of 1-6 cycle coating of 2.25% wt./vol. Agarose gel for a 100 µm diameter microsphere was experimentally investigated. The results showed that an increase in the thickness of the coating material results in an increase in sensitivity but also leads to a decrease in quality factor for the coated micro resonator. The highest sensitivity achieved in our experiments was 518 pm/% RH in the RH range from 30% to 70%. The proposed sensor offers the advantages being very compact in nature, whilst also demonstrating low hysteresis, good repeatability and a relatively low cross sensitivity to temperature.

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