Stretchable PEG-DA Hydrogel-Based Whispering-Gallery-Mode Microlaser with Humidity Responsiveness

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***Abstract*—Optical microcavities have been widely employed for sensing and detection, because of their ultrasensitive responsiveness to surrounding environment. Here we report an active disk-shaped whispering gallery mode (WGM) microcavity fabricated from a water-sensitive material of dye-doped PEG-DA hydrogel through two-photon polymerization. We demonstrate that as the environment humidity changes the microcavity can homogenously expand or shrink to keep the disk-shaped structure, producing stable WGM lasing emissions. We further show that the WGM wavelength of the microlaser shifts linearly as the humidity increases from 25% to 65%, resulting in a wavelength variation of about 1.72 nm, which indicates that the stretchable microlaser can be acted as a humidity sensor. Due to the "smart" response of humidity and biocompatibility of PEG-DA hydrogel material, our results provide a way to the design and fabrication of flexible PEG-DA hydrogel biochips for biomedical applications.**

***Index Terms*—Biosensor, humidity, hydrogel, microlaser, WGM.**

I. Introduction

WHISPERING Gallery Mode (WGM) microcavities that have a small mode volume and high quality(Q) can greatly enhance the light-matter interaction [1]–[6]. The evanescent wave around the periphery of microcavity is very sensitive to external perturbances that could lead to resonant wavelength shift or mode splitting. In the last two decades, WGM microcavities were extensively used for sensing, such as refractive index measurement and particle detection [7]–[11]. For example, a silica ring microcavity was used to measure the refractive index of a liquid [12], and a silica microsphere was employed to detect a single influenza virus [13]. However, these inorganic passive microcavities are not biocompatible, which greatly restrict their applications for biosensing. Besides, passive microcavities require embedded fiber tappers or prisms to introduce light and export signals, making the microcavity-based sensing systems complicated. Hence, nonimmune polymer microcavities, often doped with laser dyes, have been brought up as biosensors. For instance, a polystyrene divinylbenzene microcavity, doped with Firefli dye, was fabricated for intracellular tagging and tracking [14]. With rapid progress of polymer synthesis techniques, a vast of new polymer materials that can obviously change shapes under different stimulus have been synthesized and become promising for smart optical devices.

Hydrogel is one of the smart functional building materials in biological and medical science, which has been extensively used in the areas of stimulate actuator [15], drug encapsulation and release [16], [17], organ tissue engineering and microrobots etc. [18], [19]. So far, different types of hydrogels have been synthesized to have responsive to almost all stimulus in nature including water, PH value, temperature, force, light, electricity and so forth. Besides, hydrogels are transparent for light in the visible spectral range. Therefore, in this study we attempt to introduce hydrogels to be a new family of optical microcavity materials and show that the light confining ability of microcavity and the "smart" responsiveness of hydrogels could make hydrogel microcavities be good candidates for optical biosensors.

Recently, it was demonstrated that the synthetic poly (ethylene glycol) diacrylate (PEG-DA) hydrogel doped with photoinitiators allows fabrication of scaffolds and topographical patterns via polymerization by intense light irradiation, in which the acrylate groups in PEG-DA monomers can be cross-linked with each other to form a solid-state polymer [20]. On the other hand, femtosecond laser direct writing (FsLDW) that is upon two-photo absorption process has been proved to enable fabricating arbitrary complicate real 3D microcavities with relatively high spatial resolution and good morphology [21]–[28].

In this work, we choose rhodamine B-doped PEG-DA hydrogel as the photopolymerization material, in which the laser

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This paper has supplementary downloadable material available at http://ieeexplore.ieee.org. The video monitors the real time humidity responsiveness of the PEG-DA hydrogel microcavity and shows the variation of the microcavity in diameter under different humidity conditions. The size of this file is 18 MB.

Color versions of one or more of the figures in this paper are available online at http://ieeexplore.ieee.org.

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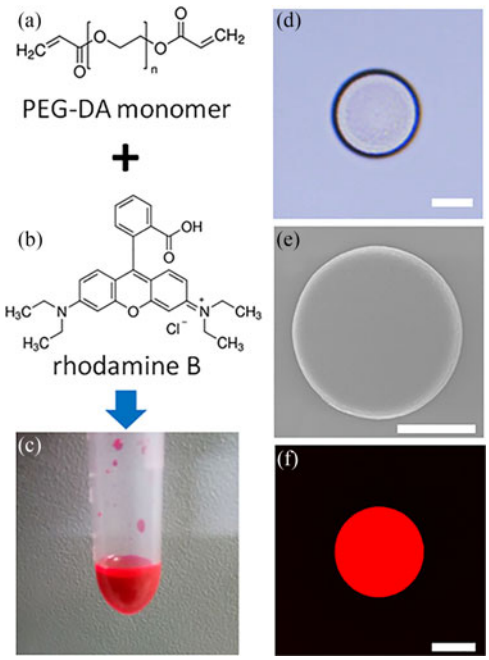

Fig. 1. Structural formulas of (a) PEG-DA monomer and (b) rhodamine B. (c) PEG-DA hydrogel photoresist composed of 100 μL PEG-DA monomer and 30 μL rhodamine B aqueous solution (20 mg/mL). (d) Optical microscope, (e) SEM, (f) two-photon fluorescence microscope images of the fabricated PEG-DA hydrogel microdisk. All scale bars are 10 μm.

dye rhodamine B serves as both the photoinitiator that triggers the formation of the polymerized structure and the gain medium that gives rise to the lasing emissions, and employ FsLDW as the microcavity processing technique to fabricate a disk-shaped WGM microcavity. We demonstrate that the dye-doped PEG-DA hydrogel microdisk possesses a relatively high-Q value of $\sim 2.8 \times 10^3$ and can produce stable WGM lasing emissions under different external conditions. Due to humidity-responsiveness of PEG-DA hydrogel, the hydrogel microdisk can swell or shrink as the environment humidity increases or decreases, resulting in obvious shifts of the WGM lasing modes. We find that the WGM wavelength of the microlaser changes as a linear function of the environment humidity, making the stretchable PEG-DA microlaser a potential smart optical biosensor.

II. Experiments

PEG-DA hydrogel monomer is a small molecule [average Mn of 700, Sigma-Aldrich, structural formulas shown in Fig.1(a)] and could not be "self-photosensitized" [18]. In this work, we chose rhodamine B laser dye [Sigma-Aldrich, structural formulas shown in Fig. 1(b)] as the photoinitiator molecule and prepared the photoresist. Firstly, 30 μL rhodamine B aqueous solution (20 mg/mL) and 100 μL PEG-DA monomer was mixed in a centrifuge tube. After shaken slightly, the photoresist mixture was placed into the water bath of an ultrasonic cleaner for about 10 minutes to mix homogeneously. 10 μL photoresist was dip-coated on a cover slide glass, which is cleaned by acetone, ethanol and deionized water in order. Since the rhodamine B laser dye was also served as the gain medium, a concentration balance of the mixture was, after tests, reached for the pre-polymer photoresist to have an accepted laser gain and a good FsLDW processing capability. Shown in Fig. 1(c) is the mixture sample that consisted of 100 μL PEG-DA monomer and 30 μL rhodamine B aqueous solution (20 mg/mL).

The sample was then placed on a 3-Dimensional stage for subsequent fabrication by FsLDW. A femtosecond laser beam from a Ti: Sapphire femtosecond oscillator (Mai-Tai HP, Spectra-Physics), characterized by a 100-fs pulse width, a center wavelength at 800 nm and an 80-MHz repetition rate, was tightly focused into the photoresist by an oil immersion lens (NA = 1.35, 100×, Olympus). After the laser irradiation, two-photon polymerization in the photoresist took place, in which rhodamine B dye molecules absorbed two photons and generate a large number of free radicals in the solution. The free radicals then attack PEG-DA monomers and induce a chain reaction process that transforms short monomers in liquid state to long polymers in solid state [20]. The sample was also illuminated by a halogen tungsten lamp, and the scattered light was collected by a CCD (DLC300) camera for monitoring the fabrication process. A 2-Dimensional galvanometer scanner and 1-Dimensional piezoelectric platform in the light path were combined to realize real 3-Dimensional scan of the laser focal spot in the photoresist sample. The laser power before the focused lens was about 20 mW and the exposure time is 500 μs. After fabrication, the sample was immersed in a mixed solution of ethanol and water with a volume ratio of 2: 1 for 10 minutes to remove unpolymerized photoresist.

This microcavity was designed to have a disk-shaped structure that has a radius of 10 μm and a thickness of 1.5 μm. The microdisk was located on a right circular cylinder supporter with a radius of 6 μm and a height of 4 μm, which can reduce the light leakage from the microcavity to the glass substrate. The morphology of the fabricated dye-doped PEG-DA hydrogel microcavity was characterized by an optical microscope, a scanning electron microscope (SEM, JSM-7500F, JEOL) and a two-photon fluorescence microscope, respectively, with the results shown in Fig. 1(d)–(f).

To control the surrounding humidity of the microcavity in the measurement process, we constructed a sealed chamber. A humidifier with an airway tube was linked to the chamber and employed for delivering water vapor into the chamber and increasing the humidity. A Silica-gel desiccant was used to decrease the relative humidity in the chamber. A hygrometer, used for external reference, was placed in the chamber to monitor the humidity in real time.

III. Results and Discussion

Shown in Fig. 1(e) is the scanning electron microscope (SEM) micrograph, from which it can be seen that the microdisk presents a good appearance and smooth surface for less scattering loss. The disk was measured to have a diameter of ~20.2 μm, showing the good fabrication capability of FsLDW for the prepared photoresist. Fig. 1(f) presents the two-photon fluorescence microscope image of the PEG-DA hydrogel microdisk, which shows a homogenous red-color distribution, indicating

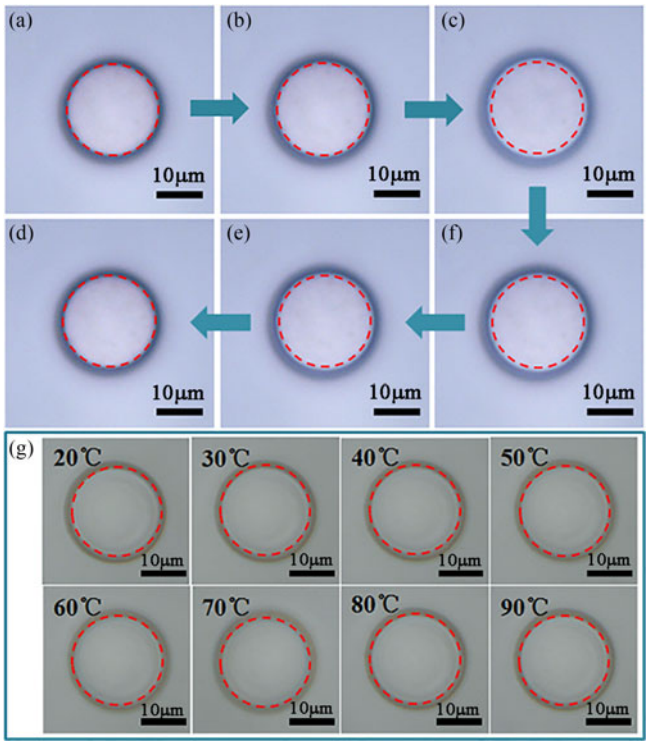

Fig. 2. The photo images of the PEG-DA hydrogel microdisk measured by a CCD camera under different humidity conditions: (a) microdisk before swelling, (b) mircrodisk being swelling, (c) microdisk after swelling, (d-e) microdisks being shrinking and (f) microdisk after shrinking. The red dashed circles are of the same size. The diameters of the microdisks in (a) and (f) are 20 μm. (g) The optical images of the PEG-DA hydrogel microdisk measured under different temperatures from 20 to 90 °C. The red dashed circles are of the same size. All scale bars are 10 μm.

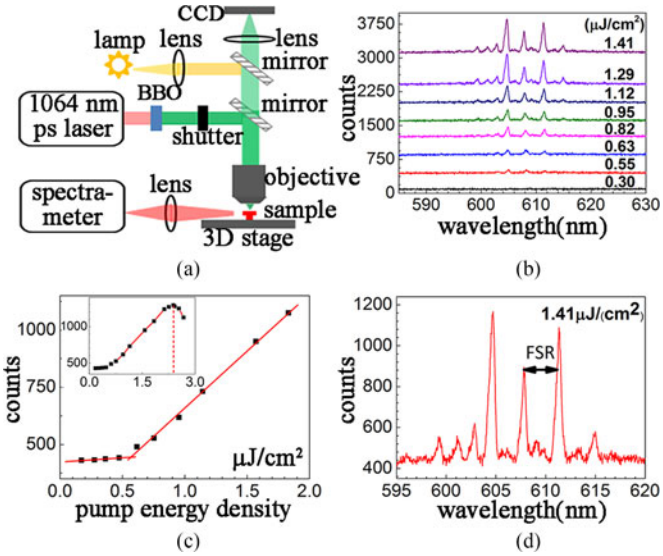

Fig. 3. (a) The schematic diagram of the optical pump and lasing spectrum measurement system. (b) WGM lasing spectra of the PEG-DA hydrogel microcavity under different pump energy densities. The spectra were shifted vertically for clarity. (c) The intensity of the 604.77 nm laser peak versus the pump power density. The inset shows the photobleaching effect of the active material. (d) Free spectral space (FSR) of microlaser determined from the lasing spectrum measured with the laser intensity of 1.41 μJ/cm2.

that the laser dye was homogenously doped into the PEG-DA hydrogel.

To exhibit the "smart" performance of the microcavity, we sprinkled water vapor from a humidifier onto the dye-doped PEG-DA hydrogel microdisk placed in an opened chamber, and observed the variation of microdisk in real time by the optical microscope. A video was taken with the CCD camera of the optical microscope to visualize the variation in the cavity diameter (see the supplementary material). As a result, we captured several graph photos of the microdisk at different times, and found, as shown in Fig. 2(a)–(c), that the microdisk swelled quickly and reached the expansion limitation in a few seconds after the water vapor was sprinkled onto it. The diameter of the microcavity shown in Fig. 2(c) was increased by about 7.6% when compared with that shown in Fig. 2(a). In this measurement, in order to monitor the diameter variation in real time, the microdisk was directly placed under a microscope, which was in an open environment, and thus the precise humidity could not be determined. The microdisk was then shrunken gradually after the water vapor was moved away, and recovered to its original size in about fifteen seconds [see Fig. 2(d)–(f)]. The results shown in Fig. 2(a)–(f) illustrate that the dye-doped PEG-DA hydrogel microdisk structure has a good humidity responsiveness, and is stretchable by absorbing/releasing water from/to the surrounding environment under different humidity conditions.

We have also investigated the temperature influence on the hydrogel microdisk. The microdisk was placed on a micro melting point apparatus that can heat the microdisk with a constant surrounding humidity. The diameter of the hydrogel microdisk was measured by the microscope. As showed in Fig. 2(g), the microdisk diameter keeps almost constant when the temperature changes from 20 to 90 °C, indicating that the diameter of hydrogel microcavity is insensitive to the temperature, that is, the influence of the temperature on the microcavity diameter can be negligible. Therefore, in the following, we only focus our attention on the experiments performed with a constant temperature of 25 °C.

Next, we show the lasing characteristics of the dye-doped PEG-DA hydrogel microdisk. As shown in Fig. 3(a), in order to pump the WGM microcavity, a picosecond laser pulse from a Nd: YLF laser (Amberpico-Q-2000, Guoke), characterized by a center wavelength of 1064 nm, a pulse width of 15 ps and a repetition rate of 50 kHz, was frequency doubled by a BBO crystal to produce the second harmonic with the wavelength at 532 nm. Then the laser beam was focused by an object lens (10×, NA = 0.25) onto the microcavity. After the optical pump, the optical emissions from the microcavity was collected by a convex lens with a focal length of 5 cm, and then focused onto the entrance slit of a spectrometer (SR303I-A, Andor), equipped with a CCD camera (DV420A-OE, Andor). The spectral resolution of spectrometer is about 0.2 nm. A halogen tungsten lamp was used to illuminate the sample and a CCD (DLC300) camera was adopted to monitor the pump process. To avoid bleaching effect of rhodamine B, a shutter with an opening time of 20 ms was placed in the pump laser beam path.

Fig. 3(b) shows the lasing spectra of the PEG-DA hydrogel microcavity with different pumping energies, from which it

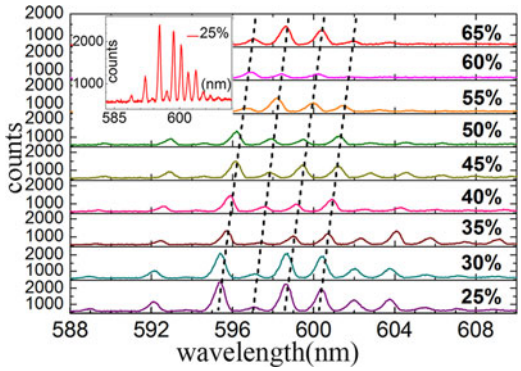

Fig. 4. Lasing spectra of the PEG-DA hydrogel microlaser obtained under different relative humidity from 25% to 65%. The black dashed lines represent the same lasing modes in the spectra under different relative humidities. The inset figure shows the same lasing spectrum measured with the humidity of 25%, showing the high signal-to-noise ratio with reliable detectability.

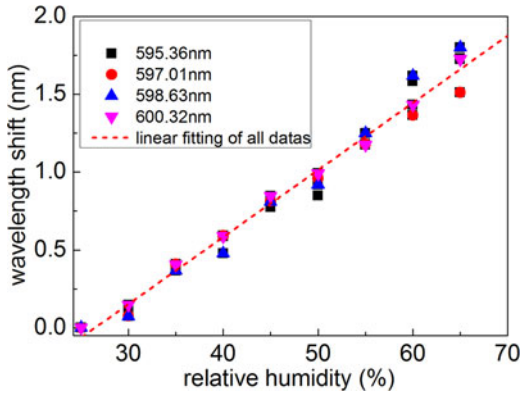

Fig. 5. The WGM resonant wavelengths shift versus the relative humidity. The lasing peaks at 595.36, 597.01, 598.63 and 600.32 nm correspond to the peaks in the 25% humidity spectrum shown in Fig. 4. The data is linearly fitted (pink dash line) with R2 = 98.16%.

can be seen that the lasing peaks become stronger as the pump power density increases. In Fig. 3(c), we plot the intensity of the 604.77 nm laser peak as a function of the pump laser energy, which exhibits an obvious lasing threshold behavior. The threshold power intensity of PEG-DA hydrogel microcavity is determined to be 0.56 μJ/cm^2. It can also be seen from the inset of Fig. 3(c) that as the pump power further increases, the lasing signal is saturated, and then decreases. This may indicate that the photobleaching of the active material occurs as the pump laser power intensity exceeds $\sim$ 2.38 μJ/cm^2.

To form a stable WGM oscillation, the resonant wavelength must satisfy the condition that the optical path for a round trip should be an integer multiple of the wavelength, described as $2\pi nr = m\lambda$, where n, r, m and λ refer to the refractive index, radius, mode number and resonate wavelength of the microcavity, respectively. As a result, free spectral range (FSR) of the WGM microcavity is defined as the spacing between two adjacent laser modes, which can be expressed by FSR $= (\lambda)^2/(2\pi nr)$. According to the spectrum shown in Fig. 3(d), the experimental value of FSR is determined to be $\sim$3.69 nm, which agrees well with the theoretical FSR value of 3.75 nm, when n = 1.55, r = 10 μm, and λ = 604.77 nm. The other important character of microcavity is the Q factor, which can be described as $Q = \lambda_0/(\lambda_2 - \lambda_1)$, where λ_0 is the center wavelength and λ_2, λ_1 refer to the wavelengths of full width at half maximum (FWHM) of a spectral line. According to the measured lasing spectrum and the instrumental broadening measured by a He-Ne laser, the measured lasing spectral profile can be deconvoluted, and the Q factor of PEG-DA hydrogel microdisk is determined to be about 2.8 $\times$ 10^3.

In order to demonstrate the characteristics of PEG-DA hydrogel microlaser as a humidity biosensor, we measured the variation of the WGMs spectra under different humidity conditions, as shown in Fig. 4. In the inset of Fig. 4, we present the lasing spectrum measured with the relative humidity of 25% to illustrate that the measured spectrum has a high signal-to-noise ratio with reliable detectability. In the measurement of humidty sensing characterisitcs, we placed the microcavity into a closed chamber to control the humidity surrounding the microcavity. The relative humidity in the chamber was increased by a humidifier and decreased by a silica-gel drier. As mentioned above, the PEG-DA hydrogel microcavity is sensitive to the humidity environment, and can swell/shrink as the surrounding humidity changes. Since the resonant wavelength is, according to the relation $2\pi nr = m\lambda$, linearly proportional to the radius of the microcavity, the change in the radius of the microcavity will lead to a shift of the WGM lasing peaks. It can be seen from the black dashed lines in Fig. 4 that as the relative humidity increases from 25% to 65%, all the peaks in the lasing spectra of microcavity are shifted to the red side, resulting in a wavelength variation of about 1.72 nm. It should be noted that the intensity variation of the modes may result from the variation of the microcavity diameter, and that no hysteresis effect has been observed for the microcavity humidity sensor.

To see closely the dependence of the wavelength shift on the relative humidity, we selected the lasing peaks at 595.36 nm, 597.01 nm, 598.63 nm and 600.32 nm from the spectrum measured at the relative humidity of 25% and depicted their changes as a function of relative humidity, as shown by the rectangle, circular, regular triangle and inverted triangle dots in Fig. 5 respectively. The data can be linearly fitted (see the red dash line), indicating that the shift of resonant wavelength is linearly proportional to the relative humidity in the range of 25% to 65%. This also indicates that the dye-doped hydrogel microcavity can swell/shrink homogenously as the relative humidity increases/decreases. Based on the above measurement, the sensitivity of the active microcavity humidity sensor is determined to be about 43.11 pm/%RH, representing that a degree of relative humidity gives rise to a wavelength shift of about 43.11 pm. This sensitivity is much better than the previously reported value (12.98 pm/%RH) obtained from a passive hybrid mcrocavity humidity sensor [29]. Clearly, these results show that the PEG-DA hydrogel microcavity can act as a humidity sensor, but it should also be pointed out that the measurable humidity range of the sensor could be limited by the moisture saturation of the hydrogel material.

IV. Conclusion

To summarize, we have fabricated an active microcavity of dye-doped PEG-DA hydrogels with two-photon polymerization by FsLDW, and demonstrated that under optical pump the microdisk can produce typical WGM lasing emissions with a low threshold of $0.56\,\mu J/cm^2$ and a high Q value of $\sim 2.8 \times 10^3$. We have further showed that the active microcavity enables to homogenously swell or shrink as the surrounding relative humidity increases or decreases, leading to the linear shift of the lasing peaks, which indicates that the stretchable microlaser can be acted as a humidity sensor. Since the PEG-DA hydrogel is biocompatable (nonimmune) and inactive with biological tissue, and rhodamine B molecules are encompassed by the polymerized hydrogel molecules in the solid phase, the rhodamine B-doped PEG-DA hydrogel sensor would not interact with other components that are present in biomedical environment [30]–[32], making the stretchable PEG-DA microlaser a potential smart optical biosensor, for example, for monitoring breath air at the mouth and measuring sweat moisture from the nostrils in the physiological examination and diagnosis [33]. However, it is clear that for practical applications of the sensor, much effort is needed, such as integrating the sensor with certain compact fiber laser pump source as well as with optical waveguide for optical collection and detection. Due to the real 3D fabrication ability of FsLDW, our results provide a way to the design and fabrication of complicate 3D hydrogel microcavity chips for biomedical science and applications.

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