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Egg white based biological microlasers

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Abstract

Biolasers made of biological materials hold great potential for implantable biosensing and cell-tracking. However, the current bio-extracted materials used for biolasers are generally required complicated synthesis process and therefore suffer from high cost. In this work, we demonstrate that low-cost natural egg white is an excellent biomaterial for a laser cavity. Using a simple dehydration method, dye-doped goose egg white microspheres are obtained with various sizes from 20 to 160 μm in diameter. These microspheres can act as excellent laser sources under optical excitation with lasing threshold of ~26 μJ/mm^2 and quality (Q) factor up to 3×10^3. The lasing mechanism is studied and ascribed to whispering gallery mode. Size-dependence of lasing spectrum and Q factor are also investigated. Owing to the ease of fabrication, the cost-effectiveness, goose egg white based microlasers are promising for biosensing and bioimaging applications.

Keywords: biolasers, microlasers, whispering gallery mode, egg white

1. Introduction

Biolasers whose gain medium and/or cavity made of biological materials is potential for cell-tracking, bioimaging and next generation biosensing applications [1-4]. Research on biolasers started five decades ago when T. W. Hansch utilized gelatin (a protein produced by the hydrolysis of collagen) as a laser cavity [5]. To date, various bio-extracted materials have been explored for biolasers such as DNA, bovine serum albumin (BSA), silk fiborin, green fluoresecent protein (GFP) [6-10]. GFP can be used as a gain material while the other materials used for a cavity matrix. However, these biomaterials often require complicated synthesis and therefore process with a high cost [11].

To compensate the high cost of bio-extracted materials, several ideas have been proposed with promising results such as the use of biocompatible synthetic polyvinyl alcohol (PVA) [12] or natural materials like starch from potato as a laser cavity [13]. Regarding these two materials, starch is lower cost and always available. However, starch also has some drawbacks such as poor mechanical properties and medium optical transparency which would affect the quality of laser cavities [14,15]. As a result, exploring other alternative natural biomaterials that are appropriate for biolasers is highly necessary.

Egg white (EW) is a natural biomaterial that has been used as a common nutritious food for a long time but it has not explored for biolasers. Compared with BSA protein and PVA polymer, egg white is supposed to have lower cost and easier to access. In general, EW composes of 88.5% water and 10.5% protein (including ovalbumin, lysozyme, ovotransferrin, ovomucin, ...) and demonstrates antimicrobial activities [16-19]. Furthermore, ovalbumin, the main protein of EW, has a high refractive index, about 1.55 at 600 nm [20], which is higher than that of PVA (around 1.45) [12] and starch polymer (1.53) [21]. As a result, EW can have a high refractive index which is crucial for making high-quality (Q) factor cavities [20,22].

In this work, we demonstrate, for the first time, the use of goose egg white (GEW) as a novel material for biolasers. By using a simple fabrication technique, dye-doped GEW microspheres are obtained. Under optical pulse excitation, these microspheres emit lasing emission with excellent properties.

2. Materials and methods

2.1 Preparing aqueous dye-doped egg white solution

A goose egg was purchased just one day after being laid. GEW was chosen for the laser cavity because it has higher transparency compared with that of other eggs such as chicken egg. GEW was separated from the yolk using a stainless steel sieve (Figure 1a). Then, dye-doped GEW solution was prepared by magnetically stirring (for 5 minutes, at room temperature) a mixture contains 3 ml GEW and 1 mL aqueous solution 2.5 wt% of Rhodamine B (RhB, > 95% dye, from Sigma-Aldrich).

2.2. *Fabrication of microsphere biolasers from dye-doped egg white solution*

Dye-doped GEW microspheres were fabricated by dehydration in a solvent [23] as shown in Figure 1b [24]. Firstly, 5 ml of 1-decanol (> 99% purity, from Sigma-Aldrich) was poured into a glass beaker where a Teflon substrate was placed at the bottom. The hydrophobicity of Teflon keeps droplets in spherical shapes which is vital for the formation of solid microspheres. Secondly, a micropipette was used to create a droplet (containing dye-doped GEW solution) on the Teflon substrate immersed in 1-decanol. Next, the droplet was dispersed into smaller droplets, the so-called microdroplets, by using a tiny needle. As 1-decanol is immiscible in water, these microdroplets are self-assembly on the Teflon substrate by surface tension. To increase the dehydration rate of water from microdroplets into 1-decanol, the whole mixture was heated to about 80°C and keep at this temperature for 6 minutes. This process dehydrates all water molecules from the microdroplets and leads to the formation of solid microspheres (Figure 1b). Finally, the obtained microspheres were taken out and subsequently heated at 100°C for 4 minutes to completely remove the 1-decanol on their surface. After cooling down to room temperature, these microspheres are ready for optical characterizations. In addition, as egg white based microspheres has been used for drug delivery [25], the current fabriation technology may also be applied for this kind of application.

2.3. Optical characterizations

We used a micro-photoluminescence (μ-PL) setup to study dye-doped GEW microspheres. The pumping source was a Nd:YAG nanosecond pulse laser (Litron Lasers) with a wavelength of 532 nm, a repetition rate of 10 Hz and a pulse duration of 7 ns. The microspheres were excited by a focus laser beam with the spot size of ~ 350 μm in diameter. Emission from the microspheres was collected by a 10× objective and subsequently delivered to an AvaSpec-2048L (Avantes) for spectral recording. The spectral resolution is ~0.2 nm. All optical characterizations were carried out in the ambient air and at room temperature.

3. Results and discussion

3.1 Dye-doped egg white microspheres

Figure 2 shows the optical microscope and scanning electron microscope (SEM) images of dye-doped EW microspheres with diameter from about 20 to 160 μm. From the optical microscope (Figure 2a), it can be seen that all microspheres exhibit round shape with a uniform red colour which confirms the good incorporation of dye molecules into the GEW. Spherical curvature of these microspheres is more evident in the SEM image (Figure 2b). A high magnification SEM image of a single microsphere indicates a smooth surface (Figure 2c) which is substantial for gaining high optical confinement and low lasing threshold.

3.2 Lasing characteristics of a typical egg white based microsphere biolaser

The obtained dye-doped EW microspheres can work as efficient laser sources under optical excitation. Figure 3a plots emission spectra of a single microsphere (about 43 μm in diameter estimated from the microscope image) under increasing pump pulse energy (PPE). Each spectrum was excited by a single pulse. At a low PPE of 1.6 μJ, the emission is characterized by the spontaneous emission with low intensity and broad spectrum. Under higher PPE of 2.72 μJ, lasing modes start to appear with wavelengths from 610 to 620 nm and a clear free-spectral range (FSR) of around 1.84 nm is observed. The intensity of these lasing mode increases sharply with increasing PPE to 5.32 μJ. In addition, under this PPE, new lasing modes are now visible. These modes are suggested

to have a different polarization compared with the original one (will be discussed later). Figure 3b shows the integrated PL intensity from the microsphere as a function of PPE, indicating a distinct laser threshold of about 2.5 μJ. The threshold is equivalent to a fluence of 26 μJ/mm^2 which is comparable with dye-doped polymer microsphere lasers [12] and significantly lower than that of starch-based biolasers [13]. The lasing threshold depends on many factors including the dye concentration thus a lower lasing threshold can be obtained with an optimized dye concentration.

Lasing mechanism is ascribed to whispering gallery mode (WGM) and the position of lasing modes can be calculated by using the explicit asymptotic formula [26]. For a WGM laser, FSR = $\lambda^2/\pi nD$, where λ is the lasing wavelength, n and D are the refractive index and diameter of the microsphere laser. As FSR and lasing wavelength can be measured from a lasing spectrum and D = 43 μm thus the refractive index can be calculated to be around 1.491. Furthermore, lasing modes can be with transverse electric (TE) and transverse magnetic (TM) modes depending on the orientation of electric field oscillation [27]. As shown in Figure 4a, the spectrum displays two separated lasing envelope, the one with higher intensity is TE modes (due to higher Q factor) while the one with lower intensity is TM modes [28]. Assuming D = 43 μm and n = 1.491, the TE and TM modes are fitted well with fundamental mode number of 312-319. To better confirm the WGM mechanism, TE lasing modes of microspheres with different sizes are compared with theoretical calculations and the result indicates an agreement between the experimental observation and WGM theory (Figure 4b).

In addition, we have studied several lasers under long term pumping and the result indicates that the lasing operation is quite stable. For instance, a 52 μm-diameter microsphere continues lasing for as many as 3×10^3 excitation pulses. It is also worth noting that all the lasing properties were measured in air. Currently, our biolasers does not work in an aqueous environment. This issue has to solve before applying these biolasers for biosensing applications.

3.3 Size-dependent lasing characteristics of egg white based microsphere biolasers

Size-dependence of lasing characteristics is investigated. Figure 5a plots lasing spectra of three different microspheres showing a red-shift of lasing wavelength with increasing microsphere diameter from 34 to 82 μm. This effect has been observed and can be explained by the out-coupling efficiency in which a longer lasing wavelength is easier to scatter out a microsphere with a larger size [29]. In addition to the red-shift of lasing wavelength, the FSR decreases with increasing the microsphere diameter. The change of FSR can be well explained by the expression FSR = $\lambda^2/\pi nD$, where λ is the lasing wavelength, n and D are the refractive index and diameter of the microsphere. Based on that equation, it is expected that FSR would be a linear function with the inverse of microsphere diameter (1/D). This hypothesis is verified in Figure 5b where FSR of 13 microspheres with diameters ranging from 34 to 160 μm is plotted versus 1/D. The result confirms that WGM is responsible for the observation of lasing emission from dye-doped GEW microspheres.

The Q factor is an important parameter of a laser cavity that describes the optical confinement. A high Q factor cavity is desired for a low lasing threshold and ultrasensitive optical sensing [30]. As shown in Figure 6c, the Q factor (calculated as Q = $\lambda/\delta\lambda$) of microsphere lasers increases from 2050 to 3100 with increasing microsphere diameter from 34 to 160 μm. The obtained Q factor is comparable with that of protein based microsphere lasers [9,24] and about three-time larger than that of the starch based lasers [13]. The Q factor increases gradually with increasing microsphere size because larger microspheres confine light better than smaller ones. The highest Q factor can be detected is 3150 (at 630 nm) due to the spectral resolution (0.2 nm) of our setup. In addition, due to lower Q factor, it is more difficult to observe lasing emission from small microspheres. The smallest lasing microsphere measured has a diameter of around 30 μm.

4. Conclusion

We have demonstrated that egg white is an excellent biomaterial for a laser cavity. By doping dye molecules into GEW and using a simple but effective method, dye-doped GEW microspheres with diameter ranging from 20 to 160 μm are fabricated. Under optical pulse excitation, these microspheres can work as efficient lasers with lasing threshold as low as 2.5 μJ or 26 μJ/mm^2 for a 43 μm-diameter microsphere. The Q factor of these microsphere lasers is proportional to its diameter and can be reached to 3×10^3 for a 160 μm-diameter microsphere. Size-dependence of lasing characteristics is studied showing a good agreement with the WGM theory. Our microsphere biolasers are very promising for biosensing and bioimaging applications.

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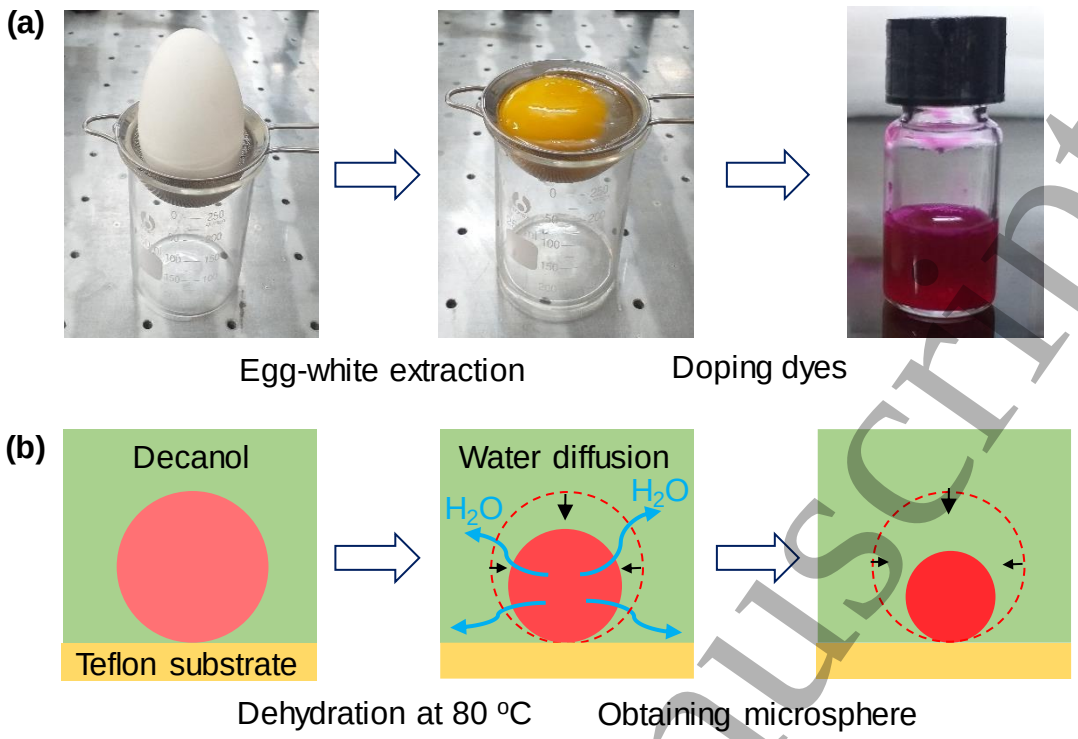

Fig. 1. Illustration of the fabrication process of dye-doped GEW microsphere.

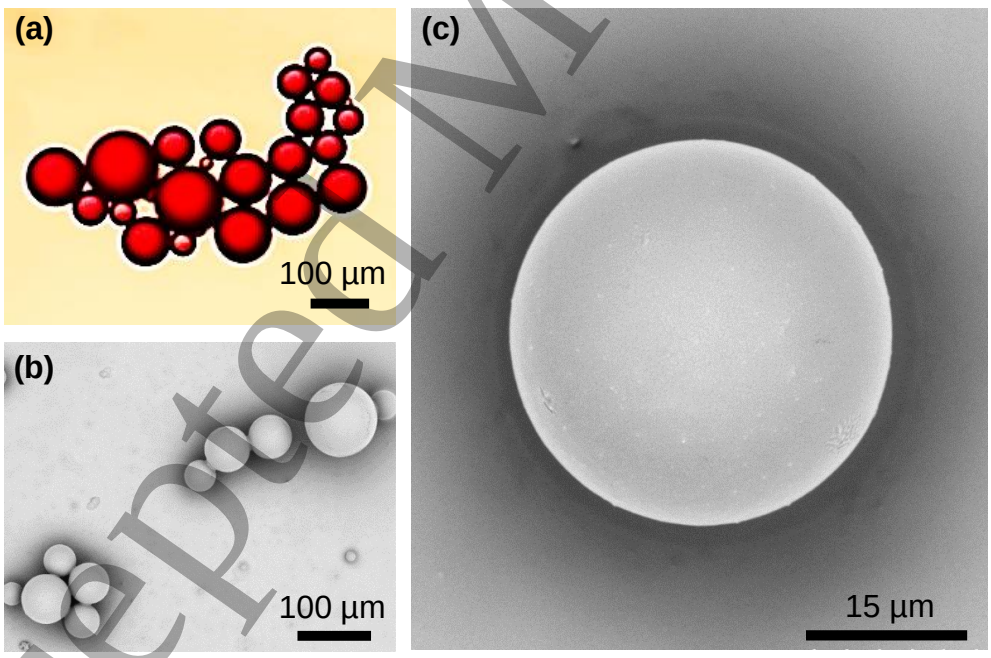

Fig. 2. Optical and SEM images of dye-doped GEW microspheres.

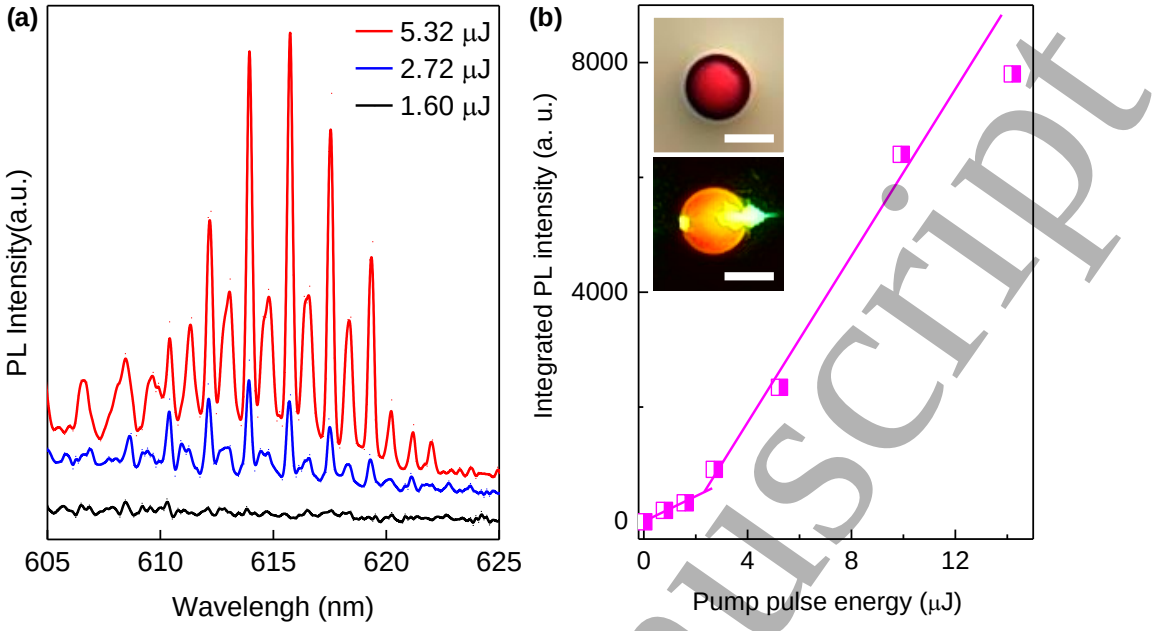

Fig. 3. a) Emission spectra from a single 43 μm-diameter microsphere under various pump pulse energies. b) Integrated PL intensity of the microsphere laser as a function of pump pulse energy. The scale bars are 50 μm.

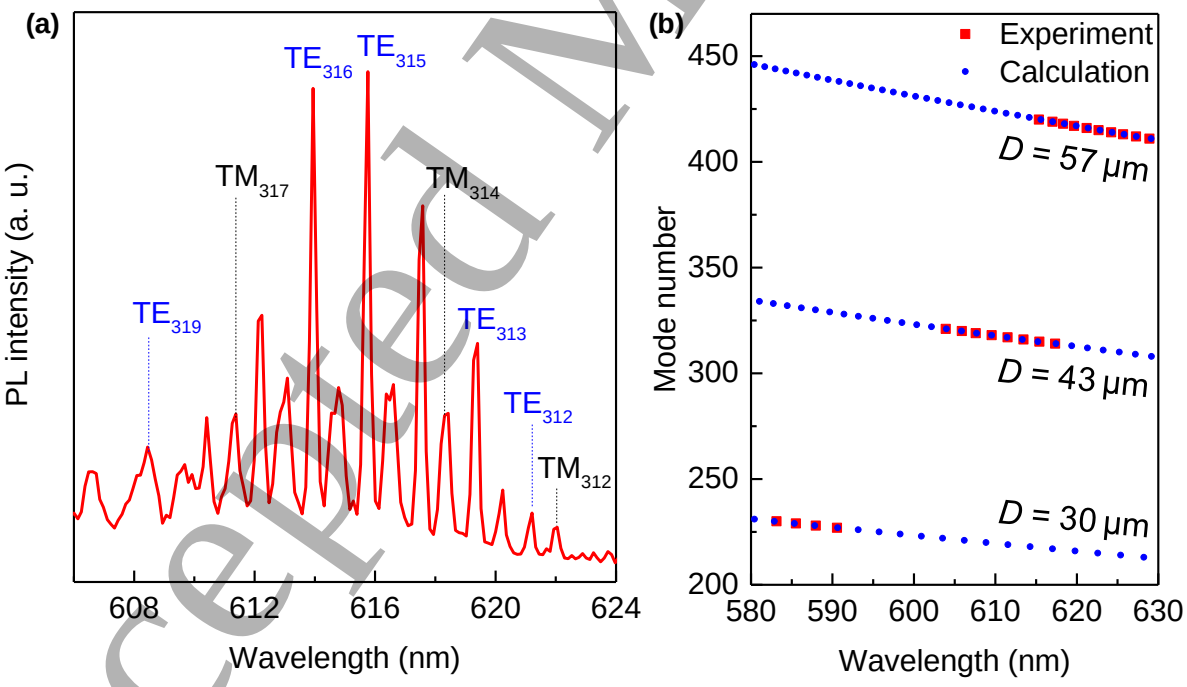

Fig. 4. a) Mode analysis of the lasing spectrum of the 43 μm-diameter microsphere. b) Agreement between theoretical calculation and experimental measurement of TE lasing modes of three different microspheres.

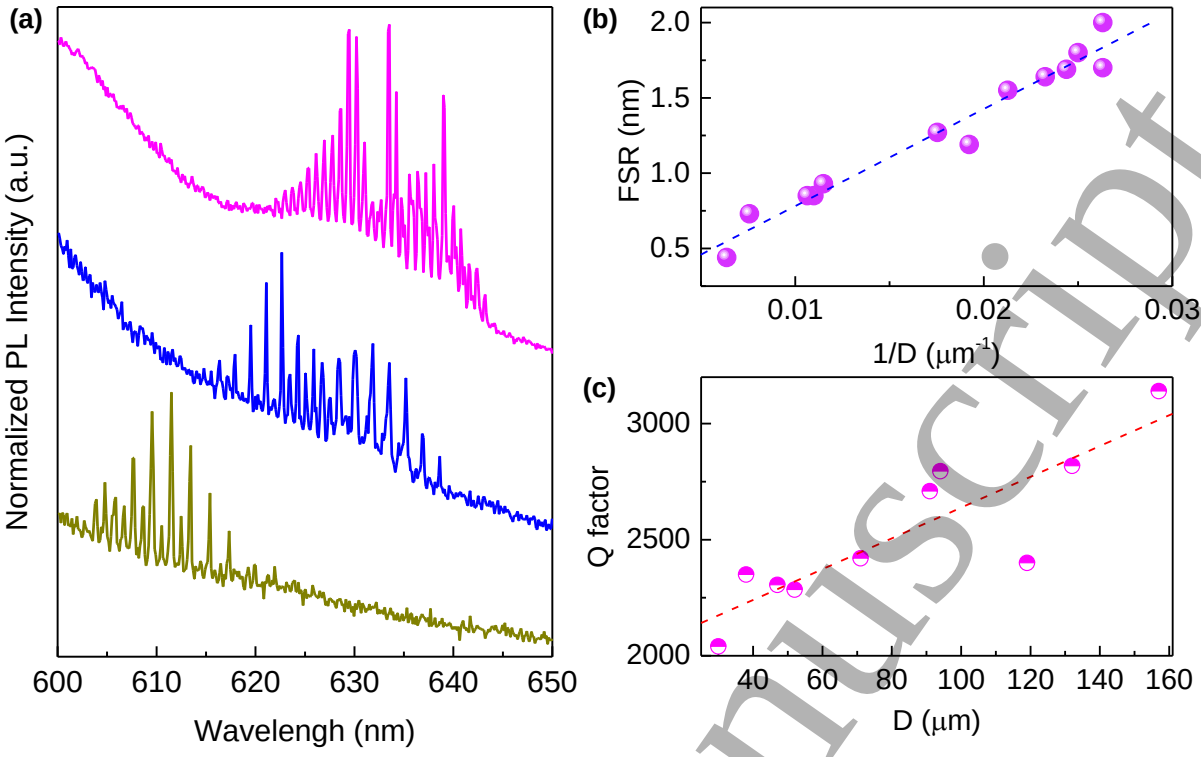

Fig. 5. a) Lasing spectra of microspheres with diameter of 34, 48 and 82 μm (from low to high), respectively. b) FSR of various microspheres as a function of their inverse diameter. c) Q factor of microsphere lasers versus their diameter.