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Fluorescence Characteristics of Random Gain Media Fabricated from Fluorescein Dye Containing Zinc Oxide Nanoparticles in Transparent Organic Hosts

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Abstract

In this work, ZnO nanoparticles with different weights (0.01-0.09 mg) were incorporated into a solution of 6×10^{-6} M Fluorescein dye dissolved in transparent resin in order to fabricate random gain media. The fluorescence and gain characteristics of these media were introduced when excited with a 405 nm laser source. The addition of ZnO nanoparticles to the Fluorescein dye dissolved in a transparent resin host leads to an observable enhancement in the fluorescence intensity of random gain media fabricated from such systems. The active role of ZnO nanoparticles as scattering centers can be observed by the enhanced optical feedback and optical gain of these media. The ZnO nanoparticles do not change the electronic structure of the Fluorescein dye and their effects specifically lie on scattering and re-absorption processes. The weights used in this work confirm that the random gain media did not cause quenching due to aggregation or excess absorption, therefore, significant losses were avoided. The accurate control of nanoparticles concertation in the random gain medium is an effective tool to control the emission or fluorescence characteristics to serve the practical purposes for which such random lasers are used.

Keywords: Random gain media; Fluorescein dye; Zinc oxide; Fluorescence

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1. Introduction

In the recent decades, laser physics has witnessed a drastic development by transforming from the conventional designs based on resonant cavities and mirrors to much more flexible and complex designs, one of them is the random laser, which is a photonic system produces optical feedback by multiple scattering within an inhomogeneous medium instead of the conventional optical cavity (resonator). This concept has established wide horizons in new applications such as sensing, biomedical imaging, advanced lighting, and optical encryption [1-5]. Random laser shows simple construction, low production cost, capability to incorporate into flexible, transparent, and bioactive materials. It can be simply constructed by a gain medium that exhibits optical gain within a scattering medium that provides complex optical paths leading to the amplified spontaneous emission (ASE). This system can reach to the threshold under specific conditions to produce output light beam with narrow lasing peaks, like the conventional laser [6-8]. The random laser provides much more free design than the strict resonant cavities in conventional lasers, which may produce laser sources with different forms, such as thin films, porous materials, and transparent polymers. It also shows high sensitivity to the environmental variations, which makes it suitable for chemical and biological sensing, as the changes in refractive index, scattering or absorption result in observable changes in emission characteristics [9-11].

The organic dyes was intensively studied and employed as gain media in random laser design due to their high gain coefficients, high absorption cross sections, and easy incorporation in different media. Fluorescein dye is one of the most common in random laser design – and generally in dye lasers – as it shows high quantum yield, high absorption in violet-blue region, wide emission in green-yellow region, relatively chemical stability, and easy dissolving and incorporating in polymeric or resin media [12-15]. However, the performance of random laser not only depends on the gain medium, but it reasonably depends on the characteristics of the scattering medium, where the nanoparticles represents the most powerful tool to control scattering, optical path, and random feedback. This control is performed by the accurate selection of type, size, shape, and concentration of nanoparticles to satisfy Rayleigh scattering condition. Zinc oxide (ZnO) nanoparticles are commonly used in random lasers due to wide bandgap, high transmittance in visible region, refractive index suitable for effective scattering without significant absorption, chemical stability, and capability to be prepared with various sizes [16-20]. Many works demonstrated that incorporating ZnO nanoparticles in organic dye solutions has resulted in drastic increase in emission intensity and lowering laser threshold due to the enhancement in the optical feedback and increase in the effective optical path length [21-25]. The nanoparticles can contribute to the local field enhancement around the dye molecules that increase the probabilities of absorption and emission, and hence increase the optical gain [26].

In a system of organic laser dye and nanoparticles hosted in a completely transparent resin, all advantages mentioned above for the random gain media can be provided when the distribution of both dye and nanoparticles is kept homogeneous as much as possible since this is a fundamental to avoid the aggregation that may cause undesired losses such as nano-radiative quenching or excess absorption [27,28]. Consequently, such photonic systems can be used in many and various applications such flexible photonic sensors, encryption systems depending on random emission modes, imaging devices requiring low-coherence light, etc. [29-31]. They also can be developed to produce tunable random lasers based on dye concentration, type of nanoparticles, host characteristics. Such tunable sources can be effectively integrated in nanophotonics and metamaterials techniques to precisely control the light paths and feedback.

In this work, ZnO nanoparticles with different weights were incorporated into solid solution of Fluorescein dye dissolved in transparent resin in order to fabricate random gain media. The fluorescence and gain characteristics of these media were introduced to show their feasibility as random lasers in the visible region.

2. Experimental Part

The Fluorescein laser dye solution was prepared by dissolving 0.01 mg of dye in 5 mL of the transparent resin (epichlorohydrin). The molar concentration (C) of the dye sample was calculated by the following equation [32]:

$$C = \frac{1000 \times w}{MW.V} \tag{3}$$

where MW is the molecular weight of the dye (332.3 g/mol), V is the volume of the solvent (mL), and w is the weight of the dye. The molar concentration of the dye sample was 6.0×10^{-6} M

The random gain media were prepared by adding varying weights (0.01, 0.03, 0.05, 0.07, and 0.09 mg) of ZnO nanoparticles with average size of 12 nm to the dye solution with continuous mixing and stirring to ensure uniform and homogeneous dispersion of the nanoparticles within the solution.

In order to produce as much as possible homogeneity of dye and nanoparticles distribution in the resin host, the mixture was sonicated in an ultrasonic bath for 30 min before the hardener is added to the mixture and manually stirred for 5 min. The samples were then placed inside a water bath at 50°C for 5 min to induce solidification reactions in the resin host and to remove air bubbles from the mixture. The mixture was then poured into cylindrical molds and left in a clean desiccator at room temperature for 24 hours to obtain the final solid shape as rods with a diameter of 1 cm and a height of 4 cm, as shown in Fig. (1).

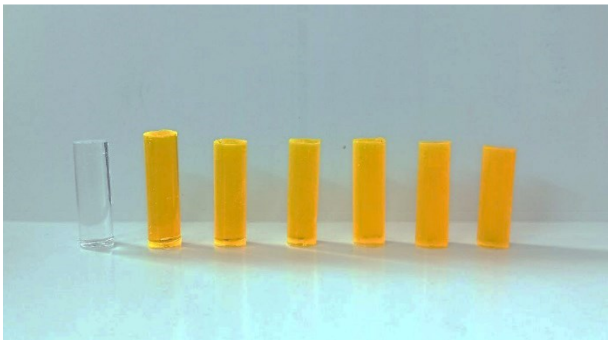


Fig. (1) The random gain media fabricated in this work with different weights of ZnO nanoparticles (from left to right: resin only, 0, 0.01, 0.03, 0.05, 0.07, 0.09, mg)

The spectroscopic measurements (absorption and fluorescence) were carried out on the fabricated samples using a Spectra Academy KMAC SV2100 spectrophotometer in the spectral range of 200-1000 nm. Fluorescence spectra were recorded as the samples were excited by a 405nm semiconductor laser source. The spectral scattering width (W) and anisotropy parameter (<cosθ>) were measured to calculate the gain coefficient (g) per unit length for the fabricated samples.

3. Results and Discussion

Figure (2) shows the fluorescence spectra of the fabricated samples when excited with a laser source operating at 405 nm, whose spectrum is apparently seen as sharp peak in this figure. The spectral response of the random gain media starts at 540 nm to a peak at 560-580 nm. The sample containing dye only shows a relatively low fluorescence intensity because the quantum yield of Fluorescein dye in the resin is limited by re-absorption, non-radiative relaxation, or reaction between dye and resin matrix. when the ZnO nanoparticles were added with small weights (e.g., 0.01 mg), the fluorescence intensity was clearly increased due to the role of these nanoparticles as strong scattering centers that increase the optical path length of photons within the gain medium. Therefore, the probability to absorb and re-emit the scattered light by the dye is increased. With increasing the weight of the ZnO nanoparticles in the sample from 0.03 to 0.05 mg, a gradual increment in the fluorescence intensity is observed, which confirms that the optical gain was enhanced due to the achieved balance between dye concentration, scattering coefficient, and effective optical path length. In addition to their role as scattering centers, the nanoparticles can support the light-dye matching throughout local field enhancement. With 0.07 mg of nanoparticles in the sample, the fluorescence intensity continues slowly to increase, which may indicate that the system is near to a partial saturation state as the number of produced photons becomes large enough to induce losses, such as self-absorption or nan-radiative collisions, to compensate the increase due to scattering.

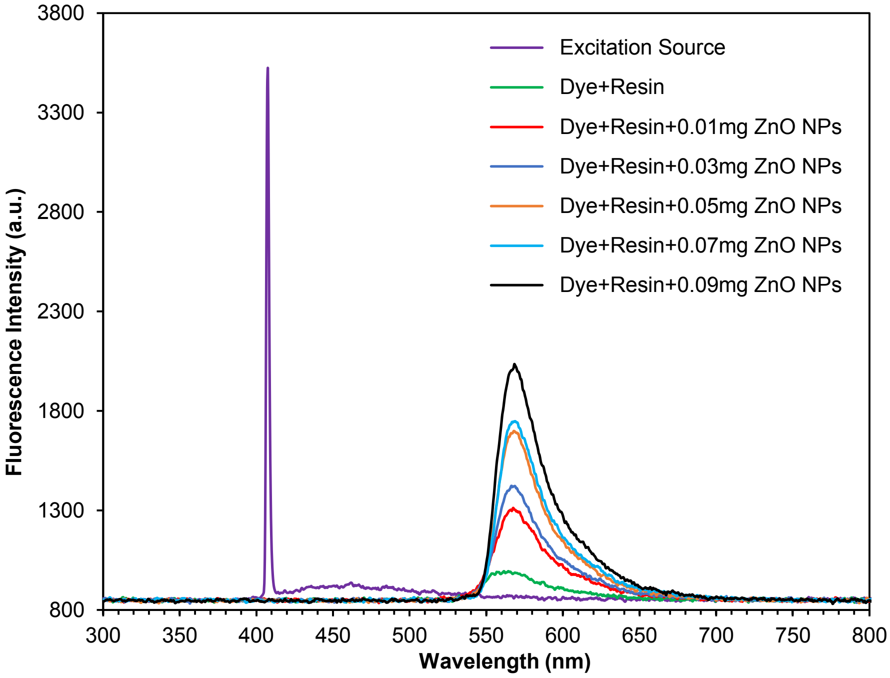
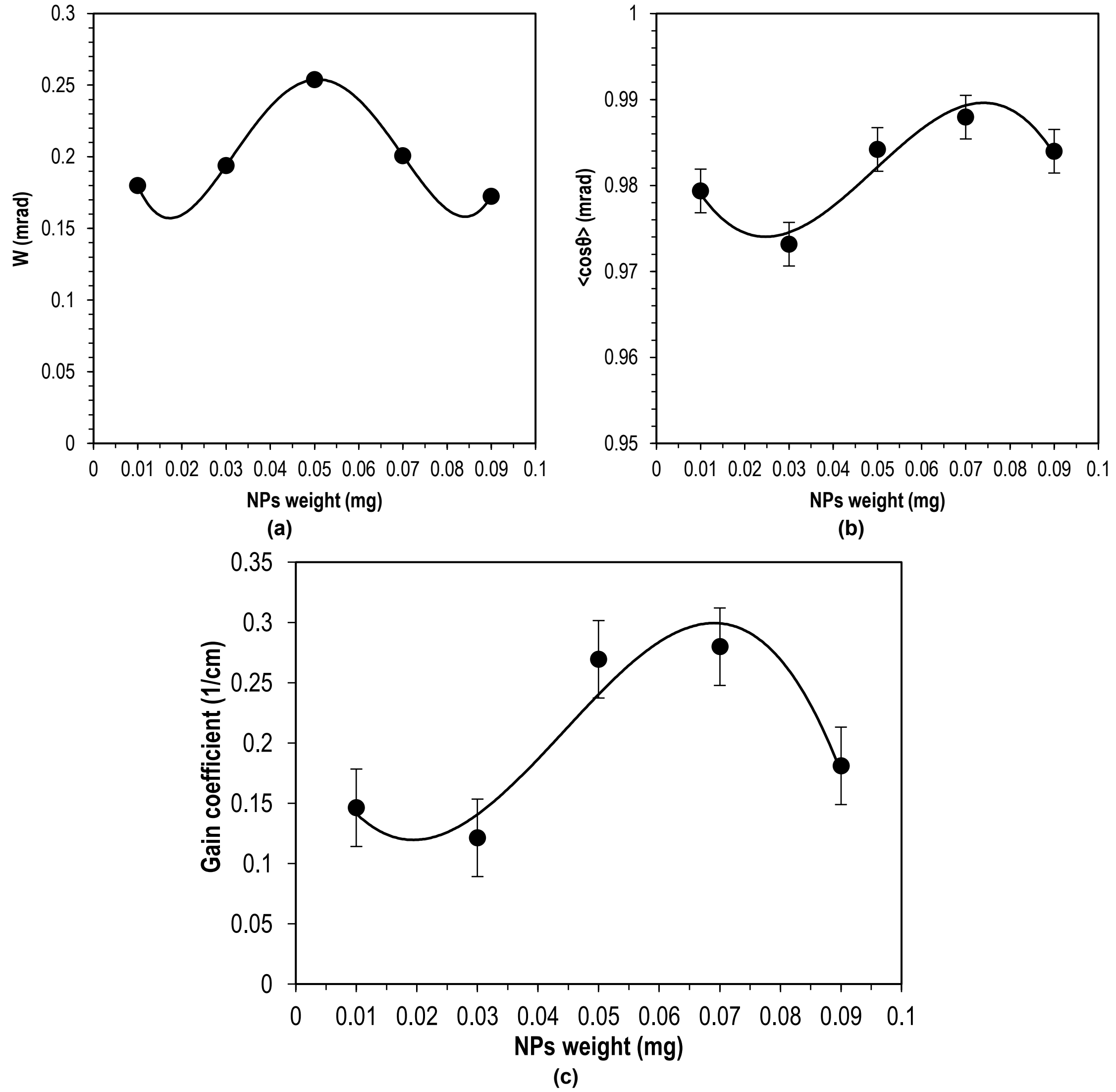


Fig. (2) Fluorescence spectra of the random gain media fabricated in this work with different weights of ZnO nanoparticles

With 0.09 mg of nanoparticles in the sample, the highest intensity was recorded with the widest spectrum, which reveals that the system has reached the optimum regime of random lasing gain without significant negative effects of aggregation quenching or excess absorption by ZnO nanoparticles themselves. The increase in the spectrum width refers to a broadband fluorescence nature rather than sharp laser linewidth. This is well expected due to the absence of a limited resonant cavity whereas the multiple scattering provides the required random feedback. Also, since the peak wavelength was approximately constant with varying concentration of ZnO nanoparticles, this reveals that these nanoparticles did not cause changes in the electronic levels of fluorescein dye, but, instead, affect scattering, path length, and re-absorption probability, i.e., they did not cause a significant spectral shift but only an intensity modulation. This can be attributed to the fact that the reaction between ZnO nanoparticles and Fluorescein dye molecules is physical rather than chemical.

The background noise can be considered low as the baselines in all spectra were very close far from emission peaks. Accordingly, the increase in the fluorescence intensity was actually caused by the effect of nanoparticles not variations in measurement or excitation source intensity. Also, the role of ZnO nanoparticles as scatterers was attributed to its wide bandgap and low absorption in the visible region, therefore, no decrease in fluorescence intensity was observed with increasing the concentration of these nanoparticles in the sample. This observation is very important to explore the random laser threshold at higher excitation energies or when the random gain medium are modified by changing sample thickness, resin type, or dye concentration, to achieve narrow lasing peaks instead of the current wide emission.

Figure (3) shows the results of measuring the spectral scattering width (W), anisotropy parameter (<cosθ>), and gain coefficient per unit length (g) for the fabricated random gain media. Figure (3a) shows the variation of the spectral scattering width (W) with the weight of ZnO nanoparticles added to the solid solution of the Fluorescein dye, as a nonlinear dependence is observed. At low weights, the scattering is relatively weak as the number of scatterers is small. With increasing the weight, the number of scatterers within the medium is increased and hence the spectral scattering width is accordingly increased and a maximum is seen at 0.05 mg. At larger weights, the value of W decreases due to the effect of agglomeration of nanoparticles or the re-absorption and optical losses, those reduce the effective random scattering.



(a) (b)

(c)

Fig. (3) Variation of (a) the spectral scattering width (W), (b) anisotropy parameter (<cosθ>), and (c) the gain coefficient per unit length (g) for the random gain media fabricated in this work

In Fig. (3b), the variation of the anisotropy parameter (<cosθ>) with the weight of the ZnO nanoparticles is shown as the converging values of <cosθ> confirm the dominance of the front scattering event. The initial addition of ZnO nanoparticles caused a slight decrease in <cosθ> due to the increase in angular and random scattering. At larger weights, the values of <cosθ> gradually increase due to the balance between the front and multiple scattering events. Further increase in the weight of the nanoparticles causes a decrease in <cosθ> as the medium was much more guided to front scattering due to the increasing effective density or the agglomeration, which both limit the random optical feedback. Figure (3c) shows that the gain coefficient per unit length of the random gain media increases with increasing weight of the ZnO nanoparticles to reach a maximum value at 0.07 mg, beyond which, the gain coefficient clearly decreases. This behavior agrees with the concept of the scattering cross section, which strongly depends on the nanoparticle radius and relative refractive index. So, the increase in nanoparticles concentration enhances the stimulated scattering, increases the light path length within the medium, and hence raise the effective gain. The excess increase in the weight (concentration) leads to additional losses due to absorption, agglomeration, and reduced pumping efficiency, which interprets the final decrease in the gain coefficient. The maximum values of W, <cosθ>, and g – observed at 0.07 mg – confirm that the optimum multiple scattering has the most effective role in optical gain within the random gain medium.

4. Conclusion

According to the results obtained from this work, the addition of ZnO nanoparticles to the Fluorescein dye dissolved in a transparent resin host leads to an observable enhancement in the fluorescence intensity of random gain media fabricated from such systems. The active role of ZnO nanoparticles as scattering centers can be observed by the enhanced optical feedback and optical gain of these media. The ZnO nanoparticles do not change the electronic structure of the Fluorescein dye and their effects specifically lie on scattering and re-absorption processes. The weights used in this work confirm that the random gain media did not cause quenching due to aggregation or excess absorption, therefore, significant losses were avoided. The accurate control of nanoparticles concertation in the random gain medium is an effective tool to control the emission or fluorescence characteristics to serve the practical purposes for which such random lasers are used.

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