

Effect of ZnO Nanoparticles Weight Content on Lasing Threshold of Fluorescein Dye-Based Random Lasers

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Abstract

In this work, ZnO nanoparticles with different weight contents (0.01, 0.03, 0.05, 0.07, and 0.09 mg) were incorporated in solid solutions of fluorescein laser dye dissolved in epichlorohydrin resin to fabricate random gain media. The effects of ZnO nanoparticles weight content on the pumping threshold and emission intensity were introduced. Lower contents showed higher threshold levels whereas the higher contents has decreased the threshold level due to increasing the scattering centers and hence amplification of random laser signal. This attempt is required to optimize the design parameters of random lasers based on fluorescein/ZnO nanoparticles nanocomposites.

Keywords: Random gain media; Emission threshold; Fluorescein dye; Nanoparticles

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1. Introduction

Random lasers are a unique category of laser sources that do not need for a conventional resonant cavity composed of two mirrors, instead, they invest the multiple scattering of light within an inhomogeneous colloidal or porous medium to provide the feedback required for the optical amplification of laser signal [1]. The main principle of this category of lasers is based on trapping photons by random closed paths within a gain medium. When this medium is optically or electrically pumped to exceed the threshold, a laser with distinct spatial and temporal features is emitted and these feature depend on the structure of the scattering medium [2,3].

The significance of the random lasers in practical applications can be highlighted by their simple fabrication and small sizes to be used in biosensing, biomedical imaging, on-chip optical communications, anti-fraud coatings, and development of flexible and implantable laser sources [4-7]. They also open new trends to study the random propagation physics as well as the interaction between light and matter in disordered systems. This makes them promising tools in modern optics and nanophotonics research [8,9].

In general, nanoparticles represent the cornerstone in the development of modern optical and photonic applications due to the phenomena of quantum confinement, amplification of surface photonic fields, and hence increasing photonic state densities, to provide a superior power to control light at the nanoscale [10]. Zinc oxide (ZnO) nanoparticles can be an ideal material for these applications, including fabrication of random lasers, due to their wide direct energy band gap (3.37 eV), high exciton binding energy (60 meV), which enable them to keep stimulated emission at high temperatures [11,12]. These nanoparticles show high ability to scatter light due to their high refractive index [13]. They also show good ability to be prepared with various nanostructures (rods, wires, flowers, spheres), which allow fine control of the random feedback paths within the laser medium [14]. Furthermore, ZnO nanoparticles can be easily prepared, very low toxicity, and biocompatibility to be optimum candidate for implantable random lasers or biosensors [15]. Consequently, ZnO nanoparticles can be successfully used to produce random lasers with low threshold, high efficiency, and controllable via changing particle size or concentration.

Fluorescein dye is one of the most optimum dyes for laser fabrication, especially the random lasers [16]. This dye shows reasonable interference between its absorption and emission spectra as its absorption peak is located at the green region of electromagnetic spectrum and extended to interfere with its fluorescence spectrum whose peak is usually located within 514-523 nm. This interference is the main parameter that determines the behavior of the random laser due to the self-absorption of the photons emitted at shorter wavelengths by the dye molecules at the ground level, which hinders their

amplification and pushes the system to emit at longer wavelengths as no absorption occurs [17-19]. Moreover, fluorescein dye shows moderate quantum efficiency and limited fluorescence lifetime those directly affect the optical gain coefficient of the lasing medium and threshold [20].

When used to fabricate random laser, the interference between absorption and emission spectra of fluorescein leads to some unique phenomena. At high concentrations, the laser spectrum may show bichromatic emission with two modes (or peaks), yellow and red. These two peaks compete with increasing pumping power as the induced amplification gradually supports the mode with higher gain. Understanding these spectroscopic characteristics and mutual dependencies of absorption and emission is crucial to control the behavior of fluorescein dye random laser, either to achieve monochromatic narrow band emission or to control the emission via changing the dye concentration of the nature of hosting medium [21-23].

Nanocomposites fabricated by incorporating nanoparticles, such as ZnO, in solid solutions of organic dyes can be considered as a qualitative transfer in modern optical, spectroscopic and photonic applications. The secret of this integration is in the investment of two synergetic phenomena: multiple scattering by the nanoparticles and effective optical gain of dyes for specific wavelengths. Therefore, these nanocomposites allow the fabrication of optical devices throughout one-touch coating techniques on different substrates [24].

In this work, the effects of ZnO nanoparticles content in fluorescein dye solid solution on the pumping threshold and emission intensity were introduced as these nanoparticles were used as scatterers within random gain media fabricated from nanocomposites based on these materials.

2. Experimental Part

In order to fabricate random gain media, shown in Fig. (1), ZnO nanoparticles were incorporated into the Fluorescein dye dissolved in transparent epichlorohydrin resin. These nanoparticles were characterized to introduce their structural and morphological characteristics. The crystalline structure of these nanoparticles were introduced X-ray diffraction (XRD) using a Malvern Panalytical AERIS diffractometer in the diffraction angle scan from 20° to 80°. The surface morphology of the ZnO nanoparticles was introduced by field-emission scanning electron microscopy (FE-SEM) using an FEI Inspect F50 instrument, which is equipped with an energy-dispersive X-ray spectroscopy (EDX) unit to perform elemental analysis.

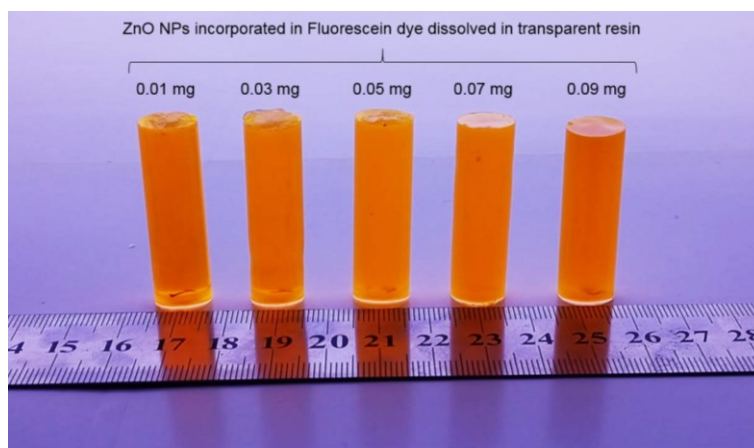


Fig. (1) A photograph of the random gain media fabricated in this work by incorporating different weights of ZnO NPs in Fluorescein dye dissolved in transparent epichlorohydrin resin

A 0.01 mg of Fluorescein dye was dissolved in 5 mL of epichlorohydrin resin, which is completely transparent in the spectral range 300-800 nm. This is important to prevent any contribution to the spectroscopic events included in this experiment. The following equation was used to determine the molar concentration (C) of the dye sample [25]:

$$C = \frac{1000 \times w}{MW \cdot V} \quad (1)$$

Here, MW represents molecular weight of Fluorescein dye ($=332.3$ g/mol), V is the solvent volume (mL), and w is the Fluorescein dye weight. Consequently, the molar concentration was 6.0×10^{-6} M

Different weights of ZnO nanoparticles (0.01, 0.03, 0.05, 0.07, and 0.09 mg) were mixed with the dye solutions to fabricate random gain media. The mixture was sonicated for 30 min before poured in the cylindrical molds to ensure uniform and homogeneous dispersion of the nanoparticles within the solution as well as complete dissolving of dye in the solvent (epichlorohydrin). The mixture was kept at 40°C to allow the reactions leading to solidification to initiate. Finally, the mixture was poured in the molds and left in a clean and dry place at room temperature for 20 hours to take its final shape. The final sample is yellow-orange cylinder of 1 cm diameter and 4 cm height, as shown in Fig. (1).

To determine the behavior of the threshold pumping power density as a function of the content of the ZnO nanoparticles in the random gain media, an 405nm laser source was used to pump these media with variable power densities and the laser-induced fluorescence (emission) intensity was measured using a KMAC SV2100 digital-controlled spectrophotometer as the sample was placed inside the cuvette of this instrument, the light source was replaced by the laser source, and the emitted signal was detected by the CCD detector of the spectrophotometer.

3. Results and Discussion

Figure (2) shows the XRD pattern of the ZnO nanoparticles used in this work. The apparent peaks seen at 31.91°, 34.51°, 36.38°, 47.71°, 56.65°, 63.09°, 66.53°, 68.00°, 69.23°, 72.64°, and 77.05° are corresponding to the crystal planes of (100), (002), (101), (102), (110), (103), (200), (112), (201), (004), and (202), respectively, according to the JCPDS card 36-1451. They express the hexagonal Wurtzite structure of ZnO, which is the most common and stable at normal conditions. The structural purity of these nanoparticles can be revealed since no peaks belonging to free Zn or other dopants or contaminants were observed. This purity is highly necessary for spectroscopic applications of nanomaterials, such as random gain media, because the spectroscopic activity of these nanoparticles should be well-known to recognize it from that of the dye as these nanoparticles are solely used as scattering centers.

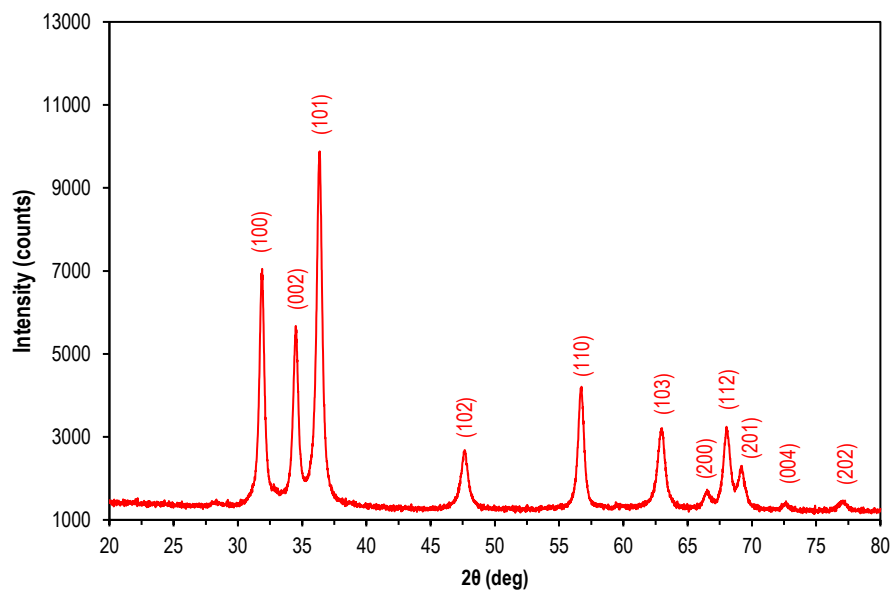


Fig. (2) XRD pattern of the ZnO nanoparticles used in this work

Figure (3) shows FE-SEM images of the ZnO nanoparticles used in this work at two different magnifications (4 kx and 16 kx). Spherical particles with size ranging in 20-100 nm can be seen in the first image with tend to aggregate due to their high surface energies. However, overall homogeneity can be confirmed in the second image with sufficient coverage of the substrate's surface and low density of pores and voids. This morphology is reasonably required for the application of random gain media to avoid any irregular interaction with the incident photons.

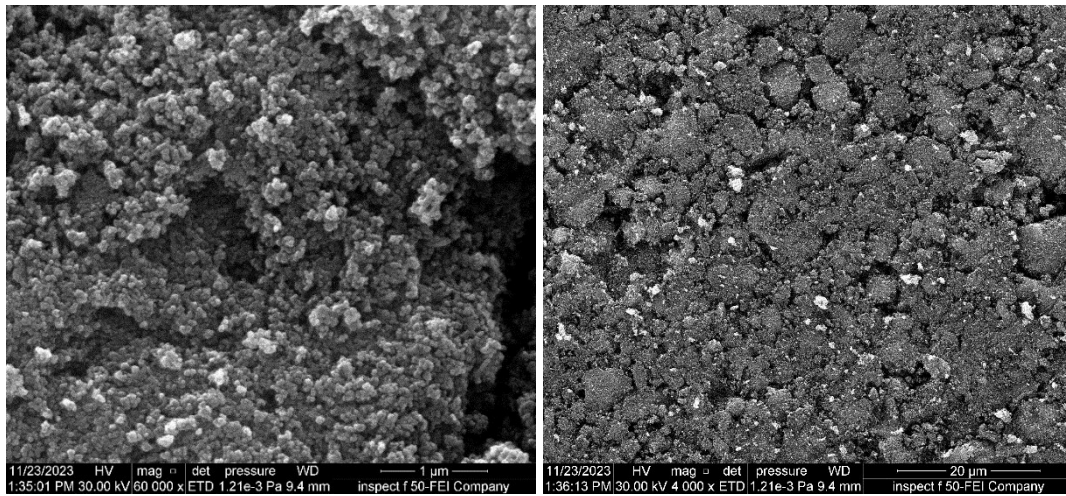
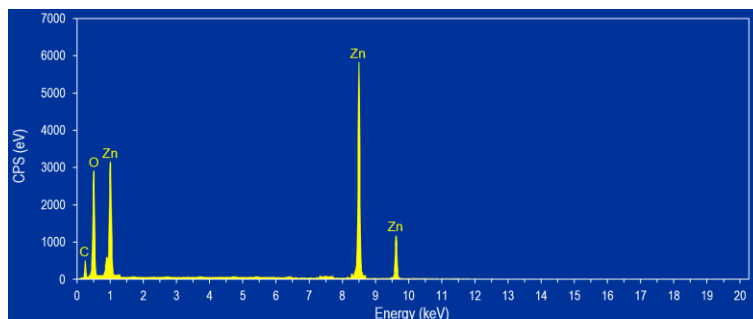


Fig. (3) FE-SEM images of the ZnO nanoparticles used in this work

Figure (4) shows the EDX spectrum of the ZnO nanoparticles used in this work. As an accurate tool to confirm the structural purity of these nanoparticles, this spectrum shows only peaks for Zn and O as the constituents of ZnO whereas the existence of carbon is inevitable due to the maintaining method of sample inside the EDX equipment. Higher peaks of Zn when compared to that of O is natural due to the difference in the dispersion cross section between two elements since oxygen is a light element and not easily detected by the EDX. According to the elemental analysis given in the table, the ZnO compound exhibits high stoichiometry (0.52:0.47), which reveals the efficiency of the preparation method employed to prepare ZnO nanoparticles.



Element	Atomic %	Atomic % Error	Weight %	Weight % Error
C	26.3	0.7	9.9	0.3
O	38.9	0.5	19.5	0.3
Zn	34.8	0.2	70.6	0.4

Fig. (4) EDX spectrum and elemental analysis of the ZnO nanoparticles used in this work

Figure (5) shows the relationship between emission intensity and pump power density of the random gain media fabricated in this work. In general, all curves show very weak response to pumping at low pump levels ($<1.5 \text{ mJ/cm}^2$), then a slow increase within the pumping range of $1.5\text{-}2 \text{ mJ/cm}^2$, and then a sharp jump in the emission intensity was measured beyond 2 mJ/cm^2 . This behavior represents a distinct feature of the random lasing emission as the medium converts from weak spontaneous emission to condensed amplified emission when the threshold pumping power density is reached. At pump level of 50 mJ/cm^2 , the increase in emission intensity got slower as the gain medium enters the saturation region. The threshold value was found to lower as the content of ZnO nanoparticles was increased. This behavior is attributed to the fact that the lower contents of nanoparticles do not provide sufficient optical gain as the concentration of scattering and amplification centers is low, while higher contents of nanoparticles provide higher concentrations of scattering centers to perform multiple scattering required

for amplification of random laser signal. However, further increase in nanoparticles content may cause much more scattering and re-absorption losses that decrease the emission intensity despite increased pumping power. These results may reveal that the contents of ZnO nanoparticles used in this work are useful to fabricate random gain media since no irregularities were observed.

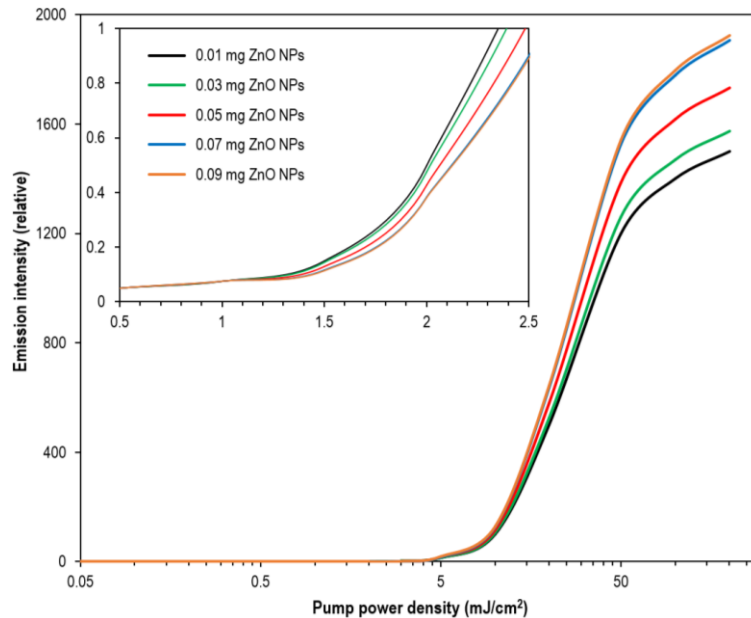


Fig. (5) Relationship between emission intensity and pump power density for the samples fabricated in this work

4. Conclusion

In conclusion, ZnO nanoparticles with different weight contents (0.01, 0.03, 0.05, 0.07, and 0.09 mg) incorporated in solid solutions of fluorescein laser dye dissolved in epichlorohydrin resin can be successfully to fabricate random gain media with emission in the green region of electromagnetic spectrum. Lower contents of ZnO nanoparticles showed higher threshold levels whereas the higher contents has decreased the threshold level due to increasing the scattering centers and hence amplification of random laser signal.

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