Journal of Physical Science, Vol. 34(2), 29–40, 2023

Random Laser Emission from Fiber coated ZnO

Abdullah Taha Ali,[1] Wan Maryam Wan Ahmad Kamil,[1*] Sheng-Chan Wu,[2] Chung-Xian Yang,[2] Hsu-Cheng Hsu,[2] Faisal Rafiq Mahamd Adikan,[3] Ghafour Amouzad Mahdiraji[3] and Fairuz Abdullah[4]

[1]School of Physics, Universiti Sains Malaysia, 11800 USM, Pulau Pinang, Malaysia
[2]Department of Photonics, National Cheng Kung University, 70101 Tainan, Taiwan
[3]Flexilicate Sdn Bhd, Faculty of Engineering, Universiti Malaya
50603 Kuala Lumpur, Malaysia
[4]Department of Electrical and Electronics Engineering, College of Engineering, Universiti Tenaga Nasional, 43000 Kajang, Selangor, Malaysia

[*]Corresponding author: wanmaryam@usm.my

Published online: 25 August 2023

To cite this article: Ali, A. T. et al. (2023). Random laser emission from fiber coated ZnO. *J. Phys. Sci.*, 34(2), 29–40. https://doi.org/10.21315/jps2023.34.2.3

To link to this article: https://doi.org/10.21315/jps2023.34.2.3

ABSTRACT: *Random lasing from a solid-state gain medium prepared on photonic crystal fibre (PCF) is observed for the first time. Vertically aligned ZnO microrods were prepared on PCF using a simple technique of chemical bath deposition (CBD). A low lasing threshold of 12.2 mJ/cm^2 was observed in sample with longer zinc oxide (ZnO) rod length. The variation in morphology and population density did not affect the lasing threshold significantly. Further investigation of the effect of fiber length revealed that a shorter fiber had a lower threshold and showed quenching of the spontaneous emission revealing better lasing output. Simulations based on the morphology of the gain medium revealed light confinement in the structure, validating the origin of the lasing emission. Overall, this study shows the potential of utilising optical fiber as random lasers with a sustainable solid state gain medium.*

Keywords: nanotechnology, energy efficient, laser materials, photonics, environmental sustainability

1. INTRODUCTION

Optical gain and multiple scattering of light produced by random-structured media set the basis for a range of phenomena, including amplified spontaneous emission and random lasing.[1–3] A random laser works differently from a traditional laser such that feedback for amplification is achieved by disorder-induced scattering. As such, having enough photons to participate in the scattering event and amplify (by means of a gain medium) is crucial to induce lasing. However, random lasing is an open cavity laser where achieving a low threshold has always been an ongoing problem. Therefore, vast amount of research was focused in designing a random laser that can achieve low threshold, compact and compatible with different applications. Various methods have been proposed to reduce the threshold in these zinc oxide (ZnO) based random lasers such as by optimising the surface morphology of ZnO nanorods, optimising the annealing conditions in ZnO nanorods, using a high concentration of ZnCdSeS/ZnS alloyed quantum dot-doped polymer-dispersed liquid crystals, forming defect pits in thin films, coupling with Fabry-Perot resonance modes induced in nanocolumns, utilising excitons, enhancing optical confinement formed by tapered nanowires and fluorescence resonance energy transfer coupled with light scattering.[4–12] However, all the above methods refer to growth or synthesis of ZnO on substrates that cannot be integrated with current applications such as fibre-based sensing system. In this work, we wish to highlight the possibility of utilising optical fibres as potential random lasers that can reduce the challenge of integrating random lasers within an optical or electrical system.

Random lasing emission from photonic crystal fibre (PCF) has been a topic of interest lately for a range of applications such as optofluidic sensing, speckle-free imaging and ultra-sensitive biosensing.[13–19] These random lasers use dyes as the gain medium to typically produce a broad lasing spectral width between 10 nm to 30 nm—which is comparable to fluorescence-based applications and are subject to degradation of the laser dye. Non-PCF based random lasers using a solid-state gain medium have shown able to provide narrow emission lines.[20–22] Hence, degradation issues may be eliminated by employing a solid-state gain medium.

In this paper, we demonstrate random lasing with narrow spectral width from ZnO microstructures prepared on PCF by a simple technique of chemical bath deposition (CBD). Changes in lasing properties were compared between two different fibre lengths. In addition, structural and morphological characteristics of the gain medium were also analysed. Further investigation on light confinement based on actual field emission scanning electron microscopy (FE-SEM) images of the samples were done to evaluate structural effects affecting light propagation. Results suggest the possibility of developing PCF random laser devices with ZnO structures as the gain medium.

2. EXPERIMENTAL

The PCF used in our experiment is specially designed by Flexilicate Sdn. Bhd. using a conventional stack-and-draw method. The fiber preform is drawn into around 2 mm diameter canes. The 2 mm cane is then re-drawn into standard fibre with outer diameter of 125 μm. The PCF created is illustrated in Figure 1(a). The gain medium (ZnO structures) was prepared by two-step CBD. First, a 100 nm thick ZnO seed layer was deposited on the outer PCF by radio frequency magnetron sputtering. Constant power and argon gas flow rate were at 150 W and 15 sccm at room temperature, respectively. The working pressure in the growth chamber is 4.63×10^{-3} Torr. Then, aqueous solutions of 0.05 M Zinc Nitrate [$Zn(NO_3)_2.6H_2O$] and 0.05 M Hexamethylenetetramine ($C_6H_{12}N_4$) were prepared. Each solution was stirred separately for 30 min and then combined before stirring for another 15 min, then poured into a glass bottle. Two sections of PCF, 3 cm and 6 cm long, were taped on a glass slide for support and then was immersed into the solution. The bottle was tightly sealed and then placed in an oven for 4 h at 96°C. The samples were labeled Sample 1 (short PCF) and Sample 2 (long PCF). The same process was repeated one more time but only with short PCF and the sample was labelled Sample 3. For clarity, fibres that were deposited with just ZnO seed layer was also tested for lasing emission.

The morphology was investigated by FE-SEM (Nova Nano SEM 450, FEI, Japan) and evaluated using ImageJ software. Confirmation of elements from the sample were obtained from energy-dispersive X-ray spectroscopy (EDX) located within the FE-SEM. Random lasing measurements were performed using a custom-built micro-photoluminescence (PL) system with a Nd:YAG pulsed laser source operating at 355 nm, 1 KHz rep rate and 350 ps pulse width. The PCF were fixed on a translational stage with one end free standing such that the portion of fibre that is under excitation was surrounded by air. The excitation light was focused on the samples through an objective lens (10×) with a spot size of about 50 μm. The lasing emission was recorded by a spectrometer (JY iHR320) equipped with a liquid-nitrogen cooled charge-coupled device (CCD) array detector. All measurements were performed at room temperature and is depicted in Figure 1(b). To estimate light propagation in the structure, a finite-element approach was utilised using a commercial solver software: COMSOL Multiphysics™.

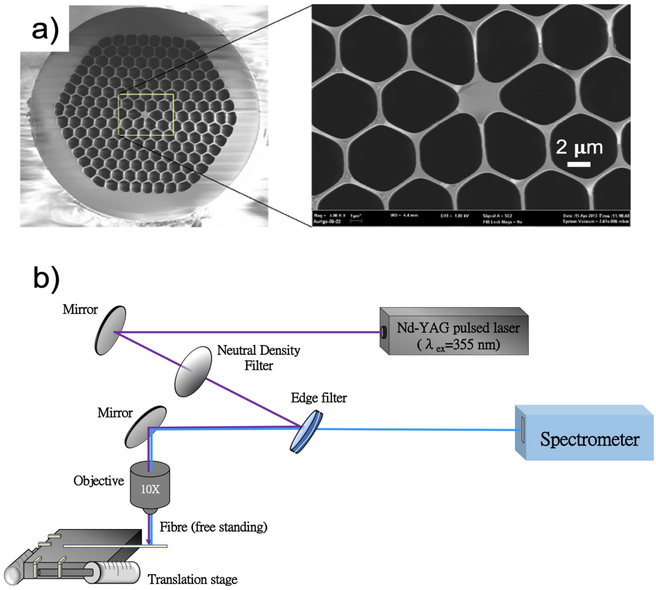
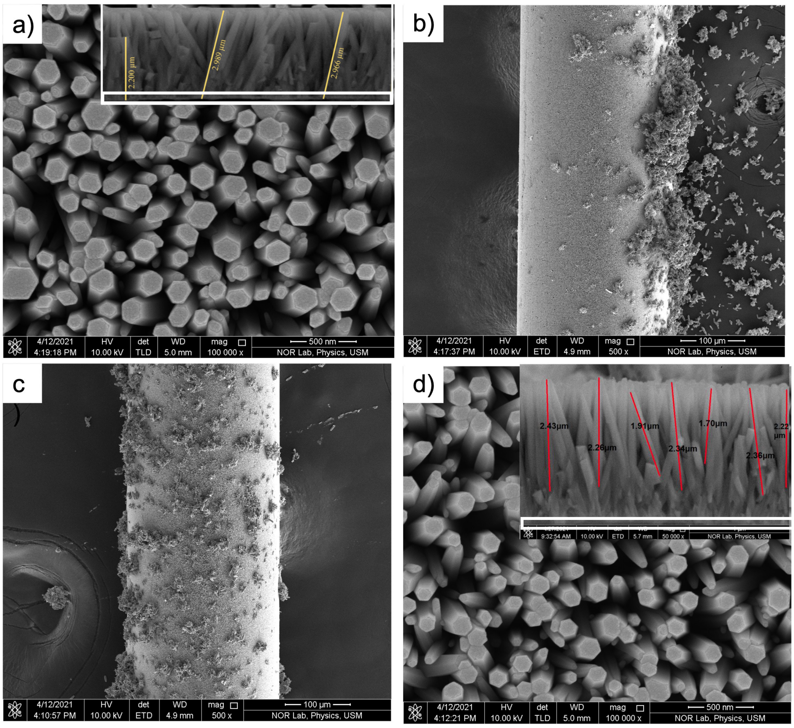


Figure 1: (a) Structure of the PCF, (b) The PL setup for random lasing measurement: The pump light was focused using a 10× objective onto free standing fibre placed on a translational stage.

3. RESULTS AND DISCUSSION

Top view of the ZnO microstructures prepared on PCF is shown in Figure 2. The seed layer can be seen at the bottom of the inset in Figure 2(a) and Figure 2(d) where it was deposited onto the PCF prior to CBD. The seed layer aids the growth of vertical rods. Figures 2(a) and Figure 2(b) were from Sample 1 whereby the inset in Figure 2(a) shows the cross section of ZnO revealing an average height of 2.81 m and average rod diameter of 195 nm. A zoomed-out image of the fiber depicted by Figure 2(b) reveals a clustered and packed ZnO structures on a surface of the outer fibre. Figure 2(c) shows a more even distribution of the ZnO structures which is obtained from Sample 3. A closeup of Sample 3 reveals ZnO microrods with average rod diameter and height of 160 nm and 2.17 m, respectively. Sample 1 has a more clustered area of growth and Sample 3 has a more uniform area of growth. Small difference in the size and distribution is expected in CBD synthesis technique.[23] The effect that this has on the lasing capabilities are discussed next.



. Figure 2: FE-SEM images of the gain medium, ZnO microstructures, prepared on optical fibres where (a) is top view image at 100k magnification with inset showing the cross section of the microrods for Sample 1, (b) is top view of the fiber for Sample 1, (c) top view of fibre for Sample 2 and (d) top view of the microstructures with inset showing the cross section of the microrods for Sample 2.

Elements from the sample were confirmed by EDX measurements and the percentage of elements are summarised in Table 1. Majority of elements are Zn and O elements. Small amounts of carbon (C) and nitrogen (N) are also detected which is a byproduct of the synthesis. The chemical equation governing the synthesis is as follows:

$$2(CH_2)6N_6 + 3Zn(NO_3)2.6H_2O \rightarrow 3ZnO(s) + 14NH_3 + 12CO_2 + 9H_2O$$

Table 1: EDX results showing atomic percentage of elements from the rods in Sample 1.

Atomic (%)			
C	N	O	Zn
16.19	8.89	39.53	35.39

Upon pumping the fibres with a pulsed light source operating at 355 nm (1 kHz rep rate), random lasing emission was observed. Lasing output obtained from Sample 1 is shown in Figure 3. Lasing threshold was observed at about 12.2 mJ/cm^2 with linewidth of about 0.282 nm. The number of lasing modes increased with increasing pump fluence—a conventional signature of random lasing.[24–26] Interestingly, lasing in this case suppressed the spontaneous emission background which indicates superior qualities of the lasing spectrum obtained.

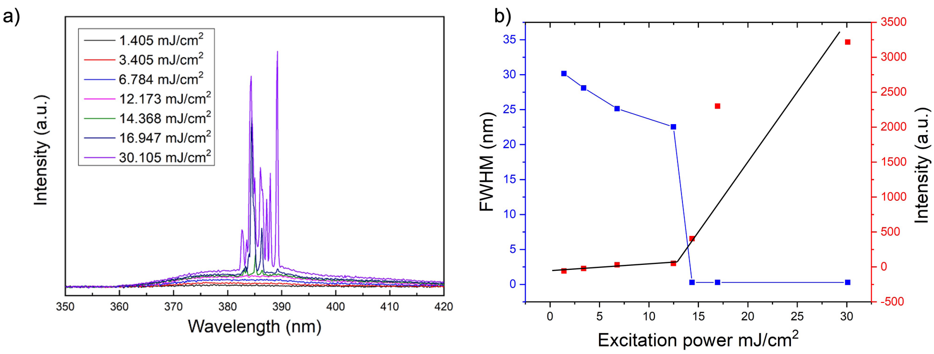


Figure 3: (a) Random lasing from Sample 1 obtained at different pump fluence and (b) changes in linewidth and intensity of the emission with respect to the excitation fluence. Threshold was observed at 12.2 mJ/cm^2.

Figure 4 shows the random lasing emission from Sample 2 whereby the synthesis of ZnO was prepared together with Sample 1, but the length of the fiber is twice as long. Threshold occurs at 16 mJ/cm^2 which was observed upon emergence of the lasing spike with linewidth as narrow as 0.284 nm. The threshold has increased compared to Sample 1 and the spontaneous emission background is high. It is likely that a longer fiber substrate contributes to more dissipated losses from the photons generated. When less photons participate in scattering within the gain medium then the probability to get random lasing to occur reduces significantly, and in some cases lasing is not even possible.[27]

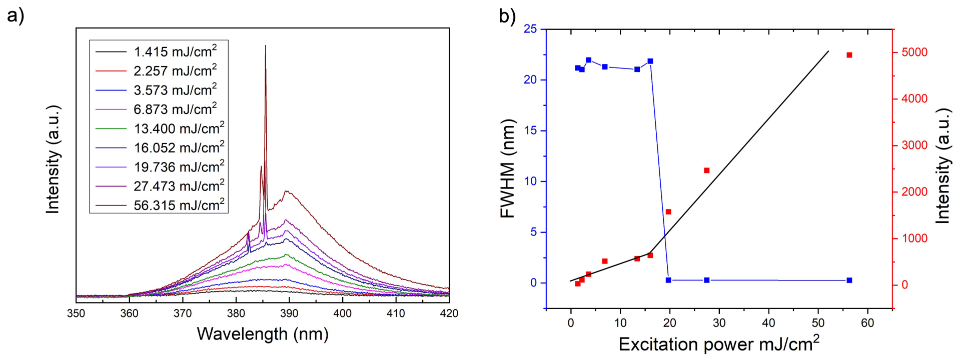


Figure 4: (a) Random lasing emission from Sample 2 at different pump fluence and (b) linewidth and intensity changes upon excitation. Threshold was observed at 16 mJ/cm^2.

Figure 5 shows random lasing emission from Sample 3 which is a repeated synthesis of the ZnO that revealed a more even distribution of ZnO microrods. The threshold recorded from this sample is 15.9 mJ/cm^2. Similarly, increasing the input pumping power increases the number of lasing modes.

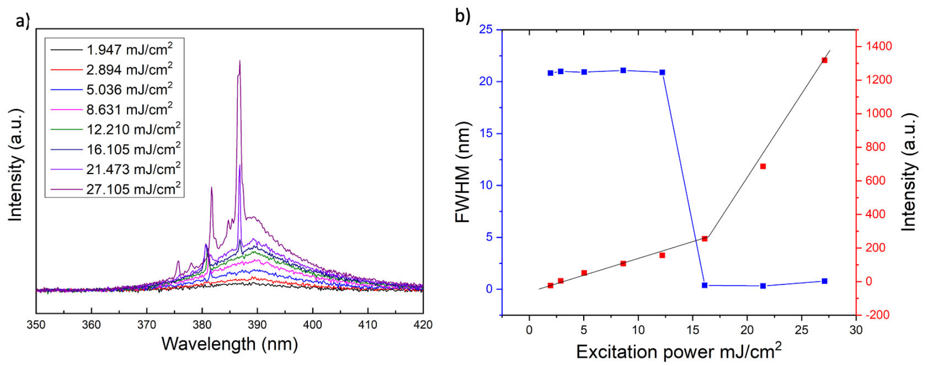


Figure 5: (a) Random lasing emission from Sample 3 at different pump fluence and (b) linewidth and intensity changes upon excitation. Threshold was observed at 15.9 mJ/cm^2.

Figure 6 shows the spectrum obtained from two fibers with only ZnO seed layer (no microstructures) and only spontaneous emission of ZnO was detected in this case. The emission at 385 nm refers to photoluminescence of ZnO.[28–30] This serves as a control sample whereby lasing is only observed when ZnO microrods are prepared on the PCF substrate.

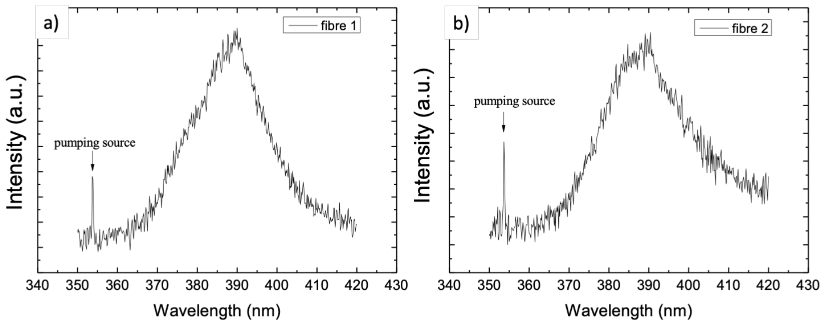


Figure 6: Spontaneous emission from PCF substrate when only ZnO seed layer is present in (a) Sample 1 and (b) Sample 2. Only spontaneous emission was observed even at high pumping powers.

For clarity on the effect of a PCF substrate in random lasing, lasing measurements were performed on ZnO structures with similar morphology that are prepared on glass substrate. The image of the sample and the lasing emission is shown in Figure 7. ZnO in this case was synthesised the same way as for the fiber samples however to match the nanorod diameter, the molarity of the chemical solution was increased to 0.08 M. Lasing was observed at 89.6 mJ/cm^2 which is at very high pump power in comparison to that observed from ZnO on PCF. Since the sample was not annealed post growth, the threshold obtained is also high when compared to other random lasing emission on glass prepared by the same method.[31,32]

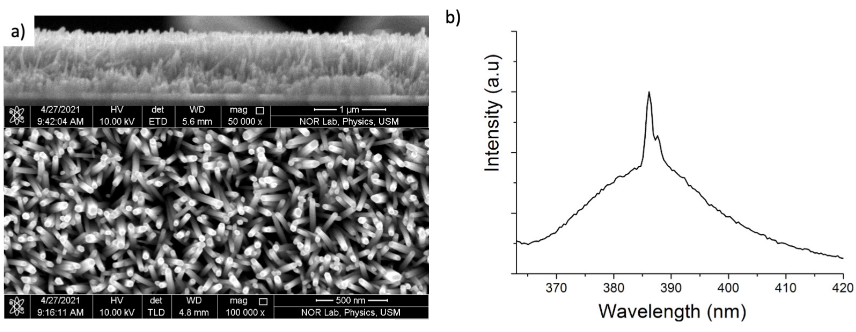


Figure 7: (a) FE-SEM image of the ZnO nanorods with upper figure showing the cross section and the bottom figure shows top view and (b) random lasing emission from the ZnO nanorods prepared on glass substrate.

To validate lasing emission observed from the samples, changes in light confinement was investigated using finite-element approach from COMSOL Multiphysics™ software. The structures were redrawn based on FE-SEM images in Figure 2. Figure 8(a) and Figure 8(c) is shading of the nanostructures from FE-SEM image of Sample 1 and Sample 3, respectively. Figure 8(b) and Figure 8(d) shows the light propagation within the structure when pumped with 355 nm pumping source. We found the scattering fields mainly locate at the edge of ZnO nanorods and the air gaps in between, which also suggests the parameters of fabrications in Sample 1 and Sample 3 are suitable for the formations of scattering loops for random lasing to occur.

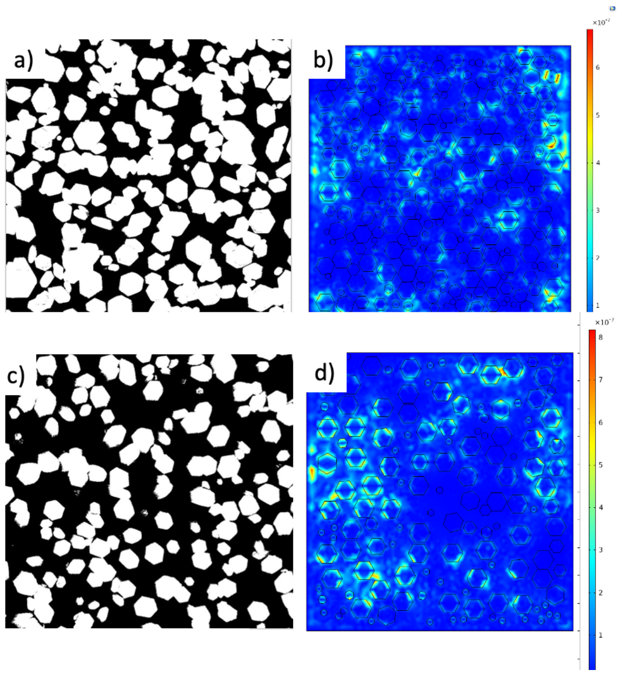


Figure 8: Simulation of light propagation within the structure of Sample 1 and Sample 3 whereby (a) and (c) refer to the FE-SEM images of Sample 1 and Sample 3, respectively; (b) and (d) refer to the light confinement from Sample 1 and Sample 3, respectively.

4. CONCLUSION

In conclusion, we have shown low threshold random lasing emission from ZnO microrods prepared on PCF substrates. The ZnO structures has an average height of 2.81 m and average rod diameter of 195 nm. Superior lasing was obtained with microrods prepared on fiber substrates without the need of annealing the ZnO structures post growth. Threshold of lasing emission was obtained at 12.2 mJ/cm^2 with a narrow spectral width of 0.28 nm. This first reported observation of random lasing from a sustainable gain medium embedded on PCF shows promising potential in making a ZnO fiber based random laser in the near future.

5. ACKNOWLEDGEMENTS

We are thankful to final year undergraduate students Aqilah, Izzati, Hadi and Aliah in preparing the ZnO gain medium. This work was supported by Universiti Sains Malaysia through Geran Khas Matching RUI under account number: 1001/PFIZIK/8011138 and 2021 SATU Joint Research Scheme (JRS) USM 04 and NCKU 44. This work was sponsored in part by the National Science and Technology Council (NSTC) of Taiwan under contract number: NSTC 111-2112-M-006-023-MY3.

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