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Blue Lasing in Poly(Octanedioxy-Co-DMPPV) Copolymer Microcavity

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Poly[octanedioxy-co-DMPPV] copolymer was synthesized to fabricate the microcavity device. The blue emitting polymers consist of strictly alternating conjugated PPV and non-conjugated alkyl group. The microcavity based on distributed Bragg reflector (DBR) and the spin-coated copolymer thin film shows promising lasing characteristics. A strong blue emission at 437.9 nm with 6.5 nm line width was due to cavity enhancement and large gain from the copolymer layer. Third harmonic of Nd:YAG laser(355 nm) was used as a pulsed pump source and its energy threshold for lasing was around few microjoules per pulse. The lasing characteristics strongly depended on the microcavity structure and copolymer film processing. Q factor was calculated to be 67 and the operational lifetime of over 7×10^5 pulses was observed.

Keywords: conjugated polymer; microcavity; blue lasing; PPV copolymer

INTRODUCTION

Among the various π -conjugated polymers, poly(*p*-phenylenevinylene) (PPV) and its derivatives enjoy a great deal of attention in photonic applications, especially in light emitting diode (LED), because of their high quantum efficiency, improved processibility and easy band gap control.^[1-3] This initial success with these materials directed toward the field of lasing and optical amplification.^[4-7] In addition to the above characteristics, low excited state absorption and high quantum efficiency for stimulated emission would be desirable for laser medium. Due to a large Stokes shift, excited state absorption is fairly low for PPV and its derivatives, however, they suffer from low quantum efficiency for stimulated emission. This lack of stimulated emission results from interchain interactions giving rise to photoinduced absorption which masks the stimulated emission.^[8,9] Another shortcoming for these materials is difficulty in obtaining a blue color. Most of PPV derivatives

show green to orange emission.

Poly(octanedioxy-co-DMPPV), a soluble dimethoxy substituted PPV-alkyl chain copolymer was chosen for laser medium to improve these

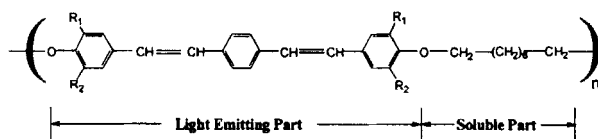


FIGURE 1. Molecular structure of Poly(octanedioxy-co-DMPPV).
 $R_1, R_2 = \text{OCH}_3$

shortcomings. This copolymer consists of PPV oligomer unit and alkyl chain as seen in Fig. 1. It has many advantages over conventional PPV in color tuning, processibility and durability.

PPV trimer was adopted to obtain the blue emission. Dimethoxy groups were substituted in phenyl rings to improve the polymer chain alignment and orientation which may minimize interchain interactions and enhance the stimulated emission. This substitution shows negligible effect on band gap, however.

In this report, we describe the characteristics of laser emitted from Poly(octanedioxy-co-DMPPV) microcavity.

EXPERIMENTAL

Poly(octanedioxy-co-DMPPV) copolymer was synthesized as described previously.^[10] Thin films were formed by spin-coating a copolymer solution in chloroform on a distributed Bragg reflector (DBR) which has a reflective stop band between 430-520 nm with better than 99.9 % reflectivity. On top of the active layer a 50 nm thick Au mirror was deposited. The microcavity was optically pumped with a third harmonic of Q-switched Nd:YAG laser (355 nm) that generated 5 ns pulses at a 10 Hz repetition rate. The pump beam was incident normally through the DBR stack which has 92 % transmission at 355 nm. The emission at the back of gold mirror was analyzed by monochromator with a PMT or CCD.

RESULT AND DISCUSSION

Fig. 2 shows the emission spectra of a poly(octanedioxy-co-DMPPV) copolymer film on a glass and a copolymer microcavity at various pumping intensity. A strong blue emission at 437.9 nm had a considerably narrower line

width of 6.5 nm. This is considerable blue shift (ca. 50 nm) from copolymer fluorescence and the fluorescence intensity at 437.9 nm is negligible. This strongly support that emission was due to cavity enhancement and large gain from the copolymer layer.

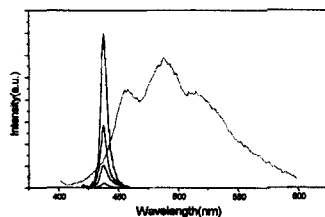


FIGURE 2. Fluorescence and lasing spectra obtained from solid state and microcavity. Input beam energy for microcav (solid line) is 4.25, 6.8, 9, 11 μ J respectively and solid phase (dotted line) is 2 mJ.

Lasing frequency could be tuned by changing the copolymer layer thickness and it was confirmed that $\lambda = 2nd/m$, where cavity mode $m=2$ and the refractive index n is determined to be 2.05 from refractometer measurement. The line broadening can be attributed to the non-uniformity of the film, because multiple coatings were necessary to obtain the copolymer film thickness to near 210 nm. It has been reported that polymer chain packing is crucial in luminescence behavior and laser action.^[11] Interchain interaction, which affects the stimulated emission quantum efficiency and gain narrowing, are strongly dependent on the morphology of the polymer chains in the film. Though dimethoxy substitution to the phenyl rings in the copolymer is expected to improve the polymer chain alignment and orientation, we could not get a benefit of it due to the multiple coating. Single coating with a proper solvent would improve the polymer chain packing.

Another supporting evidence for lasing is high directionality of the emission. As seen in Fig. 3, emissions were observed within 20° of viewing angle. The shift of emission wavelength was less than 1 nm.

Lasing was not observed until the pump energy reached a threshold level of 2.5 μ J. As the pump intensity was further increased, the laser intensity increased rapidly. Above 15 μ J, some degradation of the device performance was observed mainly due to damage of gold mirror. However, this device

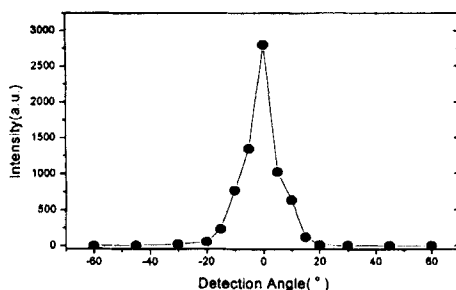


FIGURE 3. Detection angle dependence of emission from microcavity.

shows excellent durability. Slow degradation of the device was observed after 12 hours with 10 μJ pumping and the operational lifetime was over 7×10^5 pulses. It should be noted that this device was fabricated and operated under ambient condition. The copolymer was quite stable to oxidation and no degradation of the cavity performance was observed after few weeks in air.

Q factor of microcavity was calculated to be 67. This could be improved using better reflecting mirror with some sacrifice in lasing threshold power.

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