Operational lifetime improvement of poly(9,9-dioctylfluorene) active waveguides by thermal lamination

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The large research activity on the stimulated emission and lasing of organic conjugated molecules of the last 15 years resulted in a strong improvement of both the active materials and device performances [1], that are rapidly approaching the requirements for real devices applications, as shown by the recent demonstration of organic lasers optically pumped by an inorganic Light Emitting Diode, and of the use of an organic laser prototype as light source in a spectroscopy experiment. Despite these relevant advances a particularly critical point for the development of organic lasers, i. e. the improvement of the device operational stability, has received to date a limited attention. As it is well known that organic molecules suffer photo-degradation due to laser pumping in the presence of oxygen, the main strategy to improve the devices operational stability is the encapsulation in a inert environment.

To date the most effective organic lasers encapsulation consists in the protection of the active layer with a silica coverslip[2], directly bonded to the active layer with an optical adhesive, resulting in negligible threshold increase and in a remarkable device lifetime extension up to 2500 times. Encapsulation by rapid prototyping stereolithography has also been reported, resulting in a lifetime extension of 3 times, for pumping excitation density 3 times larger than the lasing threshold[3]. In order to preserve the possible mechanical flexibility parallel fabrication and encapsulation with poly(methyl methacrylate) (PMMA) and cyclic olefin copolymer (COC) flexible layers [4] and flexible Distributed Feedback Laser encapsulated by a drop casted optical adhesive layer [5] have been recently demonstrated.

In this work we show the main results about the encapsulation properties of Polyethylene terephthalate-poly (Ethylene Vinyl Acetate) (PET-EVA) laminating pouches, thermally laminated on flexible poly(9,9-dioctylfluorene)(PF8) active waveguides showing Amplified Spontaneous Emission (ASE). The complete work can be found in Ref.[6].

Two PF8 active films were realized by spin coating in air, on a standard 100 μm thick acetate transparency for photocopier, of a 10^{-4} M toluene solution with a rotation speed of 1000 rpm, leading to thickness of about 250 nm. One film was used as reference (PF8 in the following), while the second one (PF8lam) was laminated in air by a GBC A4-CL90 thermal laminator by using PET-EVA laminating pouches with a thickness of 125 μm per side (for further experimental details see Ref. [6]). As evident from the picture in Fig.1 our encapsulation preserves the sample flexibility.

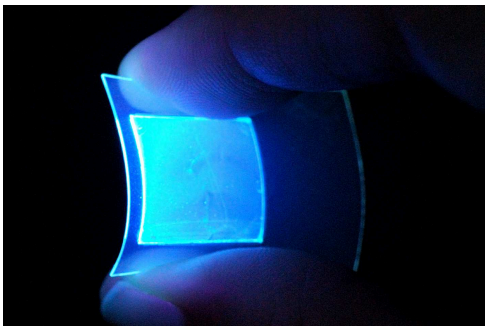

Figure 1. Photography of the laminated flexible waveguide showing the sample flexibility.

The excitation density dependence of the PF8 sample PL spectra (not shown) shows a clear ASE band at about 447 nm for high enough excitation density, that progressively dominates the sample emission. The absorbed energy density at the ASE threshold is $93 \pm 8 \mu J cm^{-2}$.

A similar excitation density dependence of the PL spectra is observed in the encapsulated sample PF8lam (see Fig.2), with absorbed energy density at the ASE threshold of $110 \pm 12 \mu J cm^{-2}$, which is only 15% larger than the reference sample one. For the sake of comparison we observe that the observed threshold increase is considerably smaller than the one (about 4.4 times) reported for flexible encapsulation by optical adhesive drop coating, due to the combination of low encapsulating layer absorption at the pump wavelength, and to its better thickness uniformity

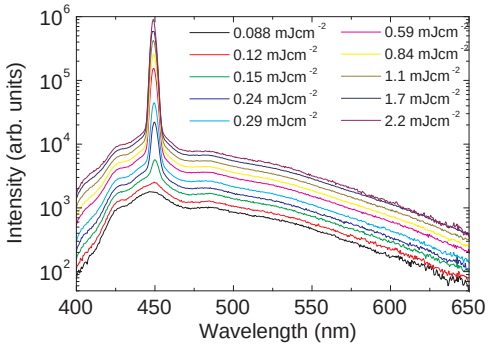

Figure 2. Excitation density dependence of the PL spectra of the encapsulated sample PF8lam.

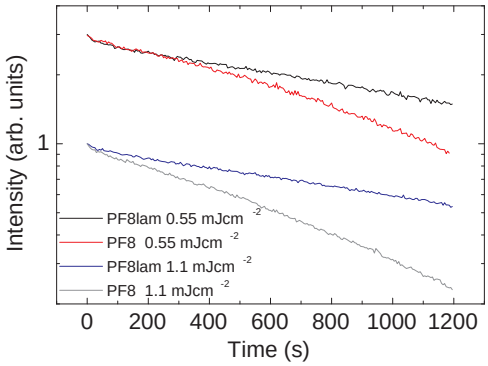

Figure 3. ASE intensity decrease during pumping at 0.55 mJcm^{-2} and 1.1 mJcm^{-2} for both samples. (The data are normalized to the initial intensity value at a given excitation density).

with respect to drop coated adhesives.

In order to determine the origin of this threshold increase we considered the variation of the waveguide propagation losses and of the Mode Confinement (MC) and the pump-mode overlap (PMO) (described in details in Ref.[7]), concluding that about 1/3 of the threshold increase is due to the MC reduction and about 2/3 are due to a weak degradation of the active layer in the lamination process takes place.

Concerning the ASE threshold stability in time after the sample deposition, due to the progressive degradation of the active layer stored in air, we repeated the ASE threshold measurements six months after the deposition. The reference sample shows an ASE threshold increase up $220 \pm 30 \mu Jcm^{-2}$, which is about 2.4 times larger than the fresh sample one, while the laminated sample shows an ASE threshold of $105 \pm 12 \mu Jcm^{-2}$, which is comparable, within the error bar, with the fresh sample one. This result clearly demonstrates that thermal lamination effectively isolates the active layer from the external environment on the scale of several months.

In order to determine the lamination effect on the ASE intensity decrease during waveguide pumping we compared the ASE intensity reduction dynamic of sample PF8lam and PF8 at an absorbed excitation density of 0.55 mJcm^{-2} and 1.1 mJcm^{-2} (see Fig. 3), which means about 5 and 10 times above the threshold excitation density.

An exponential decrease of the ASE intensity is observed for both samples at both the excitation densities, with a clearly slower intensity decay for the encapsulated device. This exponential ASE intensity decrease is due to the interplay between exciton quenching and waveguide losses variations, both due to irreversible PF8 photo-oxidation during pumping[8], and the slower ASE decay in PF8lam is a signature of the effectiveness of thermal lamination in insulating the active layer from the environment.

A quantitative investigation of the encapsulation effects on the ASE operational lifetime was done by determining the ASE intensity decay time from a single exponential fit. At the lowest excitation density the operational lifetime in the reference sample is 1100±8 s, while the encapsulation results in a lifetime increase to 1910±10 s, which is about 1.7 times larger. At an excitation density of 1.1 mJcm^{-2} the reference sample shows a more rapid intensity decay, with a lifetime reduced to 900 ± 5 s, while the encapsulated device does not show appreciable lifetime reduction, with a best fit lifetime of 2090±10 s, corresponding to a lifetime increase of 2.3 times. This lifetime increase is much larger than the one obtained by drop casted optical adhesive [5] (less than 10%), with a much lower threshold increase. Lasing lifetime increase up to 10 times have been instead reported for DFBs with thermally bonded PMMA overlayer [4], which anyway show a much stronger encapsulant absorption (about 95% against 8%).

Our results then propose thermal lamination as a simple and affective technique for active waveguides and DFB lasers encapsulation preserving the possibility to realize flexible devices.

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