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Research Article

One Dimensional Polymeric Organic Photonic Crystals for DFB Lasers

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We present a very simple method to realize a one-dimensional photonic crystal (1D PC), consisting of a dye-doped polymeric multilayer. Due to the high photonic density of states at the edges of the photonic band-gap (PBG), a surface emitting distributed feedback (DFB) laser is obtained with this structure. Furthermore, the incidence angle dependence of the PBG of the polymeric multilayer is reported.

1. INTRODUCTION

The use of polymers and dye-doped polymers for the fabrication of photonic crystal is currently being actively investigated. Photonic crystals, which consist of a periodic dielectric lattice with a periodicity in the range of optical wavelengths, are intensively studied in order to achieve control of light propagation. This property, which can be used to confine, manipulate, and guide photons, should allow the creation of all-optical integrated circuits and optoelectronic devices [1–3]. In a medium, the periodicity of the refractive index can be in one, two, and three dimension and there are many examples of one-, two-, and three-dimensional photonic crystal fabricated with several materials [4, 5]. While one-dimensional photonic crystals are inherently structurally simpler than two- and three-dimensional analogues, only few investigations have been reported on multilayer stacks of materials, known also as Bragg stacks. These Bragg stacks can be useful for various photonic and optoelectronic applications. In 1994, Dowling et al. [6] proposed and analyzed a surface-emitting laser consisting of a one-dimensional photonic crystal (1D PC) in which a gain material was incorporated. Similar to a Fabry-Pérot type laser with a single longitudinal mode, this distributed feedback (DFB) type laser can oscillate at one of the photonic band edges, where photon density is significantly enhanced [6]. Although inorganic DFB laser is commercially available, a small amount of papers reports the use of polymeric 1D PC for this application [7]. Furthermore, some properties, like incidence angle dependence of the photonic band gap (PBG) of these dye-doped structures, are usually not investigated.

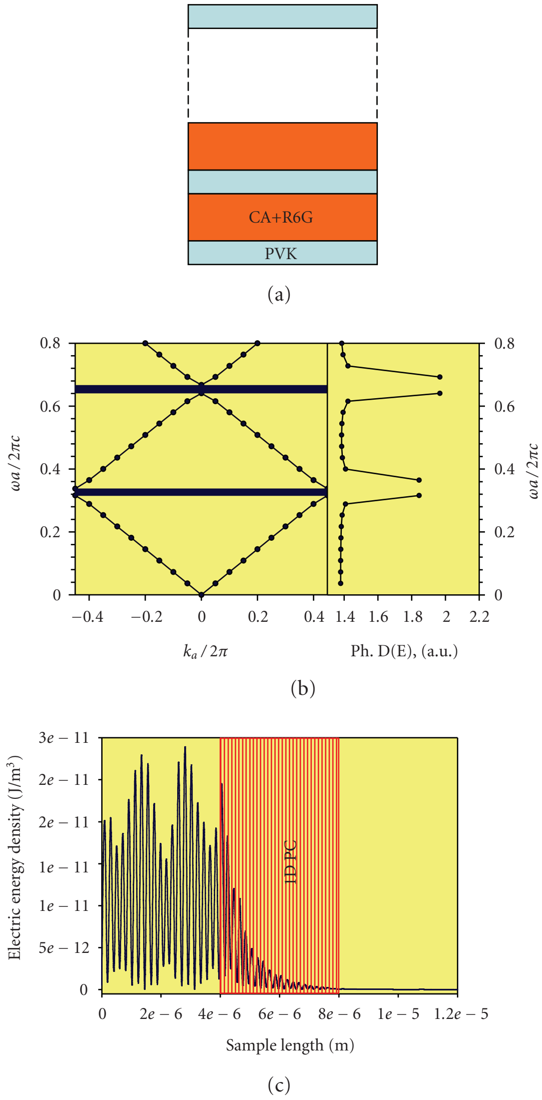
In this paper, we describe the modeling, fabrication, and optical characterization of a dye-doped one-dimensional photonic crystal, consisting in alternated stacks of polyvinylcarbazole (PVK) and cellulose acetate (CA), where each CA stack is doped with Rhodamine 6G (R6G). The laser emission of the dye in the photonic structure and the angular dependence of the PBG are also measured and discussed.

2. MODELING

In order to predict the photonic properties of the structure (Figure 1(a)) which we want to fabricate with the two polymers, we use two-computational tools: MIT photonic band gap and Comsol Multiphysics. MIT Photonic Bandgap is a freeware developed by S. G. Johnson. The program calculates ab initio the band structure of a periodic dielectric structure. The inputs for the calculation are the refractive index of the two materials (in our case $n_{\mathrm{PVK}} = 1.683$ and n_{CA}=1.475 [7, 8]) and the ratio between their thicknesses ($d_{\mathrm{PVK}}/d_{\mathrm{CA}}$). Although the maximum width of a PBG with a 1D PC is obtained by stacking layers with the same optical path ($n_{\mathrm{PVK}}d_{\mathrm{PVK}} \approx n_{\mathrm{CA}}d_{\mathrm{CA}}$), we want to fabricate a 1D PC where the dye-doped CA layer is 3/4 of the unit cell in order to increase the absorption of the pump power by the dye. The program assumes that the structure is infinite in all directions. In Figure 1(b), the calculated dispersion relation and the photonic density of states for the polymeric 1D PC are shown. The photonic density of state is very high in proximity of photonic band edges. The photonic band gap (violet in the figure) is narrow, and this is due to the relative low refractive index contrast of the two polymers (≈1.14). Nevertheless, for a DBF laser application, this fact is negligible while the zero group velocity at the photonic band edge is the main feature. The organic dye which we choose to dope the CA layer is Rhodamine 6G, that emits between 550 and 600 nm. In order to obtain a PBG centered at about 600 nm (and a photonic band edge at 590 nm), the thicknesses of the PVK layer and the dye-doped CA layer have to be 50 and 150 nm, respectively. Comsol Multiphysics is a commercially available software that solves numerically second-order differential equations. The inputs for this calculation are similar to the previous program. But in this case, we can decide the number of layers. In Figure 1(c) an e.m. wave with wavelength of 600 nm (the center of the PBG) is reflected when it passes through a 39-layers 1D PC.

3. EXPERIMENTAL

Following [7], polymers solutions of PVK (Acros Organics, Janssen-Pharmaceuticalaan 3, 2440 Geel, Belgium) and CA (Acros Organics) are prepared. The solvents are chlorobenzene (Sigma Aldrich, 3050 Spruce St, St Louis, MO, United States) and diacetone alcohol (Acros Organics) for PVK and CA, respectively. The number-average molecular weights of PVK and CA are 90 000 and 100 000, respectively. So, we have selected a concentration of 24,4 and 53,46 g/L. The CA layer is 0.5 w% doped by R6G. In order to obtain the multilayer, we have tried with dip coating, but with this technique a deposited polymer film stays too much in contact with the solvent of the other polymer and consequently the solvent of the other polymer can slightly dissolve the film. We can overcome this problem with the spin coating technique, where about 99% of the solvent is ejected in the first second of rotation. A substrate of glass (Carlo Erba, Via Generale Carlo Ferraris, 22036 Erba Como, Lombardy, Italy) has been used and the rotation speeds for PVK and for CA on the glass substrate are 3800 and 4100 rpm and the already coated layers 3600 and 4000 rpm, respectively. In this way, a $(PVK/CA)_9PVK$ multilayer is obtained. After each deposition, a baking of 5 minutes at 80°C has done. We can neglect the refractive index of the substrate because his thickness is 2 mm, and it cannot affect the photonic properties of the multilayer. The transmittance spectrum has been measured with a Cary Varian 50 spectrophotometer (bandwidth of 1 nm). For the laser emission measurement, a pulsed second harmonic of an Nd:YAG laser (Quanta System, Via Iv Novembre, 116, 21058 Solbiate Olona (VA), Italy) has been used as a pump with a repetition rate and duration of 1 Hz and 7 nanoseconds, respectively. The sample has been obliquely pumped with laser emission of the dye normal to the surface (in the direction of the refractive index periodicity). The photoluminescence was revealed by Jobin Yvon CCD camera (bandwidth of 1 nm). The angular dependence transmittance spectra have been measured with an optical set-up based on fiber optical coupled to an Avantes 2048 compact spectrometers (spectral resolution 1.6 nm) working in the range 250–1100 nm. White light was provided by a tungsten-halogen/deuterium combined lamp.



(a)

(b)

(c)

Figure 1: (a) Dye-doped 1D PC, (b) dispersion relation of the 1D PC (with an infinite number of layer) with MIT photonic band gap, and (c) propagation of an e.m. wave (λ = 600 nm) through a 39-layers 1D PC.

4. RESULTS AND DISCUSSION

In Figure 2, the transmission spectrum of the $(PVK/CA)_9PVK$ multilayer, where CA layers are 0.5 w% doped by R6G, is shown. The PBG is centered at about 610 nm and the photonic band edge at 590 nm. The band gap is shifted by 10 nm with respect to theoretical predictions; this is due to

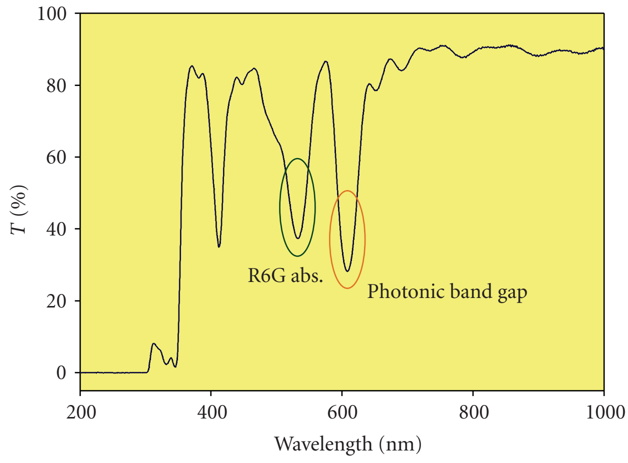


FIGURE 2: Transmission spectrum of the $(\mathrm{PVK/CA})_9\mathrm{PVK}$ multilayer. The green oval indicates the Rhodamine 6G absorption, while the orange oval indicates photonic band gap. At about 400 nm, the higher order photonic band gap is present

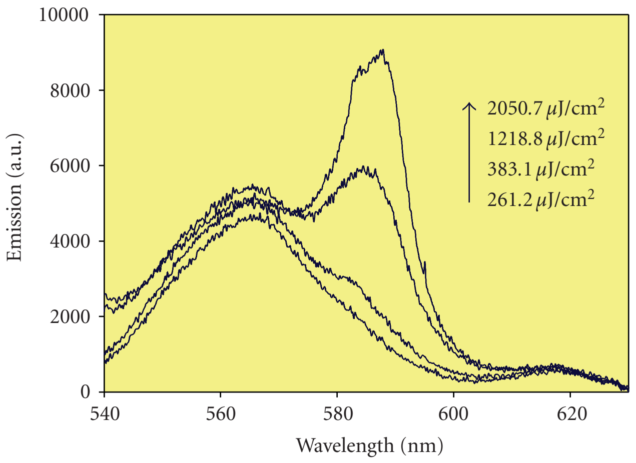


FIGURE 3: Photoluminescnce spectra of the surface emitting DFB laser made with the dye-doped 1D PC upon increasing the pump energy.

the difficult control of the thickness of each deposited stack. In the spectrum, we see also the absorption of the dye around 532 nm and the second order PBG. The depth of the PBG is about 60%. In Figure 3, the photoluminescence spectra of the R6G-doped 1D PC, at different pump energy density, are reported. The photoluminescence signal is revealed normally to the surface of the multilayer. The region between 540 and 570 nm shows the luminescence peaks of the dye, while between 570 and 630 nm remarkable effects arise from the superimposition of the luminescence of the dye with the photonic structure. In the range of energy where the photonic band gap is present, the luminescence of the dye is suppressed, while, at the energy corresponding to the first (higher energy) photonic band edge, the emission of the dye exhibits a narrow peak arising from the enhancement of the emission in the region of the high density of states.

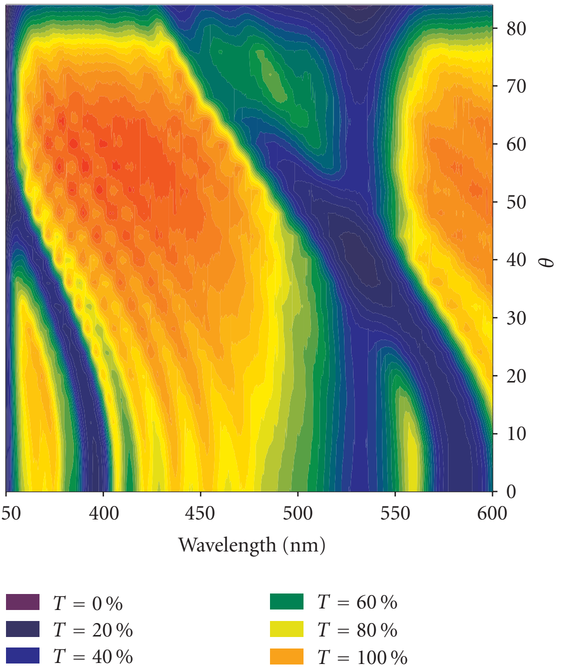


FIGURE 4: Incidence angle dependence of the transmission spectrum of the dye-doped 1D PC.

Figure 4 shows the trend of the transmission spectrum of the dye-doped 1D PC versus the incident angle θ. In the range between 520 and 540 nm, the absorption of R6G is shown, and it, obviously, does not change, whereas the photonic band gap, the higher order photonic band gap, shows a red shift of the order of 150 nm. The higher order PBG shows a red shift toward the region of PVK absorption.

From these data, we can obtain the refractive index and thickness of the multilayer by solving the equations system [9]:

$$\begin{gathered} m\lambda_{\mathrm{Bragg}} = 2D\sqrt{n_{\mathrm{eff}}^2 - \sin^2\theta}, \\ W = \frac{4}{\pi}\left|\frac{n_{\mathrm{PVK}} - n_{\mathrm{CA}}}{n_{\mathrm{PVK}} + n_{\mathrm{CA}}}\right|, \\ \frac{\varepsilon_{\mathrm{eff}} - 1}{\varepsilon_{\mathrm{eff}} + 2} = x_{\mathrm{PVK}}\frac{\varepsilon_{\mathrm{PVK}} - 1}{\varepsilon_{\mathrm{PVK}} + 2} + (1 - x_{\mathrm{PVK}})\frac{\varepsilon_{\mathrm{CA}} - 1}{\varepsilon_{\mathrm{CA}} + 2}, \end{gathered} \tag{1}$$

where the first equation is Bragg-Snell equation, the second is the expression for the PBG width for a 1D PC and the third is the Lorentz-Lorentz equation. In these equations, m is the diffraction order, λ_{Bragg} is the wavelength where the PBG is centered, x_{PVK} is the fraction, the unit cell fraction of the polymer PVK, n_{eff} is the effective refractive index, θ is the incidence angle measured off from the normal, $n_{\mathrm{PVK(CA)}}$ and $\varepsilon_{\mathrm{PVK(CA)}}$ are, respectively, the refractive index and the dielectric constant of PVK (CA). Solving the system, we calculate the refractive indexes (n) and the thicknesses (d) of the two layers and we can compare these with the values that we have found in literature and from simulations (Table 1). The results of theoretical simulations and the

Table 1: Refractive indexes and thicknesses of the polymers in the 1D PC. Comparison with literature and theoretical predictions.

	Spectroscopic data		Literature data	Theoretical sim.
	n	*d* (nm)	*n*	*d* (nm)
PVK	1.668	48.47	1.683	50
CA	1.470	149.40	1.475	150

literature data are in excellent agreement with the equation system solutions.

5. CONCLUSION

In this work, we have modeled, fabricated, and optically characterized a dye-doped polymeric 1D PC. The modeling has been useful to predict the photonic properties of a structure made with the chosen materials. Comsol Multiphysics uses finite elements algorithms and permits to study finite structures, so the prediction of the interaction between e.m. waves and aperiodic and disordered structures with this program is also possible. We have found that the spin coating deposition technique is good for easy fabrication of polymeric photonic crystals. The fabricated dye-doped 1D PC, with the photonic band edge centered in proximity of the laser dye emission, is a low threshold surface emitting DFB laser. Bleaching of the dye is observed at high pump energy density. We intend to overcome this problem by doping of the structure with novel laser dye, for example, organic complexes of lanthanides [10] in which the optically pumped excited state of the ligand is quickly depopulated by an efficient energy transfer process to the emitting ion.

Furthermore, the incidence angle dependence of the PBG of the polymeric multilayer has been measured. This behavior allows us to determine independently *n* and *d* and will be used for controlling the spontaneous emission of the dye in the 1D PC.

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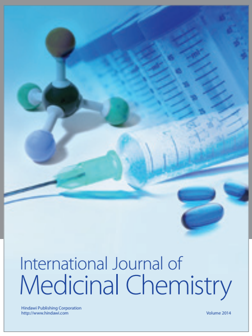
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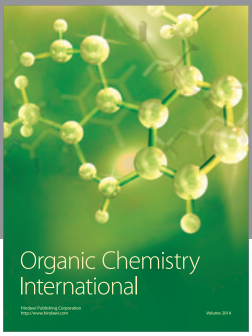
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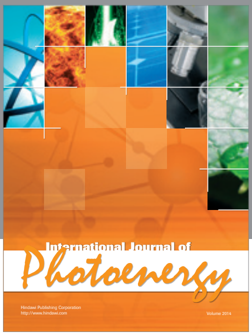
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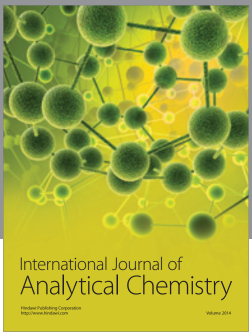
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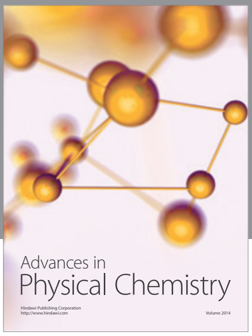
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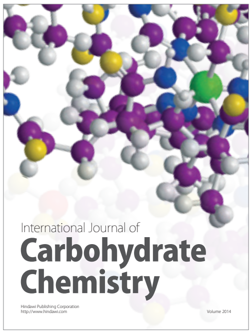
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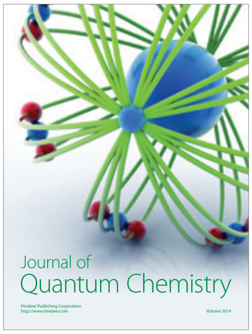
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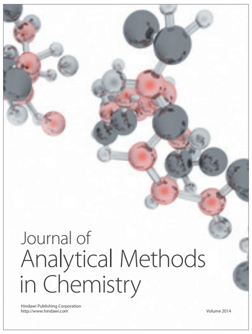
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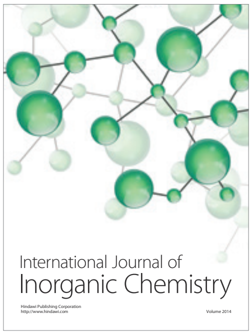
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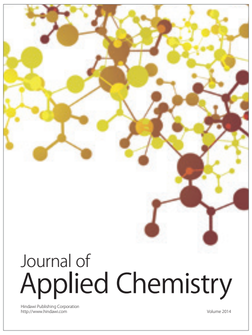
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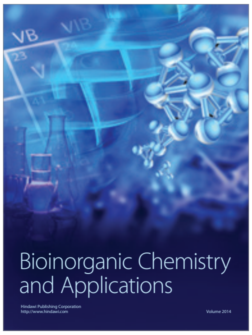
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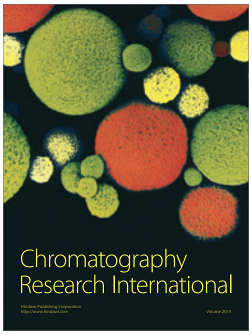
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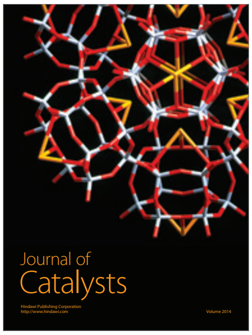
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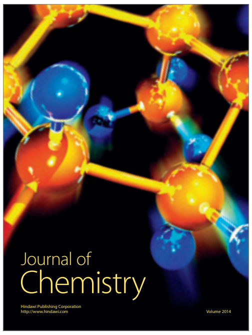
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