Journal of Luminescence 143 (2013) 288–292

Contents lists available at SciVerse ScienceDirect

Journal of Luminescence

journal homepage: www.elsevier.com/locate/jlumin

Photoluminescence manipulation based on composite photonic crystal surfaces

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ARTICLE INFO

Article history:
Received 10 January 2013
Received in revised form
15 January 2013
Accepted 26 April 2013
Available online 11 May 2013

Keywords:
Colloidal photonic crystals
Photo-crosslinkage
Photoluminescence

ABSTRACT

A series of composite PC films with different morphologies are fabricated through a facile approach. The effects of different surface morphologies and the stopband positions of composite PC films on photoluminescence (PL) behaviors are systematically investigated. We observe that the PL on composite PC surfaces can be manipulated. When dyes enter into interstitial space of PC films without photo-crosslinkage, inhibited spontaneous emission is evident and emission is enhanced at the blue edge of photonic stopband. When dyes are deposited on outer surface of composite PC films, inhibited spontaneous emission disappears, but an obvious enhancement of the emission is achieved, where the composite PC surface acted as a Bragg mirror. The study will be of great guideline for future development of highly efficient optical devices.

1. Introduction

Since photonic crystals (PCs) were proposed by Yablonovitch and John [1,2], PCs are the subject of intensive theoretical and experimental research as an impetus to manipulate the interaction between light and matter for diverse optical applications [3–8]. Their periodic dielectric structures are designed to control the propagation of electromagnetic waves via the photonic stopband effect [9,10]. The absence of propagating electromagnetic modes inside the structures can result in distinct optical phenomena such as inhibition of spontaneous emission. The control of spontaneous emission is basically and practically important for applications such as quantum optics, low-threshold lasers, high efficient LEDs, and solar cells.

To build up the environment for spontaneous emission control, scientists have adopted PCs to manipulate spontaneous emission of embedded emitters (dyes, polymers, semiconductors, etc.) [11–17]. From a general point of view, the localization of emitters in PCs allows the investigation of the stopband influence on the PL properties [18–22]. For instance, Klimov and co-workers have reported amplified spontaneous emission of semiconductor nanocrystals uniformly coated on opal surfaces to enhance the optical gain [23]. Otherwise, the angle-dependent behavior of stopbands is another crucial factor in the emission performance. Especially, Vos and co-workers have investigated the angle-dependent fluorescence spectra in opal and inverse opal PCs, whose center positions, widths and depths are analyzed and compared to stopbands from reflectivity measurements [24]. However, most of the studies on spontaneous emission control with three-dimensional PCs have been limited to the effect of photonic stopbands, little work has been paid to the configuration on PC surfaces which may be an impact on the PL performance as well, such as optically-active materials embedded in inner (opals interstice) or outer surface of PCs. Therefore, it is essential to investigate the morphology-dependent behavior of PC surfaces in PL spectra for building the relationship of PCs with the architecture and optical properties.

In this contribution, the effects of different surface morphologies and the stopband positions of composite PC films on PL behaviors are systematically investigated. To construct PC surfaces with different morphologies, different crosslinkages of latex spheres via photopolymerization of acrylamide (AAm) infiltrated PC films are fabricated. The simple photo-crosslinkage approach of composite PCs can effectively introduce different crosslinkages. Thus we can explore the PL performance influenced by engineering the morphology of composite PC surfaces. The result will provide the guidance for PL manipulation on composite PC surfaces of different morphologies.

2. Experimental

2.1. Materials

Styrenes (St), methyl methyacrylate (MMA), and arylic acid (AA) were purified by distillation under reduced pressure. The initiator of

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http://dx.doi.org/10.1016/j.jlumin.2013.04.049

$(NH_4)_2S_2O_4$ (APS) was recrystallized three times. All reagents and materials were purchased from Aldrich unless otherwise noted.

2.2. Synthesis of p(St–MMA–AA) colloidal spheres

Monodisperse latex spheres of poly(styrene–methyl methacrylate–acrylic acid) (poly(St–MMA–AA)) were synthesized via the literature [25]. The resulting latex spheres were used directly without purification. The polydispersity of the latex spheres was about 0.5%, which was detected by ZetaPALS BI-90plus (Brookhaven Instrument).

2.3. Fabrication of PC films

The PC films were fabricated on glass substrates by a vertical deposition method [26]. The glass slides were first treated with a chromic-sulfuric acid solution to ensure clean surfaces and were vertically positioned in a vial containing the monodisperse p(St–MMA–AA) aqueous suspension of 0.2 wt% at 60 °C and 60% relative humidity for 24 h. After the samples were dry, they were sintered at 85 °C for 30 min to increase the stability of the samples.

2.4. Photo-crosslinkage of composite PC films

Firstly, PC films were immersed into AAm solution for 5 min, the solutions with concentrations of 2, 5, and 10 wt% were prepared by mixing AAm, bisacrylamide (BIS) (2 wt% of AAm amount), diethoxyacetophenone (DEOP) (2 wt% of AAm amount) and H_2O. Secondly, PC films were overturned on a glass slide to keep the film under the pressure of the substrate, and then the system was exposed to ultraviolet light of 365 nm at 2.5 mW/cm^2 (from UV lamp of Jiangsu Kylin-Bell Lab Instrument Co. Ltd., GL 9406) for 15 min to make sure the photo-polymerization of AAm completed. As a result, the PAAm polymer chain would connect latex spheres altogether, and the crosslinked polymer network would form among latex spheres.

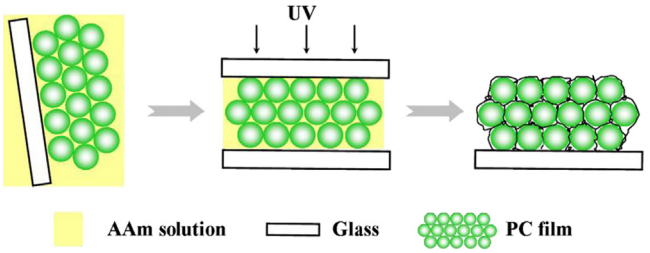

Fig. 1. Schematic illustration of the photo-crosslinkage procedure of composite PC films.

2.5. Characterization

The SEM images were obtained with a field-emission scanning electron microscope (JEOL JSM-4800, Japan), after sputtering the samples with a thin layer of gold. Atomic force microscopy (AFM) characterization was performed with an SPI 3800 N multimode scanning probe microscope (Seiko Instruments). Topographical images were obtained in the contact mode with a silicon cantilever having a nominal spring constant of 0.02 N/m and at a scan rate of 1.0 Hz. Reflection spectra were recorded on a U-4100 UV–vis spectrophotometer (Hitachi, Japan). The fluorescent dye coumarin was deposited onto composite PC films of about 20 nm thicknesses under vacuum using a VPC-260F Vacuum Deposition Equipment (ULVAC). The single spectrum mode of a Witec-Alpha scanning near-field optical microscope equipped with a liquid-nitrogen cooled (−56 °C) CCD camera detector was used for PL measurements with 442 nm excitation. The integration time was 0.1 s. All spectra were recorded normal to the hkl=111 planes of the crystal.

3. Results and discussion

3.1. Morphology of composite PC films

First, composite PC films are fabricated, which involved the self-assembly of latex spheres, the infiltration of monomers and

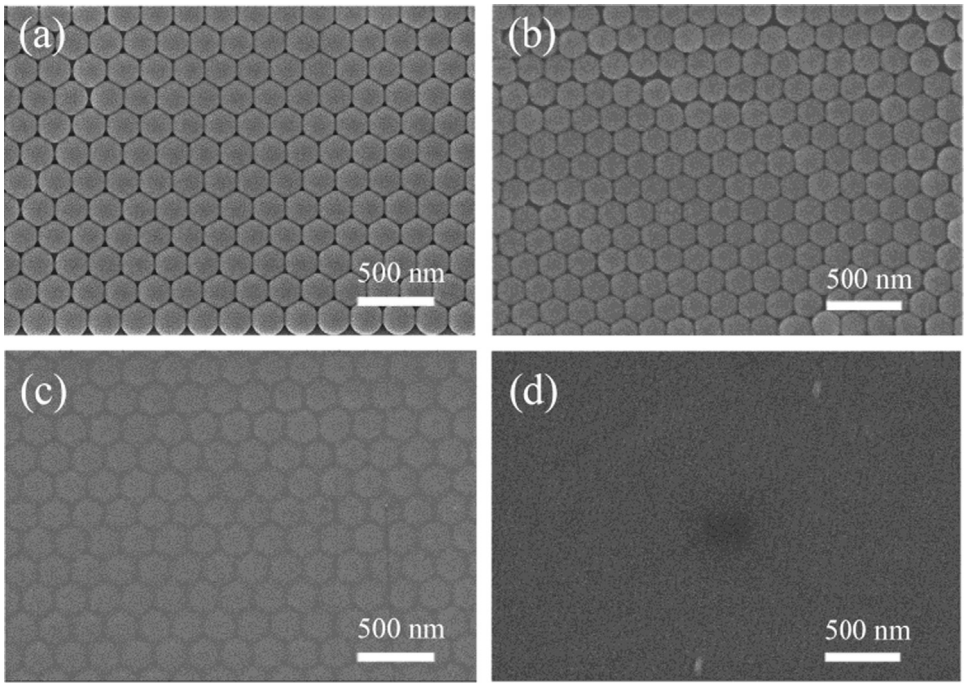

Fig. 2. SEM images of the composite PC films fabricated from different AAm concentrations: (a) 0, (b) 2, (c) 5 and (d) 10 wt% of AAm solution.