Initial Study on Thin Film Preparation of Carbon Nanodots Composites as Luminescence Material

F Iskandar[1,2], A H Aimon[1], A R Akmaluddin[1], B W Nuryadin[3], M Abdullah[1]

[1]Department of Physics, Institut Teknologi Bandung, Bandung 40132 Indonesia
[2]Research Center for Nanoscience and Nanotechnology, Institut Teknologi Bandung, Bandung 40132 Indonesia
[3]Department of Physics, Faculty of Science and Technology, UIN Sunan Gunung Djati, Bandung 40614 Indonesia

E-mail: ferry@fi.itb.ac.id

Abstract. Nowadays, the developments of phosphors materials require elements without noble metals and simple production process. Carbon nanodots (C-dots) are one of phosphor materials with wide range of emission band, and high biocompatibility. In this research thin film carbon nanodots composite have been prepared by spin coating method. Prior deposition, powder carbon nanodots were synthesized from a mixture of commercial urea as the nitrogen sources and citric acid as a carbon source by using hydrothermal and microwave-assisted heating method. The prepared powder was dispersed in transparent epoxy resin and then coated on glass substrate. The photoluminescence result for sample with 0.035 g citric acid exhibited an intense, single, homogeneous and broad spectrum with yellowish emission upon excitation at 365 nm. The Fourier Transform Infrared Spectroscopy (FTIR) result showed the existences of C=C, C-H, C=O, N-H and O-H functional groups which confirmed the quality of the sample. Further, based on UV-Vis measurement, the prepared thin film was highly transparent (transmittance 90%) with estimated film thickness around 764 nm. This result may open an opportunity for optoelectronic devices.

1. Introduction

Recently, photoluminescence-based lamps have attracted considerable attentions because they can generate useful light much more efficiently than incandescent lamp [1-3]. However, most of commercial photoluminescence materials were prepared from rare earth ions which is expensive and hazardous substances. Therefore, there is a strong demand from industrial and ecological point of views to find an alternatives materials based on inexpensive and environment friendly materials. In this regard, carbon based nanodots (C-dots), a new class of carbon nanomaterial with sizes below 10 nm, are very attractive materials to answer this challenge because of their strong fluorescence, chemical inertness, low toxicity, and versatility in their synthesis method with low-cost raw materials [3-5].

Previously reported methods to prepare C-dots include wet oxidation, ultrasonic, pyrolisis, hydrothermal and microwaved-assisted synthesis [1]. Among them, microwave-assisted synthesis method is very fascinating because this method was relatively simple and low-cost method compared to other methods [6-7]. Qu *et al*. reported that C-dots with strong photoluminescence were successfully synthesized by microwave-assisted synthesis method using citric acid as carbon source

and urea as nitrogen source [4]. Considering industrial point of views, it was wondering that analytical-grade raw materials for preparation of C-dots might be replaced with commercial-grade materials. Herein, we report a simple preparation of C-dots using microwave-assisted synthesis method with commercial urea and citric acid as precursor, followed by thin film preparation of C-dots composites using spin coating method.

2. Materials and Methods

Commercial urea $(NH_2)_2CO$ as nitrogen sources and citric acid ($C_6H_8O_7$) as carbon sources were purchased from Bratachem without further purification. Synthesis of carbon nanodots composites was follow previous method [4] with a little modification. Briefly, C-dots precursor was prepared by mixing citric acid and urea together in 10 mL of demineralized water at room temperature for 5 minutes, continued with drying in hot air oven at 100 °C for 1 hour. Then, dried precursor was heated in commercial microwave at ~800 W for 2 minutes. Thin film of C-dots composites were prepared by mixing C-dots with epoxy resin, followed by deposition of C-dots thin film onto glass substrate using spin coating method. In this experiment, the amount of citric acid was varied (0 g; 0.015 g; 0.035 g; 0.075 g and 0.125 g) to obtain knowledge about the influence of carbon source to photoluminescence quality of carbon nanodots composites.

Functional group in samples was identified by fourier transform infra red (FTIR, Bruker Optics, Germany) to predict formation of C-dots in prepared samples. Photoluminescence properties of samples were characterized by spectrofluorophotometer (RF-5300PC, Shimazu Corp., Japan), while transparency of prepared C-dots thin films were checked by UV-VIS (HR 2000CG-UV-NIR, spectrometer Ocean Optic Corp., Japan).

3. Results and discussion

A series of C-dots in thin films and powders form under white and UV light were shown in Figure 1. As can be seen in Figure 1a, prepared thin film of C-dots composites under white light were colorless and transparent. While under UV light, thin film of C-dots composites emitted homogenous blue yellowish fluorescence light (Figure 1b) which indicated photoluminescence properties of C-dots thin film. The fluorescence emission intensities were gradually increased as the concentration of citric acid raised. The maximum fluorescence intensity was observed at sample with 0.035 g citric acid. These tendencies were consistent with photoluminescence characteristic of its powder forms under white light (Figure 1c) and under UV light (Figure 1d). From these images, it can be imply that C-dots might disperse uniformly in epoxy resin as thin film of photoluminescence materials.

The FTIR spectra of C-dots powders as a function of citric acid weight can be seen in Figure 2. Generally, similar infrared spectral behaviour was observed for all of samples, except for sample prepared from citric acid 0 g. Generally, the spectra consist of three main peaks: around 1600-1770 cm^{-1} attributed to C=O bond, around 1350-1460 cm^{-1} belongs to CH_2 bond, and weak peak around 3000-3400 cm^{-1} assigned to hydroxyl and amino groups. These FTIR spectra were consistent with previous result for C-dot sample on Ref. [4]. It implied that C-dots powder was successfully synthesized from commercial urea and citric acid as the precursors by microwave-assisted method.

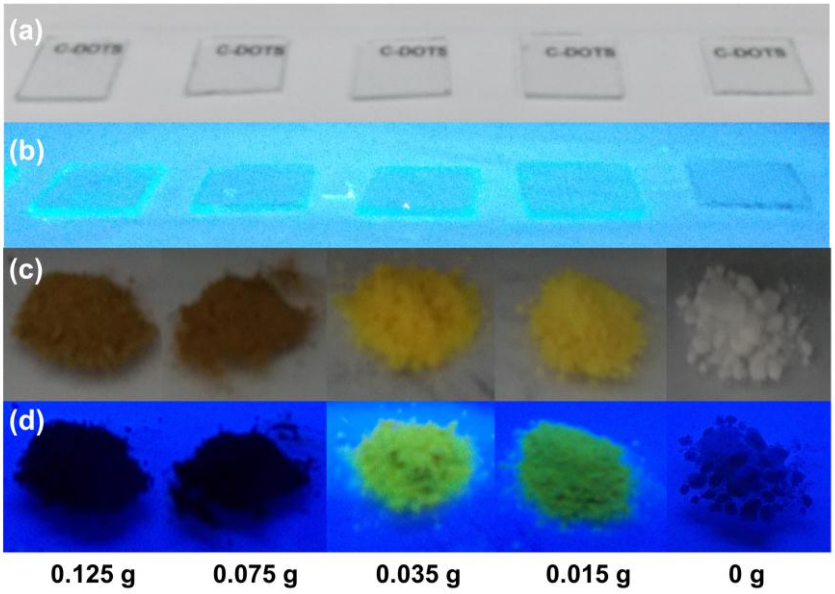

Figure 1. Photography images of the prepared C-dot embedded in the resin as a luminescence thin film (a), and irradiated under UV-light (b). Prior, the samples are produced in the powder form as C-dot luminescence material (c), and irradiated under UV-light (d).

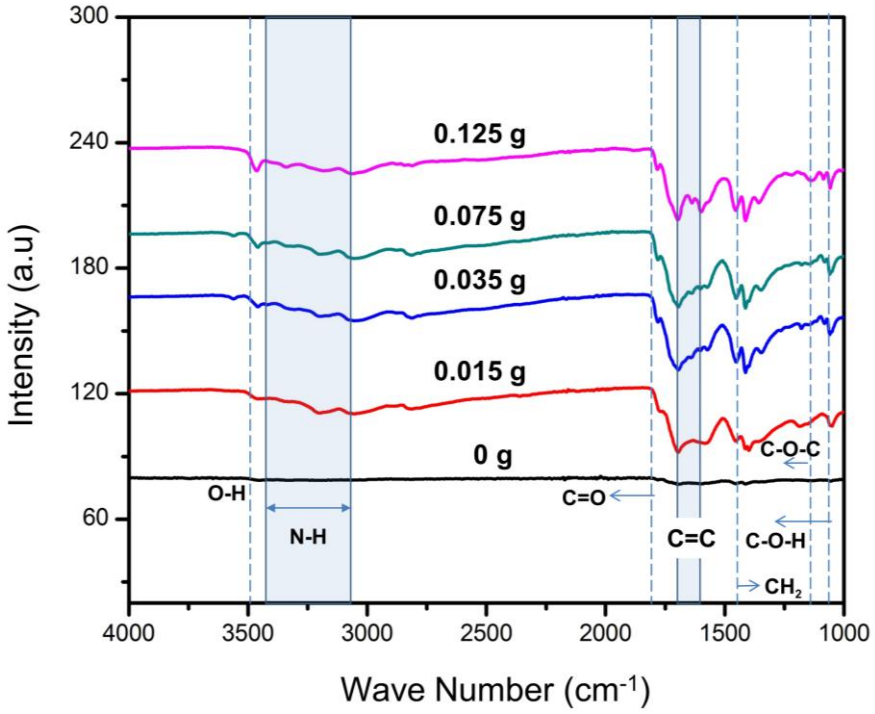

Figure 2. FTIR results of the prepared C-dot samples with citric acid mass variations: 0, 0.015 g, 0.035 g, 0.075 g, and 0.125 g.

The photoluminescence results of synthesized C-dots are shown in Figure 3. This result demonstrated that all samples have fluorescence spectrum in the visible region of 400-650 nm with different intensity as a function of citric acid weight in the samples. The addition 0.015 g citric acid into the precursor gives a high intensity with the emission wavelength at 524 nm. Increasing citric acid concentration into 0.035 g gives the highest PL intensity with emission wavelength at 541 nm under excitation at 365 nm. However, further increasing citric acid concentrations up to 0.125 g turn to reduce the PL intensity. It gives an evident that a small amount (~ 0.035 g) of carbon can give an intense PL effect. On the other hand, a lot of amount of C source may absorb the ray and did not give fluorescence effect.

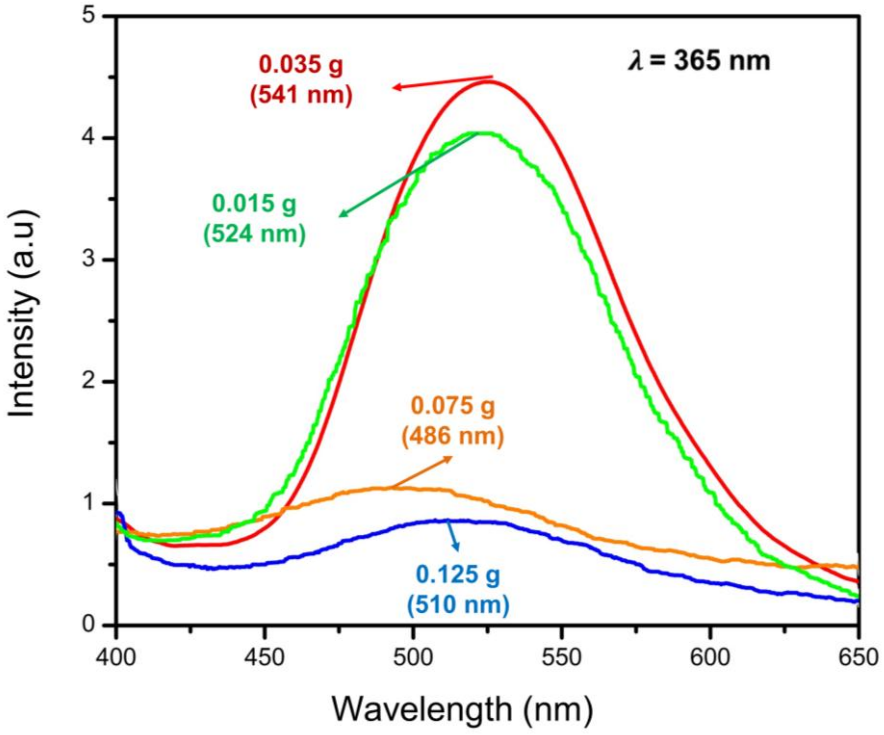

Figure 3. Photoluminescence (PL) spectra of the prepared C-dot samples with citric acid mass variations: 0, 0.015 g, 0.035 g, 0.075 g, and 0.125 g.

The transmittance measurements for the thin films are shown in Figure 4. The result confirms that the prepared thin film has a high transparency in the range 80-90%. From this data, it showed a trend that the more citric acid in the sample, the less the transparency of the sample. It is clear that carbon gives a brownish color to the films sample. Thus, it may reduce the transparency of the thin film. However, for sample whose gives a high PL intensity has 89 % of transmittance.

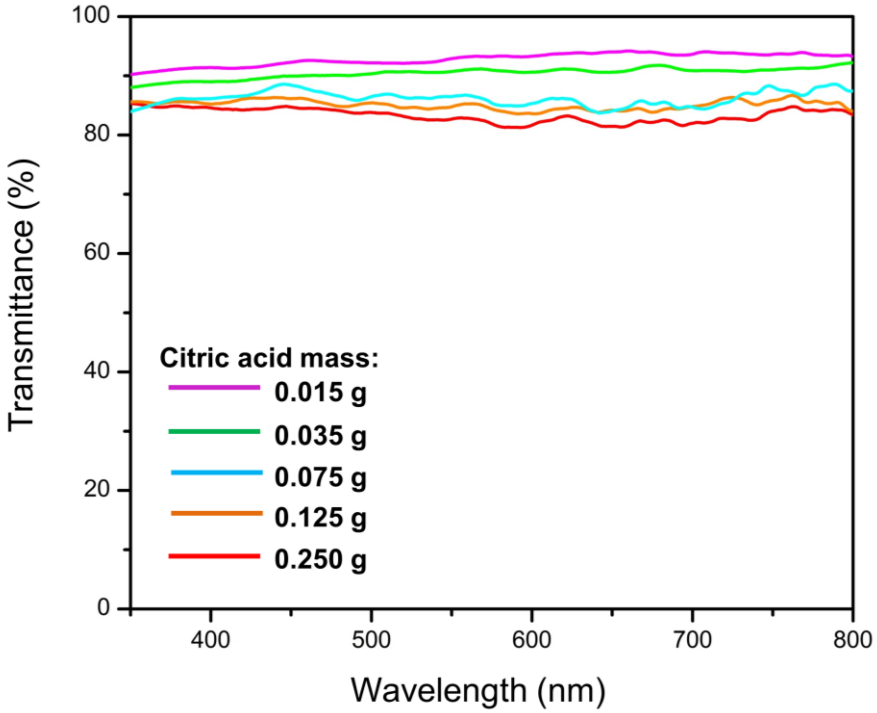

Figure 4. Transmittances result of the thin film C-dot embedded in the epoxy resin with citric acid mass variations: 0, 0.015 g, 0.035 g, 0.075 g, and 0.125 g.

4. Conclusion

Thin films C-dots-composite have been prepared by using spin-coating method on a glass substrate. It was made by embedded the C-dots powder in epoxy resin and deposited as thin film phosphor. Prior thin film deposition, the C-dots powder was synthesized by using microwave-assisted method with commercial urea and citric acid as the precursors. The prepared thin films of C-dots composites have colorless and transparent color and emit blue yellowish fluorescence light. It has a high transparency in the range 80-90%. However, it was observed that a small amount (~ 0.035 g) of carbon can give an intense PL effect. On the other hand, a lot of amount of C source may absorb the ray and did not give fluorescence effect. From these result, it may open an opportunity for optoelectronics devices.

Acknowledgement

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