Chemical Engineering Journal 287 (2016) 511–515

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Short communication

New design of hybrid remote phosphor with single-layer graphene for application in high-power LEDs

Young-Hyun Song [a,1], Eun Kyung Ji [b,1], Sang-Hwan Bak [a,1], Ye Na Kim [a], Dong Bok Lee [a], Mong Kwon Jung [c], Byung Woo Jeong [d], Dae-Ho Yoon [a,b,*]

[a] School of Advanced Materials Science & Engineering, Sungkyunkwan University, Suwon 440-746, Republic of Korea
[b] SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University, Suwon 440-746, Republic of Korea
[c] Hyosung Corporation, R&D Business Labs, Anyang 431-080, Republic of Korea
[d] LG Electronic, Material & Device Advanced Research Institute, Seoul 137-724, Republic of Korea

HIGHLIGHTS

- Remote phosphor using graphene was prepared for the generation of white LEDs.
- Remote phosphor using graphene showed higher luminous flux properties.
- Graphene plays an important role as a high thermal property.
- Remote phosphor is a good candidate of heat dissipation in optoelectronic devices.

ARTICLE INFO

Article history:
Received 2 October 2015
Received in revised form 24 November 2015
Accepted 25 November 2015
Available online 30 November 2015

Keywords:
Remote phosphor
Graphene
High power LEDs

ABSTRACT

A hybrid remote phosphor with single-layer graphene was developed for the fabrication of warm white LEDs, and with an off-the-shelf blue LED operating at 350 mA it produced a high luminous flux of ~64 lm at 100 °C. The remote use of a single layer of graphene with a phosphor has a multi-functional role that improves the luminous properties, long-term stability and thermal properties of LEDs. We suggest that it can be expected to play an important role as a promising candidate for next-generation remote phosphors of high-power LEDs.

1. Introduction

White light-emitting diodes (LEDs) with a blue LED chip and yellow emitting phosphor ($Y_3Al_5O_{12}$: Ce^{3+}) embedded in epoxy have been highly popular for solid state lighting due to their advantages of energy savings, compactness, long lifespan, high efficiency, and environmental friendliness [1–5]. The $Y_3Al_5O_{12}$: Ce^{3+} phosphor for the generation of white light possesses good luminescence properties such as excellent chemical stability, a typical broad emission band, and outstanding absorption of the light of blue LED chips [6,7]. The drawback of $Y_3Al_5O_{12}$: Ce^{3+} phosphor is its thermal dissipation characteristics, so that the correlated colour temperature is unstable with an increasing forward bias current [8]. For suitability of use in high-power LEDs, high thermal properties are required [9,10].

The alternative is a remote phosphor. Generally, the remote phosphor structure separates the phosphor layer away from the blue LED chip [11]. White LEDs using remote phosphors can avoid heat generation. As a result, a colour-stable and thermally stable design of white LEDs can be achieved.

According to Hong et al., quantum dot/polymer nanohybrid films are reported for white LEDs [12]. Generally, it is not only the quantum dot that is unstable to moisture and heat, as the polymer is also degraded with increases of temperature in blue LED chips from a forward bias current.

Graphene materials are seen as an alternative solution to the above problems. Graphene has attracted considerable attention because of two excellent physical properties, its flexibility and high intrinsic thermal conductivity, which come about because of the strong bonding of the light carbon atoms [13,14]. So, graphene has been focused on for its possible application in optoelectronic devices, to overcome the heat dissipation problems.

* Corresponding author at: School of Advanced Materials Science & Engineering, Sungkyunkwan University, Suwon 440-746, Republic of Korea. Tel.: +82 31 290 7388; fax: +82 31 290 7410.
E-mail address: dhyoon@skku.edu (D.-H. Yoon).
[1] These authors contributed equally to this work.

http://dx.doi.org/10.1016/j.cej.2015.11.093
1385-8947/

A remote phosphor used with a single layer of graphene has not previously been reported anywhere. We report on a $Y_3Al_5O_{12}$: Ce^{3+} remote phosphor that uses a single layer of graphene in application to high-power LEDs.

We fabricated graphene with $Y_3Al_5O_{12}$: Ce^{3+} phosphor to exploit the excellent thermal properties of graphene for remote use with LEDs, so as to render their output white. To the best of our knowledge, this is the first reported attempt at using such hybrid materials.

With operation at a colour temperature of 5300 K the hybrid structure respectively saves more than 17% and 37% in phosphor usage, compared to conventional remote and conformal structures. The effect on the operation of the LEDs of the down-conversion characteristics of the phosphor hybrid structure was found to be a superior luminous flux.

2. Experimental

A graphene single layer was grown on copper foils using a chemical vapour deposition (CVD) method. Firstly, the Cu foil (99.8%, Alfa-Aesar, item No. 13382) was loaded into a quartz tube of the CVD system and annealed at 1000 °C and under 90 mTorr with 10 SCCM of H_2 flowing for 60 min, to enlarge the sizes of the graphene crystals on the Cu foil. The growth was initiated using reaction gas mixtures (CH_4/H_2 = 15:10 SCCM) under 560 mTorr for 25 min, followed by rapidly cooling the sample to room temperature (10 °C min^{-1}) with flowing H_2 under 90 mTorr.

For the synthesis of $Y_3Al_5O_{12}$: Ce^{3+} phosphor, a spray pyrolysis method was utilized. The yellow emitting remote phosphor was fabricated with polydimethylsiloxane (PDMS), $Y_3Al_5O_{12}$: Ce^{3+} phosphor and hardener [15]. The mixture was dropped on the graphene/Cu foil. The deformation of the sample was at 40 °C in a vacuum oven and it was cured at 100 °C for five hours. Finally, the Cu foil of the sample was etched away. A schematic illustration of the remote phosphor using a single layer of graphene is shown in Fig. 1.

The crystalline phase of the $Y_3Al_5O_{12}$: Ce^{3+} phosphor was identified using powder X-ray diffraction (XRD, D-MAX 2500, Rigaku) with a CuKα target over the range $20° \leqslant 2\theta \leqslant 80°$. The photoluminescence properties including the external quantum efficiency of the prepared samples were analysed by room temperature photoluminescence spectrometry (PL, PSI Co., Ltd., Korea), equipped with a 500 W Xenon discharge lamp as an excitation source. The luminous properties of the hybrid remote phosphor under a 450 nm blue LED chip as a function of input current and temperature was investigated with an Instrument Systems CAS-140CT.

3. Results and discussion

Fig. 2(a) shows the XRD patterns of $Y_3Al_5O_{12}$: Ce^{3+} phosphors. All of the diffraction peaks were matched to the pure cubic $Y_3Al_5O_{12}$ phase following JCPDS Card No. 73-1370. The Ce^{3+} ions had little effect in the host $Y_3Al_5O_{12}$ materials other than bringing about an increase of the lattice parameters of the $Y_3Al_5O_{12}$ because of the ionic radii of the Y^{3+} ion (0.088 nm) and Al^{3+} ion (0.0675 nm) compared to the Ce^{3+} ions (0.118 nm) [16]. A selected SEM image of the $Y_3Al_5O_{12}$: Ce^{3+} phosphor deficits are shown as an inset of

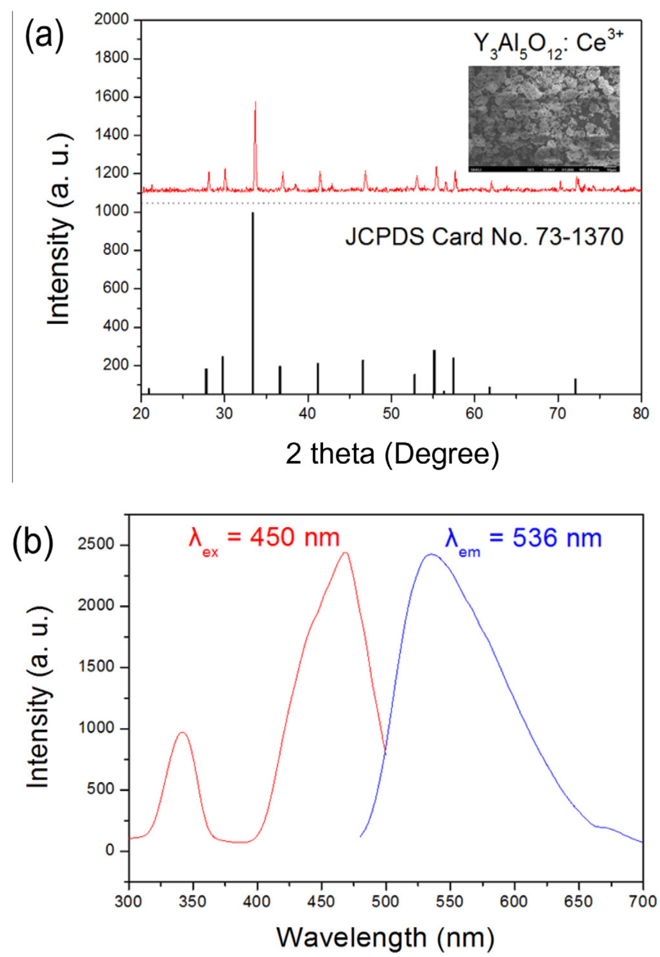

Fig. 2. (a) XRD patterns of $Y_3Al_5O_{12}$: Ce^{3+} phosphors by spray pyrolysis. The inset shows the SEM image. (b) PLE and PL spectra of the sample.

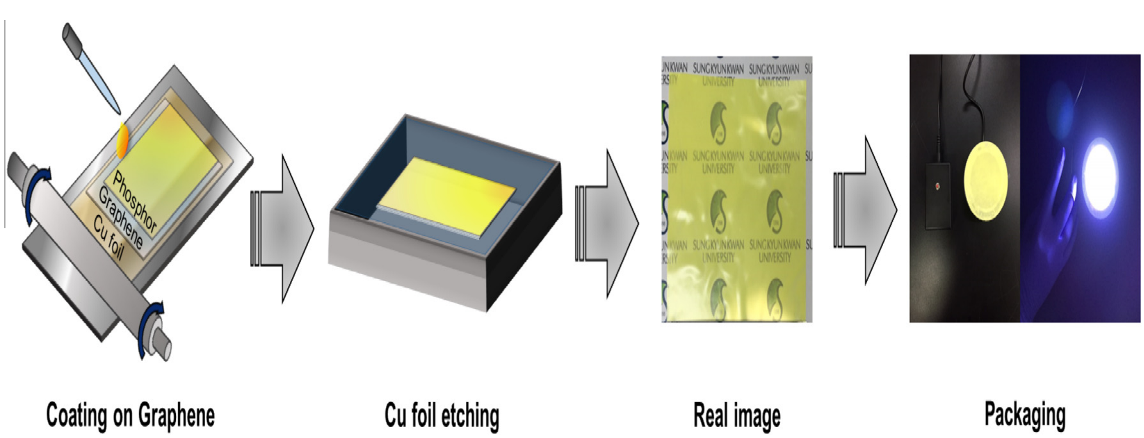

Fig. 1. Schematic of fabrication of remote phosphor using single-layer graphene.