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# Large-area color controllable remote carbon white-light light-emitting diodes

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ABSTRACT

We disperse carbon nanodots (C-dots), which can be achieved by decomposing organic acid in silane coupling agent, in epoxy oligomer to realize large-area C-dots film on any transparent substrate. Due to the similarity in solubility parameters between C-dots and epoxy resin, aggregation of C-dots in epoxy resin is suppressed. It is found that the C-dots films can support high conversion efficiency (>60%) white-light emission under 460 nm illumination. In addition, the corresponding correlated color temperature can be varied between 2500 and 10,000 K by controlling the thickness of the C-dots film. We also verify that the absolute performance of C-dots film is compatible with that of the commercial available phosphor film.



## 1. Introduction

Current trends of white-light light-emitting diodes (LEDs) design shifts to the remote phosphor technology, which required the suspension of rare-earth phosphor film over blue LEDs, has been widely adopted by the manufacturers to produce remote phosphor white-light LEDs [1,2]. As various correlated color temperature (CCT) can be obtained from white-light LEDs by using suitable composition of rare-earth materials, the remote phosphor technology standardizes the use of blue LEDs [3]. Furthermore, the application of remote phosphor technology can avoid prolong heating of rare-earth phosphor. As a result, reliable, low-cost and CCT tunable white-light LED architecture can be achieved simultaneously [4]. Nevertheless, the need of phosphor restricts the further development of remote phosphor white-light LEDs with large emission area [5]. This is due to the shortage supply of phosphor arises from the increase in the consumption of fluorescent lamps after the phase out of incandescent light bulbs [6], and the restriction of high pollution mining and refining of rare-earth materials [7].

Recently, extensive investigations had been concentrated on the use of functionalized carbon nanodots (C-dots) [8,9] to replace ‘phosphor’ in remote phosphor white-light LEDs. This is because carbon is one of the most abundance elements on earth, environmental friendly, and the fabrication procedures of free-standing C-dots film can be compatible with the manufacturing of remote phosphor [10]. In fact, direct deposition of C-dots [11], mixture of C-dots with silicone [12], C-dots embedded inside polymer matrix [13,14]

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and C-dots/agar composite [15] onto the surface of either deep-blue or ultraviolet (UV) LEDs (i.e., conventional 'phosphor on chip' design) had demonstrated white-light emission of various CCT. Furthermore, freestanding large-area C-dots films had been fabricated by either embedding C-dots inside polymer matrix [16] or casting of C-dots in water solution onto a large glass sheet [17]. However, there are two aspects of challenges hinder the realization of large-area 'remote carbon films'. Firstly, physical dispersion of C-dots in solid media may lead to low doping concentration of C-dots (i.e., low solubility in solid media) so that low photoluminescence (PL) quantum yield is unavoidable. In addition, phase separation and aggregation of C-dots (i.e., due to the high viscosity of liquid media) induce fluorescence quenching [8,9]. Secondly, the use of deep-blue and UV LEDs as the excitation sources will increase the total manufacturing cost of white-light LEDs as the most cost effective LED sources are emitted at wavelength various between 450 and 460 nm.

Here, we utilize previously developed high-efficiency white-light emission C-dots film (i.e., C-dots disperse inside epoxy) to realize large-area remote carbon white-light LEDs under 460 nm excitation [18,19]. With suitable fabrication procedures, C-dots can be homogeneously dispersed in epoxy without aggregation so that uniform white-light emission can be obtained over a large surface area. This is due to the high solubility of C-dots in epoxy. It can also be shown that CCT of the C-dots film can be adjusted from 2500 to 10,000 K through the control of its thickness. Furthermore, the absolute performance of the C-dots film is evaluated by comparing its emission characteristics with the commercial available remote phosphors (i.e., Intematix-ChromaLit™ Round XT/XTS [20]). It is noted that although the maximum emission intensity of C-dots film is about 3 times lower than that of the commercial rare-earth phosphors, the performance of C-dots film is not too far from practical applications. Hence, C-dots films can have potential to replace 'phosphor' in white-light LEDs.

## 2. Fabrication and characterization of C-dots films

Functionalized C-dots were fabricated by decomposing organic acid in silane coupling agent, then the as-prepared C-dots were mixed with commercial epoxy oligomer under vigorous stir and subsequently degassed for 10 min in vacuum (i.e., $10^{-4}$ bar) environment. Finally, the mixture was cured into solid C-dots film under continuous heating at 60 °C for 6 h. Concentration of C-dots in epoxy matrix can be controlled from 0% to 100% by varying the mass ratio of C-dots to epoxy (i.e., 50% represents both as-prepared C-dots and epoxy have the same mass). Detailed fabrication procedures of C-dots films can be found in our previous publication [18].

Morphology of the C-dots was investigated by using a JOEL JEM-2100F high-resolution transmission electron microscope (HR-TEM). Thermo gravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed by using a NETZSCH STA 449 Jupiter thermal analyzer. Photoluminescence was characterized by an EDINBURGH FLS920 fluorescence spectrometer. Emission characteristics of the remote carbon white-light LEDs were measured by using a LABSPHERE light measurement system with CDS-600 spectrometer and LMS-100 integration sphere.

## 3. Results and discussion

C-dots, which have size varies from 2.0 to 3.6 nm and lattice spacing of 0.2 nm, are uniformly dispersed inside epoxy (see Figs. S1a and b of the supporting information). The proposed C-dots consist of $sp^2$ graphite structure carbon core and surface organosilane functional groups which support effective photoluminescence under 460 nm excitation. Fig. 1a shows a schematic diagram of the silane functionalized C-dots with A-2120 functional groups. It is believed that the C=O functional group leads to board bandwidth (from 400 to 700 nm) emission under a range of excitation wavelength [21]. In addition, the presence of O=C—NH functional group enhances effective absorption of blue excitation [22] (see Fig. S2a). For the C-dots film (i.e., C-dots dispersed inside epoxy), the emission intensity and peak wavelength are a function of the concentration of C-dots. Maximum emission intensity under 460 nm excitation can be obtained with 50% concentration of C-dots (see Fig. S2b). As the location of emission peak (i.e., ~550 nm) and emission linewidth (i.e., ~120 nm) of 50% concentration of C-dots under 460 nm excitation are similar to that of the rare-earth phosphor, they are the suitable replacement of 'phosphor' in remote phosphor white-light LEDs. The conversion efficiency of C-dots is found to be larger than 60% under 460 nm excitation at room temperature. A remote carbon white-light LED can be constructed by depositing C-dots film onto a polystyrene substrate (with diameter and thickness of 60 and 1.5 mm respectively and dull polished on both surfaces) as the 'remote carbon'. Scattering and absorption coefficient of the polystyrene substrate at 460 nm is estimated to be 1 $cm^{-1}$. Fig. 1b shows the cross-section of a 400 μm thick C-dots film on the plastic substrate. It is observed that the substrate can physically support the C-dots film to avoid deformation or vibration, and acts as a light diffuser to homogenously radiate white-light emission from the C-dots film. An array of 460 nm LEDs (EPISTAR LED chip [23]), which has a maximum electrical power of 20 W, is used to excite the C-dots film. In addition, a light concentrator is placed between the blue LED array and C-dots film to concentrate light onto the C-dots film, see Fig. 1c (and Fig. S3). Fig. 1d compares the distribution of light intensity from the surface of the substrate with and without coating of C-dots film. The variation of intensity is measured along two orthogonal directions of the substrates; see also the insets of Fig. 1d. The intensity distribution observes from both substrates are almost identical and the corresponding radiation profile is mainly related to the LED array and light concentrator. This implies that light is homogenously radiated from the surface of the C-dots film.

Homogeneous light emission from the surface of the C-dots film is mainly due to the solubility of C-dots in the host matrix (i.e., uniform dispersion of C-dots without aggregation). In fact, the solubility of C-dots in the host matrix can be estimated by comparing their solubility parameters. It is noted that the aggregation of C-dots in the host can be