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Optimization of the theoretical photosynthesis performance and vision-friendly quality with multi-package purplish white LED lightings

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This study introduces the new figures of merit for photosynthesis and photo-pigmentation performances as well as vision performances for photosynthetically efficient, vision-friendly and tunable greenhouse LED lighting sources. The recently developed figures of merit, i.e., the photosynthetic luminous efficacy of radiation (PLER, plm/W), the photosynthetic luminous efficacy (PLE, plm/W), the photosynthesis illuminance (PIL), and the photosynthesis action factor (PAF) are measured and added to measurements of the widely used figures of merit of vision and color to compare the four-package white LEDs developed by our group with natural sunlight and the presently commercialized artificial lamps for greenhouse or indoor. In this study, one of the best selections of various four-package white LEDs (λ = 630, 590, 520, and 450 nm) shows excellent photosynthesis performance (PLER = 469 plm/W, PLE = 169 plm/W) and good vision performances (LER = 271 lm/W, LE = 98 lm/W, CRI = 86) at 10,000 K correlated color temperature (CCT) of blue-enriched white LED. Furthermore, this four-package white LED provides tunable capability of spectral power distribution (SPD) of lighting and excellent photosynthesis performance (PLER > 457 plm/W, PLE > 157 plm/W) even at eye-friendly vision performances (LER > 249 lm/W, LE > 87 lm/W, CRI > 85) under purplish emitting conditions. It can be expected that the four-package purplish white LEDs based on LPDF-capped, phosphor-converted monochromatic LEDs satisfy the requirements to attain high photosynthesis efficiency for the improved growth of plants, high energy savings, vision-friendly, and tunable figures of merit for the four-package LED lighting in greenhouse.

Introduction

The most important sustainable-energy production process associated with light in nature is photosynthesis, in which most plants, algae and some bacteria transform solar light into organic compounds that can be used as energy sources to maintain life. Generally, artificial light are generated for photosynthesis from traditional lighting sources, such as high pressure sodium (HPS) lamps and other metal halide (MH) in greenhouse which are production facilities for massive cultivation of vegetables or flowers. Recently, light-emitting diodes (LEDs) are an attractive solid-state lighting technology for the greenhouse industry compared with the currently commercialized lightings, such as HPS, MH and incandescent lamps due to their distinct advantages of the high energy efficiency for photosynthesis, the low radiant heat output, easy control of spectral composition and facile adaptation of light intensity to the plant photoreceptors.[1-4] In addition, an LED can be easily integrated with various sensors and digital controllers so as to manipulate smart lighting programs for photosynthesis of plants such as varying the emission ratio of blue to red colored LED, frequency, and intensity over plant species and the growth stages.[5] Therefore, over last two decades, many researchers have made a lot of effort to make LEDs an energy efficient lighting alternative to improve the flowering and photosynthetic efficiency of plants in the greenhouse.[6-10]

The impact of LEDs on photosynthesis is closely related to the match level of the spectral power distribution (SPD) of LED lamps with the photosynthesis action spectrum (PAS, P(λ)), which represents the efficiency of each wavelength in inducing photosynthesis for averaged vegetable species.[11] Actually, PAS (P(λ)) curve differs from one species to another because different plant or microorganism species use their own combination of photoreceptor pigments for photosynthesis. After we compare two of them, we selected the PAS (P(λ)) curve adopted by the German Institute for Standardization and numbered DIN 5031-10 in this study.[12] PAS (P(λ)) curve is directly related to the chlorophyll a (CL *a*) and chlorophyll b (CL *b*) pigments because they are the most common pigments for photosynthesis in green plants. These two pigments show two higher sensitivity peaks in one in the blue (B) region at ~450 nm and the other in the red (R) part of the spectrum at ~660 nm.[12, 13] Therefore, the excellent match in the blue and red wavelengths of PAS (P(λ)) curve and the emission spectrum of combined blue and deep-red LED light sources promotes photosynthesis in most green plants under these lighting environments. Moreover, additional colored LEDs, which emit at different wavelengths, can be easily added as well to enhance photosynthetic efficiency.

It is known that the range of wavelengths which most plants can use for photosynthesis has evolved over the very long time, using effectively the broad spectrum of sunlight. To date, most researchers have used the narrow-band B and R semiconductor LEDs for the application of LEDs in horticulture.[14-17] As previously reported, different semiconductor material systems such as InGaN for blue LED and AlGaInP for red LED behave very differently with respect to the surrounding temperature.[18] In addition, the low efficiency of semiconductor green and amber LED (green gap or yellow gap) limits the application of these colored LEDs into greenhouse lightings. So far, there is a little study to investigate the suitability of the other colored LED, except for blue, red, and white LED, to a dedicated light source for optimal photosynthesis of plants in the greenhouse. To solve these problems, there is one possible solution based on the use of phosphor-converted LED (pc-LED) which converts blue into any colors in the wavelength rage between green and near IR. The selection of color-by-blue approach is due to the existence of an ultra-efficient and stable InGaN blue LED, the development of excellent blue-mirror-yellow-window dichroic filter, and many of high efficient green, amber, red, and deep-red phosphors excited by blue. The choice of good phosphors in the pc-LED provides a variety of colored light, tunable emission of photoluminescence (PL) spectrum between 500 and 670 nm, the improved efficiency of green and amber LEDs in the "green gap" wavelength ranges, and the improved temperature stability of amber to deep-red range of pc-LEDs, compared with less temperature stable semiconductor amber- and red-emitting LED.[18] However, there is no concerted approach to controlling the SPDs of colored pc-LED lamps using the unique spectral tunability of various phosphor materials to optimize their effects on the efficiency of photosynthesis. Therefore, it is necessary to consider the figures of merit to estimate and control the potential impact of the SPD of monochromatic pc-LEDs or a combined multi-package of colored pc-LEDs and a blue LED on the photosynthesis action spectrum (PAS (P(λ)) and photo-pigment sensitivity action spectrum (PpAS (Pp(λ)) curve of photosynthesis. Here, p denotes the photo-pigment in photoreceptor of plants.

In this study, first, we demonstrate meaningful figures of merit required for explaining the photosynthesis efficiency and photo-pigment action efficiency of light sources. Using the same concept of defining luminous efficacy of radiation (LER, lm/W) for human vision, both the photosynthetic luminous efficacy of radiation (PLER, plm/W) and photo-pigmented luminous efficacy of radiation (PpLER, pplm/W) are suggested to explain how photosynthetically bright the radiation of the emission spectrum are as perceived by the photoreceptor system of the average plants and each pigment in different plant species. Recently, we created a series of $R_{B,M}$, $A_{B,M}$, and $G_{B,M}$ monochromatic LED which represents long wavelength pass dichroic filter (LPDF)-capped, monochromatic red, amber, and green pc-LEDs pumped by a blue LED chip to enhance the luminous efficacy of green gap LEDs, improve the temperature stability and control the peak wavelength and bandwidth of pc-LEDs.[18-21] B denotes an InGaN blue LED. Here, using our monochromatic pc-LED concept, we fabricate LPDF-capped green, amber, red, and deep-red full down-converted monochromatic pc-LEDs using various phosphors. Next, we characterize the photosynthetic and visual optical properties of LPDF-capped, monochromatic pc-LEDs as well as semiconductor type of monochromatic LEDs to determine their suitability of the application into photosynthetic reactions induced by specific photo-pigments in plants. Finally, we demonstrate the photosynthetic optical properties of sunlight at daytime, commercialized artificial lighting sources and multi-package pc-LED systems that consist of two, or four combination of $R_{B,M}$, $A_{B,M}$, and $G_{B,M}$ pc-LEDs, and a blue LED in an effort to propose the way to find the higher PLER, PLE, PpLER, and PpLE of multi-package pc-LEDs. In detail, we also dynamically control the ratio and shape of reddish and bluish emissions of both RB two-package and $R_{B,M}A_{B,M}G_{B,M}B$ four-package pc-LEDs with the change of emission wavelength of red pc-LEDs without sacrificing photosynthetic energy efficiency to evaluate the feasibility of multi-package pc-LEDs toward an excellent photosynthetic lighting source, vision-friendly color, as well as a smart tunable greenhouse lighting source.

Experimental

Fabrication of partially-converted cyan pc-LED.[18-21] To fabricate partially-converted cyan pc-LED, an InGaN blue LED (λ_{max}=445 nm) was used as an excitation source for various color phosphors of pc-LEDs. The blue LED chips were purchased from Dongbu LED, Inc. The cyan phosphor ($(Ba,Sr)_2SiO_4$:Eu) was purchased from Merck Co.. 10 wt% ~ 50 wt% of cyan phosphor ($(Ba,Sr)_2SiO_4$:Eu) were dispersed in a silicone binder to create a phosphor paste, and the same amounts of resulting phosphor pastes were dropped onto a cup-type blue LED. After dropped the phosphor paste onto a blue LED, the paste was hardened by heating in each case. The partially-converted cyan pc-LEDs were named with its color and wt% ((e.g.,) 10 wt% partially-converted cyan pc-LED: C10 wt%).

Fabrication of LPDF-capped monochromatic red/amber/ yellow/green pc-LEDs.[18-21] To fabricate LPDF-capped monochromatic red/amber/yellow/green pc-LEDs, an InGaN blue LED (λ_{max}=445 nm) was used as an excitation source for various color phosphors of pc-LEDs. The fabrication method of LPDF is in the ESI experimental section. The blue LED chips were purchased from Dongbu LED, Inc. The green/yellow phosphors were purchased from Merck Co. and amber/red phosphors were purchased from Intematix cooperation. The narrow-band red emitting phosphor, K_2SiF_6:Mn, was synthesized from our laboratory (see experimental in the ESI).[22]

Optimum amounts of each color phosphor were dispersed in a silicone binder to create a phosphor paste, and the same amounts of resulting phosphor pastes were dropped onto a cup-type blue LED to create each colored pc-LED. After dropped the phosphor paste onto a blue LED, the paste was hardened by heating. The LPDFs (L535 for yellow/green and L550 for red/amber) were capped on top of the pc-LEDs with an air gap to realize full down-converted pc-LEDs to realize monochromatic color. The green/amber/red semiconductor-type LEDs were referred to according to their colors ((e.g.,) red LED: R630S and R660S, amber LED: A590S, and green LED: G517S); the blue LED was named based on its color and wavelength (B450S); and the pc-LEDs were named based on their color and wavelength ((e.g.,) and a LPDF-capped green pc-LED with an emitting wavelength of 520 nm: G520).

Characterization of partially converted cyan pc-LEDs, LPDF-capped red/amber/yellow/green pc-LEDs, and four-package white LEDs. The emission spectra and luminous flux of the forward emission from cyan pc-LEDs, LPDF-capped red/amber/yellow/green pc-LEDs, and red/amber/green/blue semiconductor-type LEDs were measured in an integrated sphere using a spectrophotometer (PSI Co. Ltd., Darsapro-5000) with an applied current of 60 mA (rated current). The emission spectra and luminous flux of the two- and four-package LED systems were measured in an integrated sphere using a spectrophotometer (PSI Co. Ltd., Darsapro-5000) while controlling the applied current of each primary LED with a total applied current of 240 mA. A set of four-package pc-LEDs (R630, A590, G520, B450S) was selected for the reference primary LEDs to compare the optical properties with a set of four CCTs (10,000 K, 6,500 K, 3,500 K, and 2,000 K) while changing each colored pc-LED.

Results and discussion

As shown in the PAS (P(λ)) and PpAS (Pp(λ)) curve in Figs. 1a and 1b, chlorophylls (CL *a* and CL *b*) play an important role in the photosynthesis and absorb strong blue and red wavelength of light.[23] However, plants have other antenna pigments, such as the carotenoids β-carotene (*β*-CT), phycoerythrin (PE), and phycocyanin (PC) etc., to absorb different colors from both blue and red light for supporting the growth of plants (Fig. 1b). In addition, several reports have shown that plant growth, flowering, and metabolite to the color of absorbing light is plant species specific. It is also reported that green light can contribute to the plant growth and development.[24-26]

To select the proper SPD of phosphors and monochromatic pc-LEDs, the effects of light quality of colored pc-LEDs on the photosensitivity curves of all photo-pigments for contributing photosynthesis of plants should be considered. Nearly all of the photosynthetic performance indicators of monochromatic pc-LED and LED light, such as the photosynthesis and photo-pigment luminous efficacy, and illuminance, can be calculated on the basis of P(λ) and Pp(λ), as using the same concept as the calculation of figures of merit of vision performance of artificial lightings based on photopic sensitivity action curve (V(λ)).[27,28] Similarly, the photo-pigment action efficiency of each pc-LED can be also calculated on the basis of sensitivity curve of all photo-pigments as shown in Fig. 1b.

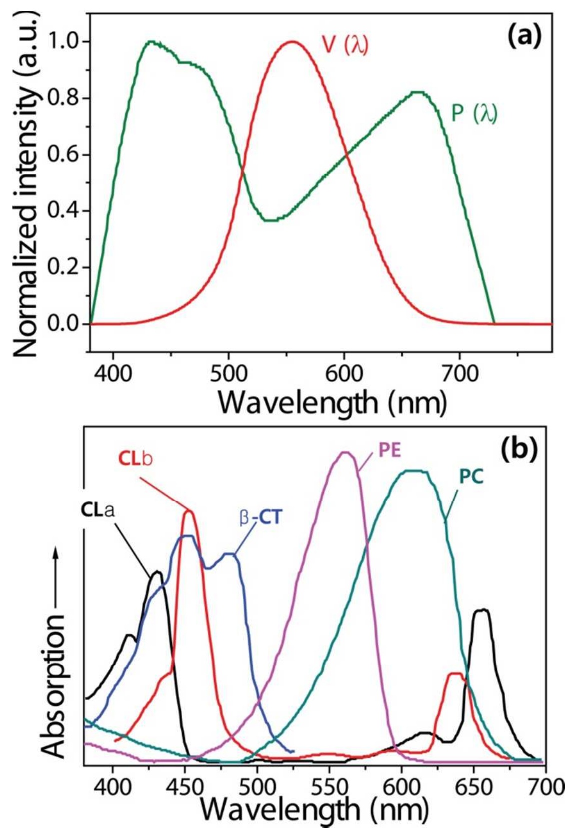

Fig. 1 (a) Normalized spectra of photopic sensitivity action curve (V(λ)) and photosynthesis action spectrum (PAS, P(λ)). (b) Absorption spectra of photo-pigment sensitivity action spectrum curve of photosynthesis (PpAS, Pp(λ)). CL*a*: Chlorophyll a, CL*b*: Chlorophyll b, *β*-CT: *β*-carotene, PE: phycoerythrin, and PC: phycocyanin.

Accordingly, the photosynthetic luminous efficacy of radiation (PLER, K_p) and photo-pigmented luminous efficacy of radiation (PpLER, K_{pp}) can be defined as the ratio of the photosynthesis luminous flux to the radiant flux (S(λ)) and photo-pigment luminous flux to the radiant flux (S(λ)), respectively, using the same concept to calculate the luminous efficacy of radiation (LER), in lumens per watt (lm/W), is a parameter explaining how visually bright the radiation of the emission spectrum is as perceived by the average human eye (See Eq. 1 and Eq. 2).[29]

$$K_p = K_{p0} \int_{380\text{nm}}^{780\text{nm}} \mathrm{P}(\lambda)\mathrm{S}(\lambda)\mathrm{d}\lambda / \int_0^{\infty} \mathrm{S}(\lambda)\mathrm{d}\lambda \qquad \text{Eq. 1}$$

$$K_{pp} = K_{pp0} \int_{380\text{nm}}^{780\text{nm}} \mathrm{Pp}(\lambda)S(\lambda)\mathrm{d}\lambda / \int_0^{\infty} S(\lambda)\mathrm{d}\lambda \qquad \text{Eq. 2}$$

Where P((λ) is the photosynthesis action spectrum and Pp(λ) is the photo-pigment action spectrum of each pigment. K_{p0} and K_{pp0} is the maximal spectral photosynthesis and photo-pigment efficacy value for a photosynthetic system (= 683 plm/W and 683 pplm/W). Therefore, PLER and PpLER were clearly defined as parameters explaining how photosynthetically bright the radiation of the emission spectrum are as perceived by the photoreceptor system of the average plants and each pigment in the most plant species. As previously reported, the photosynthesis action factor (PAF) and the photo-pigment action factor (PpAF) are defined Eq. 3 and Eq. 4 as the ratio PLER to LER and PpLER to LER, respectively.[11]

PAF (plm/lm) = PLER (plm/W) / LER (lm/W) Eq. 3

PpAF (pplm/lm) = PpLER (pplm/W) / LER (lm/W) Eq. 4

To assess the vision and photosynthesis performances of actual monochromatic LED, multi-package LED, and general artificial lighting systems, several figures of merit should also be considered. The first important figure of merit for the visual energy efficiency of real lighting source is the luminous efficacy, as measured in lumens per watt (lm/W). In addition, the external quantum efficiency (EQE, η_e) of each lighting system is defined as the ratio of the luminous efficacy (LE, η_{lm}) to the LER. With the same logical sense, the photosynthesis and photo-pigmented luminous efficacy (PLE, η_p, and PpLE, η_{pp}) are defined by multiplying the PLER and EQE (and the PpLER and EQE) of each lighting source, respectively (See Eq. 5, Eq. 6 and Eq. 7).[21]

$\eta_e = \eta_{lm}$ / LER, Eq. 5

η_p = PLER x η_e Eq. 6

η_{pp} = PpLER x η_e Eq. 7

It is possible to evaluate monochromatic LED, multi-package LED, and pc-LED lightings of alternative light source by acquiring the spectral distributions and photosynthetic (photo-pigmented) luminance per watt (plm/W (or pplm/W)) and computing the corresponding figures of merit, in this case the photosynthesis luminous efficacy and photo-pigmented luminous efficacy.

To determine the minimum amount of light from individual artificial light sources to improve the photosynthetic efficiency of greenhouse system, it is also important to measure the photosynthetic (or photo-pigmented) illuminance (PIL (or PpIL)) of the light source instead of relying on the visual illuminance (VIL) value. Similar to the illuminance measurement method, the PIL (or PpIL) can be obtained by measuring the total photosynthetic (or photo-pigmented) luminous flux incident on a surface per unit area. The only different concept when measuring the PIL (or PpIL) from the VIL is the use of the photosynthesis (or photo-pigment) action spectrum (P(λ) (or Pp(λ))) instead of the CIE photopic action curve (V(λ)) while calculating the photosynthesis luminous flux.[30] Simply, the PIL and PpIL can be defined from the equation below:

PIL (plm/m^2, plx)
= PAF (plm/lm) x VIL (lm/m^2, lx) Eq. 8

PpIL (pplm/m^2, pmlx)
= PpAF (pplm/lm) x VIL (lm/m^2, lx) Eq. 9

If we know the PAF or PpAF of any type of light source and the VIL, we can calculate the PIL or PpIL of light source under a specified condition. Consequently, one must bear in mind that a high PAF (or PpAF) and a high PIL (or PpIL) value for individual artificial light source are achieved under the same VIL conditions. Furthermore, the LED lamp spectra, which can have a high PAF (or PpAF) value, have the potential to concentrate the energy in a spectral region that is photosynthesis-sensitive (or photo-pigment-sensitive), thus having a strong impact on photosynthesis of plants in greenhouse system. A variety of efficient monochromatic and multi-package LEDs were characterized to determine the photosynthetic performances as well as vision performances of their pc-LEDs. Figs. S1a-S1f show the normalized photoluminescence (PL) spectra and color coordinates of six RAGB semiconductor LEDs, five partially converted cyan phosphor-coated blue LEDs, and sixteen $R_{B,M}$, $A_{B,M}$, and $G_{B,M}$ LPDF-capped, monochromatic pc-LEDs which fabricated by simply capping of LPDF on top of the InGaN blue LED with each corresponding phosphor. These figures indicate that the emission spectrum and color coordinates of all monochromatic pc-LEDs is well matched with those of the corresponding phosphors in our previous publications.[21] Here, all monochromatic LEDs are denoted as a combination of the color and peak wavelength of the emission spectrum. In addition, S denotes the semiconductor-type of monochromatic LEDs. Also, partially converted cyan LEDs denote color and the weight % of phosphor in paste. All LERs, LEs, PLERs, PpLERs, PLEs, PpLEs, and PAFs of monochromatic colored LEDs for both the visual performance of eye and photosynthesis performance of plants are measured at 60 mA (rated current) and displayed with the change of the peak wavelength of the monochromatic pc-LEDs and semiconductor LEDs in Tables S1a and S1b in electronic supplementary information (ESI). Unfortunately, to date, there are several different photosynthesis action spectra (P(λ)) to evaluate the photosynthetic performance in the different species, for example: Aube et al.,[11] and Purves et al.,[13] are proposed the different P(λ)s, depending on the plant species and measuring conditions. Among these, we compared the different types of photosynthesis action spectra (P(λ)) from Aube et al. (P(λ)), and Purves et al., (P'(λ)). Figs. 2a and 2b show the photosynthesis action spectra and PAF of each blue LED and cyan/green/amber/red LPDF-capped pc-LEDs from 450 nm to 670 nm. Also, we compare the PLERs and PLEs of monochromatic LEDs with different wavelengths of EL emission. As shown in Figs. 2b-2d, both PAFs has similar values regardless the photosynthesis action spectra, but PLER and PLE calculated from P(λ) are higher than those from P'(λ), owing to the higher sensitivity of P(λ) over the wavelength ranges from 430 to 600 nm. So, we used the photosynthesis action spectra (P(λ)) from the German Institute for Standardization and numbered DIN 5031-10 in this experiment.[11,12] Here, the figures confirm that the shapes of the dependent graphs of PLER and PLE on the peak wavelength are identical in appearance to the photosynthesis action spectra (PAS; P(λ) and P'(λ)). The figures also indicate that bluish and reddish LEDs have higher PAF values (the ratio of PLER to LER) and that the greenish LEDs have lower PAF values as is expected. This indicates that bluish and reddish LEDs are more effective than the greenish LEDs on the photosynthesis system of plants. Figs. S2a and S2b show that the trends of PpLERs and PpLEs of five different photo-pigments with the peak wavelength resemble the wavelength dependence of the photo-pigment sensitivity action spectrum PpAS (Pp(λ)) of each photo-pigment (see Fig. 1b).

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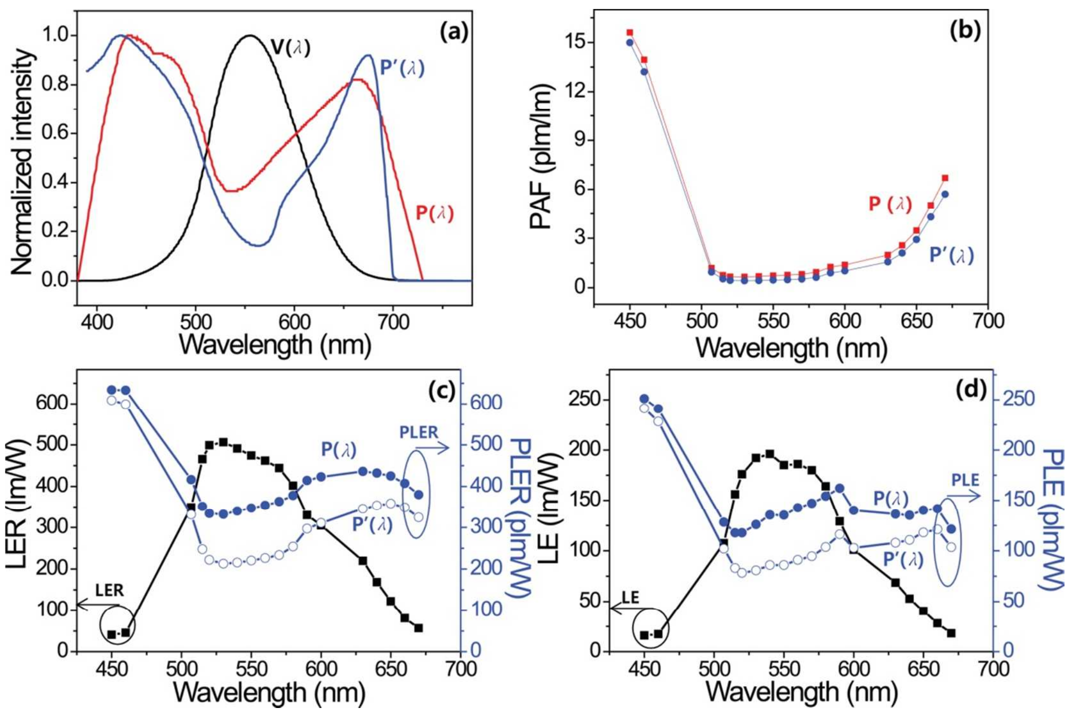

Fig. 2 (a) Normalized spectra of photopic sensitivity action curve (V(λ)) and two kinds of PAS (P(λ) and P'(λ)). Figures of merits of each blue LED and cyan/green/amber/red LPDF-capped pc-LEDs form 450 nm to 670 nm (b) two kinds of PAF, (c) LER and two kinds of PLER, (d) LE.

As expected, the different photo-pigment has the maximum luminous efficacy at different EL wavelength of monochromatic LEDs. These figures show that the bluish LEDs have the highest photo-pigmented efficiency of CL *a*, CL *b* and *β*-CT. Otherwise, the greenish (530 nm) LED and the amber (590 nm) LED have the maximum photo-pigmented efficacy of PE and PC photo-pigment, respectively. Most plants and major pigments for photosynthesis respond strongest to red and blue light for photosynthesis, otherwise, other antenna pigments (such as PE and PC), which participate in light absorption and play a significant role in photosynthesis, respond strongest to green and amber light. Therefore, the use of green and amber LEDs, as well as blue and red LEDs, should be considered to have a balanced effect on shape, development and flowering (photomorphogenesis) while growing plants. The characteristics of artificial lighting types that are ideal for vision are fairly different from than those that are maximally effective for the photosynthesis system. The most general lighting and greenhouse lighting sources used in the present market were not developed exclusively for improving the photosynthesis performance and photo-pigment performance by considering the match SPDs of light sources and P(λ) and Pp(λ)s of plants. Figure S3 and Tables S2a and S2b show the reported or measured SPDs and figures of merit of 10 commercialized types of lighting and daylight. These data indicate that the SPDs of all 10 artificial lighting types used presently general lighting and greenhouse lighting are fixed with a specific correlated color temperature (CCT). Here, we also measured and calculated the figures of merit of daylight as a standard source for photosynthesis in order to know the requirements for mimicking daylight. As reported in our previous publication,[21] the amounts of visual and photosynthesis light from the sun, those are, VIL and PIL (or PpIL), simply increase from early in the morning until 12:30 p.m. in the afternoon and then simply decrease until sunset. Figs. 4k-4l indicate that the PAF values of daylight remain nearly similar values with small variations near 1.83 ~ 2.05. This indicates that the photosynthesis brightness of daylight is around 1.9 fold higher than the value of the visual brightness of daylight. Because P(λ) covers blue and red parts of visible light but V(λ) covers only green part of skylight from sun. As shown in all photosynthesis performance data of artificial lightings and skylights in Fig. S3 and Tables S2a and S2b, there is no ideal lamp to meet all of the requirements to attain high photosynthesis performance and good photo-pigment performance, while having a highly positive vision effect. Because both figures of merit, such as visual luminous efficacy and photosynthesis luminous efficacy for artificial lighting sources are in a tradeoff relationship, managing the light of an artificial lighting source to optimize the photosynthesis performance of plants, and vision performance of humans become critical. Therefore, it is clear that acquiring the capability of controlling all figures of merit of lighting sources for photosynthesis and vision is the most important prerequisite for excellent photosynthesis and smart lighting systems.

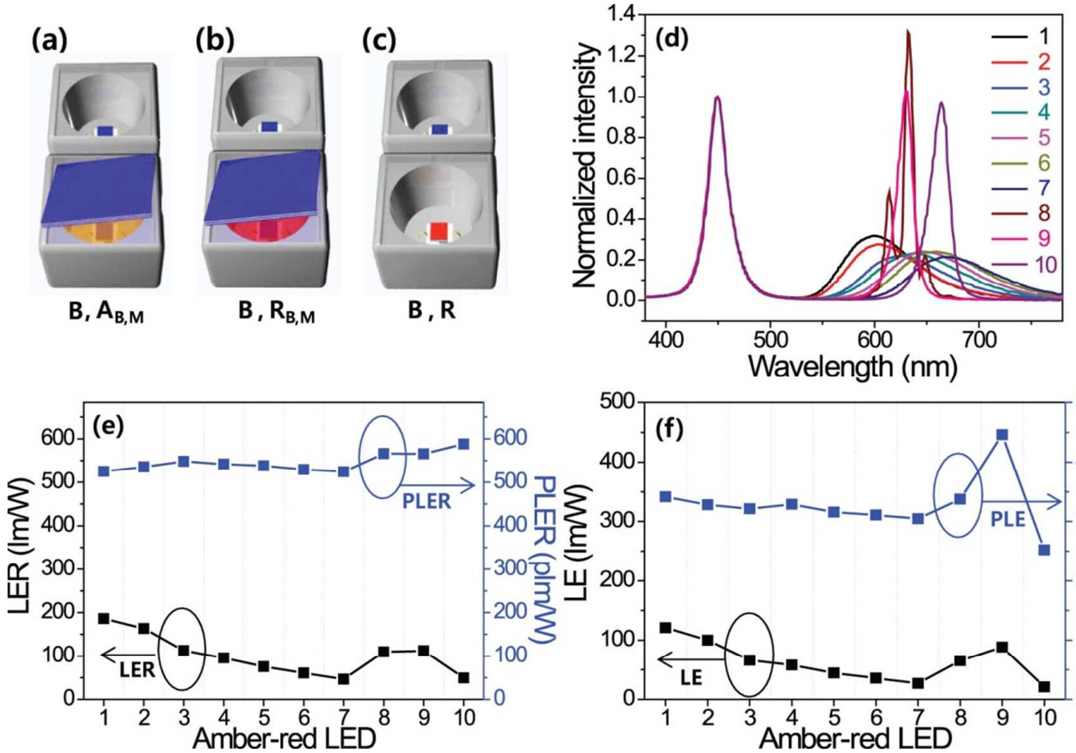

Fig. 3 Schematic illustrations of two color LEDs combining with (a) $A_{B,M}B$, (b) $R_{B,M}B$, and (c) RB. (d) blue normalized spectra, (e) LER and PLER, (f) LE and PLE of blue and amber or red dichromatic LEDs with changing the amber and red LEDs. Sample number 1, A590/B450S; 2, A600/B450S; 3, R630/B450S; 4, R640/B450S; 5, R650/B450S; 6, R660/B450S; 7, R670/B450S; 8, RKSF/B450S; 9, R630S/B450S; 10, R660S/B450S.

To date, various two colored LEDs of combining blue and red LED have been widely studied for the use of greenhouse lighting to enhance the growth of various vegetables in greenhouse.[14-17] Although the conventional R and B two-color approach allows for the facile dynamic control of R-to-B ratio points and provides high PLER, this approach has disadvantages, such as the different temperature/current dependence of each semiconductor-type R and B LED (see Figs. S1 and S2 in ESI), the visually-unfriendly violet color to human, and the reduced light quality due to the unbalanced colors for all process of photosynthesis. Fig. S4 and S5 indicate that amber and green pc-LEDs have reduced or at least similar variations of the efficacy and color coordinates with the current/temperature compared to the wide variation in the levels of current/temperature stability among red monochromatic III-V LED that do not contain phosphors. It means that the blue-to-red ratio of $R_{B,M}B$ two-color LEDs remain almost constant with a small variations as functions of the applied current and temperature in the ranges of the conditions of use. Here, we demonstrate a RB, $R_{B,M}B$ and $A_{B,M}B$ two package violet LED system that consists of a series of amber and red pc-LEDs and a blue LED (λ = 450nm) in an effort to compare their all photosynthetic figures of merit with RB two-color light. Figs. 3a-3c show the schematic illustration of RB, $R_{B,M}B$, and $A_{B,M}B$ two-color LEDs. Based on the emission spectrum of each two-color LED (see Fig. 3d) and meaningful figures of merit (the LER, LE, PLE, PLER, PpLER, and PpLE) of all types of RB, $R_{B,M}B$, and $A_{B,M}B$ two-color LEDs for the photosynthesis applications are calculated and summarized, as displayed in Figs. 5e and 5f with the change of amber and red LEDs. Although the highest PLE (~ 446 plm/W) is obtained by a combined two-color LED of InGaN blue and AlGaInP red LED, it can be speculated that the combined two color LED of InGaN blue LED and red pc-LED including narrow-band $K_2SiF_6:Mn^{4+}$ phosphor (denoted as RKSF) is appropriate for maintaining the stable light quality and showing the high PLE value (~ 337 plm/W). However, blue and red two color LEDs have some limitations on the induction of photoreaction of some photo-pigments which have peak of photo-pigment sensitivity action spectrum (Pp(λ)) at green and yellow wavelength because the match level between LED emission spectrum and Pp(λ) is very low. It needs green light even in small amounts to activate antenna pigment, such as a PE for balanced photosynthesis in the growth of plants, as considering the very low PpLE of red and blue two color LEDs in Figs. S6a and S6b. Therefore, it can be simply speculated that the wide band SPD of possible greenhouse LEDs must include green emission in order to stimulate healthy and balanced growth of plants.

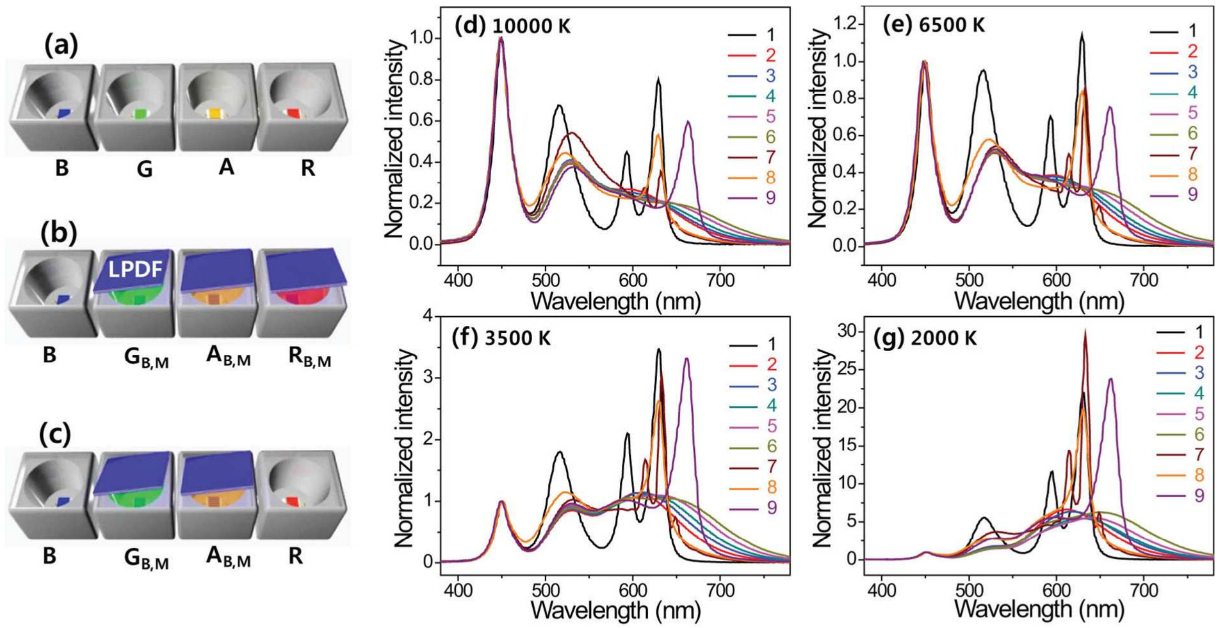

Fig. 4 Schematic illustrations of four-package white LEDs of (a) RAGB LED, (b) $R_{B,M}$,$A_{B,M}$,$G_{B,M}$,B, and (c) $RA_{B,M}$,$G_{B,M}$,B. the overlapped integrated emission spectra of four-package white LEDs at CCTs of (d) 10,000 K, (e) 6,500 K (cool white), (f) 3,500 K (warm white), and (g) 2,000 K (firelight) with changing six $R_{B,M}$ pc-LEDs and two red semiconductor LEDs. Sample number 1, R630S/A590S/G517S/B450S; 2, R630/A590/G520/B450S; 3, R640/A590/G520/B450S; 4, R650/A590/G520/B450S; 5, R660/A590/G520/B450S; 6, R670/A590/G520/B450S; 7, RKSF/A590/G520/B450S; 8, R630S/A590/G520/B450S; 9, R660S/A590/G520/B450S.

Both the conventional RAGB four-package approach using different colored semiconductor LEDs and the $R_{B,M}A_{B,M}G_{B,M}B$ ($R_{B,M}A_{B,M}G_{B,M}$ representing a LPDF-capped, full down-converted, monochromatic red, amber and green pc-LED pumped by a blue LED chip) approach using full-down converted colored LEDs are possible candidates for the control of SPDs and the intensity of white LEDs. In this experiment, a RAGB multi-package white LED is characterized as standard samples for comparison. Furthermore, we characterize the figures of merit of various $R_{B,M}A_{B,M}G_{B,M}B$ four-package white LEDs in terms of the vision performance and photosynthesis performance, such as LER, LE, CRI, PAF, PLER, and PLE. As the schematic illustrations of the $R_{B,M}A_{B,M}G_{B,M}B$ four-package white LEDs in the Figs. 4a-4c show, to assemble RAGB, $R_{B,M}A_{B,M}G_{B,M}B$ and $RA_{B,M}G_{B,M}B$ four-package LEDs. $R_{B,M}A_{B,M}G_{B,M}B$ four-package LEDs using one blue, five cyans, six greens, four amber-yellows, and six reds, the total number of combination provides a great number of different types of white LEDs. Numerous combinations present too many four-package LEDs to be assembled and characterized.

Consequently, one four-package $R_{B,M}A_{B,M}G_{B,M}B$ LED is selected as a control white LED in this study. As considering the photo-pigmented luminous efficacies of blue, green and amber LEDs, a set of primary LEDs with peak wavelengths of 590 nm (amber-yellow, A590), 520 nm (green, G520), and 450 nm (blue, B450S) is selected for the $R_{B,M}A_{B,M}G_{B,M}B$ four-package white LEDs by analyzing the effect of varying two narrow-band semiconductor-type LED reds, five broad-band pc-LED reds and on narrow-band pc-LED red on the photosynthesis performance of the four-package white LEDs. Figs. 4d-4g show the overlapped integrated emission spectra of each LED in a RAGB, $R_{B,M}A_{B,M}G_{B,M}B$ and $RA_{B,M}G_{B,M}B$ four-package white LED at CCTs of 10,000 K, 6,500 K (cool white) 3,500 K (warm white), and 2,000 K (firelight) along with five tunable wide-band red pc-LEDs, one narrow-band red pc-LED and two narrow-band semiconductor red LEDs. The reference of RAGB white LED systems show a narrow spectrum of each colored LED, while the other systems, combined with pc-LEDs, show a broad spectrum at all CCTs.

As shown in Fig. 5, the narrow-band RAGB LED and wide-band $RA_{B,M}G_{B,M}B$ or $R_{B,M}A_{B,M}G_{B,M}B$ LED with narrow-band red LED are superior in terms of the LER and PLER values but the wide-band $R_{B,M}A_{B,M}G_{B,M}B$ four-package LED with broad-band red LED is superior in terms of the LE and PLE. These figures also indicate that the blue and green portions of the white color decrease while the red portion of the white color increases with an increase of the red emitting wavelength at all CCTs. As a result, the LER, LE, PLER, and PLE of the $R_{B,M}A_{B,M}G_{B,M}B$ decrease slowly with an increase of the emitting wavelength of the red pc-LED at all CCTs in the red series.

This also shows that the PAFs of the $R_{B,M}A_{B,M}G_{B,M}B$ white LED have similar values with an increase in the red emitting wavelength at one CCT. As is similar to the circadian action factor (CAF),[21] the PAF values of the $R_{B,M}A_{B,M}G_{B,M}B$ white LED depend mainly on the variations of the CCTs except for 2,000K. Accordingly, the PAFs of firelight (2,000 K) of $R_{B,M}A_{B,M}G_{B,M}B$ LEDs increase from 1.41 to 1.83 with an increase of the emitting wavelength of the red pc-LED. This wide control of PAF values suggests that four-package white LEDs are good artificial lighting candidates that can simultaneously function as lamps for human vision and photosynthesis of plants for activating photosynthetic reaction. Furthermore, the PLE values of four-package $R_{B,M}A_{B,M}G_{B,M}B$ and $RSA_{B,M}G_{B,M}B$ white LEDs are higher than those of HPS and MH lamps, which are used for greenhouse lighting. In different from the LE and PLE trends, the color quality property, i.e., the color rendering index (CRI) of the four-package white-light LED show nearly constant values over 85 with an increase in the emitting wavelength of the red pc-LED at CCTs of either 6,500 or 3,500 K and values that exceed 80 at 10,000 or 2,000 K (see Fig. 5c).

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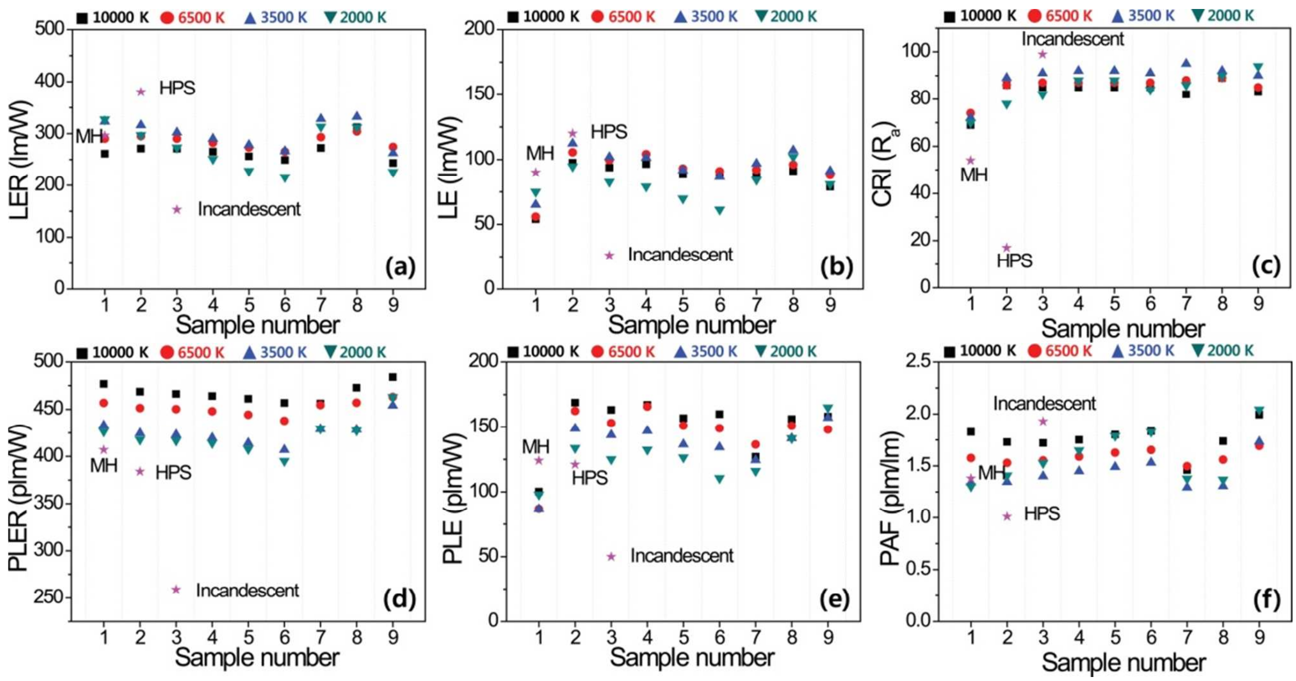

Fig. 5 igures of merit of four-package white LED at 10,000 K, 6,500 K (cool white), 3,500 K (warm white) and 2,000 K (firelight). (a) LER, (b) LE, (c) CRI, (d) PLER, (e) PLE, and (f) PAF. Sample number 1, R630S/A590S/G517S/B450S; 2, R630/A590/G520/B450S; 3, R640/A590/G520/B450S; 4, R650/A590/G520/B450S; 5, R660/A590/G520/B450S; 6, R670/A590/G520/B450S; 7, RKSF/A590/G520/B450S; 8, R630S/A590/G520/B450S; 9, R660S/A590/G520/B450S.

This indicates that the four-package white LEDs show good color quality even at 10,000 K where gives highest photosynthesis luminous efficacy. It is well known that the LE and CRI exist in a trade-off relationship in a single-package white pc-LED. Otherwise, Fig. 5 indicates that the PLE and CRI values have no direct trade-off relationship in four-package $R_{B,M}A_{B,M}G_{B,M}B$ and $RA_{B,M}G_{B,M}B$ white LEDs. Among the nine different $R_{B,M}A_{B,M}G_{B,M}B$ and $RA_{B,M}G_{B,M}B$ four-package white LEDs shown in Figure 5, the highest vision and color performance of human eye and an excellent photosynthetic effect are attained from the different combination of a red, amber, green, and blue LED in four-package white LED. In this figure, a combined four-package LED of R630, A590, G520, and B450S is selected as having the best PLE at CCT of 10,000K. This four-package offers excellent color qualities (CRI > 86), excellent vision performances (LER > 295 lm/W, LE > 105 lm/W), and a tunable photosynthesis performance (PLER = 451, 425 plm/W, PLE = 162, 149 plm/W, PAF = 1.53, 1.34) at cool- and warm-white CCTs of 6,500 and 3,500 K. In addition, blue-enriched emission (10,000 K) of $R_{B,M}A_{B,M}G_{B,M}B$ (R630, A590, G520, and B450S) LEDs provide reasonable color quality (CRI = 86), good vision performances (LER = 271 lm/W, LE = 98 lm/W) and highly efficient photosynthesis effect (PLER = 469 plm/W, PLE = 169 plm/W, PAF = 1.73). Consequently, it can be simply considered that it is not a difficult step to select a good combination of four differently colored LEDs to attain high LE, CRI, and tunable PLER, PLE and PAF values with variation of the CCT value in the $R_{B,M}A_{B,M}G_{B,M}B$ white-light system. Therefore, $R_{B,M}A_{B,M}G_{B,M}B$ four-package LEDs can be considered as a possible artificial lighting candidates to mimic daytime sunlight, reduce the green portion of white light in the green house, and maintain good vision performances, good color qualities and controllable photosynthesis effects.

Although two color LED system of InGaN blue and red pc-LED shows the much higher PLE (~320 plm/W) than that of 10,000K $R_{B,M}A_{B,M}G_{B,M}B$ (R630, A590, G520, and B450S) LEDs (~169 plm/W), the absence of green and amber color is detrimental to the balanced growth of plants and the visual comport of workers in greenhouse. The required level of green and amber photons for optimum plant growth is different from plant species to species. Our four-package LED lights with tunable and different wavelength of red, amber, green, and red four colors would be beneficial in determining the species specific optimal wavelength for plant growth. To study the photosynthesis luminous efficacy of four package LEDs with the change of SPD of $R_{B,M}A_{B,M}G_{B,M}B$ LED lamp, we decrease the ratio of green/amber portion from the ratio value of 10,000 K white (blue; 75 mA, green; 105 mA, amber; 30 mA and red; 32 mA) to the enriched blue/red portion with the constant blue/red applied current (blue; 75 mA, red; 32 mA).

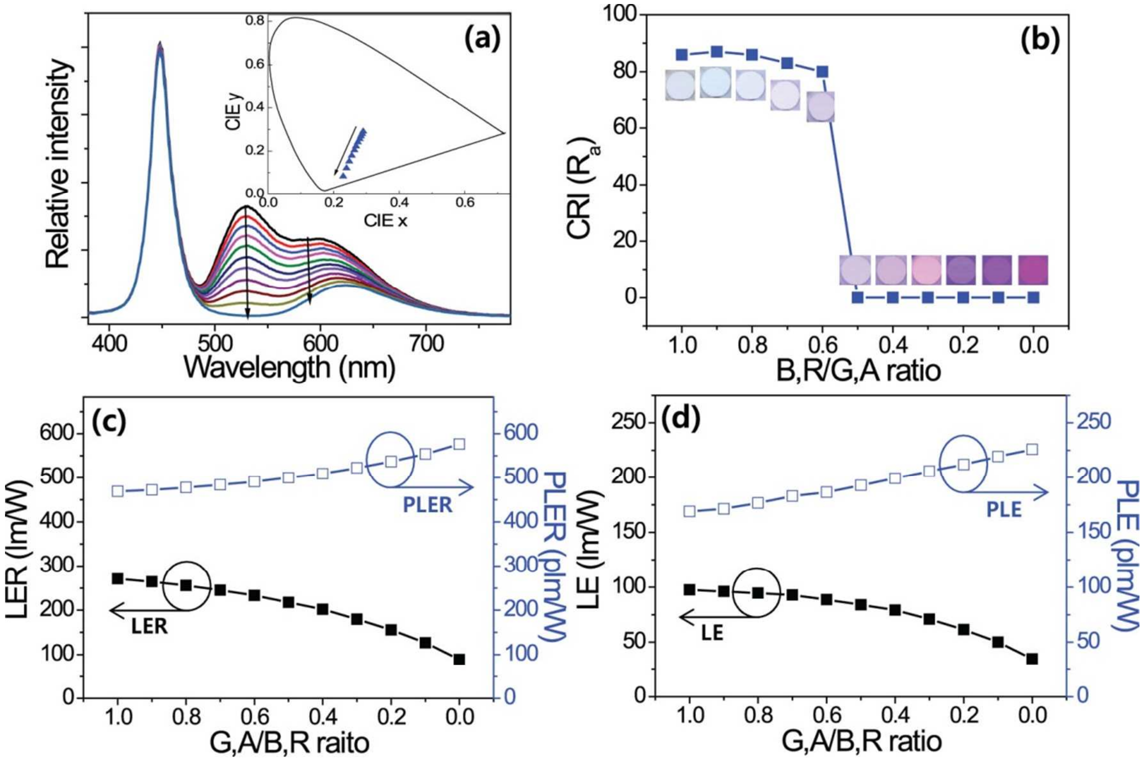

Fig. 6 (a) EL spectra and CIE color coordinates, (b) CRI (insets; photographs of emitting color), (c) LER and PLER and (d) LE and PLE of R630/A590/G520/B450S four-package by decreasing of green and amber portion with 10 steps starting at 10,000 K.

Fig. 6a shows the change of the overlapped integrated emission spectra of a $R_{B,M}A_{B,M}G_{B,M}B$ four-package with decrease of green/amber portion. As shown in Fig. 8a inset, the whitish emission is changed to the purplish color when the blue and red portions of emitting spectrum increase with the decrease of green and amber portion. Therefore, the CRI value of $R_{B,M}A_{B,M}G_{B,M}B$ four-package comes down to zero from 80 when the 0.5 green/amber portion (See Fig. 6b). Figs. 6c and 6d also show that the PLER and PLE of photosynthesis for plants increase from 469 plm/W to 577 plm/W, and 169 plm/W to 226 plm/W, respectively but the LER and LE of human vision decrease from 271 lm/W to 89 lm/W, and 98 lm/W to 35 lm/W, respectively, with the increase of blue and red portions. As expected, the optimum SPD of $R_{B,M}A_{B,M}G_{B,M}B$ four-package purplish LED for photosynthesis efficiency is totally reversed from the optimum SPD of whitish LED for efficiency of human vision. Therefore, the spectrum distributions of $R_{B,M}A_{B,M}G_{B,M}B$ four-package LED should be controlled by the presence or absence of workers in the greenhouse for balancing and maximizing the photosynthesis efficiency of plants and the cultivating and managing efficiency of workers.

Conclusions

The technological advancements for developing good greenhouse LED lighting sources have about two-decade history since a single-package white pc-LED was first commercialized by Nichia Co. Ltd. To date, all current reports that study the development and optimization of blue and red two color and/or white pc-LED systems have focused on the productivity of plants. The figures of merit, which are widely used in the LED lighting society, are the LER and the LE for vision performance as well as the CRI and CCT for color quality. However, these properties are insufficient when seeking to represent all performances required when searching excellent artificial lighting sources for greenhouse lighting. Here, the possible figures of merit are proposed and explained to describe the photosynthesis performance and the photo-pigment performance. As above explained, the PLER, PLE, PAF, PpLER, PpLE, and PIL are added to the visual figures of merit of lighting, in this case the PLER and PLE values and the PIL in order to assess the quality of commercialized artificial lighting sources along with our four-package $R_{B,M}A_{B,M}G_{B,M}B$ LED lighting source and standardize the sunlight with respect to the vision performance and photosynthesis performance. This analysis of the optical data of sunlight, MH lamps, and HPS lamps can provide newly developed artificial greenhouse lighting sources with guidelines for attaining photosynthetically efficient, vision-friendly, and tunable multi-package LEDs by comparing the optical properties of the photosynthesis performance and vision performance among our four-package $R_{B,M}A_{B,M}G_{B,M}B$ LEDs, natural skylight, and presently commercialized light sources. An additional important characteristic for optimizing all figures of merit for good greenhouse LED lightings is the capability of individual colors to control and adjust the SPDs of LED lighting sources to the optimum SPD for photosynthesis of plants. The distinct color control of $R_{B,M}A_{B,M}G_{B,M}B$ LEDs combined with a narrow InGaN blue LED and three wide-band LPDF-capped green, amber, red pc-LEDs provides the capability to create tunable figures of merit while also ensuring excellent photosynthesis performances and good vision qualities. In this study, one of the best choices of four-package $R_{B,M}A_{B,M}G_{B,M}B$ LED (R630, A590, G520, and B450S) shows excellent photosynthesis performance (PLER = 469 plm/W, PLE = 169 plm/W) and good vision performances (LER = 271 lm/W, LE = 98 lm/W, CRI = 85) at CCT of 10,000 K. Furthermore, this $R_{B,M}A_{B,M}G_{B,M}B$ LED provides tunable capability of SPD of lighting and excellent photosynthesis performance (PLER > 457 plm/W, PLE > 157 plm/W) even at eye-friendly vision performances (LER > 249 lm/W, LE > 87 lm/W, CRI > 85) under blue- and red-enriched purplish emitting conditions. More elaborate experiments are required to realize a photosyntheically efficient, vision-friendly, and tunable multi-package LED lighting system with higher photosynthesis performances in which the quantum efficiency of the InGaN blue LED chip and phosphors are enhanced and the combination of each colored pc-LED is properly selected in a multi-package greenhouse LED system. This study, which defines important figures of merits for efficient photosynthesis lighting systems, which compares the optical properties of natural skylights and commercialized greenhouse lamps, two-package (or two-chip) RB LED lamps, and which utilizes four-package $R_{B,M}A_{B,M}G_{B,M}B$ systems with LPDF-capped pc-LEDs, can lead to the creation of high-quality smart lighting systems for high efficiency of photosynthesis, energy savings, low heat generation, good thermal stability, and the realization of vision-friendly purplish white color.

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Notes and references

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