Effects of Green - Emitting Phosphor (La,Ce,Tb)PO_4:Ce:Tb on Luminous Flux and Color Quality of White LED Lamps

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Abstract. *In this paper, we present and analyze the influence of (La,Ce,Tb)PO_4:Ce:Tb green phosphor (LaTb) toward the performance of the multi-chip white LED (MCW-LEDs) lamps including color uniformity, lumen output, Color Rendering Index (CRI), and Color Quality Scale (CQS). By mixing the LaTb green phosphor and the YAG:Ce yellow phosphor compounding under condition of 7000 K MCW-LEDs, this new approach can produce a huge meaningful change in lumen output and the angular color distribution of MCW-LEDs. We also study the interaction between the concentration and size of the LaTb particles with output flux, color uniformity, CRI, and CQS. The obtained results demonstrate that the higher lumen output, the higher color uniformity enhancement could be attained by adding the LaTb particles with a size range around 6-8 μm and the concentration around 1.5 % in phosphor layer. Meanwhile, the decrease of the color rendering value (CRI) and the Color Quality Scale (CQS) tend to be stable and insignificant. In other words, the obtained results provide a prospective method which plays an important role in the development of MCW-LED manufacturing technology.*

Keywords

(La,Ce,Tb)PO_4:Ce:Tb phosphor, Color Quality Scale, Color Rendering Index, color uniformity, luminous flux, multi-chip white LED lamp.

1. Introduction

Recently, MCW-LED is becoming one of the most widely used lighting sources in illumination applications since its characteristics such as high light intensity, energy-efficient, low power consumption, fast response, and expended durability outperform conventional lighting sources (Michael Cantore et al. 2015 and Yu et al. 2016). Nevertheless, there are challenges in improving those performance indicators. That leads to further research and development. Two important properties of white LEDs that attract great attention of researchers are lumen output and color uniformity which have been studied and published in previous papers of Ying et al. (2016) and Chiang et al (2015). To improve these two features, these papers were mainly focused on scattering enhancement of the incident and the converted light of phosphor particles in MCW LED.

White light, which is produced by white LED, is the result of mixing in the incident blue light from the chips and converted yellow light. YAG:Ce yellow phosphor particles are the preferred material for white LEDs. This phosphor is used to absorb exciting blue light from the chips and to emit the yellow light (Liu et al. 2008, Liu et al. 2011). However, it is perceived that the blue light intensity is not in balance with the yellow light intensity due to the backscattering problems. Consequently, the MCW-LEDs do not offer uniform color distribution and thus are not suitable for some applications (H-C Chen et al. 2013 and K-J Chen et al. 2012).

One of the alternative methods to enhance the color uniformity and luminous flux of white LEDs is to add green $Ce_{0.67}Tb_{0.33}$ $MgAl_{11}$ O_{19}:Ce,Tb phosphor into phosphor compounding with the suitable phosphor concentration as proposed by Anh et al. (2016). In addition, Hakeem and Park (2015) introduced $Na(Sr_{072}Ca_{0.25})PO_4{:}0.03Eu^{2+}$ phosphor that showed stronger emission spectra intensity in white LEDs and could be employed as a blue emitting phosphor for LEDs. Other studies found that the use of appropriate concentration and thickness as well as suitable location of phosphor layer can help achieve better LED performance (Shuai et al. 2011). Moreover, the effect of LED lens on optical qualities have also been studied recently. For example, Enguo Chen (2014) demonstrated that a structure with a freeform microlens array could improve the luminous flux of white LEDs. The lens with the droplet evaporation exhibits a stronger light intensity (Xing Guo et al. 2016). Although there were different approaches to intensify the efficiency of white LEDs, the angular color uniformity and the lumen output of white LEDs are still not high enough and insufficient to satisfy requirements and increasing demand of applications in the commercial LED lighting market. Therefore, advanced research should be conducted to achieve the desired color uniformity and higher lumen output.

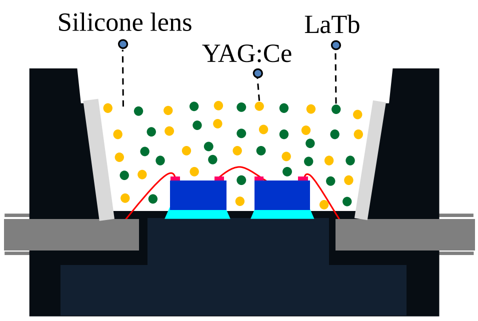
In this study, we propose the incorporation of the new green LaTb particles in the phosphor layer of the MCW-LEDs to enhance the lumen output and the correlated color temperature uniformity. Through the examination of the effect of green $(La,Ce,Tb)PO_4$:Ce:Tb phosphor on the white LED illumination in In-cup Phosphor Packages (IPP), we found that LaTb particles can provide better light distribution in the MCW-LED package. The simulation of this package is carried out on various weight percentages and different sizes of LaTb phosphor particles. The scattering properties of phosphor layer are investigated by considering both light absorption and light conversion in LEDs when LaTb particles are injected into phosphor layer. In other words, this work investigates how both concentration and size of LaTb green phosphor particles will impact on luminous flux, angular color distribution, CRI, and CQS.

2. Simulation and Computation

The main purpose of this research is to reveal the effect of green LaTb phosphor on the angular CCT uniformity and the lumen output efficiency of the MCW-LEDs, so we used a 7000 K MCW-LED with in-cup phosphor structure to simulate. The MCW-LED models used in the simulation processes were the commercial MPBGA (Multi-Package Ball Grid Array) provided by the Siliconware Precision Industries Co. Ltd., Taiwan.

The model structure, as shown in Fig. 1, was built up according to the MCW-LED samples which consists of a blue chip, a reflector cup, a phosphor layer, and a silicone layer. Each blue chip was designed with 1.16 W radiant power, a peak wavelength of 453 nm, and a dimension of 1.14 mm × 0.15 mm. These chips were securely attached to the reflector which has a 2.07 mm

(a) the sample LED used in this study



(b) with in-cup phosphor configuration

Fig. 1: Illustration of MCW-LEDs structure.

depth, 8 mm bottom length, and 9.85 mm surface length. The chips were covered by a 0.08 mm thick phosphor layer, as shown in Fig. 1. In order to better understand the effect of using this green phosphor, the optical simulation process is conducted with variation of LaTb particle concentration and size. It is worth noting that if the concentration of green LaTb phosphor varies, the YAG:Ce phosphor concentration needs to be suitably adjusted to maintain the color temperature of white LEDs at 7000 K.

LightTools 8.1.0 optical software was used in this simulation process to model the LED structure and analyze its optical performance. The LaTb phosphor with the wavelength at 546 nm was added to the MCW-LED with the several alterations in concentration and size to optimize the optical properties of LEDs. Therefore, these MCW-LEDs contain the green phosphor LaTb, the yellow phosphor YAG:Ce, and the silicone glues in their phosphor layer. In this structure, green LaTb phosphor particle sizes were ranging from 1 μm to 10 μm, while the radius of the YAG:Ce yellow phosphor particle is set to 7.25 μm.

The scattering of the LaTb phosphor particles is calculated and verified by following the Mie-theory (Lai et al. 2016). The relationship among the angular scattering amplitudes and the weight as well as the size of the LaTb particles is described by the following equations:

$$S_1 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n(x,m)\pi_n(\cos\theta) + b_n(x,m)\tau_n(\cos\theta)\right], \tag{1}$$

$$S_2 = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} \left[a_n(x,m)\tau_n(\cos\theta) + b_n(x,m)\pi_n(\cos\theta)\right], \tag{2}$$

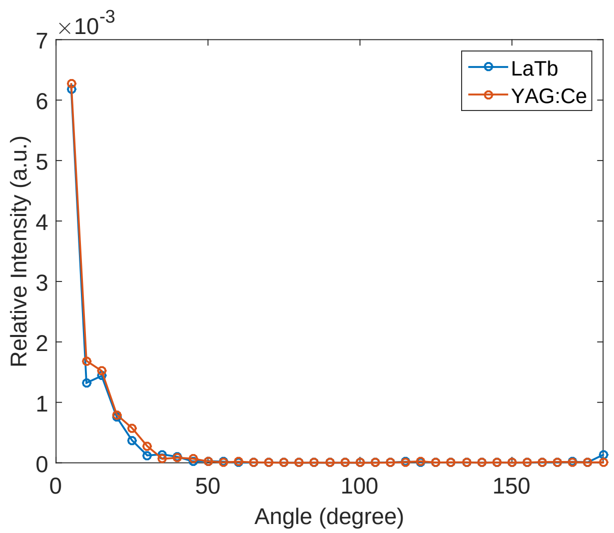


Fig. 2: The angular scattering diagram of LaTb and YAG:Ce particles in the phosphor layer of MCW-LEDs for 546 nm and 555 nm.

where S_1, S_2 are the scattering intensity functions, x is the particle size parameter, m is the phosphor particle refractive index. Their values are determined by the angular functions $\pi_n(\cos\theta)$ and $\tau_n(\cos\theta)$, the parameters m, x, and the expansion coefficients a_n, b_n. The refractive index of phosphor particle is set to be 1.85 and 1.83 for LaTb and YAG:Ce. The scattering intensity S_1, S_2 of MCW-LEDs are simulated and computed at the peak of 555 nm of yellow YAG:Ce phosphor and the wavelength of 453 nm of blue chips.

Figure 2 depicts the angular scattering intensity of LaTb and YAG:Ce particles on the Cartesian coordinate system. The Matlab simulation results in Fig. 2 express that the angular scattering intensity of LaTb diffuse particle is similar to the yellow YAG:Ce phosphor particle. Hence, the combination of the LaTb particles and the YAG:Ce phosphor layer will produce a scattering enhancement of the incident light in MCW-LED.

3. Results and Discussion

To determine the color uniformity quality of LEDs, we use the Correlated Color Temperature deviation (△CCT) which is the difference between the maximum CCT value and the minimum CCT value. This parameter was also characterizes the angular CCT uniformity of LEDs. Thus, lower △CCT value corresponds to the better light quality of LEDs (K-C Huang et al. 2013).

Figure 3 shows the relationship among △CCT, the concentration, and size of LaTb particles. When the LaTb particle concentration increases, the CCT deviation remarkably decreases all LaTb particles with different sizes. If the concentration of LaTb phosphor increases from 0 to 1.4 % for small LaTb particles and from 0 up to 1.8 % for bigger sizes, LED shows an ex-

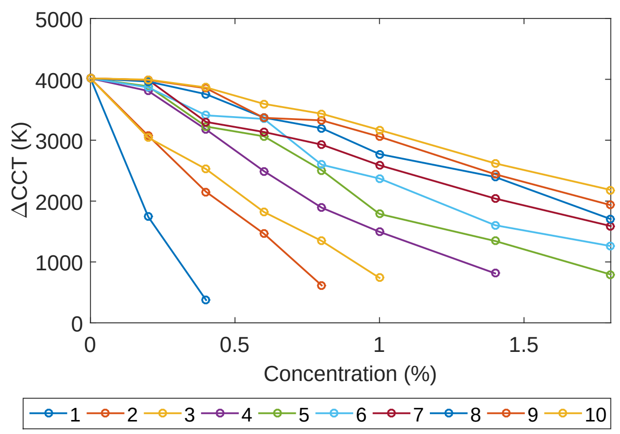


Fig. 3: The CCT deviation (△CCT) of MCW-LEDs is at average CCT 7000 K with different LaTb particle sizes and concentrations.

cellent color uniformity in its optical property. In some cases, △CCT significantly reduces from 4000 K to less than 500 K. The lowest CCT deviation value can be achieved for less than 450 K at the LaTb particle concentration of 0.4 % and particle size of 1 μm. Hence, LaTb phosphor could provide great scattering capability for LEDs and it makes the spatial color distribution much flatter than those without LaTb. In other words, the scattered light is able to move in many different directions as the concentration of LaTb particles pumped into yellow YAG:Ce phosphor increases.

Besides inspecting the influence of LaTb concentration, this study also focuses on investigating the impact of LaTb particle radius on the color uniformity of the in-cup package MCW-LEDs. The LaTb particle radius ranging from 1 to 10 μm is surveyed and analyzed for different concentration of LaTb particles. With the small particle sizes from 1 μm to 4 μm, the CCT deviation of white LEDs, which is shown in Fig. 3, rapidly decreases at around 500 K as the LaTb particle concentration increases. Considering these facts, it can be deduced that the small particles could scatter more light within the wider viewing angle. As for the greater size range 5–10 μm, the tendency of curves expressed by the color uniformity is similar to the small particle case.

This is associated with the fact that the incident light tends to be scattered, and hence, the decay level of CCT deviation for these large particles is slower than those of small particles. Generally, after adding the new phosphor LaTb into YAG:Ce phosphor, the color uniformity can increase significantly. In particular, LaTb particles can improve the color uniformity of LEDs for all particle sizes with range within 1–10 μm, even if the large particles are employed as seen in Fig. 3.

Furthermore, the simulation results shown in Fig. 4 also demonstrate that LaTb particle size and weight percentage have a strong impact on the lumen output. The higher weight percentage of the green LaTb phosphor, the better quality of the green lights, and the higher luminous flux of white LED. To study of the influence of the LaTb weight percentage on lumen output, we continuously varied the LaTb concentration from 0 to 1.8 % for each fixed size in the simulation process. As shown in Fig. 4, as for 1–4 μm LaTb particles, luminous flux significantly rises with LaTb weight percent from 0 % to 1.5 %. Obviously, when the LaTb particle size is small, the lumen output will decrease because the unwanted backward scattering has been demonstrated in many previous papers. It means that there is more light trapped inside a package and the lumen output escaping from that. However, mixing the new LaTb phosphor into phosphor layer, lumen output could overcome the restriction with the small size of phosphor particles and is rapidly improved to achieve the best luminous flux. With this novel feature, the utilization of LaTb phosphor becomes one of the most promising methods for improving the optical properties of LEDs. With particle sizes larger than 5 μm, the light tends to propagate stronger in the forward direction and restricts backward light absorption of LED chips, thereby the luminous flux at large size particles will be higher. Hence, a mixture of LaTb particles with different sizes and YAG:Ce phosphors tremendously improves the lumen output of the in-cup phosphor package MCW-LEDs.

Accordingly, LaTb green phosphor can be used to yield higher lumen output due to a supplement of green light in MCW-LEDs. However, the CRI values can decrease as increasing LaTb concentration. It can be observed that with the increasing luminous flux, the CRI value insignificantly decreases in size ranging from 6 to 8 μm as shown in Fig. 5. The reason is that in order to

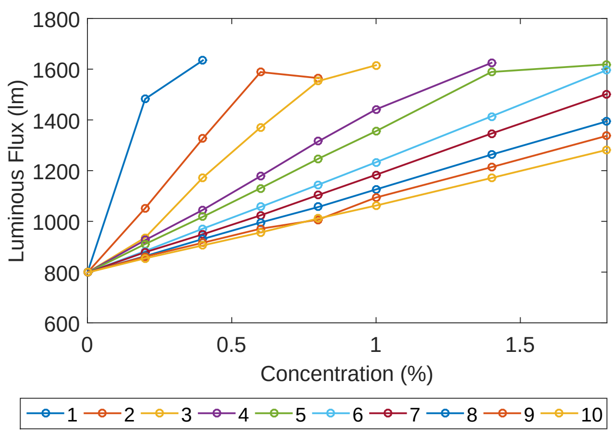


Fig. 4: The luminous flux of MCW-LEDs is at average CCT 7000 K with different LaTb particle sizes and concentrations.

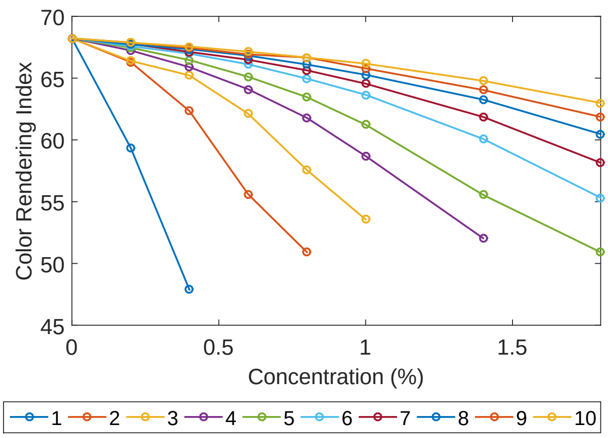


Fig. 5: The Color Rendering Index (CRI) value of MCW-LEDs is at average CCT 7000 K with different LaTb particle sizes and concentrations.

achieve better CRI values, LEDs must have a broader source spectrum to respond, and thus, the efficiency of white LED would be reduced. In addition, one of the most important factors used to evaluate the color quality of white LEDs is the CQS factor, which is also investigated in this study. This factor is characterized by the color fidelity level of MCW-LEDs and it is assessed by some terms of color quality including CRI, chromatic discrimination, and observer preferences (Davis and Ohno 2010). In the simulation results (see Fig. 6), there is a slight lessening in CQS values with concentration around 1.5 % and the particle 6–8 μm.

We found that the 1–4 μm LaTb particles show greater performance capability for lumen output and color uniformity after adopting LaTb particles into phosphor layer. However, we conclude that the most appropriate are phosphor particles having a size range from 6 to 8 μm and the concentration around 1.5 %. The reason is that color uniformity and optimal lumen output can be achieved for the low CRI, CQS values in the same size and concentration range.

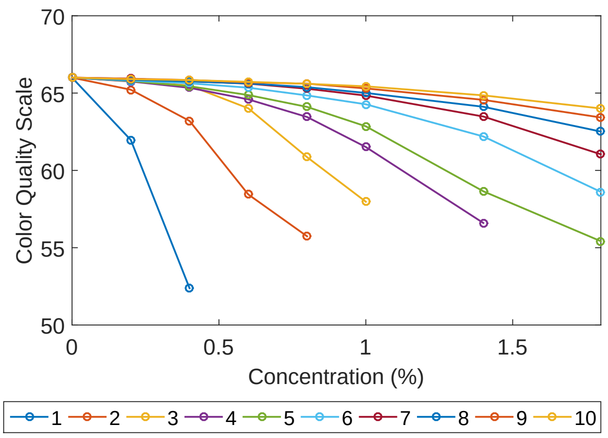


Fig. 6: The Color Quality Scale (CQS) value of MCW-LEDs is at average CCT 7000 K with different LaTb particle sizes and concentrations.

4. Conclusion

In this article, a research was conducted to find out the impact of green (La,Ce,Tb)PO_4:Ce:Tb phosphor particles with the various concentrations and sizes on the MCW-LED performance including the △CCT, luminous flux, CRI, and CQS. The obtained results indicate that the green LaTb phosphor can improve both the color uniformity and the lumen output. The optimum LaTb concentration and size are determined to obtain the lowest value of the CCT deviation as well as the highest luminous flux accompanying with a negligible decrease in CRI, CQS values. In fact, the obtained CRI and CQS values can insignificantly decrease when applying around 1.5 % LaTb concentration and 6-8 μm particles. It could be concluded that by pumping a small amount of LaTb phosphor has the concentration ranging from 0–1.8 % and the particle sizes from 6–8 μm, we can obtain better performance of white LEDs. Therefore, LaTb particles should be a prioritized solution to be chosen for improving the optical properties of MCW-LEDs.

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References

[1] CANTORE, M., N. PFAFF, R. M. FARRELL, J. S. SPECK, S. NAKAMURA and S. P. DENBAARS. High luminous flux from single crystal phosphor-converted laser-based white lighting system. *Optics Express*. 2016, vol. 24, iss. 2, pp. A215–A221. ISSN 1094-4087. DOI: 10.1364/OE.24.00A215.

[2] YU, S. D., Z. T. LI, G. W. LIANG, Y. TANG, B. H. YU and K. H. CHEN. Angular color uniformity enhancement of white light emitting diodes by remote micro-patterned phosphor film. *Photonics Research*. 2016, vol. 4, iss. 4, pp. 140–145. ISSN 2327-9125. DOI: 10.1364/PRJ.4.000140.

[3] YING, S. P. and J. Y. SHEN. Concentric ring phosphor geometry on the luminous efficiency of white. *Optics letters*. 2016, vol. 41, iss. 9, pp. 1989–1992. ISSN 0146-9592. DOI: 10.1364/OL.41.001989.

[4] CHIANG, C. H., H. Y. TSAI, T. S. ZHAN, H. Y. LIN, Y. C. FANG and S. Y. CHU. Effects of phosphor distribution and step-index remote configuration on the performance of white light-emitting diodes. *Optics letters*. 2015, vol. 40, iss. 12, pp. 2830–2833. ISSN 0146-9592. DOI: 10.1364/OL.40.002830.

[5] LIU, Z. Y., S. LIU, K. WANG and X. LUO. Analysis of Factors Affecting Color Distribution of White LEDs. In: *Proceedings of International Conference on Electronic Packaging Technology & High Density Packaging*. Shanghai: IEEE, 2008, pp. 386–393. ISBN 978-1-4244-2740-6. DOI: 10.1109/ICEPT.2008.4607013.

[6] LIU, S. and X. B. LUO. *LED Packaging for Lighting Applications: Design, Manufacturing and*

Testing. Beijing: Chemistry Industry Press and John Wiley & Sons, 2011. ISBN 978-0-470-82840-3.

[7] CHEN, H. C., K. J. CHEN, C. C. LIN, C. H. WANG, H. V. HAN, H. H. TSAI, H. T. KUO, S. H. CHIEN, M. H. SHIH and H. C. KUO. Improvement in uniformity of emission by ZrO_2 nano-particles for white LEDs. *Nanotechnology*. 2012, vol. 23, iss. 26, pp. 1–5. ISSN 0957-4484. DOI: 10.1088/0957-4484/23/26/265201.

[8] CHEN, K. J., H. C. CHEN, M. H. SHIH, C. H. WANG, S. H. CHIEN, H. H. TSAI, C. C. LIN and H. C. KUO. Enhancement of angular CCT by hybrid phosphor structure in White Light-emitting Diodes. In: *Proceedings of International Nanoelectronics Conference (INEC)*. Singapore: IEEE, 2013, pp. 30–32. ISBN 978-1-4673-4842-3. DOI: 10.1109/INEC.2013.6465944.

[9] ANH, Q. D. N. and H. Y. LEE. Improving the angular color uniformity and the lumen output for multi-chip white LED lamps by green $Ce_{0.67}Tb_{0.33}MgAl_{11}O_{19}$:Ce,Tb phosphor. *Journal of Display Technology*. 2016, vol. 39, iss. 7, pp. 871–875. ISSN 0253-3839. DOI: 10.1080/02533839.2016.1208062.

[10] HAKEEM, D. A. and K. PARK. Synthesis and Luminescence Properties of Blue $Na(Sr_{097-x}Ca_x)PO_4$:$0.03Eu^{2+}$ Phosphors for White Light Emitting Diode Applications. *Journal of Nanoscience and Nanotechnology*. 2015, vol. 15, iss. 7, pp. 5074–5077. ISSN 1533-4840. DOI: 10.1166/jnn.2015.10385.

[11] SHUAI, Y., Z. HE, N. T. TRAN and F. G. SHI. Angular CCT Uniformity of Phosphor Converted White LEDs: Effect of Phosphor Materials and Packaging Structures. *IEEE Photonics Technology Letters*. 2011, vol. 23, iss. 3, pp. 137–139. ISSN 1941-0174. DOI: 10.1109/LPT.2010.2092759.

[12] CHEN, E., R. WU and T. GUO. Design a freeform microlens array module for any arbitrary-shape collimated beam shaping and color mixing. *Optics Communications*. 2014, vol. 12, iss. 3, pp. 75–78. ISSN 1551-319X. DOI: 10.1016/j.optcom.2014.01.032.

[13] GUO, X., X. LEI, H. ZHENG, C. LI, J. C. CHU and S. LIU. Fabrication of Lens With Large View Angle Through Droplet Evaporation for High-Power Light-Emitting Diodes. *Journal of Display Technology*. 2016, vol. 12, iss. 3, pp. 288–293. ISSN 1551-319X. DOI: 10.1109/JDT.2015.2480420.

[14] LAI, M. F., N. D. Q. ANH, H. Y. MA and H. Y. LEE. Scattering effect of SiO_2 powders on correlated color temperature uniformity of multi-chip white light LEDs. *Journal of the Chinese Institute of Engineers*. 2016, vol. 39, iss. 4, pp. 468–472. ISSN 0253-3839. DOI: 10.1080/02533839.2015.1117950.

[15] DAVIS, W. and Y. OHNO. Color quality scale. *Optical engineering*. 2010, vol. 49, iss. 3, pp. 288–293. ISSN 1551-319X. DOI: 10.1117/1.3360335.

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