NANO EXPRESS   **Open Access**

Nanoparticle-Doped Polydimethylsiloxane Fluid Enhances the Optical Performance of AlGaN-Based Deep-Ultraviolet Light-Emitting Diodes

Zhi Ting Ye[1], Yung-Min Pai[2], Chih-Hao Lin[2], Lung-Chien Chen[3*], Hong Thai Nguyen[4] and Hsiang-Chen Wang[4*]

Abstract

This paper proposes a new encapsulation structure for aluminum nitride-based deep UV light-emitting diodes (DUV-LEDs) and eutectic flip chips containing polydimethylsiloxane (PDMS) fluid doped with SiO_2 nanoparticles (NPs) with a UV-transparent quartz hemispherical glass cover. Experimental results reveal that the proposed encapsulation structure has considerably higher light output power than the traditional one. The light extraction efficiency was increased by 66.49% when the forward current of the DUV-LED was 200 mA. Doping the PDMS fluid with SiO_2 NPs resulted in higher light output power than that of undoped fluid. The maximum efficiency was achieved at a doping concentration of 0.2 wt%. The optical output power at 200 mA forward current of the encapsulation structure with NP doping of the fluid was 15% higher than that without NP doping. The optical output power of the proposed encapsulation structure was 81.49% higher than that of the traditional encapsulation structure. The enhanced light output power was due to light scattering caused by the SiO_2 NPs and the increased average refractive index. The encapsulation temperature can be reduced by 4 °C at a driving current of 200 mA by using the proposed encapsulation structure.

Keywords: DUV-LEDs, PDMS, Nanoparticle, Light extraction efficiency, Flip chip

Background

Aluminum nitride-based deep UV-emitting diodes (DUV-LEDs) with a eutectic flip chip and a wavelength range of 200–300 nm have been used in curing engineering, communication security, sterilization engineering, chemical decomposition, water purification, air purification, forgery detection, and sensing [1–10]. DUV-LEDs are considered a near-future replacement for traditional UV light sources because they are free from mercury and highly reliable [11–14]. However, the output power of the flip chip DUV-LED remains low mainly because of quantum well defects, light absorption, and total internal reflection (TIR) at the sapphire–air interface [15–17]. The light extraction efficiency (LEE) of visible-light LEDs has been improved by reducing TIR loss using a silicon encapsulation layer [18–30]. In this paper, we propose a fluid encapsulation method by using polydimethylsiloxane (PDMS) with high refractive index ($n = 1.43$) and transmittance at a wavelength of 275 nm. The PDMS fluid has excellent properties, such as nontoxicity and resistance to oxidation, chemicals, and heat [31, 32]. The proposed encapsulation method enhances the light output efficiency of DUV-LEDs and reduces the adverse effects of LEDs on people and the environment. Mixing SiO_2 NPs into the PDMS fluid can also improve the light efficiency.
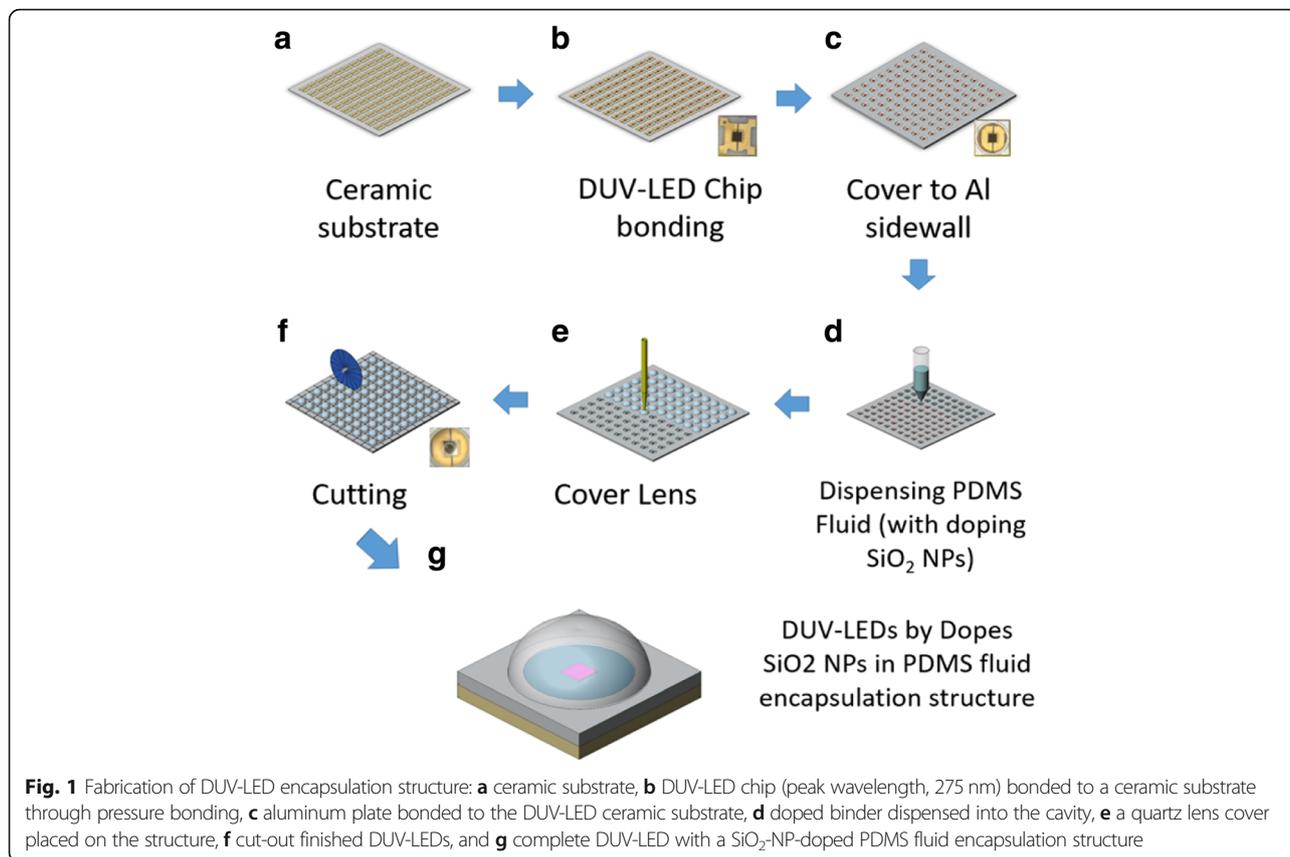
Methods and Materials

Figure 1 shows the schematic of the proposed DUV-LED encapsulation process consisting of the following steps: (a) a ceramic substrate is prepared with alumina as the electrode material; (b) the DUV-LED chip (peak wavelength 275 nm) is bonded to the ceramic substrate through hot pressure bonding; (c) the aluminum reflector sidewall cavity is bonded to the DUV-LED

* Correspondence: ocean@ntut.edu.tw; hcwang@ccu.edu.tw
[3]Department of Electro-Optical Engineering, National Taipei University of Technology, No.1, Sec. 3, Chung-Hsiao E. Rd, Taipei 10608, Taiwan
[4]Department of Mechanical Engineering and Advanced Institute of Manufacturing with High-tech Innovations, National Chung Cheng University, 168, University Rd., Min-Hsiung, Chia-Yi 62102, Taiwan
Full list of author information is available at the end of the article

Fig. 1 Fabrication of DUV-LED encapsulation structure: **a** ceramic substrate, **b** DUV-LED chip (peak wavelength, 275 nm) bonded to a ceramic substrate through pressure bonding, **c** aluminum plate bonded to the DUV-LED ceramic substrate, **d** doped binder dispensed into the cavity, **e** a quartz lens cover placed on the structure, **f** cut-out finished DUV-LEDs, and **g** complete DUV-LED with a SiO_2-NP-doped PDMS fluid encapsulation structure

ceramic substrate, and the chip is placed at the center of the opening; (d) PDMS fluid is dispensed into the aluminum reflector sidewall cavity; (e) coating binder and a hemispherical UV-transmissive glass with a diameter of 3 mm and height of 1.3 mm are placed on the outer ring of the aluminum reflector sidewall cavity; (f) individual DUV-LEDs are cut out along the scribe lines; and (g) a complete DUV-LED with a SiO_2-NP-doped PDMS fluid encapsulation structure is obtained. Figure 2a illustrates a conventional DUV-LED, and Fig. 2b shows a DUV-LED encapsulated with PDMS fluid proposed in this study. The intermediate layer comprises PDMS doped with SiO_2 NPs. The traditional method uses a vertical ceramic sidewall on the left- and right-hand sides of the DUV-LED flip chip, planar UV-transmissive glass on the top, and air as the medium between the DUV-LED flip chip and glass. The middle layer of the proposed design was an encapsulated structure of SiO_2 NPs in PDMS fluid with a hemispherical UV-transmissive glass structure above. Figure 2c plots the transmittance of the PDMS fluid at different wavelengths as obtained using an optical spectrophotometer measurement system (Hitachi, Tokyo, Japan). The graph reveals that the PDMS fluid transmittance was 85% at 275 nm. Figure 2d presents a photograph of the DUV-

LED with a surface area of 0.78×0.75 mm^2 (Dowa Co. Ltd., Tokyo, Japan) and its emission spectrum was captured at 200 mA forward current. The chip's dominant wavelength was 275 nm with a full width at half maximum of 12 nm. All data were obtained using an optical system SLM-20 integrating sphere (Isuzu Optics, Hsinchu, Taiwan). Table 1 lists the specifications (surface and material properties) of all the components of the proposed encapsulated DUV-LED.

A transmission electron microscopy image of the SiO_2 NPs (AEROSIL hydrophobic fumed silica, Frankfurt am Main, Germany) is presented in Fig. 2e. The NPs were prepared by first removing the moisture at 150 °C in an oven and then placing the NPs in a N_2 tank for 48 h to dry their surfaces. The average size of the NPs was set at 14 nm to prevent them from sticking together due to moisture.

Results and Discussion

Four types of DUV-LED encapsulation were employed and are shown in Fig. 3. Figure 3a shows DUV-LED (I) with a DUV-LED chip and aluminum reflector sidewalls at an angle of 60°. Figure 3b shows DUV-LED (II) in which the aluminum reflector sidewall cavity was filled with PDMS fluid. Figure 3c shows DUV-LED (III) in which the aluminum reflector sidewall cavity was filled
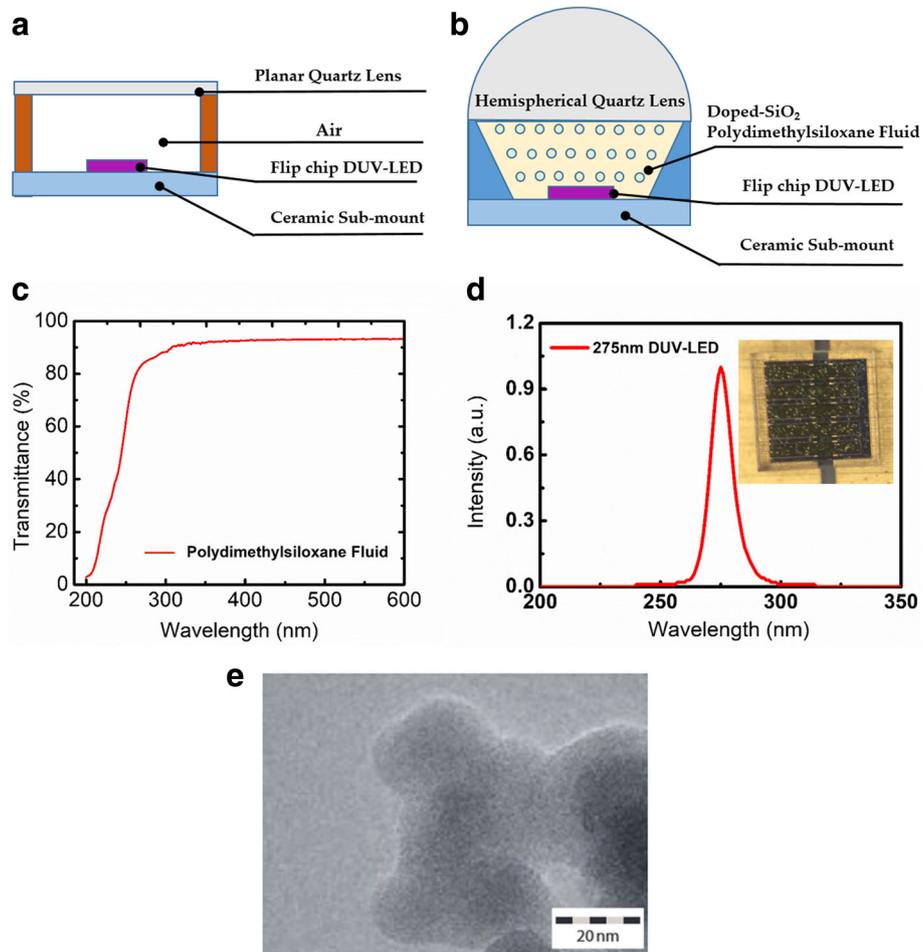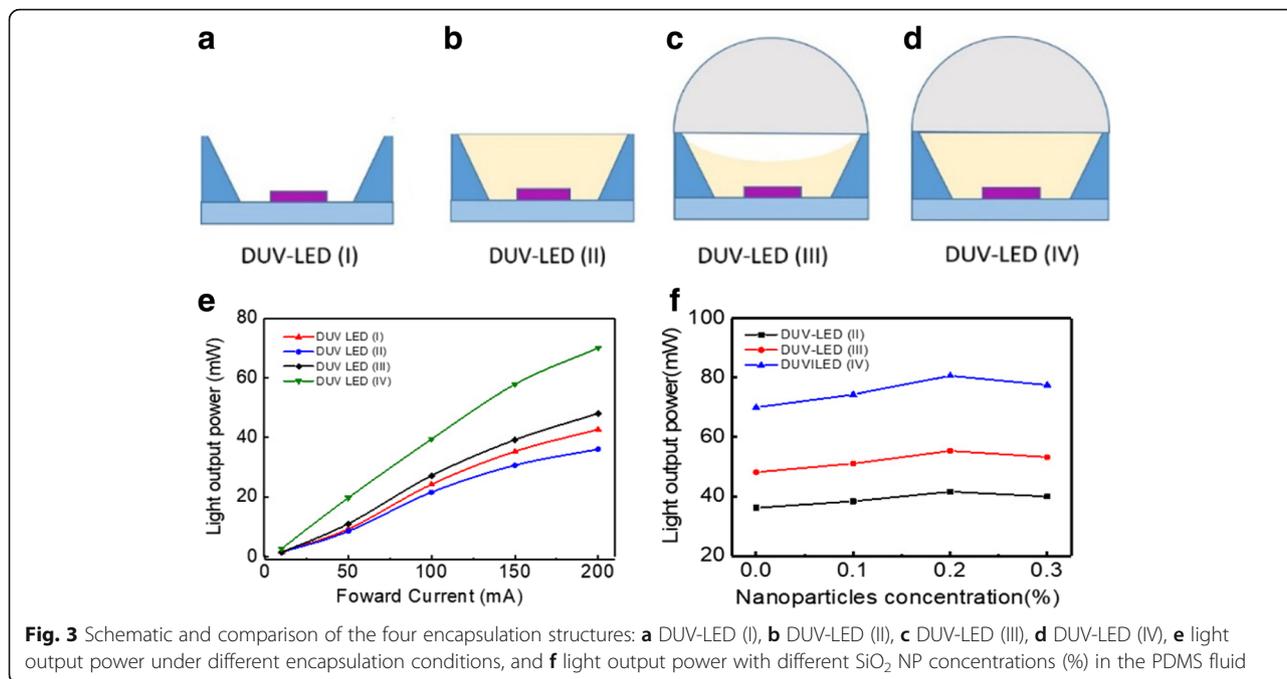
Fig. 2 DUV-LED encapsulation structure: **a** Schematic of conventional flip chip DUV-LED, **b** encapsulation structure and SiO$_2$ nanoparticle (NP)–doped polydimethylsiloxane (PDMS) fluid DUV-LED encapsulation structure, **c** transmittance of the PDMS fluid from 200–600 nm, **d** photograph of the DUV-LED and emission spectrum captured at a forward current of 200 mA for the proposed DUV-LED, and **e** high-resolution transmission electron microscopy image of SiO$_2$ NPs[26]

with slightly less PDMS fluid than that in DUV-LED (II) and with a hemispherical UV-transmissive glass cover. Figure 3d shows DUV-LED (IV) in which the aluminum reflector sidewall cavity was completely filled with PDMS fluid and a hemispherical UV-transmissive glass cover was used. Integrating sphere measurement was performed for the four types of DUV-LED encapsulation (Fig. 3e). When the driving current of the DUV-LED (I) chip was 200 mA, the light output power was 42.07 mW. By contrast, when the drive current of the DUV-LED

Table 1 Materials and characteristics of DUV-LED

Component	Characteristics	Material
DUV-LED	Peak wavelength = 275 nm	AlGaN
Nanoparticles	Average primary particle size = 14 nm Refractive index (n) = 1.45 Energy gap (Eg) = 9 eV	SiO$_2$
Tilt angle reflector sidewall cavity	Tilt angle = 60° Diameter = 2 mm	Aluminum
Hemispherical Quartz glass	Diameter = 4 mm High = 1.3 mm	Quartz
Encapsulation structure	Polished	Al$_2$O$_3$ ceramics
Encapsulation material	Refractive index (n) = 1.45	Polydimethylsiloxane fluid

Ye *et al. Nanoscale Research Letters* (2019) 14:236

Page 4 of 6

Fig. 3 Schematic and comparison of the four encapsulation structures: **a** DUV-LED (I), **b** DUV-LED (II), **c** DUV-LED (III), **d** DUV-LED (IV), **e** light output power under different encapsulation conditions, and **f** light output power with different SiO_2 NP concentrations (%) in the PDMS fluid

(II) chip was 200 mA, the light output power was 36.11 mW, which was 14.16% lower than that for DUV-LED (I). This condition occurred mainly because TIR transpired when PDMS fluid filled the aluminum reflector sidewall cavity. The extraction efficiency ratio of UV light coupled into the PDMS fluid to UV light coupled into air is given by the following equation [12]:

$$\frac{\eta_{PDMSfluid}}{\eta_{air}} = \frac{1 - cos_{\theta c, PDMS\ fluid}}{1 - cos_{\theta c, air}}, \qquad (1)$$

where $\theta_{c,PDMS\ fluid}$ and $\theta_{c,air}$ are the critical angles for TIR at the PDMS fluid DUV-LED and air UV-LED interfaces, respectively. When the driving current of the DUV-LED (III) chip was 200 mA, the optical output power was 48.126 mW, which was 14.39% higher than that for DUV-LED (I). This condition occurred mainly because the concave lens reduced the TIR but increased the LEE. However, DUV-LED (III) had an air gap, which hindered it from having the highest light output power among all the fabricated devices. When the driving current of the DUV-LED (IV) chip was 200 mA, the output power was 70.045 mW, which was 66.49% higher than that of DUV-LED (I). The DUV-LED (IV) encapsulation structure yielded the highest light output power because no air gap was present in the encapsulation, thus enabling the full transmission of DUV light from the DUV-LED. The light output power was also determined for DUV-LED (II), DUV-LED (III), and DUV (IV)

encapsulation when the PDMS fluid was doped with SiO_2 NPs (Fig. 3f). The DUV-LED (I) structure was not included in the comparison because it did not contain PDMS fluid. The weight percentage concentrations (%) of NP were set to 0, 0.1, 0.2, and 0.3 wt%. When the driving current of the DUV-LED (IV) chip was 200 mA, the light output power was 70.04, 74.32, 80.58, and 77.44 mW. Thus, a SiO_2 NP doping concentration of 0.2 wt% resulted in the highest LEE. Doping the PDMS fluid with SiO_2 NPs increased the amount of scattered light but decreased the amount of TIR. Doping with 0.2 wt% SiO_2 NP resulted in 15% higher LEE than doping with 0 wt% SiO_2 NP. Compared with that of DUV-LED (I), the LEE was 81.45% higher for a driving current of 200 mA. DUV-LED encapsulation was performed using the manufacturing methods outlined in Fig. 3. Table 2 shows the images of the operation at a driving current of 200 mA of the DUV-LED (IV) with PDMS fluid doping at 0.2 wt% SiO2 NPs. Figure 4 provides a comparison of the average interface temperatures of DUV-LED (I) and DUV-LED (IV) containing SiO_2 NP-doped PDMS fluid at different driving currents. When the driving current was 200 mA, the interface temperature in the DUV-LED (IV) device was 4 °C lower than that in the DUV-LED (I) device, revealing that the encapsulation structure effectively weakened the thermal temperature. Table 2 shows a temperature map of the DUV-LED (I) and DUV-LED (IV) that was obtained using an infrared thermal imager (ChingHsing Co. Ltd., Taipei, Taiwan). At the driving current of 140 mA, the DUV-LED (IV) had lower

Table 2 Image of DUV-LED (IV) containing PDMS fluid doped with SiO$_2$ NPs and its operation. Infrared thermal images of (a) the normal image of DUV-LED (I), (b) the normal image of DUV-LED (IV) in normal image, (c) the infrared thermal image of DUV-LED (I), and (d) the infrared thermal image of DUV-LED (IV) in normal image

	DUV-LED (I)	DUV-LED (IV)
Normal image		
Infrared thermal image		

operating temperature than the DUV-LED (I). For DUV-LED (I) without PDMS fluid, the temperature was the highest on the surface of the chip. The results in Fig. 4 and Table 2 reveal that the encapsulation structure with PDMS fluid doped with SiO$_2$ NPs has superior heat dissipation capability.
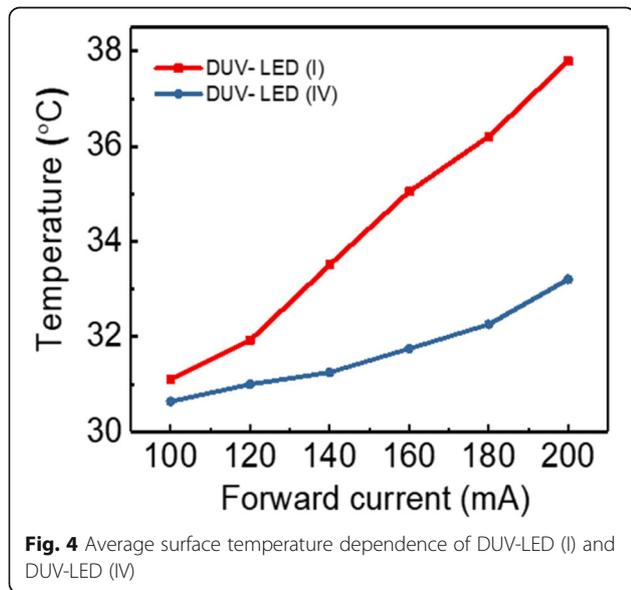
Conclusions

This paper proposes a new encapsulation method for improving the LEE of DUV-LEDs by doping the PDMS fluid with SiO$_2$ NPs. A considerably high light output power was achieved by using the SiO$_2$ NP-doped PDMS fluid encapsulation structure. IN particular, the light output power was 81.45% higher when the PDMS fluid doped with 0.2 wt% SiO$_2$ NPs was placed in the cavity rather than in the air. This enhancement is attributed to the reduced TIR and the additional light scattering in the PDMS fluid because of the addition of SiO$_2$ NPs. The average interface temperature was 4 °C lower at a driving current of 200 mA. The proposed architecture was compact and feasible for fabricating high-LEE AlGaN-based DUV-LEDs in the future.

Fig. 4 Average surface temperature dependence of DUV-LED (I) and DUV-LED (IV)

Ye *et al. Nanoscale Research Letters*   (2019) 14:236

Page 6 of 6

Authors' Contributions
ZTY and YMP performed the experiments and fabricated the samples. YMP
and HCW coordinated the project. CHL fabricated the DUV-LED. CPL and
CHL obtained the spectra and performed the optical measurements. CPL
performed the infrared thermal measurements. ZTY, CPL, HTN, and HCW
wrote the draft of the paper. All the authors read and approved the final ver-
sion of the paper.

Availability of Data and Materials
Not applicable

Competing Interests
The authors declare that they have no competing interests.

Author details
[1]Department of Electro-Optical Engineering, National United University, 2,
Lienda, Miaoli 26063, Taiwan. [2]Department of Photonics, National Chiao
Tung University, 1001 University Road, Hsinchu 300, Taiwan. [3]Department of
Electro-Optical Engineering, National Taipei University of Technology, No.1,
Sec. 3, Chung-Hsiao E. Rd, Taipei 10608, Taiwan. [4]Department of Mechanical
Engineering and Advanced Institute of Manufacturing with High-tech
Innovations, National Chung Cheng University, 168, University Rd.,
Min-Hsiung, Chia-Yi 62102, Taiwan.

References
1. Lin CH, Huang CH, Pai YM, Lee CF, Lin CC, Sun CW, Chen CH, Sher CW, Kuo
   HC (2018) Novel method for estimating phosphor conversion efficiency of
   light-emitting diodes. Crystals. 8:442
2. Lin CH, Pai YM, Kang CY, Lin HY, Lee CF, Chen XY, Tu HH, Yang JJ, Chen CH,
   Lin CC (2018) Square column structure of high efficiency, reliable, uniformly
   flexible LED devices. Crystals. 8:472
3. Muramoto Y, Kimura M, Nouda S (2014) Development and future of
   ultraviolet light-emitting diodes: UV-LED will replace the UV lamp.
   Semicond. Sci. Technol. 29:084004
4. Shatalov M, Sun W, Jain R, Lunev A, Hu X, Dobrinsky A, Bilenko Y, Yang J,
   Garrett GA, Rodak LE (2014) High power AlGaN ultraviolet light emitters.
   Semicond. Sci. Technol. 29:084007
5. Shur MS, Gaska R (2010) Deep-ultraviolet light-emitting diodes. IEEE Trans.
   Electron Devices 57:12–25
6. Sun W, Adivarahan V, Shatalov M, Lee Y, Wu S, Yang J, Zhang J, Khan MA
   (2004) Continuous wave milliwatt power AlGaN light emitting diodes at
   280 nm. Jpn. J. Appl. Phys. 43:L1419
7. Taniyasu Y, Kasu M, Makimoto T (2006) An aluminium nitride light-emitting
   diode with a wavelength of 210 nanometres. Nature 441:325
8. Wu IC, Syu HY, Jen CP, Lu MY, Chen YT, Wu MT, Kuo CT, Tsai YY, Wang HC
   (2018) Early identification of esophageal squamous neoplasm by
   hyperspectral endoscopic imaging. 8:13797
9. Wang HC, Nguyen NV, Lin RY, Jen CP (2017) Characterizing esophageal
   cancerous cells at different stages using the dielectrophoretic impedance
   measurement method in a microchip. 17:1053
10. Wu IC, Weng YH, Lu MY, Jen CP, Fedorov VE, Chen WC, Wu MT, Kuo CT,
    Wang HC (2017) Nano-structure ZnO/Cu$_2$O photoelectrochemical and self-
    powered biosensor for esophageal cancer cell detection. 25(7):7689-7706
11. Kneissl M, Yang Z, Teepe M, Knollenberg C, Johnson NM, Usikov A, Dmitriev
    V (2006) Ultraviolet InAlGaN light emitting diodes grown on hydride vapor
    phase epitaxy AlGaN/sapphire templates. Jpn. J. Appl. Phys. 45:3905
12. Liang R, Wu F, Wang S, Chen Q, Dai J, Chen C (2017) Enhanced optical and
    thermal performance of eutectic flip-chip ultraviolet light-emitting diodes
    via AlN-doped-silicone encapsulant. IEEE Trans. Electron Devices 64:467–471
13. Pai YM, Lin CH, Lee CF, Lin CP, Chen CH, Kuo HC, Ye ZT (2018) Enhancing
    the light-extraction efficiency of AlGaN-based deep-ultraviolet light-emitting
    diodes by optimizing the diameter and tilt of the aluminum sidewall.
    Crystals. 8:420
14. Tsuzuki H, Mori F, Takeda K, Iwaya M, Kamiyama S, Amano H, Akasaki I,
    Yoshida H, Kuwabara M, Yamashita Y (2009) Novel UV devices on high-
    quality AlGaN using grooved underlying layer. J Cryst Growth. 311:
    2860–2863
15. Su YK, Wang PC, Lin CL, Huang GS, Wei CM (2014) Enhanced light
    extraction using blue LED package consisting of TiO2 - doped silicone layer
    and silicone lens. Electron Device Lett. 35:575–577
16. Wang PC, Su YK, Lin CL, Huang GS (2014) Improving performance and
    reducing amount of phosphor required in packaging of white LEDs with
    TiO2-doped silicone. Electron Device Lett. 35:657–659
17. Yin WJ, Chen S, Yang JH, Gong XG, Yan Y, Wei SH (2010) Effective band gap
    narrowing of anatase TiO 2 by strain along a soft crystal direction. Appl.
    Phys. Lett. 96:221901
18. Zhao P, Zhao H (2012) Analysis of light extraction efficiency enhancement
    for thin-film-flip-chip InGaN quantum wells light-emitting diodes with GaN
    micro-domes. Optics Express. 20:A765–A776
19. Zheng H, Li L, Lei X, Yu X, Liu S, Luo X (2014) Optical performance
    enhancement for chip-on-board packaging LEDs by adding TiO2/silicone
    encapsulation layer. Electron Device Lett. 35:1046–1048
20. Gnani E, Reggiani S, Colle R, Rudan M (2000) Band-structure calculations of
    SiO/sub 2/by means of Hartree-Fock and density-functional techniques. IEEE
    Trans. Electron Devices. 47:1795–1803
21. Tan C, Arndt J (2000) Temperature dependence of refractive index of glassy
    SiO2 in the infrared wavelength range. J. Phys. Chem. Solids 61:1315–1320
22. Xi JQ, Schubert MF, Kim JK, Schubert EF, Chen M, Lin SY, Liu W, Smart JA
    (2007) Optical thin-film materials with low refractive index for broadband
    elimination of Fresnel reflection. Nat. Photonics 1:176
23. Bradley S, White K, McCay J, Brown R, Musgrove D, Wilson S, Stern M, Luster
    M, Munson A (1994) Immunotoxicity of 180 day exposure to
    polydimethylsiloxane (silicone) fluid, gel and elastomer and polyurethane
    disks in female B6C3F1 mice. Drug Chem. Toxicol. 17:221–269
24. Carrillo F, Gupta S, Balooch M, Marshall SJ, Marshall GW, Pruitt L, Puttlitz CM
    (2005) Nanoindentation of polydimethylsiloxane elastomers: Effect of
    crosslinking, work of adhesion, and fluid environment on elastic modulus. J.
    Mater. Res. Technol. 20:2820–2830
25. Hshieh FY (1998) Shielding effects of silica-ash layer on the combustion of
    silicones and their possible applications on the fire retardancy of organic
    polymers. Fire and Materials 22:69–76
26. Huang YR, Chou YC, Wu ZL, Huang KC, Ting SY, Yao YF, Chiang PJ, Tseng
    SH, Wang HC (2018) Light extraction efficiency enhancement of flip-chip
    blue light-emitting diodes by anodic aluminum oxide. 9:1602-1612.
27. Huang YR, Luo WC, Wang HC, Feng SW, Kuo CT, Lu CM (2017) How smart
    LEDs lighting benefit color temperature and luminosity transformation. 10:518.
28. Wang HC, Chiang YT, Lin CY, Lu MY, Lee MK, Feng SW, Kuo CT (2016) All-
    reflective RGB LED flashlight design for effective color mixing. 24(5):4411-4420
29. Wang HC, Chen YT (2012) Optimal lighting of RGB LEDs for oral cavity
    detection. 20:10186-10199
30. Wang HC, Yu XY, Chueh YL, Malinauskas T, Jarasiunas K, Feng SW (2011)
    Suppression of surface recombination in surface plasmon coupling with an
    InGaN/GaN multiple quantum well sample. 19:18893.
31. Kanellopoulos A, Owen M (1971) The adsorption of polydimethylsiloxane
    polyether ABA block copolymers at the water/air and water/silicone fluid
    interface. J. Colloid Interface Sci. 35:120–125
32. Wilson K, Goff J, Riffle J, Harris L, St Pierre T (2005) Polydimethylsiloxane-
    magnetite nanoparticle complexes and dispersions in polysiloxane carrier
    fluids. Polym. Adv. Technol. 16:200–211

Publisher's Note
Springer Nature remains neutral with regard to jurisdictional claims in
published maps and institutional affiliations.