Silane-Functionalized Carbon Dots for Full-Spectrum White Light-Emitting Diodes

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Experimental details

Synthesis of SiCDs and their glasses

The SiCDs were synthesized using a solvothermal method. Initially, 0.192 g of CA (99.0%, Innochem) was dissolved in 5 mL of a selected solvent (water, ethanol, DMF, or acetone). Subsequently, 5 mL of DEAS (98%, Bide Pharmatech) was added to the resulting solution under stirring to ensure homogeneity. The mixture was then transferred to a 25-mL Teflon-lined stainless-steel autoclave and subjected to heating at 180 °C for 12 h.

The crude SiCDs were purified using a two-step protocol. First, the crude product was subjected to dialysis against ethanol using a dialysis membrane with a MWCO of 1000 Da for 3 days to eliminate low-molecular-weight byproducts. Subsequently, the dialyzed solution was filtered through a 0.22-μm pore-size membrane to remove residual aggregates or particulates.

The SiCD glasses were fabricated through the self-polymerization of SiCDs. The liquid-phase SiCDs were transferred into a Petri dish (3 cm in diameter) and then dried at room temperature (~25 °C) for 5 days to remove residual solvent, followed by thermal treatment in an oven at 80 °C for 2 days to promote further crosslinking and consolidation into a glassy state.

Fabrication of WLEDs

The WLEDs were fabricated by the combination of a violet LED chip (λ_{EL} = 395 nm) and these SiCDs. Specifically, the liquid-state B-, G-, and R-SiCDs were sequentially dispensed onto the chip and then dried in an oven at 60 °C for 1 h. The mass ratio is around 1:1:1.5:2, respectively.

Characterization

Transmission electron microscopy (TEM) analysis was conducted using a JEOL 2100F instrument, performing at an acceleration voltage of 200 kV. X-ray diffraction (XRD) patterns were acquired using a Bruker D8 Discover diffractometer using a Cu Kα irradiation (1.54056 Å). Fourier-transform infrared spectra (FTIR) were recorded from SiCDs dispersed in KBr (IR grade, ≥99%, Sigma-Aldrich), using a Shimazu IRTracer 100 spectrometer. X-ray photoelectron spectroscopy (XPS) analyses were performed on a Thermo Scientific ESCALAB 250Xi with a monochromatized micro-focused Al Ka X-ray source (1486.68 eV). Photoluminescence (PL) spectra were measured using an Edinburgh FLS980 fluorescence spectrometer, with a 450 W ozone-free xenon arc lamp as the excitation source. Time-resolved PL was measured with a standard photomultiplier (PMT-900, Edinburgh) as the detector, using a pulsed diode laser (λ_{ex} = 445 nm, pulse width = 95 ps) as the excitation source. Absorption spectra were obtained by a Hitachi U-3010 UV-Vis spectrophotometer. The PLQY was measured with a spectrometer (C9920-02G, Hamamatsu) equipped with an integrating sphere. The properties of the WLED were measured by Everfine HASS-1200 LED system.

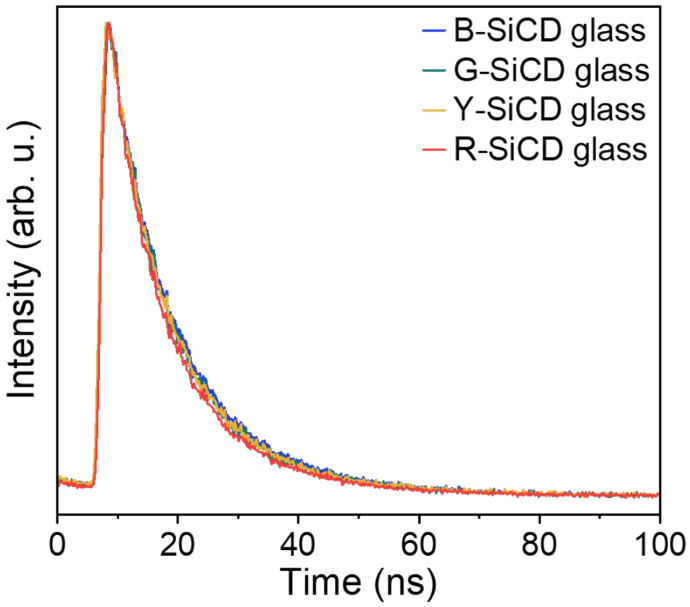

Figure S1. Time-resolved PL decay curves of the SiCD glasses.

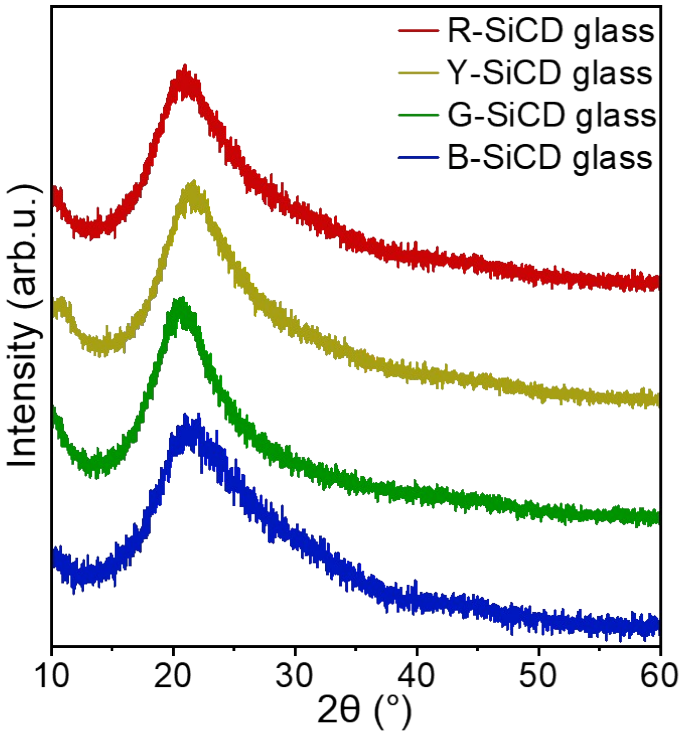

Figure S2. XRD patterns of the SiCD glasses in the form of powders.

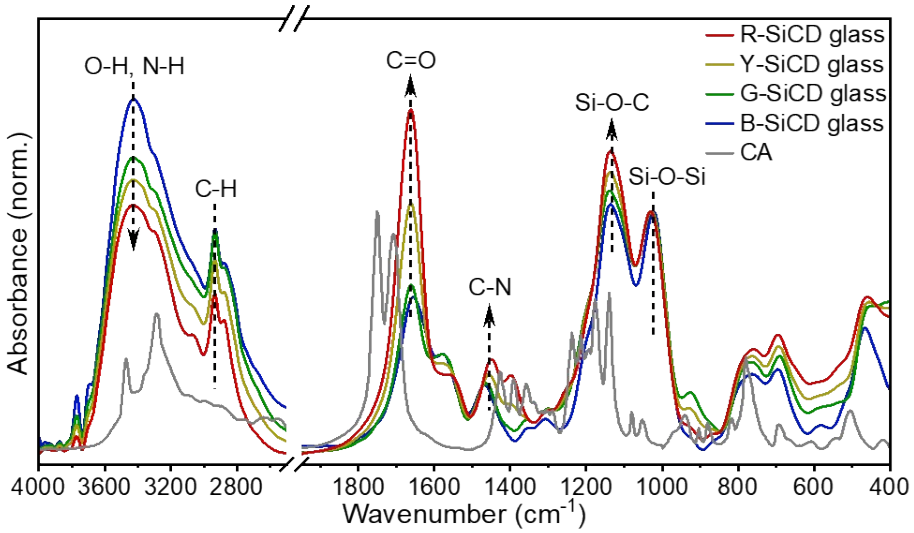

Figure S3. FTIR spectra of the SiCD glasses in the form of powders and CA (as a reference).

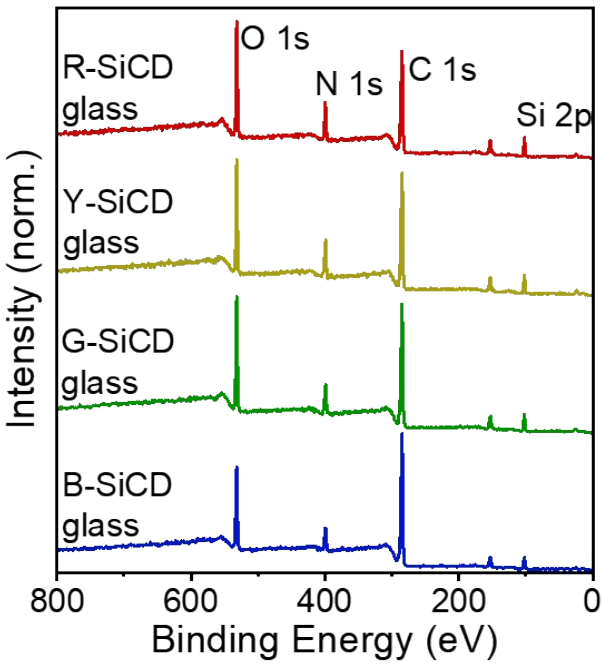

Figure S4. XPS survey spectra of the SiCD glasses.

Table S1. Atomic ratios of Si 2p, C 1s, N 1s and O 1s in the SiCD glasses from their XPS survey spectra.

Sample	Si 2p	C 1s	N 1s	O 1s
R-SiCD glass	8.10 at.%	58.23 at.%	12.25 at.%	21.42 at.%
Y-SiCD glass	7.89 at.%	59.55 at.%	11.86 at.%	20.70 at.%
G-SiCD glass	7.41 at.%	61.64 at.%	10.08 at.%	20.87 at.%
B-SiCD glass	6.19 at.%	67.86 at.%	8.68 at.%	17.27 at.%

Table S2. Atomic ratios of the deconvoluted components derived from the high-resolution C 1s spectra of the SiCD glasses.

Sample	C-Si	C-C	C-N	C-O	C=O
R-SiCD glass	283.6 eV 9.56 at.%	284.5 eV 51.40 at.%	285.5 eV 20.00 at.%	286.1 eV 7.02 at.%	287.6 eV 12.02 at.%
Y-SiCD glass	283.5 eV 8.00 at.%	284.5 eV 54.54 at.%	285.6 eV 19.64 at.%	286.2 eV 7.42 at.%	287.9 eV 10.40 at.%
G-SiCD glass	283.4 eV 6.17 at.%	284.5 eV 57.51 at.%	285.7 eV 19.14 at.%	286.5 eV 9.08 at.%	288.2 eV 8.35 at.%
B-SiCD glass	283.4 eV 4.20 at.%	284.5 eV 59.47 at.%	285.4 eV 18.20 at.%	286.3 eV 11.73 at.%	288.0 eV 6.40 at.%

Table S3. Atomic ratios of the deconvoluted components derived from the high-resolution N 1s spectra of the SiCD glasses.

Sample	Pyridinic N	Pyrrolic N	Graphitic N	Amino N
R-SiCD glass	398.4 eV 30.76 at.%	399.1 eV 32.33 at.%	399.8 eV 19.07 at.%	400.9 eV 17.84 at.%
Y-SiCD glass	398.3 eV 32.50 at.%	398.9 eV 27.87 at.%	399.8 eV 23.32 at.%	400.9 eV 16.31 at.%
G-SiCD glass	398.4 eV 25.40 at.%	398.9 eV 32.96 at.%	399.8 eV 28.01 at.%	400.9 eV 13.63 at.%
B-SiCD glass	398.4 eV 31.05 at.%	399.0 eV 22.88 at.%	399.8 eV 35.40 at.%	401.1 eV 10.67 at.%

Table S4. Atomic ratios of the deconvoluted components derived from the high-resolution O 1s spectra of the SiCD glasses.

Sample	Si-O	C-O	C=O
R-SiCD glass	531.0 eV 33.61 at.%	531.9 eV 32.29 at.%	532.6 eV 34.09 at.%
Y-SiCD glass	531.0 eV 30.81 at.%	531.9 eV 37.13 at.%	532.6 eV 32.06 at.%
G-SiCD glass	531.0 eV 24.32 at.%	531.9 eV 47.68 at.%	532.7 eV 28.00 at.%
B-SiCD glass	531.0 eV 18.62 at.%	531.9 eV 55.93 at.%	532.8 eV 25.45 at.%

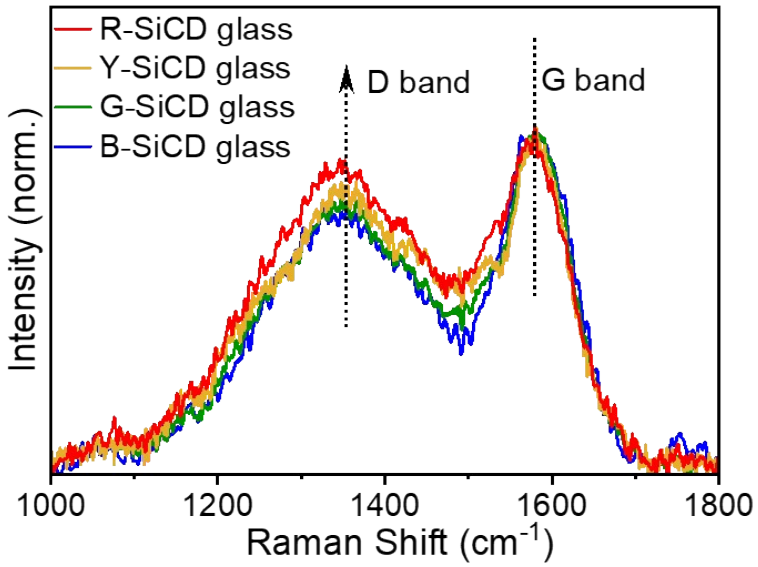

Figure S5. Raman spectra of the SiCD glasses in the form of powders.

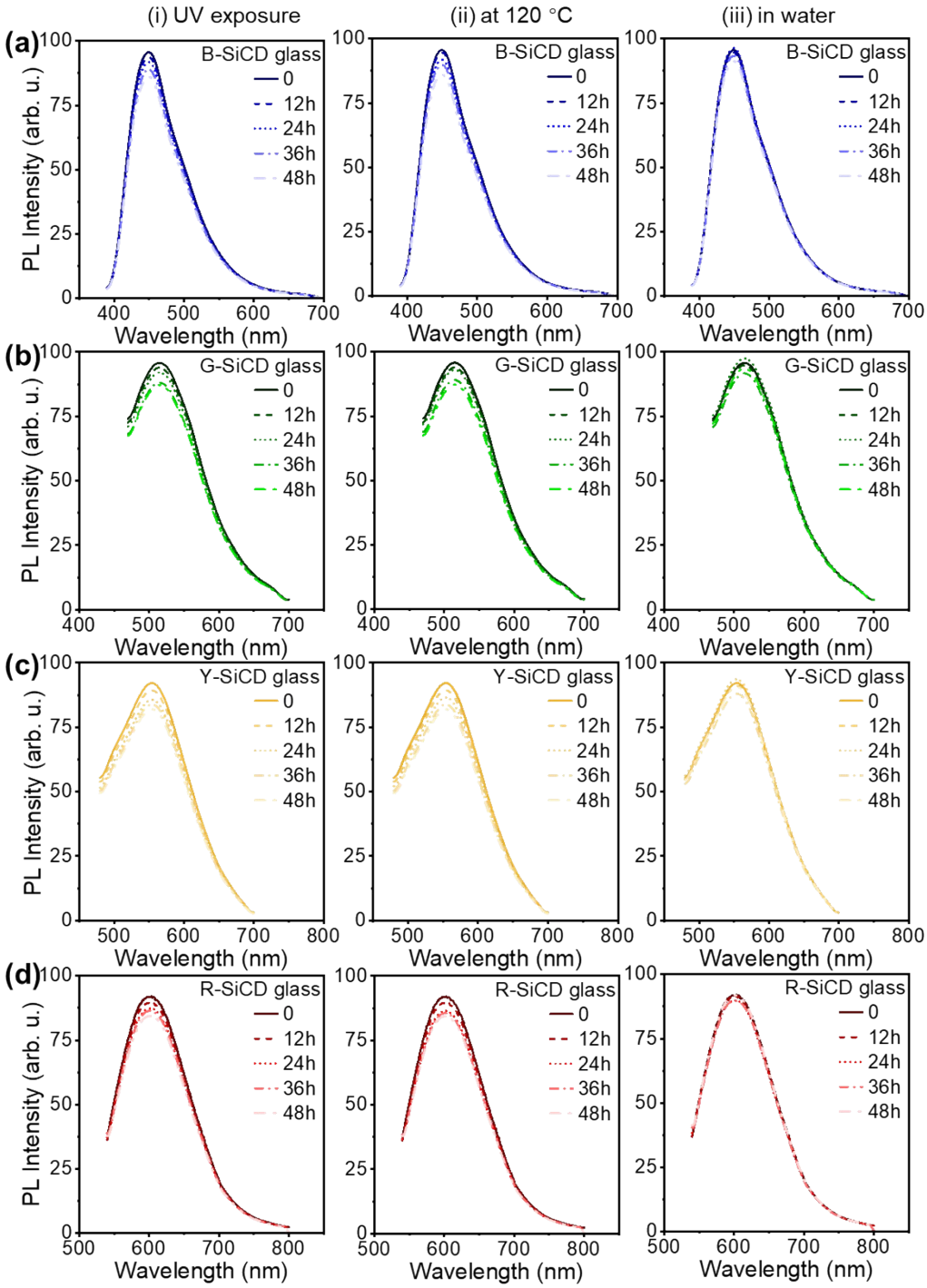

Figure S6. PL spectra of **(a)** B-SiCD glass, **(b)** G-SiCD glass, **(c)** Y-SiCD glass and **(d)** Y-SiCD glass as a function of time under (i) continuous UV irradiation at 365 nm, (ii) thermal stress at 120 °C, and (iii) underwater immersion. The excitation wavelengths for B-, G-, Y-, and R-SiCD glasses are 370, 450, 460, and 520 nm respectively.

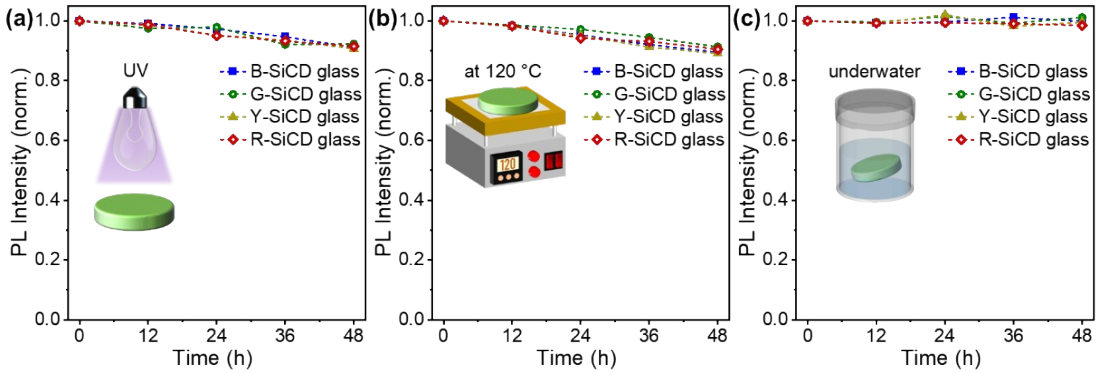

Figure S7. Normalized PL intensities of SiCD glasses as a function of time under **(a)** continuous UV irradiation at 365 nm, **(b)** thermal stress at 120 °C, and **(c)** and underwater immersion.

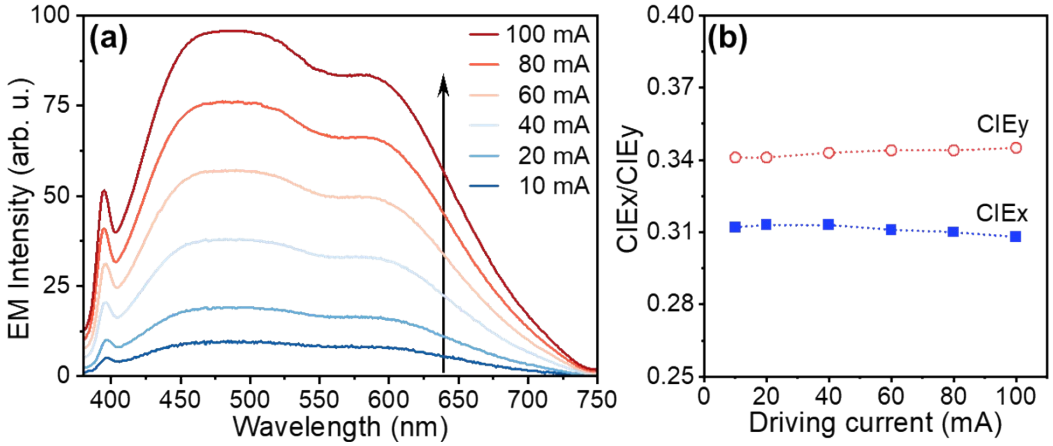

Figure S8. (a) EL spectra and **(b)** corresponding CIE coordinates of the fabricated WLEDs under different driving currents.