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Supporting Information:

Highly Luminescent Flexible Amino-Functionalized Graphene Quantum Dots@Cellulose Nanofiber-Clay Hybrids for White Light-Emitting Diodes

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Figure S1. Comparison of energy levels and HOMO–LUMO gaps in armchair-edged $C_{54}H_{18}$ with different functional groups.



Figure S2. a) Bright-field TEM image of green-emitting afGQDs. b) The size distributions of green-emitting af-GQDs.



Figure S3. a) CV curves (anodic scan) of afGQDs: blue line, blue-emitting afGQDs; green line, green-emitting afGQDs; orange-line, orange-emitting afGQDs. The LUMO

levels can be calculated by the following empirical equation: $LUMO = -e(E_{red} + 4.4) \text{ eV}.$

b) Energy level diagram for afGQDs: (i), blue-emitting afGQDs; (ii), green-emitting afGQDs; (iii), orange-emitting afGQDs. The HOMO levels were determined by subtracting excitation energy from LUMO energy, where the excitation energy was estimated by the direct conversion of emission peak into eV.



Figure S4. C1s (a) and N 1s (b) X-ray photoelectron spectra for afGQDs; blue line: blueemitting afGQDs, green line: green-emitting afGQDs, yellow line: yellow-emitting afGQDs, and orange line: orange-emitting afGQDs. c) The table summarises the C/N ratios calculated from the atomic concentrations: I, blue-emitting afGQDs; II, greenemitting afGQDs; III, yellow-emitting afGQDs; IV, orange-emitting afGQDs.



Figure S5. Comparison of FT-IR spectra: (a) green-emitting afGQDs, (b) starting OGSs. New peaks at 1243, 1617, and 3300–3600 cm⁻¹ appeared after the amino-hydrothermal treatment. These peaks were assigned, respectively, to C-N in-plane, N-H out-of-plane, and N-H in-plane stretching of the amine groups. Additionally, characteristic amide–carbonyl (–NH-CO–) stretching vibration was observed at 1650 cm⁻¹, which implies the formation of amide groups through interactions with the carboxylic groups as Lewis acids.



Figure S6. XRD pattern from green-emitting afGQDs@CNF-clay film.



Figure S7. TG-DTA curve with the weight loss and exo-endothermal reaction of the green-emitting afGQDs@CNF-clay film. Two stages of weight loss were apparent: (i) substantial weight loss attributable to the release of water molecules adsorbed on the clay (<100 °C) and (ii) thermal decomposition of the cellulose nanofiber (320 °C).



Figure S8. Luminescence spectra of blue LED with CNF-clay films without afGQDs under various forward currents.



Figure S9. Excitation power dependence of PL intensity for green-emitting afGQDs@CNF-clay hybrids. The PL measurements were carried out using a 355 nm Nd:YAG laser and a high-sensitive photomultiplier tube detector at room temperature. The PL intensity increased linearly as the laser power was below 20 mWcm⁻² and then showed slight saturation at higher powers.