Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Electronic Supplementary Information

Synthesis of Single-particle Level White-Light-Emitting Carbon Dots via a One-Step Microwave method

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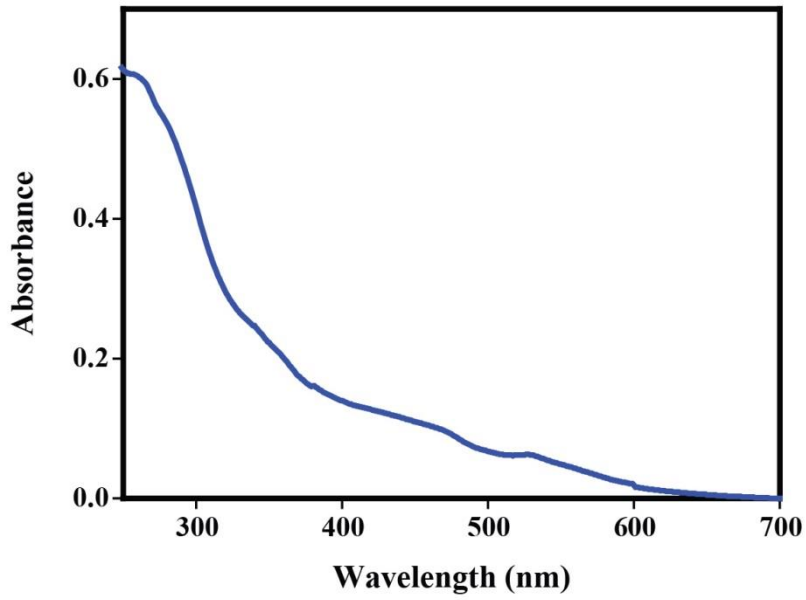

Fig. S1 Absorption spectrum of Cdots in water before subjecting to column chromatography.

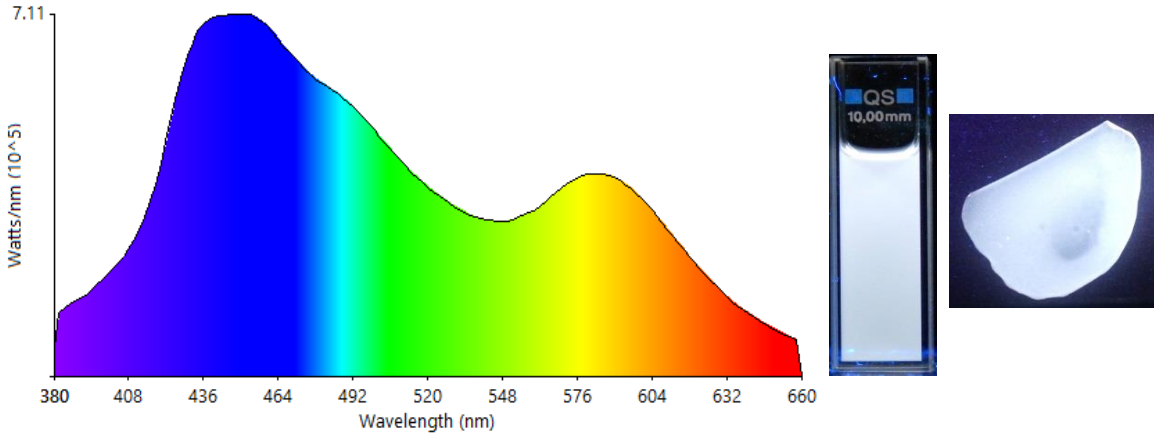

Fig. S2 Emission spectrum of Cdots prior to column chromatography when excited by 340 nm wavelength light and corresponding digital images in water and PVA matrix under 365 nm excitation in a UV lamp.

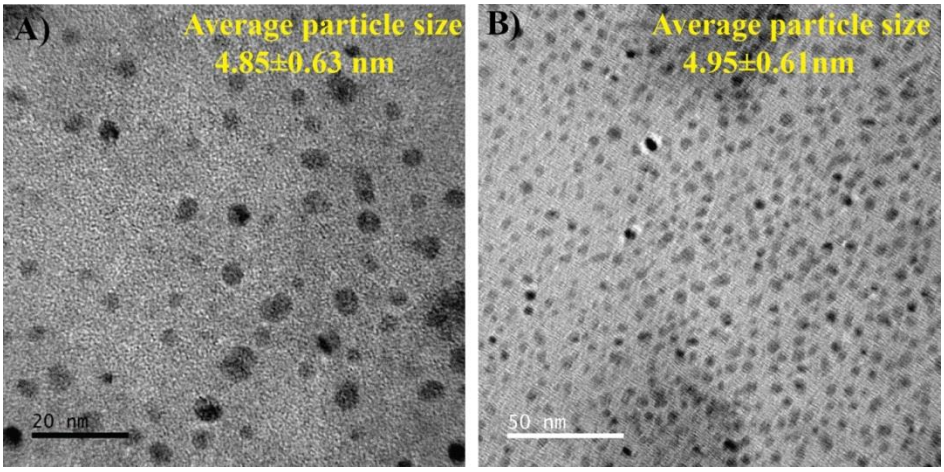

Fig. S3 TEM images of two different fractions: (A) fraction 2 and (B) fraction 4 of Cdots as obtained from column chromatography.

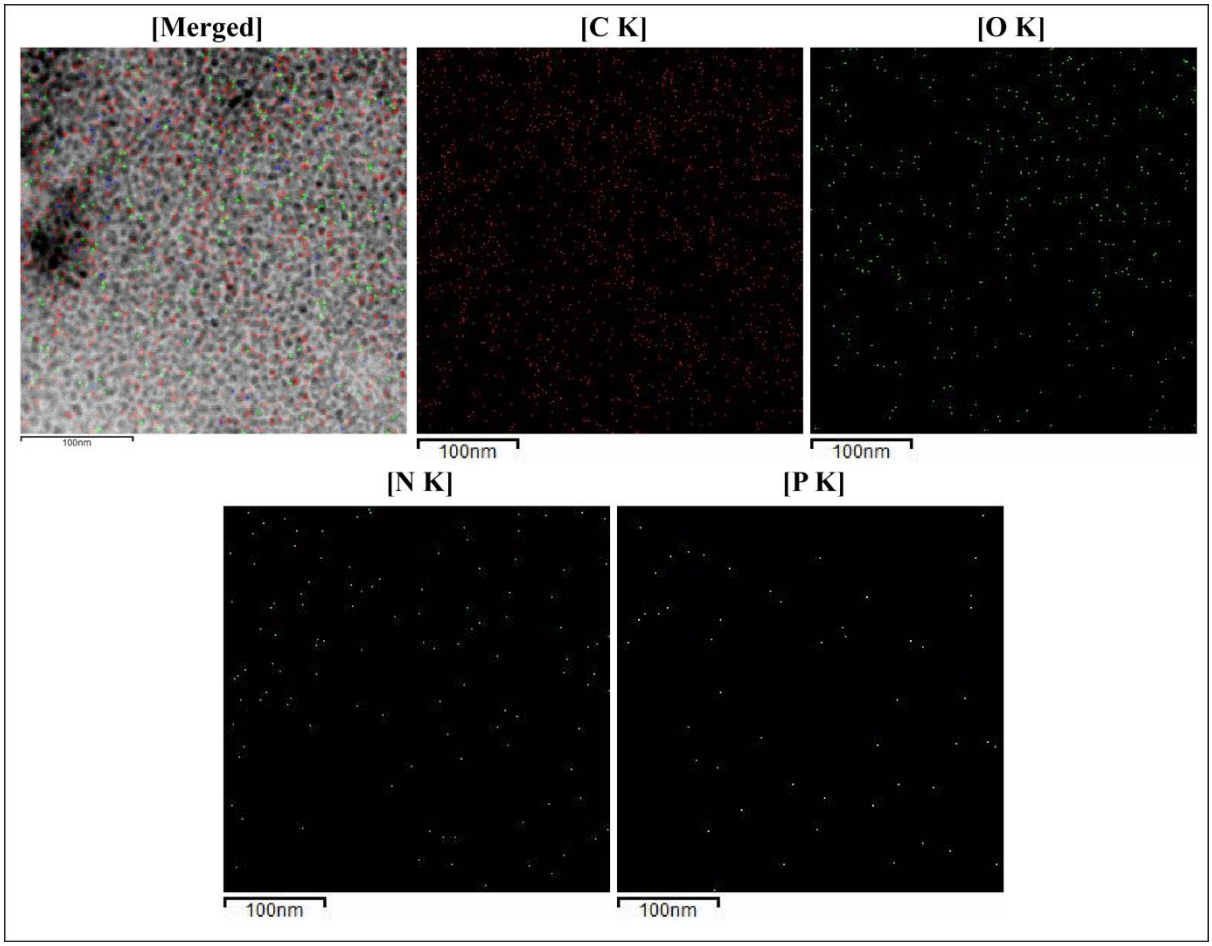

Fig. S4 Bright-field TEM image (merged) and STEM-EDS mapping of Cdots in the same area C K edge, O K edge, N K edge and P K edge revealed uniform presence of C, O, N and P.

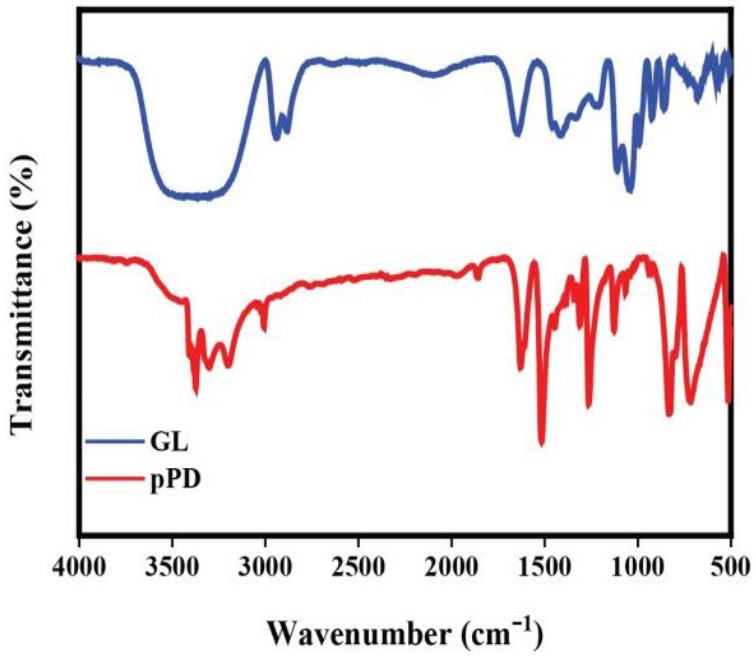

Fig. S5 FT-IR spectra of starting materials glycerol (GL) and p-phenylenediamine (pPD).

Table. S1 FT-IR peak assignment for Cdots, glycerol (GL) and p-phenylenediamine (pPD).[1, 2]

	Cdots		GL		pPD	
Sl. No.	**Center (cm^{-1})**	**Assignment**	**Center (cm^{-1})**	**Assignment**	**Center (cm^{-1})**	**Assignment**
1	3430	O-H/N-H stretching	3385 (broad)	O-H stretching	3409 & 3375	NH_2 stretching
2	2920 & 2847	C-H stretching	2940 & 2878	C-H stretching	3028 & 3008	CH stretching
3	1730	C=O stretching	1643	HOH bending(due to water)	1630	NH_2 scissoring
4	1600	C=C/C=N	1461 & 1417	CH_2 bending	1517 & 1311	C-C stretching
5	1448	C-N/C-P	1326	CH_2 wagging	1444	C-C stretching / C-N bending
6	1222	C-O-C	1110 & 1040	C-O stretching	1262	C-NH_2 stretching
7	1114	P=O	992 & 924	C-O-H stretching	1127	C-C bending
8	1056	P-O-R (R =alkyl)			1065 & 1041	NH_2 twisting
9	930	P-O-R (R=aromatic)			825 & 799	C-H bending

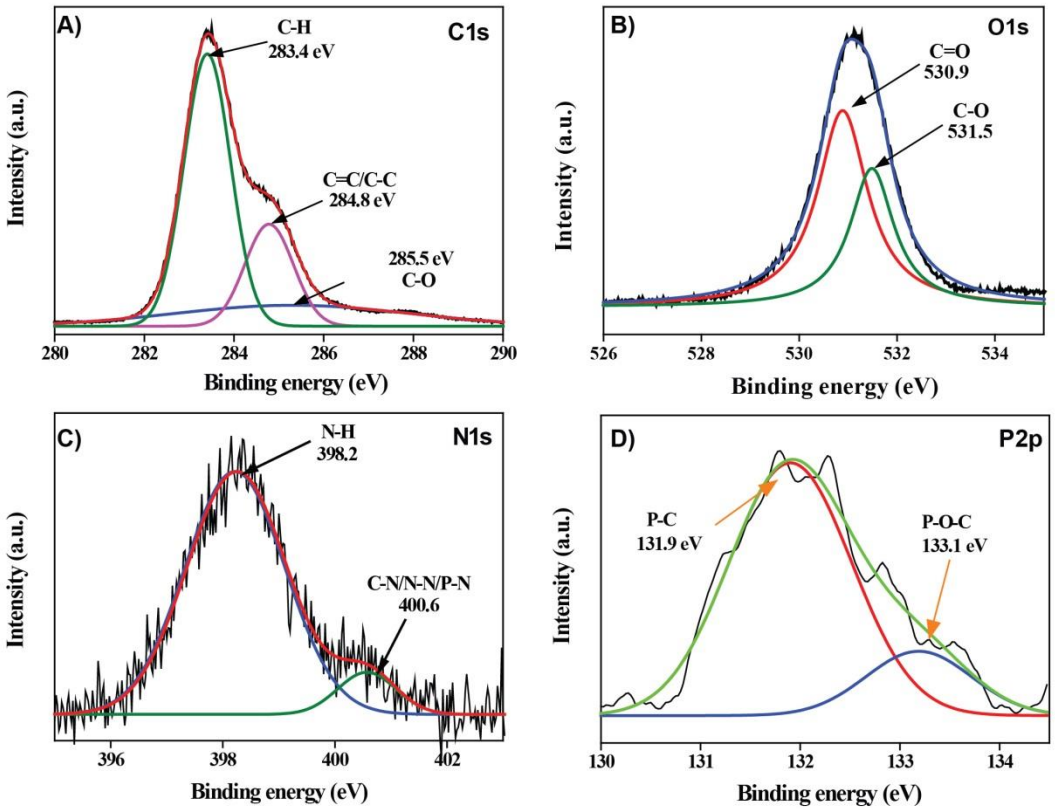

Fig. S6 Deconvoluted XPS spectra for (A) C_{1s}, (B) O_{1s}, (C) N_{1s} and (D) P_{2p} peaks of Cdots corresponding to Fig. 2B in the manuscript.

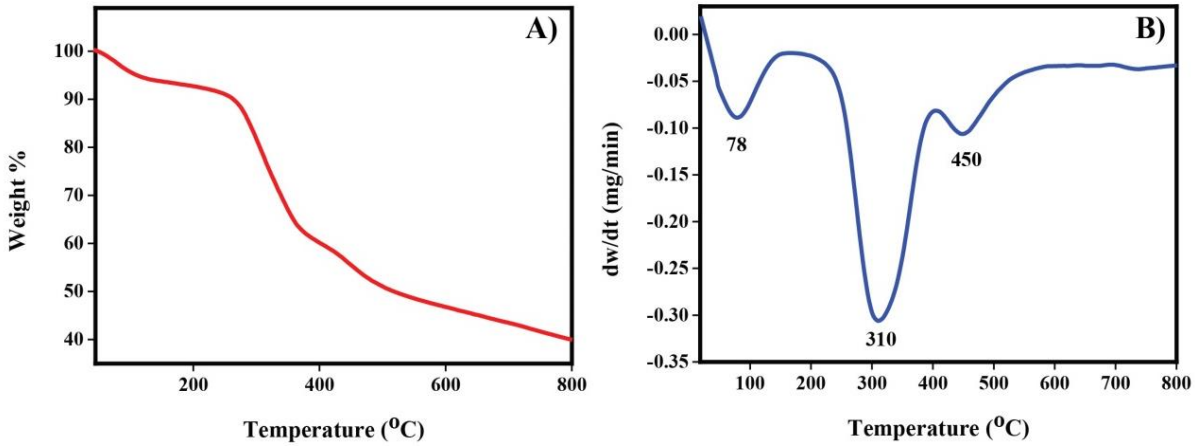

Fig. S7 A) Thermogravimetric analysis (TGA) and B) differential thermal analysis (DTA) curves of Cdots.

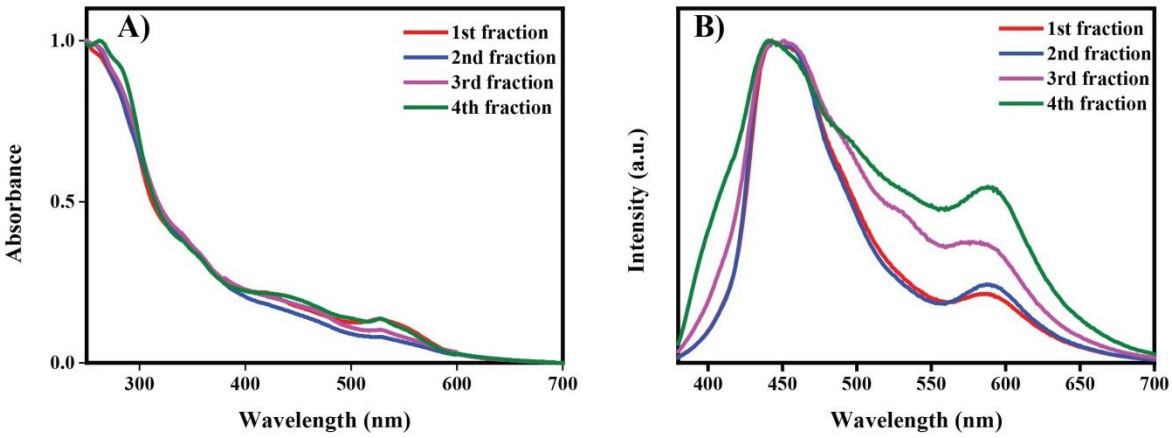

Fig. S8 A) Normalized absorption and B) normalized emission spectra (λ_{ex}=365 nm) of various fractions of Cdots in water as obtained from column chromatography.

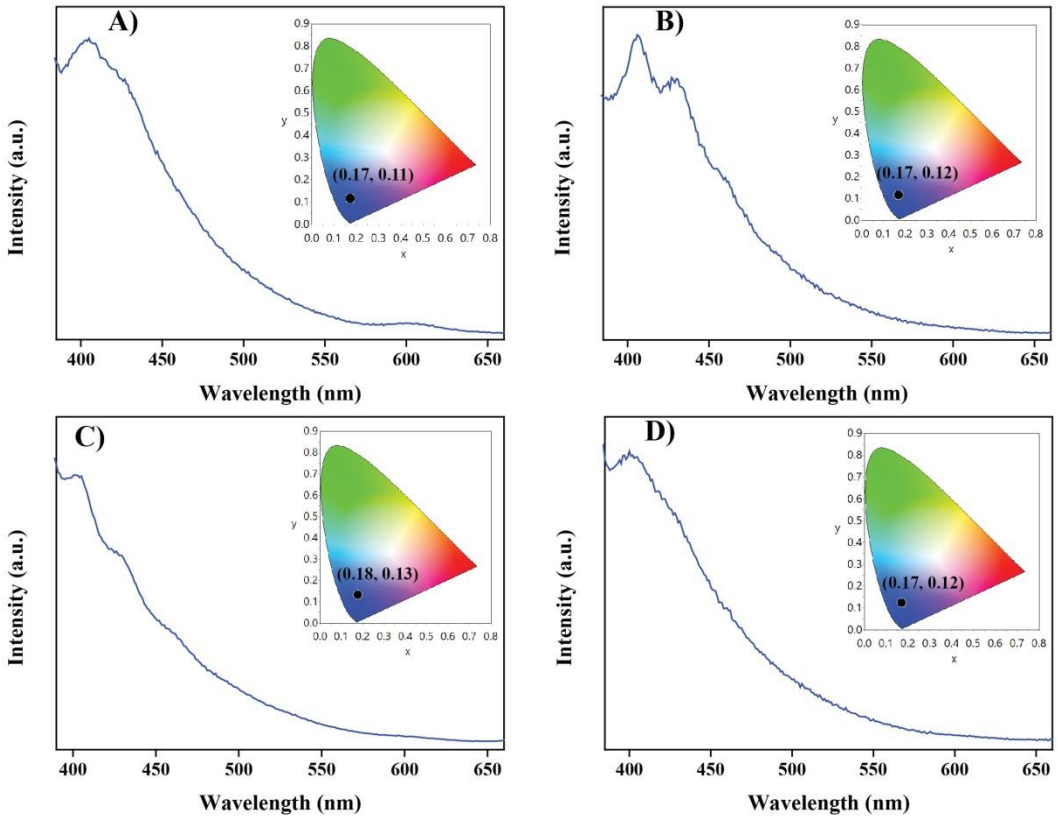

Fig. S9 Photoluminescent spectra and corresponding CIE chromaticity diagrams of WLE Cdots in toluene (A), o-xylene (B), m-xylene (C) and p-xylene (D) with λ_{ex} =340 nm.

The Cdots were found to be very weakly dispersible in non-polar solvents and the resulting dispersion was found to exhibit very weak blue emission lacking the emission in green and red regions, which could be due to a small fraction of Cdots devoid of polar functional groups giving rise to the intrinsic blue emission.

Quantum Yield Calculation

We have calculated Quantum yield with respect to quinine sulphate (QS) in 0.1 M H_2SO_4, using the following formula.[4]

$$Q_S = Q_R \times \frac{I_S}{I_R} \times \frac{A_R}{A_S} \times \frac{\eta_S^2}{\eta_R^2} \quad [S1]$$

Where,

Q_S = quantum yield of sample; Q_R =quantum yield of reference; I_S=area under the emission curve of sample; I_R= area under the emission curve of reference; A_R = absorbance of reference; A_S = absorbance of sample, η_S = refractive index of sample; η_S = refractive index of reference.

Quantum yield of Q_R =0 .54; Refractive index of water = 1.33.

(The concentration of all samples and the reference quinine sulphate were adjusted so that the optical densities of all samples were 0. 10 ± 0.03 at the excitation wavelength (365 nm)).

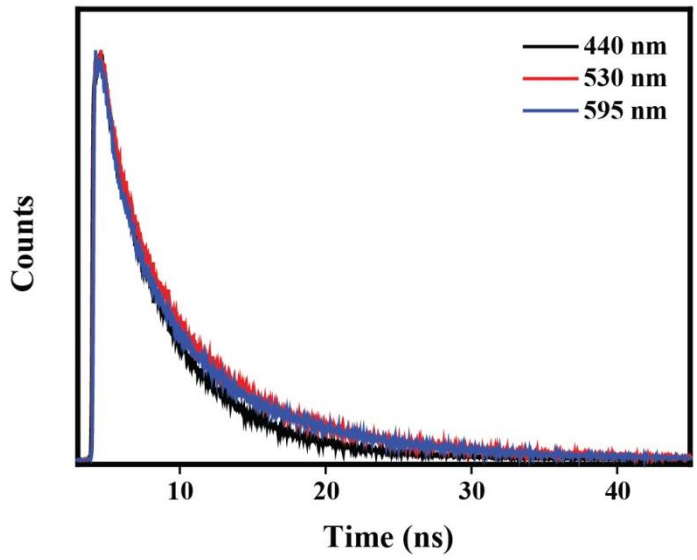

Fig. S10 Time-resolved photoluminescence spectra (λ_{ex}=375 nm) of Cdots at different emission wavelengths (λ_{em}) as mentioned in the legends.

The decay profiles observed for Cdots can be fitted with multi-exponential functions as shown in equation (S2), where a_i and τ_i are the amplitude and decay time of the i^{th} components, respectively

$$\langle \tau \rangle = \sum_i a_i \tau_i^2 / \sum_i a_i \tau_i \qquad [S2]$$

Table. S2 Decay parameters as obtained from the time-resolved photoluminescence spectra of Cdots (λ_{ex}=375 nm).

λ_{em}	α_i (%)	τ_i(ns)	<τ> (ns)	χ^2
440	38.81 61.19	3.057±0.005 6.35±0.001	5.58	1.007
535	4.37 53.20 42.43	0.848±0.077 4.648±0.003 11.53±0.001	9.12	1.015
595	5.00 36.01 58.99	0.722±0.072 3.84±0.006 9.60±0.0079	8.43	1.040

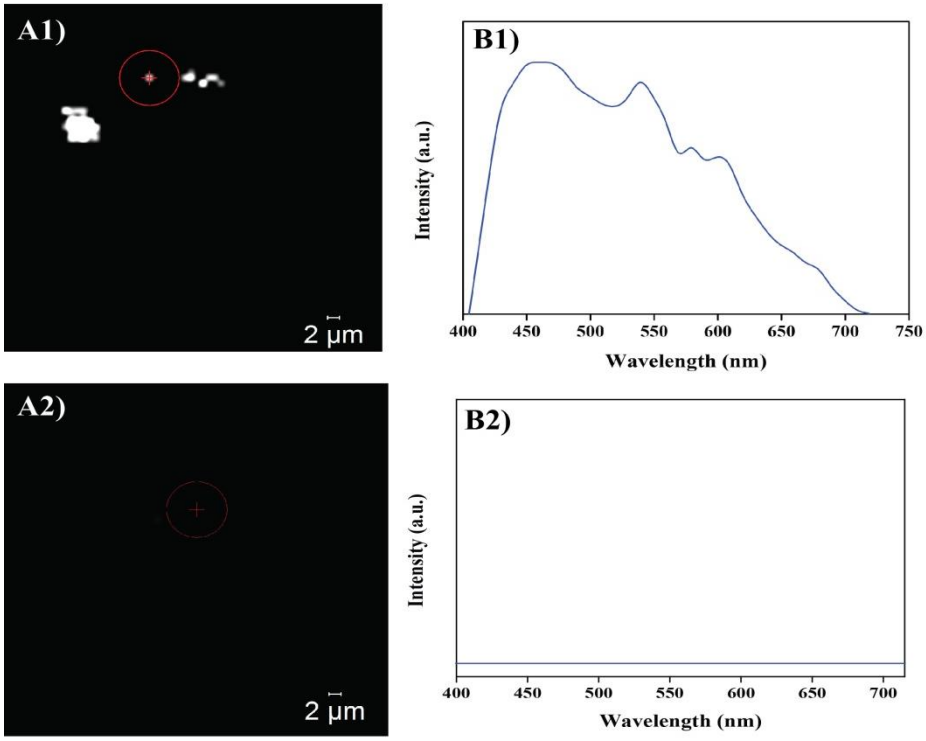

Fig. S11 A) Super resolution images of (A1) single Cdot particles in PVA matrix and (A2) only PVA matrix and their corresponding emission spectra of the assigned single particle B1 and B2 respectively (λex = 355 nm).

The single localised Cdot particles showed broad emission spectrum while the emission from PVA only matrix under the same conditions showed no emission further confirming the broad emission as obtained due to Cdot particles.

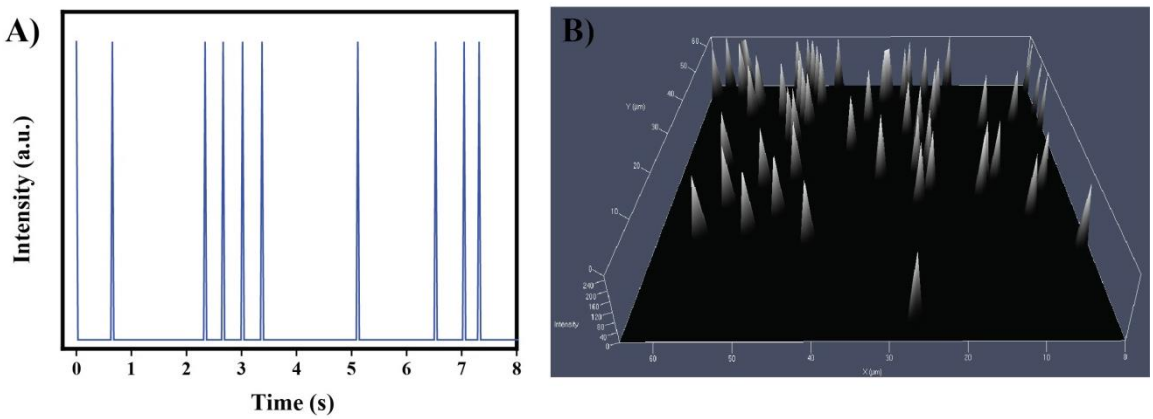

Fig. S12 A) Blinking profile and B) Point spread function of single Cdots highlighting their specific localization in PVA matrix recorded at 355 nm excitation wavelength.

The photon distribution was fitted with a point spread function (PSF) with the purpose of localizing single Cdot particles. As the density of the sample was low during these CLSM measurements each single emitter in the image is represented as an isolated PSF.

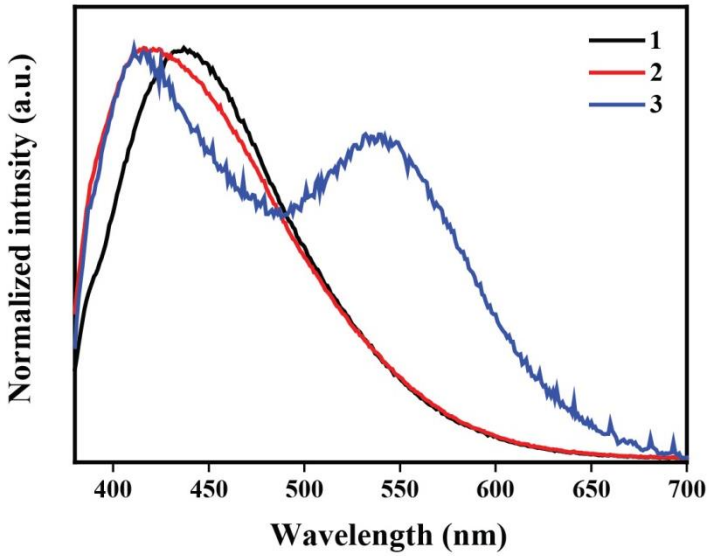

Fig. S13 Emission spectra of Cdots prepared from bare glycerol (1), glycerol and phosphoric acid (2) and glycerol and p-phenylenediamine (3) recorded at 340 nm excitation wavelength.

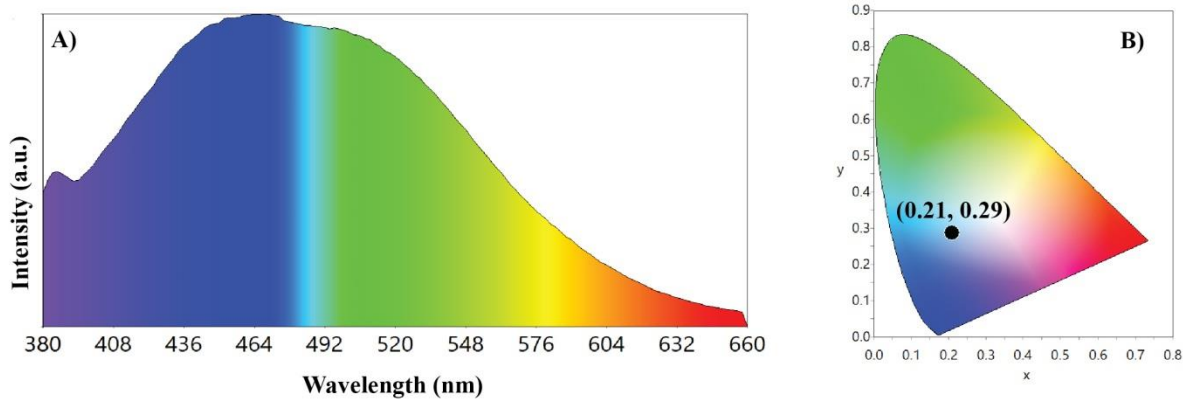

Fig. S14 A) Photoluminescent spectrum of Cdots synthesized using ethylene glycol in the presence of p-phenylenediamine and phosphoric acid and B) the corresponding CIE chromaticity diagrams of Cdots. The solvent used was water with λ_{ex} =340 nm.

The synthetic procedure involves microwave irradiation of the precursors which results in high temperature carbonization based on dipole moment of the reactants. The absence of RGB emission may be attributed to the lower dipole moment of ethylene glycol as compared to glycerol which resulted in lower carbonization temperature leading to absence of additional surface states and/or different chromophores responsible for the red emission.

Reference:

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4 M. P. Sk and A. Chattopadhyay, *RSC Adv.*, 2014, **4**, 31994–31999.