Deep eutectic solvents (DESs) are transparent liquids composed of two or three components, which when mixed in an appropriate ratio form a eutectic with a melting point much lower than that of individual components, and such behavior has been ascribed to the generation of intermolecular hydrogen bonds (H-bonds). 1-3 A large number of HBAs have been exploited in combination with HBDs to have DESs with desired properties, however, most commonly used HBA is choline chloride. 4 Easy preparation of DESs with high purity, low cost, their stability towards water, easy biodegradability, and pharmaceutically acceptable toxicity 4-11 have made them versatile solvents for several applications, be it catalysis, extraction, biomass dissolution as well as processing, enhanced oil recovery, enzyme activity, extraction processes or drug delivery and materials synthesis.12-16

Apart from several advantages, 4-11 DES have some common properties with ionic liquids, such as a high thermal and chemical stability and supports self-assembly of amphiphilic molecules, therefore the investigation of colloidal systems involving DES has recently experienced a major upsurge.17-23 In a further development, an IL in DES colloidal formulation was constructed by Tan et al wherein aggregation of 1-alkyl-3-methylimidazolium chloride with different alkyl chain lengths in a deep eutectic solvent (DES, composed of choline chloride and glycerol) for the first time.24 Herein, we have constructed an even greener colloidal formulation comprising of a bio-based IL in DES, demonstrated its practical utility for in situ generation and stabilization of photoluminescent CDs and their application as bio-labeling and bio-imaging agents. Compared to luminescent semiconductor quantum dots, which have known toxicity,25 photoluminescent carbon dots have attracted growing interest in biological labeling, bioimaging, drug delivery and optoelectronic device applications due to biocompatibility and low toxicity.26-39 In recent years, exciting work has been done on synthesis of these benign materials through various routes, references to these are nicely compiled in a recent article by Xiaohui et al.37 Many of these routes have severe limitations in terms of tedious synthesis process, harsh reaction conditions, requirement of surface passivation reagents, expensive starting materials and low quantum yields, and developing an efficient method still remains a challenge.40-42 In this regard, continuous efforts are being made by several researchers to tune the intrinsic properties of CDs via chemical doping with heteroatoms in order to improve the quantum efficiency.40-46

DESs have the ability to donate and accept protons and electrons, which confers DESs the ability to interact with biomass components via establishing H-bonding network for enhanced dissolution. In fact, DESs have been reported

**DES-N-Doped Oxygenated Carbon Dots Colloidal Solutions for Light Harvesting and Bio-imaging Applications**

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as good solvents and extraction agents for the extraction/dissolution of biomass/biopolymers,\textsuperscript{47,48} as well as self-assembly media for amphiphilic molecules.\textsuperscript{17,22} The ability of DESs to dissolve biomass and support self-assembly media prompted us to create a colloidal system for establishing a new and sustainable methodology for preparation of carbon dots (CDs). Herein we have developed a new simple and greener route to prepare N-doped CDs-choline oleate ([Cho][Ola]) in choline chloride-ethylene glycol hybrid system by direct dissolution of “gelatin” a model protein as single precursor (a rich source of carbon and nitrogen) in colloidal solution.

Initially, we synthesized choline oleate ([Cho][Ola]) by mixing equimolar amounts of choline bicarbonate and oleic acid in toluene with continuous stirring at 100°C for 24 h under reflux conditions. The purity of [Cho][Ola] the was ensured from \(^1\)H NMR and LC-MS techniques (Fig. S1). Colloidal solutions were then prepared by addition of [Cho][Ola] in choline chloride-ethylene glycol, (1:2 mole ratio). Molecular structure of components used in construction of colloidal solutions is shown in Fig. 1.

![Molecular structure of surface active ionic liquid (SAIL, [Cho][Ola]) and deep eutectic solvent (DES, choline chloride-ethylene glycol).](image)

**Fig. 1.** Molecular structure of surface active ionic liquid (SAIL, [Cho][Ola]) and deep eutectic solvent (DES, choline chloride-ethylene glycol).

Aggregation behavior of [Cho][Ola] in DES (choline chloride-ethylene glycol) was characterized using surface tension, DLS and 2D NOESY measurements (Fig. 2a-d).

[Cho][Ola]-DES system exhibited a minima in surface tension profiles (Fig. 1a), marking the saturation of air-solution interface, and onset of aggregation of [Cho][Ola] in bulk (critical aggregation number, cac). This phenomenal aggregation in DES is as good as aqueous system (Fig. S2) and is likely due to cholinium ions which creates the common ion as well as solvophobic effect wherein the oleate alkyl chain group spontaneously come closer (induce aggregation). Structural organization in aggregates was envisaged from 2D NMR NOESY correlation spectra (Fig. 2c, 2d). The NOEs originate due to interactions between protons. NOE cross peaks (3.8 ppm and 4.7 ppm) corresponds to interaction between surfactant choline cation (C2) with 3.44 ppm choline cation of DES indicating the dispersion of [Cho][Ola] in DES (full spectra in supporting information Fig. S3). Inference also indicate that the aggregation behavior arises from the hydrophobic effect of SAIL alkyl chains.

Dynamic light scattering measurements performed on solutions of [Cho][Ola] in DES established the presence of aggregates, and the average hydrodynamic radius \(R_h\) was found to be 42±20 nm. Large size of aggregates in DES is in accordance to the earlier observations of aggregation of amphiphilic molecules in ionic liquid medium.\textsuperscript{49} The large aggregate size is due to clustering of smaller micellar structures which are loosely arranged in DES medium. In ionic medium such as DES, ions exists as positively or negatively charged ion clusters, instead of separate single ions, and makes an electrical double layer surrounding the micellar surface thereby keeping them apart through electrostatic interactions.\textsuperscript{50}

![Dynamic light scattering measurement](image)

**Fig. 2** (a) Surface tension, (b) DLS plot of [Cho][Ola] and (c) 2D NMR NOESY spectra in choline chloride-ethylene glycol (1:2 mole ratio) deep eutectic solvent (DES).

[Cho][Ola]-DES colloidal solution was used to generate N-doped, carbon-rich, CDs by direct dissolution of gelatin (1 wt%) for 2-4 h at 100°C (Fig. 3). Here, [Cho][Ola] micelles acted as a template for the synthesis of CDs. Turning of

![Schematic representation for the preparation of colloidal luminescent CDs in [Cho][Ola]-DES micellar system.](image)

**Fig. 3** Schematic representation for the preparation of colloidal luminescent CDs in [Cho][Ola]-DES micellar system.
reaction mixture from pale yellow to brown colour indicated the formation of CDs. For further confirmation of CDs in DES based micelles, we examined the DES-CDs pure solid and colloidal solutions under FT-IR (Fig. 4a, FigS4a) and Raman Spectroscopy (Fig.4b, Fig.S4b). From the FT-IR spectra, broad and intense peak centred at 3351 cm$^{-1}$ is assigned to the –OH bending. The stretching frequencies at 2932 cm$^{-1}$, 2340 cm$^{-1}$ are attributed to -C-H and -C≡N group peaks respectively. The bands in the range of 1653 cm$^{-1}$, 1562 cm$^{-1}$, and 1457 cm$^{-1}$ are -C=O, -N=H, and -C=C stretching frequencies respectively. The peak at 1086 cm$^{-1}$ represents -O-H bending vibrations. This implies that the existence of large number of residual hydroxyl groups at the pure CDs and at interface of the DES-CDs composite material. Appearance of two bands in Raman spectra in both pure CDs and micellar stabilized CDs (Fig. 4b, Fig. S4b) i.e. at 1352 cm$^{-1}$ (D band, due to breathing modes of sp$^2$ atoms only in the rings) and 1602 cm$^{-1}$ (G band, due to bond stretching of all pairs of sp$^2$ atoms in both rings and chains) also indicate stacked nanocrystalline graphene/graphene oxide domains in the colloidal system. From this FT-IR and FT-Raman results we strongly confirm that the CDs are formed and stable in micellar form and exhibiting its fundamental properties.

High stability of CDs in colloidal solution is likely due to strong electrostatic interactions of carboxylate group of [Cho][Ola] with CDs at the micellar interface. Since, the hybrid system, i.e., CDs-in-micelles formed a very stable colloidal system, we devised a separation method wherein CDs could be extracted with ease. For that we added toluene in excess to the hybrid system. CDs migrated into the toluene slowly after vigorous shaking due to their hydrophobic nature. The extracted CDs were then characterized through HR-TEM and PXRD which conformed the formation of highly crystalline ultra-small (1 to 3 nm) CDs (Fig. 5a). From the HR-TEM (Fig. 5b,c), the d-spacing value, 0.216 nm corresponds (002) phase, which indicates the graphite lattice of the carbon dots. Formation of graphitic crystallinity inside the CDs structure has been observed. Generation of sp$^2$ bonded C–C domains, organized into a 2D honeycomb lattice inside the carbon core, which was revealed by the TEM broad and multiple d-spacing values. The lattice phase of the CDs was also confirmed by XRD diffraction (20) at 22.69$^\circ$ which corresponds to (002) phase of graphite lattice (Fig. 5d). N doped CDs when dispersed in toluene showed intense blue light emission under UV-Visible light (365 nm) indicating passivation of surface with oleate ions (Fig. S5). The presence of interaction of [Cho][Ola] carboxyl groups with CDs in DES offers various surface modifications for potential applications in sustainable energy conversion materials and in bioimaging purpose. Therefore, N doped CDs-[Cho][Ola]-DES colloidal solutions were further examined for the optical properties. UV-Vis spectroscopy and emission spectra strongly corroborated the formation of CDs. Broad absorbance band (300 to 415 nm) and slight broad adsorption peak from ~400 to 580 nm originates from both n-π* and π-π* charge-transfer transitions and indicated the N-doped oxygen-functionalized graphitic CDs (Fig. 6a).

Using the maximum absorbance ($A_{max}$) value, we recorded the emission spectra at different excitation values such as...
spectra at 300, 360, 380, 390, 400, 430, 450, 460, 480 and 500 nm and which showed emission of the light (bluish to red) at 416, 456, 458, 462, 471, 514, 527, 544 and 556 nm (Fig. 6b and Fig. S6) indicating formation and stabilization of CDs in DES-based micelles wherein different emissions originated from different luminescence centres.\(^{51}\) Blue emission normally originate from the surface defects and the green emission arises from molecular states with distinct energy levels. The CIE colour coordinates \([(0.22, 0.21), (0.23, 0.28), (0.24, 0.29), (0.24, 0.31), (0.25, 0.33), (0.31, 0.46), (0.35, 0.52), (0.37, 0.54), (0.41, 0.56)\) and \((0.45, 0.54)\)] calculated from the different emission spectra indicated the blue to red emission from the CDs-DES hybrid colloidal system at different excitation values (Fig. 6c).

Time-resolved photoluminescence (TrPL) was employed to understand the excited-state lifetime of colloidal CDs emitting blue light by monitoring the decay profiles at 456 nm (Fig. 6d, IRF spectra provided as Fig. S7). The emission of blue light under ultra violet rays is likely due to surface-related emission, and attributed to the presence of surface energy traps that become emissive upon stabilization as a result of the surface passivation in micelles.\(^{52}\) The decay dynamics of the ensembles of colloidal CDs usually exhibit multiexponential dynamics and the kinetics follows the equation, \(A + B_1e^{-\tau_1} + B_2e^{-\tau_2} + B_3e^{-\tau_3}\). This is a tri-exponential decay function, where \(\tau_1\), \(\tau_2\) and \(\tau_3\) represent the shorter lifetime and longer lifetime, respectively, and \(B_1\), \(B_2\) and \(B_3\) are the amplitude of the components at \(t = 0\). The shorter lifetime is the intrinsic recombination and the longer time is due to radiative recombination of electrons and holes. The fitting data of micelles-CDs hybrid system shows \(\tau_1\), \(\tau_2\) and \(\tau_3\) equals to 3.07, 7.34 and 18.02 ns respectively. Therefore, the major surface-related emission is due to the radiative recombination of charge carriers involving surface states.

Quantum efficiency (QE) of the surface-state emission of micelles-CDs hybrid system was determined by relative method with the area under its curve with the area under the band-edge peak emission (supporting information). The obtained QE is remarkably very high ca. 82% and is much larger than previously reported for light emitting CDs or doped CDs (Fig. 7 and Table S1).

![Fig. 7 Comparison of quantum yield of CDs synthesized by different research groups with present work. (ref. = reference).](image)

Very high QE is likely due passivation of quenching defects on the surface and suppression of non-radiative surface trap sites existing on the surface of the CDs by the oleate ions in a manner similar to that accounted for a dramatic increase in fluorescence QE of CdSe nanocrystals (8% to 45%).\(^{53}\) Confinement of CDs with large number of DES ions in micelles which creates the polarization effects at the interface between CDs and surrounding ions is also contributing towards increase of QE. The previous reports on antimicrobial activity of DES(choline chloride-ethylene glycol) and SAIL, [Cho][Ola] checked against gram positive and gram negative bacteria and EC50 values comes under practically harmless.\(^{54,55}\) CDs synthesized from biomass show nontoxic effects against bacteria and fungus in antimicrobial study.\(^{56}\) In this study four different bacteria, *Escherichia coli*, *Vibrio owensii*, *Bacillus cereus*, and *Vibrio alginolyticus* of OD\(_{600}\) 0.6 were grown in Luria broth.

![Fig. 8 Fluorescence microscopic images of different types of bacteria under normal light and through green filter (a) Escherichia coli, (b) Vibrio owensii, (c) Bacillus cereus, and (d) Vibrio alginolyticus of OD\(_{600}\) 0.6, labelled with the N-doped carbon dots.](image)
supplemented with colloidal-CDs (4:1) for 16 h at 30°C with 180 rpm. Bacterial cells were pelleted by centrifugation (5000 rpm for 4 min at 4°C), washed in with phosphate-buffered saline (PBS; 5000 rpm, 4 min) and suspended in PBS. Bacterial cells were observed under an epifluorescence microscope at a wavelength $\lambda_{max}$ 575 nm and normal light. Commonly, colloidal CDs interact with bacteria through electrostatic interaction, and some colloidal CDs may lead to ROS generation in bacterial cells, whereas other cannot. ROS inhibition activity was studied with all four bacterial strains treated with different dilutions of DES-N-CDs containing colloidal solution. No or negligible ROS inhibitory activity was observed up to $\frac{1}{4}$ dilution, whereas about 50-60% ROS inhibition was estimated with higher concentrations of colloidal CDs. Therefore, it may be concluded that low concentration of CDs does not generate ROS in bacterial cells, while higher dose leads to ROS generation. A very good fluorescence indicated that the CDs were easily internalized in bacterial cells (Fig. 8 and Fig. S8, S9, S10 and S11). The results also confirmed that due to greener nature of all the components, the colloidal solutions of N-doped CDs-choline oleate ([Cho][Ola]) in choline chloride-ethylene glycol can be directly applied in growth media up to very high concentrations, and CDs have the capability to retain with bacterial cells.

In conclusion, the ability of deep eutectic solvents to dissolve biopolymers and to support self-assembly of amphiphilic molecules has been exploited to develop a novel green colloidal formulation (choline oleate ([Cho][Ola]) in choline chloride-ethylene glycol system) wherein N-doped carbon dots have been generated and stabilized in micelles by direct dissolution of biopolymer gelatin. The colloidal CDs exhibited varying light emission (bluish to red) depending upon excitation wavelength with a very high quantum yield (ca.82%). The CDs also showed a very good biocompatibility with different bacteria for cell labelling. Such a system holds promise as potential material for light harvesting and bioimaging applications suitable for single molecule resolution due to very small size (1 to 3 nm) of CDs. In future, there is a brighter prospect of developing more such tailor-made colloidal CDs as an inexpensive, stable, and biocompatible marker with tuneable light emission and quantum efficiency by choice of bio-friendly ionic liquids or deep eutectic solvents, amphiphilic molecules and biopolymers.

Materials and Methods:

Materials:

Oleic acid (>99%) was obtained from TCI India, Choline chloride (≥ 99%), Choline bicarbonate (≥ 80%) and Gelatin (>98%) type A (300 Bloom, IEP=9.0) were purchased from Sigma-Aldrich. Ethylene glycol, Hexane, Ethyl acetate and Toluene were purchased from SRL India and SD fine chemicals Ltd. India respectively. Milli-Q water was used wherever required. All the chemicals were of AR grade and were used as received.

Methods:

Synthesis of SAIL: Equimolar amounts of choline bicarbonate and respective oleic acid is dissolved in toluene and stirred at 100°C for 24h under reflux conditions and the progress of the reaction was monitored by TLC. The product was obtained by concentrating the reaction mixture (removal of toluene). After washing with ethyl acetate and hexane the product was dried under vacuum for several hours to remove the moisture present and was stored in desiccator. The purity of the [Cho][Ola] SAIL was ensured from 1H NMR, LC-MS techniques (Fig. S1).

FTIR, FT-Raman and FT-NMR measurements: FT-IR measurements were carried out at 25°C using a Perkin Elmer FT-IR spectrometer using BaF$_2$ windows and a Teflon spacer. The optical path length was 0.02 mm. For each spectrum, at least 20 scans were made with a resolution of 0.5 cm$^{-1}$. The Raman shift were characterized using LabRAM HR Evolutions (Horiba Jobin Yvon) Raman Spectroscopy.

NMR Measurements: $^1$H & 2D NOESY NMR experiments were performed on JEOL 600 MHz NMR spectrometer.

Surface tension: The surface tension measurements were performed in DES and water systems using SAIL to measure the surface tension using an Attention Sigma 700 Biloin scientific instrument with automated titrants employing the Du Noüy ring method. The solutions (SAIL in water/DES) were prepared 10 times higher concentration than that of $cmc$ and left for at least 15 min. Then, the solutions were titrated against the pure desired solvent which was taken in standard vessel having diameter 70 mm. The data was noted with an accuracy within ±0.1 mN m$^{-1}$.

Dynamic Light Scattering (DLS): DLS measurements of solutions with known viscosity and refractive indices were carried out in a quartz cuvette of 1 cm path length, at 298.15 K on Spectro Size™ 300 (NaBiTec, Germany) light scattering apparatus with a He–Ne laser at a wavelength of 660–670 nm, and power 4 mW) as a source at an angle of 90°. The data evaluation of the DLS measurements was performed with the inbuilt CONTIN algorithm. The temperature of the measurements was controlled with an inbuilt thermoelectric peltier device with an accuracy of ±0.1 K.

Ultra Violet-Visible Spectroscopy: The UV-Vis spectra of synthesized materials in micelles have been measured using a UV 3600 Shimadzu UV-vis-NIR spectrophotometer at 298.15 K. In a typical experiment, the colloidal solutions were taken in quartz cuvette of 1 cm path length with different concentrations.

Fluorescence Spectroscopy: Fluorescence measurements were performed using Edinburgh Instruments Xe900 (µF 920H) spectrophotofluorometer using a quartz cuvette of path length 1 cm. The intrinsic fluorescence of Cs, in colloidal formulations in DES was measured at their respective excitation/emission wavelength. The maximal values of fluorescence are the average of three measurements.

Time Resolved Photoluminescence spectroscopy (TrPL): Fluorescence decay profiles were measured using a time–correlated single-photon counting spectrometer i.e., Edinburgh instruments OB 920 fluorescence spectrophotometer using a pulsed diode laser (Laser - EPLED-375 nm; emission: 456 nm) as an excitation source.
Fluorescence lifetimes were measured by plotting fluorescence decay curves as a function of time (t).

Transmission electron Microscopy (TEM): The images were taken using a JEOL JEM-2100 electron microscope at a working voltage of 80 kV. Samples were prepared by putting a drop of sample solution on the carbon / lace-coated copper grid (300 mesh) prior to analysis it was dried for 24 hours in a vacuum desiccator.

Powder X-ray diffraction (PXRD): X-ray measurements were performed using a XRD, Philips X'pert MPD system with CuKα radiation (λ = 1.54056 Å).

Bio-imaging studies: Four different bacteria, Escherichia coli, Bacillus cereus, Vibrio owensii, and Vibrio alginolyticus of OD600 0.6 were grown in Luria broth supplemented with ILS CDs containing colloidal solutions (4:1) for 16 h at 30ºC with 180 rpm. Next day, bacterial cells were pelleted by centrifugation (5000 rpm for 4 min at 4 ºC), washed in with phosphate-buffered saline (PBS; 5000 rpm, 4 min) and suspended in PBS. Bacterial cells were observed under an epifluorescence microscope (Axio Imager, Carl Zeiss AG, Germany) and documented.

ASSOCIATED CONTENT
Supporting Information. Materials characterization, Surface tension plots, Calculations of QE, Comparative QE and fluorescence images are provided.

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DES based colloidal solution → CDs, DES → [Cho][Ola]

Gelatin → 2-4h, 100°C

CDs stabilized colloidal solution

Light harvesting

Bio-Imaging