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Comprehensive Review on Hydrophobic Carbon Dots and Their Multifunctional Applications

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Hydrophobic carbon dots have been considered a novel class of functional nanomaterials that integrate surface hydrophobicity with remarkable optical tunability and chemical stability. These nanostructures exhibit excellent physicochemical, photophysical and biocompatible properties, making them promising candidates for optical electronics, anticounterfeiting dual encryption, latent fingerprint detection, bioimaging, sensing and oil water separation applications, *etc*. This review critically consolidates various preparation methods, fundamentals of hydrophobicity, brief insight into fluorescence mechanism into hydrophobic carbon dots. Overall, the review provides a comprehensive overview of the current status of hydrophobic carbon dots, highlighting the challenges and future opportunities for translating their unique hydrophobicity character into practical performance and technologies.

1. Introduction

Nature has engineered many hydrophobic species to execute a special function, for instance, the lotus leaves exhibit remarkable hydrophobic properties, where their leaves repel water and prevent dust or microbial growth, ensuring self-cleaning and long-lasting vitality. In fact, these natural hydrophobic properties devise from micro/nanostructures present in natural species and makes them useful for sustainable hydrophobic coatings. This natural strategy of water repellence has inspired scientists to design artificial materials with similar properties, where hydrophobicity provides resistance to moisture, stability in harsh environments, and protection against degradation.[1,2] Among them, carbon dots (CDs) have emerged as excellent hydrophobic nanomaterials, combining fluorescence with water-repelling characteristics, making them ideal for advanced applications.[3,4] The hydrophobic nature not only improves stability and durability in solid-state devices but also enhances performance in optoelectronics, coatings, and biomedical fields. Thus, the journey of materials development from natural plants to engineered carbon dots, hydrophobicity proves to be a powerful and multifunctional properties, well connecting the important domains such as sustainability, innovation, and future technology.

The history of these promising materials can be traced back to the early 2000s, with an accidental discovery made in 2004 by Xu *et al.* during the electrophoretic purification of single-walled fluorescent material, which they identified as a mixture of fluorescent nanoparticles derived from the nanotubes.[5] Building upon this initial discovery, in 2006, Sun *et al.* reported a method to produce such material and termed "carbon dots".[6] These nanoparticles are celebrated for being both environmentally friendly and exceptionally versatile. Indeed, it was an important discovery having a winning combination of characteristics; they are tiny, measuring less than 10 nm, yet remarkably stable and safe for living cells. The CDs have successfully drawn the significant scientific attention because their emission wavelengths are easily tunable, they possess high photoluminescence brightness, and they can be synthesised through simple techniques using a wide variety of readily available starting materials. This unique set of advantages makes them promising approach for numerous applications, often outshining other materials in the carbon family. The carbon core in this family plays a major role, decorated with a wide variety of chemical attachments, such as amine, carboxyl, or hydroxyl groups. Scientists can even swap out some carbon atoms with other elements like nitrogen or sulphur, to further tailor their properties. This ability to modify their surface is incredibly powerful because it allows us to control how the carbon dots interact with their environment.[7–9] In fact, it cannot be considered only as new-generation smart materials, but also has emerged as a replacement for multi-step, tedious and over-expensive synthetic organic materials, particularly in applications such as optoelectronics, textiles, bioimaging, oil-water separation, and anticounterfeiting ink *etc*.

The history of hydrophobicity has been evolved over more than two centuries through fundamental theoretical and experimental milestones. The earliest scientific foundation dates back to 1805, when Thomas Young introduced the concept of contact in his work ‘Essay on the Cohesion of fluids’, providing the first physical explanation of wetting and non-wetting behaviour at the solid-state interface, which later became the basis of hydrophobicity.[10] Furthermore in 1936,

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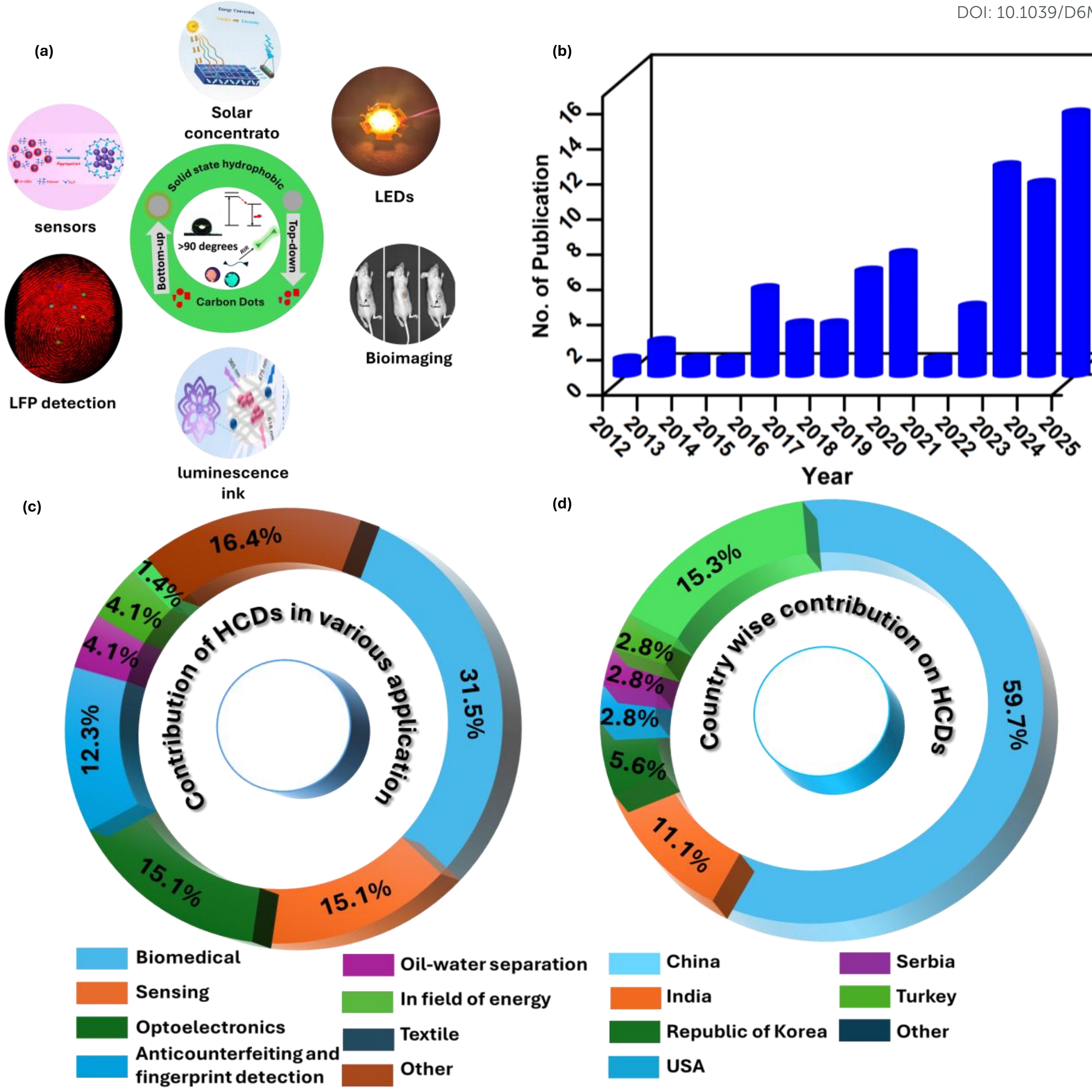

Fig. 1 (a) The overview of hydrophobic carbon dots (HCDs) application (reproduced with permission from ref. 11 © The American Chemical Society 2023, 12 © The American Chemical Society 2023, 13 © The American Chemical Society 2020 , 14 © The Elsevier 2024, respectively).[11–14] (b-d) The general progress trend of publications on HCDs (data acquired from Scifinder)

Wenzel demonstrated that surface roughness significantly amplifies intrinsic wettability, showing how a rough hydrophobic surface can enhance water repellency.[15] In 1959, Walter Kauzmann coined the term ‘hydrophobic bond’ to explain nonpolar interactions in aqueous systems.[16] Finally, Barthlott and Ehler reported the self-cleaning properties of plants' leaf surface with hierarchical micro-nano structures due to the superhydrophobic behaviour of plant leaves.[17]

The carbon dots also show hydrophilic, hydrophobic and superhydrophobic behaviour. Based on the contact angle, if the surface of CDs with water is greater than 90 degrees, considered as hydrophobic CDs, while a contact angle of more than 150 degrees is accepted as superhydrophobic CDs. However, there is no sharp boundary between hydrophobic and superhydrophobic behaviour. The proportionate contribution by different countries in development of HCDs have been charted in figure 1, where China and India are the leading countries, accounting for more than 60% contribution of the total reported publication on HCDs. Correspondingly HCDs have shown potential across various application domains such as biomedical, sensing, anticounterfeiting, fingerprint detection and optoelectronics (Fig. 1).

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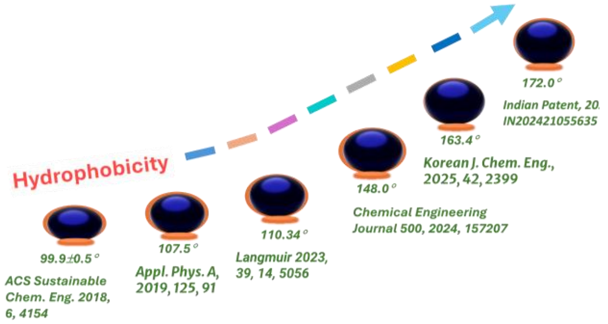

Fig. 2 A general trend of progress in HCDs with contact angle demonstration from hydrophobicity to super hydrophobicity.[11,18–22]

Despite substantial advances, a dedicated review covering all possible aspects of HCDs has not been reported to date. This review aims to provide a thorough look into the world of these versatile hydrophobic carbon dots. We explored the methods used to synthesise them, the reason behind the hydrophobicity in CDs, the fluorescent mechanism of HCDs, and showcase their applications in various fields. By bringing these insights together, the current review will serve as a valuable guide for anyone looking to harness the unique capabilities of HCDs. Moving forward, the key to unlocking their full potential will lie in developing synthesis methods, perfecting their surface design, and application is in focus because a single prepared HCD may show potential in different fields, as discussed in the application section.

2. Classification of CDs

Carbon dots exist in two states either dispersive state or solid state. Based on interaction with water, CDs are further classified into three categories: hydrophilic, hydrophobic, and amphiphilic.[4]

2.1 Hydrophilic Carbon Dots

Hydrophilic carbon dots are well dispersed in aqueous medium, because CDs have abundant polar or charged groups (e.g., -OH, -COOH, -NH_2, etc.) towards outer surface which facilitate strong binding interactions with water molecules (hydrogen bonding, ion-dipole interaction, etc). These hydrophilic moieties make the CDs remain colloidally stable in water, good biocompatibility, and can easily adapt with biological environments. For instance, C. Dai et. al., synthesised high-hydrophilic carbon dots from petroleum coke, which showed enhanced oil recovery applications using the concept of water affinity.[23] In general, hydrophilic CDs are often used in bioimaging, sensing, aqueous phase catalysis, and biomedical applications because of their favourable solubility and stability in water.[23–27]

2.2 Hydrophobic Carbon Dots

Hydrophobic Carbon Dots (HCDs) are engineered to be dispersible in nonpolar or organic solvents and to have minimal affinity with water. HCDs' surfaces are decorated with nonpolar moieties (alkyl chains, aromatic rings, *etc.*) or reduced quantities of polar groups to minimize the interaction with water. Such rational design strategy makes CDs insoluble in water but stable in organic media or in dry states.[4,24,28,29] Scientists also utilized hydrophobicity concept to achieve aggregation due to concept that molecules present in CDs organize itself in hydrophobic and hydrophilic arrays to minimize the contact from water, leading to aggregated nano/micro structures of CDs. In addition, water driven aggregation of CDs leads to quenching/enhancement of fluorescence. All aspects of HCDs are explored like preparation method, mechanism, and application, and discussed in further sections.

2.3 Amphiphilic Carbon Dots

The combined nature of hydrophilic and hydrophobic carbon dots is also reported by researchers as amphiphilic carbon dots. But there is no clear boundary in reference of contact angle, however they typically exhibit intermediate or context-dependent wetting behaviour, with contact angles generally in the range of 40-80° depending on solvent polarity, surface morphology, roughness and measuring conditions.[30] They can interact with both water and organic media, and often form stable dispersions in both aqueous and organic solvents. This dual affinity is achieved by engineering the surface of CDs by attaching both polar and nonpolar groups. Amphiphilic CDs can act as interfacial or surfactant-like agents, residing at water/organic or water/air interfaces, and are especially useful for applications like drug delivery, phase transfer, and sensing in mixed solvents. For instance, B. Zheng et al. synthesised amphiphilic CDs that dispersed in both aqueous and organic solutions and used them as fluorescence probes for iodine species in saline water.[31] Amphiphilic carbon dots have recently been used for enhanced oil recovery (EOR).[32–36] However, this customisation brings its own set of challenges, particularly, for the water-loving versions of amphiphilic CDs. Another, a significant problem arises when structure. When these dots are packed closely together, they have a natural tendency to aggregate and stack on top of each other, often cause the quenching of fluorescence, which limits the use of CDs.[37–39] Overcoming this issue is a major hurdle for researchers aiming to unlock the full potential of carbon dots in practical solid-state devices.

To tackle the problem of fluorescence quenching, scientists have devised a clever solution, making the carbon dots solid-state, water-repelling by attaching specific organic chains to their surface, these dots can be easily dispersed in organic solvents, which makes them highly compatible with the materials used in organic electronic devices. This shift has sparked a wave of research into hydrophobic carbon dots, focusing on how to best create and tailor their properties for a multitude of technological applications. One of the most promising aspects of HCDs is their safety profile.

Researchers are trying to use them in solid-state applications, such as electronic screens, advanced sensors, as well as in characterization of their Studies have consistently shown that they are generally non-toxic to mammalian cells, paving the way for exciting biomedical applications.[40] They can be seamlessly blended into various polymer structures, like those used in food packaging, medical implants, and sophisticated drug delivery systems. Beyond medicine, HCDs are proving to be highly sensitive sensing materials.[41–44]

3. Synthesis of Hydrophobic Carbon Dots

Various preparation methods of carbon dots have been reported since 2004. All these synthesis methods are classified into two categories, top-down and bottom-up. The top-down approach, similar to sculpting, begins with a large piece of carbon and breaks it down into tiny nanoparticles. The bottom-up method is more like building with LEGOs where you start with small molecules and piece them together to create the dots.[45–47] When the goal is to create specifically hydrophobic carbon dots with solid-state fluorescence or dispersive state, the strategy shifts slightly with the same preparation methods. Researchers might start with water-repelling materials from the get-go, partially burn organic substances, or take already-made water-soluble carbon dots and give them a new, hydrophobic "coat" by modifying their surface with specific molecules. These varied approaches allow scientists to craft HCDs with the precise properties needed for different applications. The commonly explored techniques for HCDs are discussed in the sub-sections.

3.1 Hydrothermal/Solvothermal Approach

A popular and efficient method to synthesize the hydrophobic carbon dots is the solvothermal approach, wherein, solution of precursor materials is placed inside a sealed container under high pressure and temperature. The main difference between the two names comes down to the liquid used; if the broth is water-based, it's called a hydrothermal process, whereas when an organic solvent is used, the process is referred to as a solvothermal approach. By precisely controlling the cooking conditions, such as the temperature, pressure, and reaction time, scientists can effectively craft a wide variety of carbon dots with desired properties. This approach is favoured because it is simple, cost-effective, and offers fantastic control over the final product, allowing for the tuning of the dot's size, shape, and surface as per the desired properties. Another advantage of this technique, it is accepted as environmentally friendly.

In 2019, Solid-state hydrophobic carbon dots were synthesised straightforwardly and efficiently using a one-pot solvothermal method by H. Yang *et al*. This process involved heating the precursor, 2,2'-dithiodibenzoic acid (DTSA), and melamine in an organic solvent. These HCDs are converted into HCD aggregated powder upon dispersion in an appropriate solvent, as shown in figure 3.[48] Similarly, X. Zhao *et al.* further synthesized red, yellow, and green AIE HCDs by using the solvothermal method. DSTA was selected as the primary carbon source, where acetic acid not only served as the solvent for the reaction, but also helped in the carbonisation during the reaction. Melamine, urea, and sulfonamide were used as nitrogen and sulphur sources.[49] In this sequence, W. Gao *et al*. again developed red fluorescent solid-state hydrophobic carbon dots by using DTSA and urea in acetic acid by a one-step solvothermal method.[12] Very recently, metal-doped hydrophobic carbon dots were reported by Gong's group, and they successfully replaced acetic acid with ethanol, resulting in red fluorescence as shown in figure 3. This approach makes CDs more environmentally friendly by avoiding acidic media.[50] Furthermore, hydrophobic carbon dots explored with super hydrophobicity by M. Ali *et al.,* they made two different combinations of solid-state fluorescence HCDs using DTSA and melamine. To get super hydrophobicity in CDs, they increased the carbon chain length by using hexane. Hexane is a nonpolar solvent, and used as a worthy candidate to increase hydrophobicity.[21] Another unsealed hydrothermal method, solid phase hydrophobic carbon dots were prepared by D. Zhao *et al*. using citric acid and L-cys·HCl in 2 mL water upon heating at 70 °C for 12h.[51] Moreover, people are trying to prepare hydrophobic carbon dots with solid-state fluorescence using the solvothermal method, which is emerged as a very helpful approach to create HCDs along with aggregation-induced emission, as highlighted in table 1.

To study synthesis methods other than the solvothermal technique, various alternative methods have also been

Table 1 The preparation methods of HCDs along with precursors, photoluminescence, quantum yield, hydrophobic contact angle and applications in various fields.

S. N.	Precursors	Synthesis method	Extraction/ purification	Emissions	Application	Contact Angle	PL Yield	Ref.
1	Pluronic F-68	Microwave assisted	Solvent extraction	Green		122°	7%	52
2	2,2-dithiosalicylic acid, adenine/4-amino benzene thiol	Solvothermal	Filtration	Orange, Green	Orange and green LEDs	137.7°, 117.8°		53
3	Dithiosalicylic acid (DTSA), urea	Solvothermal	Filtration	Orange-Red	Anticounterfeiting ink	112.9°	7.4%	12
4	Soyabean oil, H_2O_2	Solvothermal	Dialysis	Blue		104.8°		28
5	Polyoxyethylene–Polyoxypropylene–	Stirrer Heating	Filtration	Blue	Antibacterial activity	99.9°, 98.9°		18

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	Polyoxyethylene (Pluronic 68)							
6	2,3-Diaminonaphthalene	Hydrothermal	Filtration	Orange	Sensing of water in THF	127.9˚	35.1%	54
7	Citric acid, urea, octylamine, N, N′-dicyclohexylcarbodiimide, N-hydroxysuccinimide	Solvothermal, Stirrer heating	Column Chromatography	Red	Cancer cell detection	106°	15%	55
8	Spirulina powder	Solvothermal	Column chromatography	Red, NIR	Water detection and live cell imaging	110⁰	9.8%	56
9	8-amino-2-naphthol (8N2OH)/ 2,3-dihydroxynaphthalene (DHN), KIO_3	Solvothermal	Vacuum filtration	Blue	In reverse-phase capillary liquid chromatography	131.28°, 121.89°	3.8%, 1.8%	57
10	1,3,6-trinitropyrene, acetonitrile, dicyanobenzene	Solvothermal	Filtration	Red	CO_2 reduction to CH_4	148°		20
11	Dried flower of Magnolia Denudata	Ultrasonication	Column chromatography	Bright NIR fluorescence	Fluorescent Imaging	106.0°	12.18%	58
12	f-Si powder	Hydrothermal	Dialysis	Blue	Pressure Sensing Hydrogel	91°		59
13	Methyl methacrylate, 2,2′-azobisisobutyronitrile, dimethyl diallyl ammonium chloride	Reflux Heating	Filtration	Blue	LEDs	100°–110°		19
14	melamine and 2,2-dithiobenzoic acid, oleic acid, octadecene	Hydrothermal	Filtration	Red	Coated fabric	122.3°,134°,161.6°, 163.4°		22
15	DTSA, OPD	Hydrothermal	Filtration	Blue, Red	CDs/PVA Hydrogel, Information Encryption	129.2°		60

developed, such as chemical oxidation, microwave synthesis, pyrolysis, and solid-state oven heating. *etc.* A brief discussion of these methods is stated below.

3.2 Microwave-assisted

Rapid synthesis of HCDs with high quantum yield is possible through this approach. In this method, electromagnetic waves are absorbed by precursor molecules, and the heat generated by these waves induces the rapid carbonisation and thereby producing the CDs. Researchers are attracted towards the method due to its high efficiency and simplicity. For example, A. Goswami *et al.* for the first time mentioned a facile synthesis of luminescent HCDs using a household microwave oven. Their group used poloxamer Pluronic F-68 and *o*-phosphoric acid at optimum power 450W for time 4 min to synthesize HCDs. It avoided multiple functionalization stages as well as costly raw materials.[52] Y. Shu *et al.* prepared HCDs with 58% yield by combining 1,3-dibutylimidazolium dicyanamide ionic liquid and ethyl alcohol via same method at 1300 W for 5 min. Cationic moieties having N- heteroaromatic rings or derivatives of ammonium group present in ionic liquids are prone to decomposition, thereby greatly enhancing the effectiveness of carbonization.[61] In one more study, P. Wang *et al*., synthesized a carbon dot using pheophytin and DMF by irradiating microwaves for 30 min under 150 ⁰C operated at 400 W.[62]

3.3 Pyrolysis treatment

Pyrolysis involves the thermal decomposition of organic precursors at high temperature for the preparation of CDs. It overcomes the problems present in other synthesis methods as in solvothermal/hydrothermal. Pyrolysis has no need of solvent, avoiding dangers of high pressure, economic viability, ease of operation, scalability and quick reaction time. Thus, it has become preferable choice for the researchers.[63,64] To prepare pyrolytic HCDs a lot of polymeric feedstocks have been explored as demonstrated in figure 3. For example, M. Bodik *et al*. synthesised hydrophobic CDs below a size of 5 nm by heating the mixture of polyoxyethylene-polyoxypropylene-polyoxyethylene triblock copolymer (Pluronic PF-68) and phosphoric acid at 250°C with stirring for 2 h.[65] E.P. Giannelis *et al*. synthesised HCDs *via* pyrolysis of lauryl gallate powder and investigated the nonlinear optical response of HCDs for the first time. In their experiment, no modifier or secondary agent was used, and the reactant molecules were directly placed in a porcelain crucible and subjected to heat treatment at 270 °C for

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2 h.[66] V. Guttena *et al*. prepared a hydrophobic CD powder having solid-state emission and applied it for latent fingerprint visualisation. N-doped carbon dot was synthesized using homogeneous mixture of *o*-phenylenediamine and pyridinedicrboxylic acid (mole ratio, 1:1) in a ceramic crucible in an oven at 180°C for 4 hours.[67] M.Z. Fahmi *et al.* reported carbon dots by pyrolyzing tartaric acid and L-tyrosine at 220°C for 30 min, as demonstrated in figure 4.[68] In subsequent years, P. Purkayastha *et al*. synthesised a photostable, non-cytotoxic HCDs *via* a solvent-free, one-step thermal treatment using biocompatible precursors, namely dodecylamine and sucrose, at 210 °C for 10 min, as shown in figure 3. These HCDs were employed as fluorescent lipid raft markers.[69]

3.4 Solid phase synthesis

Another approach to get rid of from organic solvents and strong acids/bases is solid-phase synthesis that make it more eco-friendly over other synthesis techniques. D. Zhao *et al.* synthesised HCDs utilising a small volume of water with L-cysteine, citric acid, and N, N′-dicyclohexylcarbodiimide (DCC) as main precursors. The prepared carbon dot demonstrated the outstanding fluorescence properties and utilized in white LEDs as an illumination source.[51] H. M. Xiong, *et al*. used Resorcinol, *o*-phenylenediamine, and *o*-aminophenol as a carbon source and zinc sulphate as a catalyst. The mixture was thermally treated at 200 °C for 3h to produce carbon dots with blue, green and yellow emission, respectively.[70]

3.5 Chemical oxidation approach

It is a prominent approach for the preparation of carbon dots at industrial scale, in which the organic raw materials are dissolved in a chemical oxidant solution to proceed a chemical reaction. However, some drawbacks of this method are non-uniformity in the size of carbon dots, harsh reaction conditions and low yield. H. T. Chang *et al.* used chemical oxidation method for the synthesis of HCDs by mixing the triolein with concentrated sulphuric acid, in turn, precipitation took place after liquid - liquid extraction and organic phase is neutralized with the help of $NaHCO_3$ solution.[71]

D. Xiao *et al*. reported the synthesis of HCDs using cetylpyridinium chloride monohydrate (CPC) and sodium hydroxide. In the typical synthesis procedure, CPC was combined with NaOH (0.0-360 mM) at room temperature for different reaction time (28-180 h). They demonstrated that higher yield of HCDs can be obtained by increasing reaction time and concentration of NaOH. In 2019, this group reported the same HCDs again and used it to modify the natural cotton fibre for efficient adsorption of radioactive iodine in water.[72] H. Hou *et al.* prepared carbon dots through mixing of acetaldehyde aqueous solution and sodium hydroxide at room temperature under intense agitation. After reacting for 1 h, the mixed solution was aged at 25 °C for 72 h; afterwards, the alkaline solution was neutralised with 1M HCl solution.[72]

The solvothermal method is considered the best among available techniques because it offers the most precise control over reaction parameters such as temperature, pressure, solvent environment, and reaction time, which directly govern the size, surface chemistry, and optical properties of HCDs. Unlike microwave or pyrolysis methods, which are fast but often compromise on uniformity and reproducibility, while the solvothermal process ensures high-quality, tunable, and stable fluorescence, especially enabling AIE in the solid-state HCDs.

Solid-state heating is simple but lacks the flexibility to finely tailor emission properties, making it unsuitable for advanced optoelectronic applications. Chemical oxidation uses strong oxidants that lead to high cost as well as pollute the environment. Thus, solvothermal synthesis balances simplicity, scalability, cost-effectiveness, and precise control, making it the most versatile and widely adopted approach.

4. Hydrophobicity in Carbon Dots

Traditionally, most CDs are hydrophilic in nature, and they show properties of luminescence in the liquid state. Unfortunately, the property of luminescence of these CDs can not be easily transferred to their solid-state counterpart, due to the severe quenching effect, named aggregation caused quenching (ACQ). With the time, luminescence properties of most aqueous or solvent-dispersed carbon dots get diminish, thus, to retain the fluorescence properties, an extra matrix of polymer needed. But usually, polymer substrate is not suitable for applications like oil-water separation, coating, electrode deposition in energy storage devices *etc*. because the polymer matrix may affect the properties and real structure of CDs. Another problem with these polymer chain or matrices, they restrict the concentration of doped CDs. Overall, the hydrophilic nature of CDs makes them vulnerable to water exposure and challenging to disperse in solid state matrices, thereby limiting their applications in various field, where water free conditions are strictly required. Therefore, new approach is ongoing for the CDs, is known as solid state emissive carbon dots.[73,74]

Nowadays, researchers' attention attracted towards the hydrophobic carbon dots because they have ability to exhibit strong fluorescence even in the solid state, unlike conventional carbon dots. The HCDs are useful for real practical application purposes, such as bioimaging, sensing,

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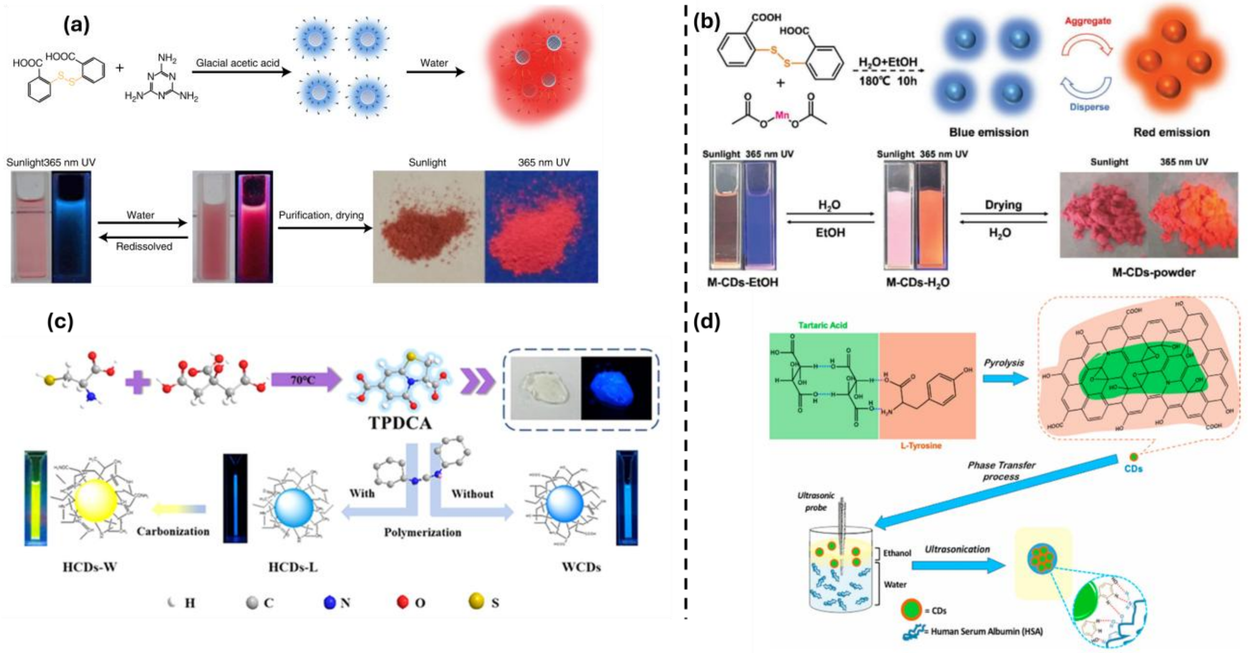

Fig. 3 Preparation route for HCDs through solvothermal (a) dual-mode luminescence solid-state HCDs. (reproduced with permission from ref. 48 © The Springer Nature 2019) .[48] (b) Synthesis of metal-doped dual-mode switchable fluorescent HCDs (reproduced with permission from ref. 50 © The Wiley 2024).[50] (c-d) pyrolysis synthesis methods explored for HCDs preparation with stable fluorescent properties (reproduced with permission from ref. 51 © The American Chemical Society 2020, ref. 68 © The Elsevier 2020 respectively).[51,68]

optoelectronics, anticounterfeiting, hydrophobic coatings for textiles, oil-water separation, and corrosion protection of metals *etc*. The origin of hydrophobicity in the CDs is of considerable interest and a subject of curiosity; thus, based on literature available and principles of chemistry, a critical overview of the factors responsible for the hydrophobicity of HCDs have been articulated in the subsequent section of the current review.

4.1 Surface chemistry by functional groups

In the study of surface chemistry, functional groups are a responsible factor for tailoring the surface properties because some functional groups form hydrogen bonds with water and reveal a hydrophilic nature, and some functional groups do not have any interaction with water, as a result, they show hydrophobic behaviour. Typically, the polar groups like hydroxyl (-OH), carboxyl (-COOH), and amino (-NH_2) on the surface of CDs readily form hydrogen bonds with water molecules, in turn, CD shows excellent dispersibility in aqueous and biocompatibility.

In contrast, the hydrophobic carbon dots show a slightly different architecture from the hydrophilic carbon dots. For example, Y. Wang *et al.* reported hydrophobic carbon dots using *o*-phenylene diamine (OPD) and DTSA, whereas FTIR suggested the presence of the NH_2 group in OPD and the OH group in DTSA precursor materials. Interestingly, characterization of synthesized CD revealed the absence of NH_2 and OH peaks, because of the reduction of hydrophilic moieties which makes carbon dots powder hydrophobic in nature.[75] Similarly, hydrophobic carbon dots reported by J. Hao *et al.*, using aliphatic chain as one terminal functional group, which undergo carbonisation and surface condensation during synthesis. As a result, the carbon core becomes encapsulated within nonpolar hydrocarbon layers composed of C-H-rich alkyl chains. The surface enrichment with long aliphatic moieties replaces or minimises the presence of polar functional groups (-OH, -COOH, -NH_2), which are otherwise responsible for hydrophilicity in conventional CDs. The comparative study of C_{12}-NH_2 and CDs showed disappearance of the NH_2 peaks in the IR spectra of CDs.[76] To date, many other AIE hydrophobic carbon dots have been proposed with the contact angle measurement, while some researchers have concluded the hydrophobicity of CDs based on their interactions with water molecules. Thus, a common conclusion from AIE HCDs can be drawn that there is less intensity of OH peaks in the IR study, but C-H and S-S stretching in bonds have been revealed in most of the reported results.[12,48,51]

Another surface study of hydrophobic carbon dots was reported by the J. Wang *et al.*, the hydrophobicity of the prepared CDs is directly linked to systematic surface chemical modification arising from the structure of the ionic liquid precursors. The authors demonstrated that by increasing the alkyl chain length in the precursor ionic liquids, the resulting CDs acquire surfaces rich in long hydrocarbon chains (C-H and C-C bonds), while the density of polar functional groups such as hydroxyl (-OH), carboxyl (-COOH), and nitrogen-containing groups decreases significantly. The FTIR spectra showed weakened O-H and C=O stretching vibrations, whereas strong C-H stretching bands confirmed the dominance of nonpolar

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moieties. Thus, hydrophobicity in these CDs arises from the replacement of polar oxygenated functionalities by extended alkyl chains, resulting in a nonpolar, C-H-rich surface with reduced surface energy and enhanced compatibility with organic environments.[77]

4.2 Effect of precursors and solvents

The nature of the precursor and solvents is also a significant factor for the introducing hydrophobicity in CDs. Many researchers have used DTSA to achieve hydrophobic carbon dots with AIE. For example, it is very surprising that a DTSA molecule, along with different precursors, namely melamine, urea, sulphonamide, phenylenediamine, *etc.*, have produced hydrophobic CDs. Therefore, the understanding role of DTSA in generation of hydrophobic carbon dots becomes important. The DTSA has aromatic benzene ring and sulphur-rich molecular structure, which strongly influences the surface properties, thus properties of DTSA have been discussed briefly in the following sub-sections.

4.2.1 Enrichment of aromatic structure

The benzene rings in DTSA promote the formation of extended π-conjugated carbon frameworks. These aromatic domains are intrinsically nonpolar, lowering surface polarity and suppressing hydrogen bonding with water molecules.

4.2.2 Surface passivation through sulphur

Cleavage of the disulfide bond introduces sulphur atoms into the carbon lattice at surface sites, leading to the formation of C-S-C and C=S linkages. These sulphur functionalities are less polar than oxygenated groups and act as hydrophobic surface terminations, decreasing the surface free energy.[78]

These properties of the DTSA molecule make it suitable to use in the preparation of hydrophobic carbon dots. For instance, H. Yang *et al*. reported hydrophobic red emissive solid state carbon dots in 2019 using DTSA, melamine and acetic acid under solvothermal conditions, to achieve nitrogen- and sulphur-doped carbon dots with disulfide- and pyridine-rich aromatic surfaces. During carbonisation, most hydrophilic groups (-OH, $-NH_2$) are eliminated, while nonpolar C–S, S–S, and C=N functionalities dominated, making the surface highly hydrophobic. Acetic acid plays a crucial role as both solvent and catalyst, promoting amidation and controlled carbonisation while limiting the formation of oxygenated polar groups. This results in carbon dots with low surface polarity with tendency to aggregate in water due to hydrophobic interactions, exhibiting red aggregation-induced emission.[48]

A similar combination was used by M. Ali *et al.*, to introduce the super hydrophobicity, but by using slightly different solvent conditions. The melamine and 2,2-dithiobenzoic acid, undergo condensation to form an sp^2-hybridized carbon core doped with N and S atoms. These heteroatoms facilitate crosslinking with the passivating agent (oleic acid), leading to the growth of hydrophobic polymer chains covalently anchored to the CQD surface. The mixture of hexane and octadecene is nonpolar and directs the oleic acid's long alkyl tails outward while the hydrophilic head towards carbon dots nuclei. This alignment ensures complete surface passivation, eliminating polar oxygenated groups (-OH, -COOH) and leaving a surface rich in C-H and C=C bonds. Compared to acetic acid-based synthesis and the mechanistic change from hydrophobic to superhydrophobicity, the nonpolar solvent system promotes dense alkyl chains grafting and nanoscale roughness, thereby reducing surface energy and trapping air pockets at the solid-liquid interface. The resulting contact angle of ~163° confirms superhydrophobicity. Thus, the transition from polar to nonpolar solvent directly modulates the extent of polymer chain encapsulation and surface functional group suppression, which together are responsible for the observed superhydrophobic behaviour.[22]

4.3 Aggregation behaviour

Beyond the surface chemistry and solvent-precursor effect, the aggregation behaviour is also responsible for hydrophobicity in addition to aggregation-induced emission (AIE, *vide supra*). Aggregation effect is not only responsible for AIE but also the ACQ effect. If HCDs glow under high-energy electromagnetic radiation, it is considered an AIE effect; if there is no fluorescence, it is a simple ACQ effect. In the ACQ effect, fluorescence will be lost due to π-π stacking, whereas in AIE, due to the restricted intramolecular motion, a favourable non-radiative decay pathway is suppressed, resulting in enhanced fluorescence in the solid-state aggregated phase and the same for the liquid state, but at the same time, quantum yield and wavelength might be changed. Hydrophobicity and aggregation behaviour in carbon dots are interrelated phenomena that stem from the surface energy and interfacial interactions of the nanostructures. Hydrophobic CDs are typically characterised by nonpolar surfaces dominated by alkyl chains, aromatic hydrocarbons, or sulphur-containing moieties, which significantly reduce their affinity towards polar solvents such as water. When dispersed in aqueous media, these nonpolar surfaces minimise unfavourable water-surface interactions by self-associating through van der Waals forces and π–π stacking, leading to spontaneous aggregation. This aggregation is energetically favourable, as it reduces the exposed hydrophobic surface area and thereby lowers the total Gibbs free energy of the system. Interestingly, such aggregation does not necessarily lead to fluorescence quenching, as is common for hydrophilic CDs. Instead, hydrophobic CDs often exhibit AIE, where restriction of intramolecular

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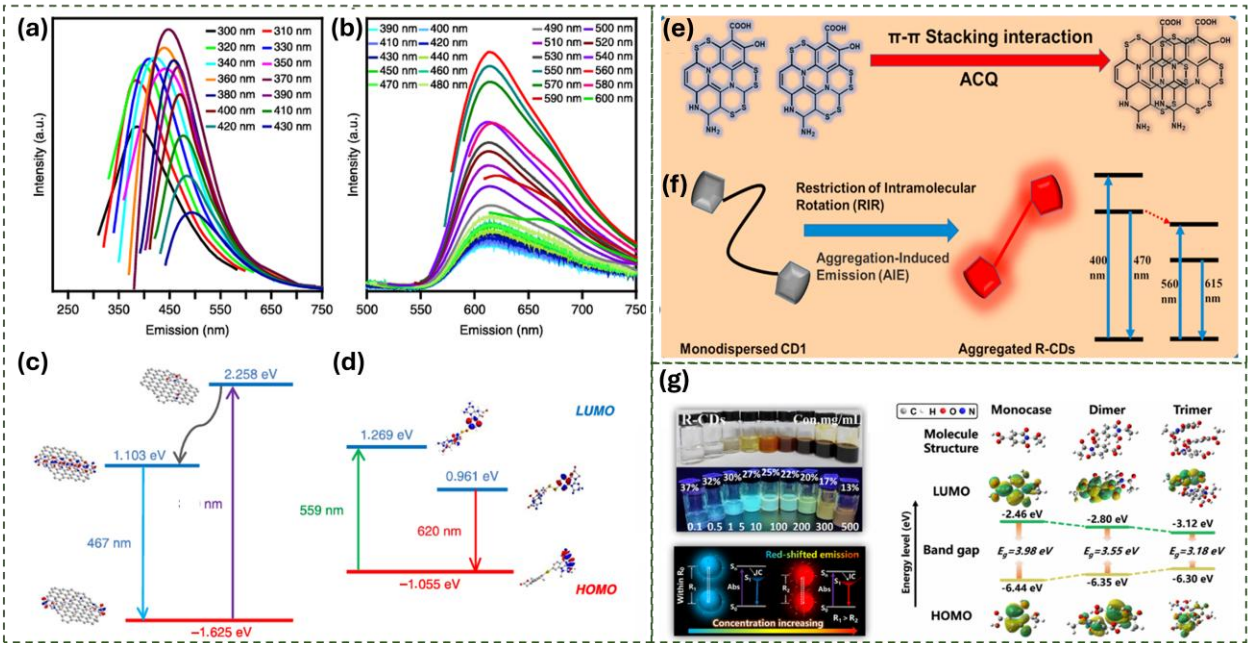

Fig. 4 (a,b) PL emission spectra with different excitation wavelengths of H-CD powder and as-prepared solution. (c,d) Energy level diagram of the proposed H-CDs' graphitizing core, and the surface symmetrical heterocycle's molecular orbital (reproduced with permission from ref. 48 © The Springer Nature 2019).[48] (e) Aggregation quenching effect of CDs. (f) Aggregation induction emission of R-CDs (reproduced with permission from ref. 49 © The Elsevier 2023).[49] (g) photographs of R-CDs in ethanol at different concentrations with the energy band gap between the HOMO and LUMO levels of the potential R-CDs structure (reproduced with permission from ref. 79 © The American Chemical Society 2024).[79]

rotation and vibration in the aggregated state suppresses nonradiative decay pathways, resulting in enhanced luminescence. The AIE behaviour further confirms that aggregation is not merely a byproduct of hydrophobicity but a functional manifestation of it, stabilising emissive states through molecular packing and steric confinement.[49,54,77,80,81]

4.4 Determination of hydrophobic behaviour

Another intriguing feature of hydrophobicity is that any observer may verify it just by looking at how the water interacts with it, eliminating the need for an expensive apparatus. In the morning, one may observe the hydrophobic qualities of grass as dew drops adorn its blades. Similarly, the 'arbi' leaves (Taro leaf or Colocasia leaf) are an excellent example of hydrophobicity. Hydrophobicity could be seen by the naked eye, but one need to prove scientifically or must be a parameter for comparative study of hydrophobic and superhydrophobic materials. The contact angle measurement of water on hydrophobic surface has been widely accepted to prove the hydrophobicity. As basic From CDs' perspective, CDs are consistently supported by multiple characterisation techniques, such as FTIR, XPS, NMR, TEM, and XRD studies indicate that CDs possess small crystalline sp² carbon cores surrounded by disordered shells enriched in carbon moieties. These structural features correlate directly with their optical properties, particularly solvent polarity-dependent photoluminescence and enhanced emission in the aggregated or solid state in HCDs. At first sight, hydrophobicity can be observed with the naked eye, but scientific validation and comparative assessment of hydrophobic and superhydrophobic materials require quantitative parameters. Contact angle measurements is most effective techniques for assessing the hydrophobicity of carbon dots. However, the observed contact angle is an intrinsic property of the material but can vary with sample preparation methods such as pressed powders, thin films or drop-cast coatings, where roughness and film uniformity significantly influence wettability. for example, A. Goswami et al. reported a contact angle of 122° with good hydrophobicity.[52] J. Wang et al. report contact angles of 94.4 - 97.1° for HCDs by controlling the structural features of precursor ionic liquids with respect to the particle size of CDs.[77] H. Wang et al. get the contact angle (left 109.56° and right 110.34°) with good hydrophobic behaviour made by organic pollutant Rhodamine B. as shown in figure 2.[11] M. Ali et al. transformed hydrophobic to superhydrophobic CDs, proved through contact angles from 122.33° to 163° by varying the concentration of oleic acid (10 g/L to 30 g/L). The mechanism of hydrophobicity has also been explored, melamine and 2,2-dithiodibenzoic acid trigger the formation of graphitic carbon core while the surface of CDs is fully covered by the polymeric chain of oleic acid that provides a significant roughness on the surface, contributing to superhydrophobicity.[22] Moreover, examples of hydrophobic carbon dots with contact angle have been tabulated in table 1.

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5. Fluorescent mechanism in hydrophobic carbon dots

The fluorescence mechanism in HCDs is a fascinating interplay of molecular design, environmental interactions, and photophysical principles that collectively overcome the common issue of fluorescence quenching. The central theme that emerges from these studies is the concept of AIE, which ingeniously transforms the usually detrimental process of aggregation into a powerful tool for generating bright and often colour-tunable solid-state luminescence as well as dispersion state aggregation-induced emission. Traditionally, most fluorescent molecules and nanomaterials, including conventional CDs, suffer from a phenomenon known as ACQ effect when they go from liquid to solid state, their hydrophobicity can be either reduced or enhanced; it's hard to predict. When these materials are concentrated in solution or packed into a solid, their fluorescence intensity decreases dramatically. This quenching is primarily caused by strong intermolecular π-π stacking interactions between the aromatic structures of adjacent nanoparticles. These interactions create non-radiative pathways, allowing the excited-state energy to be dissipated as heat or vibrations rather than being released as light. This has been a major obstacle for the practical application of CDs in solid-state devices like light-emitting diodes (LEDs) and sensors**.** A remarkable feature of the HCDs is their dual-emission capability, which is directly linked to their state of aggregation. Generally, in DTSA-based CDs, shows dispersed State (Blue Emission), but when the HCDs are dissolved suitable organic solvent (like ethanol, acetone, or DMSO), they typically exhibit a bright blue or green fluorescence.[11,22,37,73,74,82] This emission is attributed to the intrinsic electronic transitions within the carbonized core of the individual dots, which is rich in sp²-hybridized carbon domains. In this dispersed state, the individual HCDs are segregated or isolated from one another, and the disulphide bond of the DTSA molecule is free for intramolecular rotation. The liquid state blue fluorescence can be changed to red, yellow, and green or any other wavelength upon the aggregation into solid powder. Now the restriction of intramolecular rotation needs further explanation in aggregated carbon dots.[67,80,83–85]

5.1 Restriction of Intramolecular Rotation (RIR)

AIE phenomenon in CDs is governed by principle of Restriction of Intramolecular Rotation. The mechanism relies on the specific chemical groups attached at the surface of the carbon dots. Across the majority of the provided articles, the synthesis of these AIE-active HCDs strategically employs precursors containing disulfide bonds (-S-S-), such as 2,2'-dithiodibenzoic acid (DTSA) or dithiosalicylic acid (DSTA). These disulfide bonds act as flexible molecular rotors. In the dispersed state, these surface groups can freely rotate and vibrate. However, when the HCDs aggregate in the solid state or in a poor solvent, the physical packing and steric hindrance prevent these disulfide bonds from rotating freely. This restriction of motion effectively closes the non-radiative decay channels. With the non-emissive pathways blocked, the excited-state energy has no other option but to be released radiatively, resulting in the strong fluorescence in the aggregated state. For instance, H. Yang *et al.* and another group, X. Zhao *et al.* have reported restriction of intramolecular rotation in aggregation-induced emissive carbon dots. Figure 4 shows a clear visual of the photoluminescence demonstration and mechanism in the dispersion state and aggregation state. The demonstration of π-π stacking and restriction of intramolecular rotation, or locking the molecular rotors in place activates the luminophore (Fig. 4). A group Y. Li *et al.* prepared solid-state fluorescent carbon dots with hydrophobic modification using acetone, induced red emission and applied them in LED applications. They used DFT model for geometry optimisation and the gap between HOMO and LUMO (Fig. 4). By adjusting the proportion of citric acid and urea 1:0, 0.25:1, 0.5:1, and 1:1, four types of blue-fluorescent carbon dots were obtained in the acetone solution, exhibiting maximum fluorescence wavelengths ranging from 430 to 440 nm. However, in the solid state, these four types of CDs exhibited characteristic fluorescence as blue, green, yellow, and red, with respective red shifts ranging from 30 to 190 nm. According to the FT-IR and XPS results, the solid fluorescence changes from blue to red, and the intensity ratio of O−H bonds to C−H bonds and the content ratio of O and C elements gradually decreased, indicating that the hydrophilicity of carbon dots gradually weakens, and the involvement of acetone gradually increases. AFM testing also shows that along with the fluorescence redshifts, the particle size of the CDs nanoparticles gradually increases at the same concentrations.[48,49,85,86]

5.2 Influence of Doping and Core Structure

The alignment of the carbon core in CDs structure plays a crucial role in elucidation of properties. For instance, X. Gong *et al.*, tried to dope manganese (Mn) to achieve hydrophobic carbon dots. The Mn acts as a catalyst during the synthesis, promoting the formation of a more graphitic carbon structure within the core. A well-formed graphitic core can influence the overall electronic properties of the CDs and contribute to the efficiency of the solid-state emission, complementing the surface-state effects driven by the AIE mechanism. In summary, the fluorescence mechanism of the solid-state HCDs described in these articles is a sophisticated process that overcomes the fundamental problem of ACQ. It is primarily driven by the AIE phenomenon, activated by the RIR of specific surface groups, most notably disulfide bonds. The process is initiated by the hydrophobic interaction of the CDs, which forces them to aggregate.[25,50,87]

6. Applications

The distinctive characteristics of hydrophobic carbon dots (H-CDs), especially aggregation-induced emission (AIE), have facilitated numerous creative applications that are frequently unattainable with traditional hydrophilic variants. Leveraging the unique properties of HCDs, in this section we discuss an overview on the current applications in various fields.

6.1 Anti-Counterfeiting and Information Encryption

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The most prominently featured application for these H-CDs is in the field of advanced security, including anti-counterfeiting and multi-level information encryption. The core principle relies on the solvent-switchable fluorescence of the H-CDs, which typically emit blue light when dispersed in an organic solvent and a distinct red or orange-red light when they are present in an aggregated state, However, researchers have not extensively discussed the number of reversible cycles achievable or the long-term stability of dual-mode ink systems. Hydrophobicity is the direct and essential driver of this security mechanism. When an ink formulated with H-CDs is applied to a substrate (e.g. filter paper) and wetted with water, the hydrophobic dots are repelled by the water molecules, forcing them to cluster together. This aggregation activates the AIE mechanism, revealing a red fluorescent pattern under UV light. Conversely, when the same ink is wetted with an organic solvent, such as ethanol, the H-CDs disperse perfectly, and the pattern emits a blue fluorescence. This creates a "two-switch-mode" luminescent ink that can display different information depending on the solvent used for verification, offering a dynamic and highly secure anti-counterfeiting property. Numerous studies demonstrate the creation of complex patterns, QR codes, and even multistage encryption systems where information is encoded in different colours and can only be revealed by a specific sequence of solvent treatments.[14]

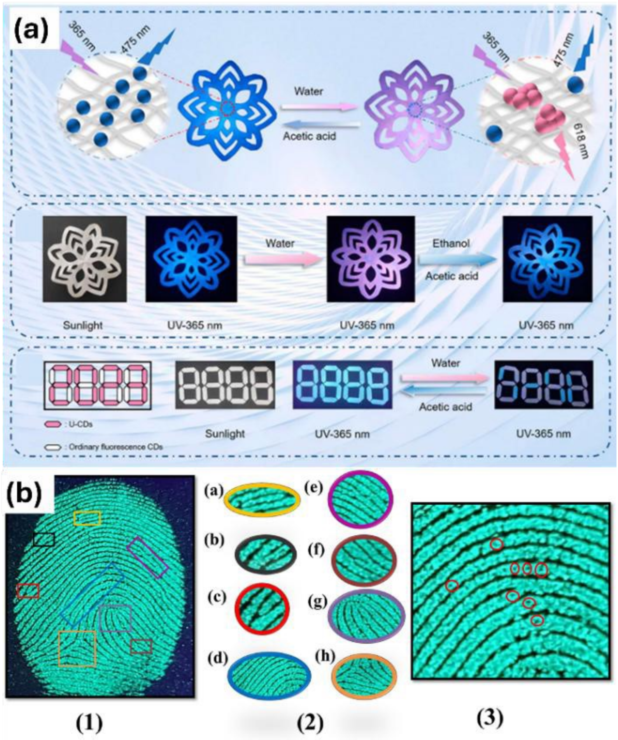

Fig. 5 (a) Dual switch luminescence and information encryption (reproduced with permission ref. 12 © The American Chemical Society 2023).[12] (b) Latent fingerprint detection (reproduced with permission from ref. 67 © The Elsevier 2020).[67]

This extends to multidimensional encryption, where information can be stored not just in colour but also in fluorescence lifetime, creating an exceptionally high level of security. For instance, W. Gao *et al.* prepared HCDs and successfully used in anticounterfeiting application and its dual nature reversibility depends on the solvent. It showed the blue color in the acetic acid, and on the contrary, when water was sprayed, then it showed pink colour in the aggregated state under 365 nm UV lamp on the paper, while in the visible light, there is no appearance of color as demonstrated in figure 5.[12] Similar effects with hydrophobic behaviour reported are by X. Zhao *et al*., they prepared HCDs at different temperatures, and as a result, they discovered blue to red aggregation-induced emissive carbon dots powder. These HCDs can be used as an anti-counterfeiting application.[88]

6.2 Latent Fingerprint Detection

The visualisation of latent fingerprints is a cornerstone of forensic science. The unique surface properties of H-CDs make them highly effective fluorescent dusting powders for latent fingerprints. Fingerprint visualisation operates on the "like dissolves like" principle, in which the hydrophobic surfaces of H-CDs exhibit strong affinity for lipid-rich residues in fingerprints while repelling hydrophilic substrates. Upon application and gentle removal of excess powder, H-CDs selectively adhere to the ridge patterns. Under UV excitation, the aggregated H-CDs display intense solid-state fluorescence, enabling high-contrast and high-resolution fingerprint imaging with minimal background interference. For instance, V. Guttena *et al.* demonstrated obvious visualisation of hydrophobic carbon dots in latent fingerprint detection on different material surfaces such as Auto fluorescent lid, floppy, Glass slide, leather keychain, Magazine paper, disk, compact disc, Aluminium surface and Journal cover page. The subpart of a finger impression is shown in figure 5.[67] Recently, M. Dubey *et al.* reported superhydrophobic AI-enabled solid-state carbon dots with green, red, orange and yellow fluorescence with exceptional contact angle of 172°. In this patent, a Python program was used in fingerprint detection, and the sample score matched 90% which shows a significant similarity to the standard control.[21] So this application of HCDs has great potential to be used in real criminal cases. Moreover, many other researchers are developing and working on advancements in the HCDs and non-HCDs carbon dots for this application.[3,73,75]

6.3 Hydrophobic coating on textile

Hydrophobic coatings on textiles provide excellent water repellence, preventing liquid absorption and keeping fabrics dry and clean. Such coatings enhance durability by protecting fibres from moisture-induced degradation, stains, and microbial growth. They also maintain fabric breathability while improving comfort in wet conditions. Additionally, hydrophobic treatments reduce the need for frequent washing, contributing to energy and water conservation. These coatings are widely applied in outdoor clothing, medical fabrics, and smart textiles for advanced functionality. In these types of applications, CDs offer certain characteristic properties because of their

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hydrophobicity. M. Dubey *et al*. have successfully converted hydrophobic carbon dots into superhydrophobic carbon dots (172°) and applied them on textile and mild steel, with very interesting contact angle 161° and 158°, respectively, as reported in their work as patent. Another researcher, M. Ali's group, gave a clear demonstration of the superhydrophobic contact angle of 163.4° on the textile sample.[21,22]

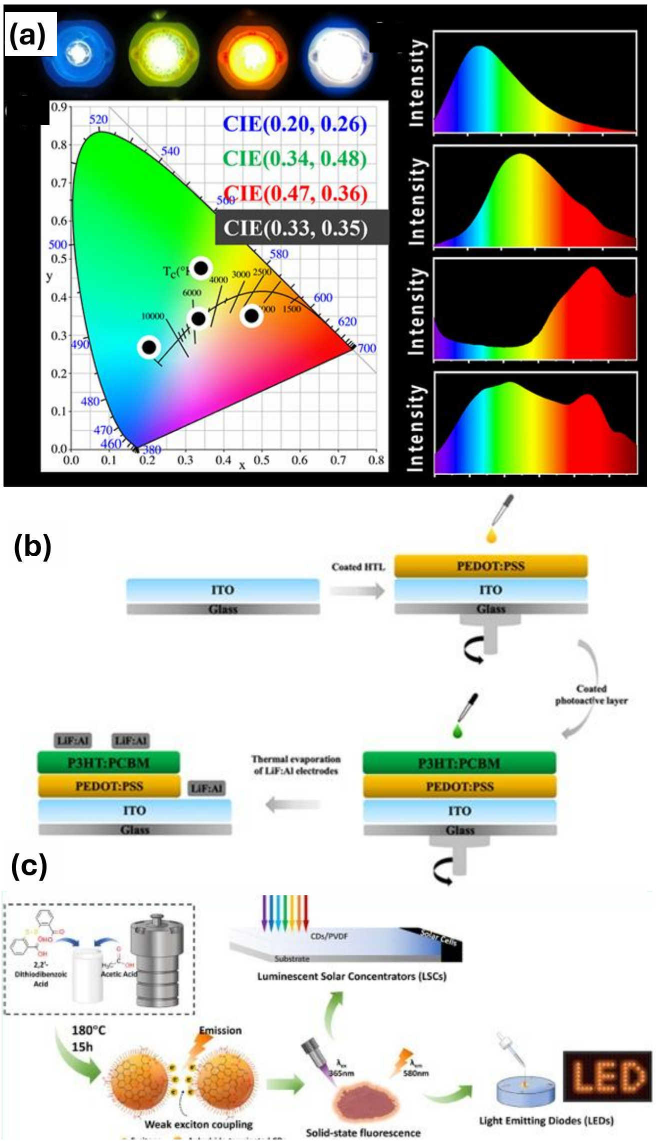

Fig. 6 HCDs used in optoelectronic application, (a) different color LEDs preparation with CIE coordinated demonstration (reproduced with permission from ref. 79 © The American Chemical Society 2024).[79] (b) Photovoltaic application (reproduced with permission from ref. 30 © The American Chemical Society 2022).[30] (c) HCDs in solar concentrators (reproduced with permission from ref. 89 © The American Chemical Society 2022).[89]

6.4 Light-Emitting Diodes (LEDs)

The development of efficient and stable lighting is one of the major goals in materials science. Many researchers are adding their contributions to the field of optoelectronics in application of LEDs using various materials such as phosphors *etc.*[90–93] To fulfil this requirement, H-CDs have emerged as promising phosphors for white light-emitting diodes (WLEDs). Traditional fluorescent materials often suffer from severe quenching when used in a solid device, whereas AIE-active H-CDs overcame this limitation.[73,94] Hydrophobicity provides critical advantages; on one hand, it ensures good compatibility and dispersibility within the organic polymer matrices (such as epoxy, silicone or PDMS, etc.) that are used to fabricate LED devices. On the other hand, it preserves excellent fluorescence in the aggregated solid state as well as in polymer matrix. LED devices are typically classified by two mechanisms: electroluminescence and exhibit n-type or p-type charge transport behaviour. Now to solve this problem, carbon dots are often sandwiched between hole transport layers (Spiro-OMeTAD, PEDOT:PSS, *etc.*) and electron transport layers (TiO_2, ZnO etc.). In contrast, the photoluminescent HCDs are mixed in the appropriate polymer matrix, and this matrix is coated on a UV or blue chip. When the voltage is applied to the UV chip, then the coated materials absorbed this radiation and emit the light in visible region. This type of fabrication is very easy and useful because carbon dots show properties in polymer matrix then after coating result will always be in favour. Furthermore, in advantages, there is no need to change the band gap of the semiconductor to produce different color wavelength light. These properties were utilised by X. Zhao *et al*. and who mixed HCDs powder in epoxy resin and reported red, green, and yellow LEDs. After it by help of these emission properties, warm white light LED is also prepared with a high colour purity of 98.8% and significant CRI coordinates over 60%.[49] It is not single study, other researchers are also performing work on HCDs, and they are reporting many other multicoloured LEDs day by day using different precursors, and solvents.[50,51] Here an example is shown in Figure 5 with corresponding plots of luminescent properties and CIE coordinates reported by X. Lin's group. They produced HCDs on large scale using very cheap precursors, citric acid and urea in acetone. The blue, green, and red LEDs were fabricated using a PVA polymer matrix. Later, a white light LED was also prepared using a similar strategy with exhibition of very high CRI(96.6) value.[79,85]

6.5 Solar cell

Carbon dots (CDs) have emerged as a promising material for next-generation solar cell applications due to their unique optoelectronic and surface properties. Their strong and tunable photoluminescence, excellent light-harvesting ability, and up-conversion photoluminescence enable efficient utilization of the solar spectrum, including near-infrared light. Unlike traditional semiconductor quantum dots that often contain toxic heavy metals, CDs are environmentally friendly, cost-effective, and their tunable surface functionalities allow effective charge transfer and interface engineering, enhancing power conversion efficiency when incorporated into perovskite, dye-sensitized, or hybrid solar cells. Thus, carbon dots represent a sustainable and efficient alternative to conventional quantum dots for solar cell fabrication, combining green chemistry with advanced photovoltaic performance.[95] Moreover, very little

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work is explored in HCDs as photovoltaic applications, but these types of materials may also be used in solar cell applications. With the advantage of hydrophobicity in these types of devices are more stable in moisture. For instance, C. Ünlü *et al.* have made highly luminescent water-insoluble hydrophobic/ amphiphilic carbon dots, so these carbon dots are not truly hydrophobic but partially sustain a hydrophobic nature. The device fabrication is shown in Figure 6. In addition, the productivity of all the parameters in the device was increased after incorporating the CP-DOTs in the photoactive layer. In doped OSCs, FF increased up to 52.15% from the undoped device's FF, which was 48.51% and short circuit current density rose from 7.842 to 9.113 mA cm^{-2} after introducing 3 vol% carbon dots. CDs doping led to an increase in PCE from 2.66 to 3.39%. The addition of CP-DOTs to solar cell systems is a master approach for enhancing device efficiency that can contribute to the scale-up of solar cell technology and commercialization of Device.[30]

6.6 Solar concentrators

Carbon dots (CDs) are promising materials for luminescent solar concentrators (LSCs) due to their strong absorption, tunable emission, and excellent photostability.[96] Their broad absorption in the UV-visible region allows efficient harvesting of sunlight, while their narrow and stable fluorescence enables effective guiding of re-emitted light toward photovoltaic cells placed at the edges. CDs also exhibit low toxicity, easy synthesis, and compatibility with polymer matrices, making them ideal for large-area, flexible, and low-cost LSC devices. Additionally, their high Stokes shift reduces re-absorption losses, improving overall light-conversion efficiency and offering a sustainable route for next-generation solar energy harvesting technologies. HCDs are also efficient materials for this application.[41,79]

C. Wang *et al.* reported an anhydride-terminated CDs with solid state emission in orange region (580 nm) with good photoluminescence quantum yield (PLQY) of 28%. For the fabrication of LSC, typically, 0.3 g of purified anhydride-terminated CDs, DMF with a volume of 7 mL and 0.84 g of PVDF were mixed to form gel that was drop casted on the glass plate (thickness: 2 mm) and scraped into a uniform thin film with a coater of 50 and 200 µm, respectively. After drying, the Solar concentrator was ready for the application. A pictorial demonstration is shown in Figure 6.[89]

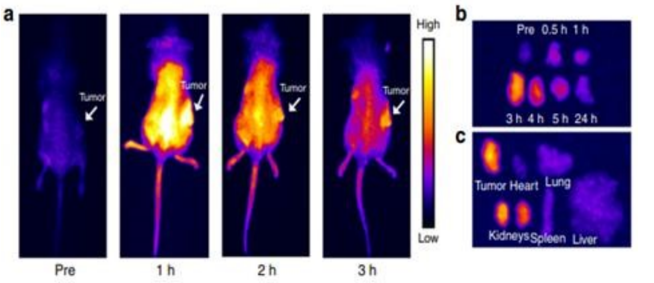

Fig. 7 Tumour cell imagination (reproduced with permission from ref. 97 © The Springer Nature 2018).[97]

6.7 Bioimaging and biosensing

Bioimaging has emerged as a versatile technique for visualizing intracellular compartments, cells, and tissues, thereby providing valuable insights into biological functions under both *in vitro* and *in vivo* conditions. Carbon dots (CDs) have attracted considerable attention in this field owing to their strong biocompatibility and suitability for long-term cellular tracking. While most of the articles focus on materials applications, the principles of controlling fluorescence through molecular interactions are also relevant to bioimaging.[98,99] The *in vivo* bio-distribution of the CDs was studied by S. Qu *et al.* using near-infrared (NIR) fluorescence imaging in both tumour-bearing and normal mice to explore their suitability for tumour diagnosis and therapy. After intravenous injection of the CDs into H$_{22}$ tumour-bearing mice, strong NIR fluorescence gradually appeared across the whole body. Approximately three hours post-injection, the overall fluorescence intensity declined noticeably, while a distinct NIR signal persisted in the tumour region, clearly contrasting with the surrounding tissues (Fig 7).[97] H. F. Wu *et al.* employed hydrophobic carbon dots for imaging breast cancer stem cells (BCSCs). The control group, consisting of BCSCs treated with commercial CdSe quantum dots, displayed bright blue fluorescence. Similarly, the HCDs synthesized using paraplast as a precursor *via* hot injection method produced comparable fluorescence images and no noticeable difference in fluorescence intensity was observed after 6 h and 24 h of incubation with the cells. Owing to their hydrophobic nature, these C-dots can more readily penetrate the cellular membrane than hydrophilic counterparts, making them highly suitable for bioimaging applications.[100]

6.8 Sensing

6.8.1 Chemical Sensing

Small size, large surface-to-volume aspect ratios, and functionalization provide more reactive nature to CDs, allowing them to interact with chemical compounds, which eventually results in changes to their luminescence properties. The tunable emission spectra, narrow spectral bands, and easy functionalization make CDs a good candidate for fluorescence-dependent sensing applications.[101,102] Various chemicals are detected by nano sensors made up of HCDs. For example, X. An *et al.* utilized the efficiency of synthesised HCDs to identify 2,4,6- trinitrophenol (TNP). The dissolved HCDs showed a strong emission at λ_{em} = 495 nm (λ_{ex} = 410 nm) in tetrahydrofuran and the intensity of PL was quenched linearly (limit of detection [LOD] = 1.8 µM) when 1-110 µM TNP was added due to inner filter effect. In addition, they also prepared portable fluorescent test strips that showed the intensity of fluorescence quenched gradually under 365 nm UV-light, with increasing TNP concentrations by 2, 20, 200, and 400 µM, respectively.[103] Recently, X. Gong and his coworkers published a portable sensing system with efficient sensitivity for ethyl alcohol detection in Chinese Baijiu using a smartphone. In explanation of procedure, HCDs, exhibiting bright red emission, were synthesised *via* a hydrothermal method using *o*-phenylenediamine, p-aminobenzoic acid, manganese chloride,

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along hydrochloric acid. The authors visualized that the addition of ultra-pure water to an ethanol solution containing HCDs led to aggregation of HCDs, which changed the PL intensity and absorption behaviour. The changes in colour of HCDs in the water-ethanol mixture under both daylight and UV light allow the creation of a smartphone-based colourimetric detection system for simple and real-time monitoring of ethanol. A schematic overview of the mechanism of HCDs for the detection of ethyl alcohol is represented in figure 8. In the process, captured images of probes exposed to visible and UV spectra of light were successfully analysed, and R, G, and B values were determined. Finally, established a linear correlation between the concentration of ethanol and R, G, and B values. Conclusively, the sensor showed better sensitivity, a wide range of detection and remarkable resistance to interference. Further, some other observations, like interesting reproducibility with standard deviation < 6.6% and high recovery rates (96.5-104.5%), validated its effectiveness.[14]

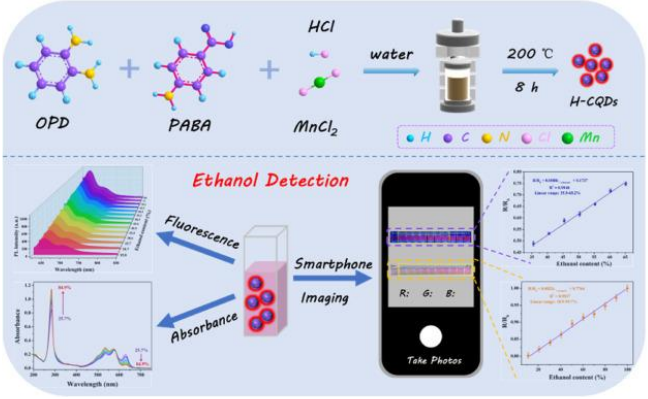

Fig. 8 Schematic illustration of the synthesis process and ethanol detection of HCQDs (reproduced with permission from ref. 14 © The Elsevier 2024).[14]

6.8.2 Physical sensing

Earlier, few research groups synthesized fluorescent gels and put effort to enhance its conductive properties by incorporating metal ion or metal nanoparticle with an objective to utilize in optoelectronic and energy storage devices.[104–107] Recent advances in fluorescent metallogels and supramolecular emissive systems highlight how self-assembly, fluorescence switching and stimuli responsiveness can be integrated within soft materials. These studies collectively underscore the broader importance of adaptive fluorescent platforms for sensing and functional materials research. In recent years, the conductivity, surface-engineered multifunctionality and fluorescent properties of gels is imparted by CDs that enable its use in physical sensing.[108–114] These nanoparticle- immobilized gels incorporated into the external polymer matrix to enhance physical stability. In another case, CD-derived hydrogels have been used for pressure sensor applications. Deformation of hydrogels occurs under various mechanical stimuli, such as vibrations, pressure and tensile stress that bring changes in the electric signal. The signal is recorded by an instrument, and meaningful data helps to use it for different purposes. For instance, S. Y. Park *et. al.* designed hydrophobic carbon dot nanoparticles (f-CD) mixed with polyvinyl alcohol and catechol-conjugated chitosan to obtain a hydrogel suitable for pressure and vibration sensor applications. First, the author utilised the hydrogel for alphabet handwriting sensing, as demonstrated in different letter patterns on hydrogel showed specific electric signals, so the fabricated system can be implemented as the writing pattern decoder as shown in figure 9. In addition, the hydrogel sensor was evaluated through a vocal cord monitoring experiment. the hydrogel sensor was employed to monitor the vibration and transient air gaps generated during the pronunciation of the syllable "Ha." As illustrated in figure 9. Moreover, a smartphone-based analysis was performed by interfacing the hydrogel with a custom-built Bluetooth module (Arduino platform). A clear electrical signal was detected immediately following the articulation of "Ha," whereas no measurable response was recorded during the idle or resting state.[59] S. Y. Park *et. al.* has already fabricated a wireless film hydrogel from a TiO_2-CD-loaded PVA/chitosan matrix to achieve a controllable swelling ratio upon irradiation with UV and visible light. The thin-film hydrogel effectively detected finger movements during both stretching and relaxation, displaying stable oscillations in capacitance. It also demonstrated the potential for real-time monitoring through a wireless transmission setup.[115]

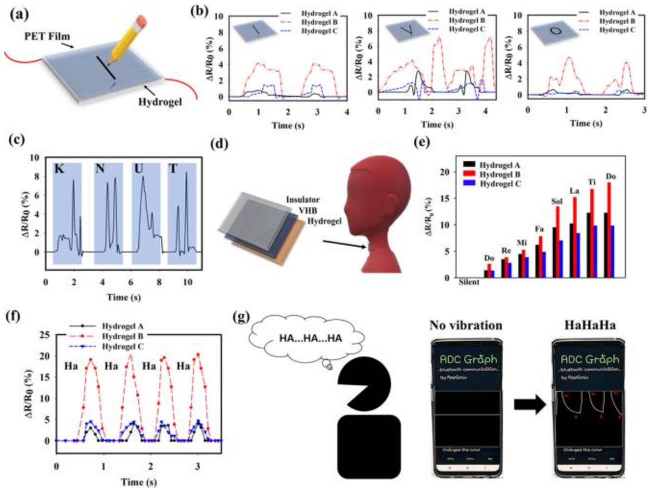

Fig. 9 (a) Illustration of the handwriting analysis using fabricated hydrogel. (b) Relative resistance response during handwriting analysis of I, V, and O. (c) K-N-U-T resistance signal gain when writing on the surface of hydrogel B. (d) Schematic of the vocal cord vibration using a different PVA/C-Chitosan/f-CD hydrogel. (e) Analysis of the vocal cord resistance during one octave vocal range. (f) Resistance response of the novel hydrogel when speaking "Ha." (g) Wireless communication of hydrogel B during connection to smartphone (reproduced with permission from ref. 59 © The American Chemical Society 2020). [59]

6.9 Oil water separation

Oil spill accidents in seawater and industrial oily wastewater have become a major concern because they not only contaminate drinking water sources and foul shorelines, but also lead to significant loss of natural resources. Therefore, effective technologies are needed for oil-water separation.

Several studies have focused on developing materials capable of separating oil from oil-water mixtures. Various superwetting materials-such as sponges, aerogels, and filtration membranes, have been explored for this purpose. In particular, hydrophobic filtration membranes have long been considered promising materials for achieving efficient oil-water separation. G. Chen *et al.* fabricated a membrane having superhydrophobic wettability as well as an antifouling surface that not only achieved efficient oil-water separation (up to 99%), but also performed water desalination to generate fresh water for an emergency water supply in water-scarce regions such as the Middle East and North Africa. It is noteworthy that the membrane exhibited high performance for oil in water emulsion by creating a Span of 80 surfactants for water-in-oil emulsions was quantitatively evaluated through water droplet removal consequently purification of emulsion, indicating effective but more challenging separation behaviour. They synthesised hydrophobic C_{18}-CQDs using citric acid and octadecylamine *via* thermal decomposition method. Superhydrophobic membranes were prepared by cross-linking the synthesized C_{18}-CQDs with cotton fabric using tolylene-2,4-diisocyanate (TDI). This process forms covalent bonds between the C_{18}-CQDs and the cellulose fibres of the cotton, thereby minimizing the potential release of C_{18}-CQDs into the atmosphere. Their single-membrane system effectively achieved simultaneous oil-water separation and saltwater desalination, showing excellent separation efficiency, high salinity rejection rate (~99%), maintaining high flux (~145 $kg \cdot m^{-2} \cdot h^{-1}$) and strong antifouling and recyclability. Antifouling property of C_{18}-CQDs was analysed by measuring the contact angles during cycles of separation process. The membrane showed no significance change in water and oil contact angles over 30 separation cycles, indicating good antifouling properties and stable performance. Furthermore, the membrane efficiently separated various water-in-oil emulsions and maintained high performance even under harsh environmental conditions.

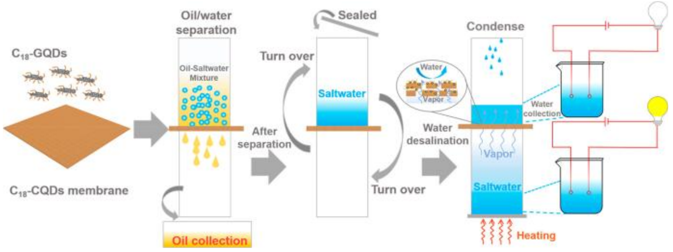

Fig. 10 Separation and Saltwater Desalination (reproduced with permission from ref. 116 © The Americal Chemical Society 2019).[116]

Likewise, porous thermoplastic polyurethane (PTPU) has emerged as a promising material for oil/water separation owing to its intrinsic high porosity, elasticity, and structural recoverability. However, enhancing its surface affinity toward organic contaminants remains essential for achieving higher separation efficiency.[116]

Accordingly, Y. Tian *et. al.* synthesized hydrophobic carbon dots using the same thermal decomposition approach reported earlier, employing citric acid and n-octadecylamine (ODA) as the precursor materials. Now, the prepared HCDs were employed to modify PTPU, yielding PTPU/CDs composites with markedly improved performance. Through optimization of both the PTPU preparation process and CD incorporation, the composites exhibited enhanced hydrophobicity (water contact angle increasing from ~120° to 138.1°), significantly higher compressive strength (186 to 233 KPa), and greater saturated oil absorption capacity (2.4-23 g^{-1} to 4-39 g^{-1}) relative to the unmodified PTPU. The introduction of CDs also imparted bright yellow fluorescence to the composites, enabling direct visualisation of the oil uptake process under UV illumination. The PTPU/CDs materials further demonstrated strong mechanical durability and environmental stability during repeated oil–water treatment cycles. Absorption kinetics indicated that the modified material achieved an approximately 1.5-fold faster absorption rate than pristine PTPU. When utilised as a filtration membrane, the PTPU/CDs composite effectively separated surfactant-stabilised toluene-in-water emulsions with efficiencies up to 99%, underscoring its potential for handling a wide range of oily wastewater streams.[117] Furthermore, M. E. Mohamed *et al.* reported CDs using banana leaves through a hydrothermal route and subsequently used the CDs for superhydrophobic coating to fabricate a textile fabric, coated for efficient oil/water separation. The resulting H-CQD-based superhydrophobic membrane showed amazing surface wettability, with a high-water contact angle of 163° and an exceptionally low sliding angle of 1°. The membrane showed strong performance in terms of oil absorption capacity, separation efficiency, and flux permeativity when evaluated with three representative oils: n-hexane, petroleum ether and silicone oil. Among them, petroleum ether displayed the highest efficiency of separation (99.5%) and permeation flux (13500 $Lm^{-2}h^{-1}$), whereas silicone oil recorded the lowest values in these categories. In contrast, silicone oil demonstrated the highest absorption capacity (218.9 g^{-1}), while petroleum ether showed the lowest (194.8 g^{-1}). A one-way ANOVA was performed to evaluate differences in absorption capacity and separation efficiency among the three oils, and the results confirmed statistically significant variations, underscoring the capability of the superhydrophobic membrane to handle oils with different physicochemical properties. The membrane also exhibited strong mechanical durability, retaining its superhydrophobic behaviour even after an abrasion length of 850 cm, and showed excellent chemical resistance, maintaining its performance across a pH range of 1-13. Overall, the CQD-based superhydrophobic membrane demonstrates considerable potential for practical oil/water separation, combining high efficiency, robustness, and stability under diverse environmental conditions.[118]

7. Future scope/suggestions

As discussed, the potential applications of HCDs in various fields, such as anticounterfeiting dual-mode ink, latent fingerprint detection and information encryption. The stability of CDs has also been optimised for forensic purposes because of its application in the identification of criminals and secure message transfer technology.

Hereby, researchers are continuously exploring ways to enhance the hydrophobicity in the textiles. The optimum variation in contact angles between 90° and 163° has been explored. There is a need for further optimisation in their stability and reproducibility for use in the textile industry.

The hydrophobic properties of HCDs can be utilised in coating application for corrosion protection over metal surfaces. Furthermore, HCDs with good CRI coordinate and fluorescence efficiency, have been utilized for LED devices fabrication. Notably, HCDs also solved the tedious problem related to band gap tuning in semiconductor materials for achieving a particular wavelength to fabricate expensive LEDs. The different coloured LEDs can be fabricated easily by using cost effective UV chip and polymer-HCDs matrix.

In the field of bioimaging, HCDs are very promising materials, and many researchers reported the research work at a lab scale and demonstrated successful results in tumour detection. Despite of their vast potential applications, the development of HCDs at large-scale remain a challenge. The lack of green synthesis approaches, complex multi-stage functionalization, broad particle size distribution, and time-intensive purification processes are key limitations that must be addressed. Furthermore, comprehensive toxicological studies and environmental safety evaluations are essential to ensure their regulatory acceptance and safe utilisation in commercial applications.

In order to explore the future scope in the application of HCDs, scientists are exploring various fields, like energy conversion devices where HCDs can be utilised, like in solar cells and solar concentrators, but these applications need more study because the efficiency of solar cell devices is often poor. But, HCDs can show great performance, if they are used as doping/composite with other well-explored materials like semiconductor quantum dots, perovskite semiconductor materials, *etc.* Furthermore, HCDs need exploration in the field of sensing and oil-water separation owing to cost effective utilization. To address more futuristic applications, HCDs should be explored in the paint industry to achieve superhydrophobic fluorescent paints with an objective to increase the durability of paint by protecting from UV and moisture.

8. Conclusions

In this review, we systematically explained the various preparation methods of hydrophobic carbon dots in an appropriate way, introducing their promising electronic, optical and environmentally friendly properties. In addition, we primarily summarized the factors responsible for hydrophobicity in HCDs. Properties of HCDs are strongly correlated with preparation method, solvent, temperature, reactants, *etc*. The unique luminescence and surface properties provide the ability to HCDs for various applications. The optical properties are suitable for optoelectronics applications like LEDs, Solar cells, solar concentrators, etc. Some researchers used hydrophobic carbon dots to enhance the luminescence properties of polymers. Since the surface properties and emission properties can be tuned by adjusting the extent of carbonization, doping of heteroatoms, and chemical group modifications on the surface, the HCDs are extensively used as physical sensors in motion sensing and chemosensors, for example ethanol detection, various explosive compounds, toxins, etc. HCDs are utilised in bioimaging due to their low cytotoxic nature. Additionally, the properties of HCDs are utilised in fingerprint detection and security ink, crucial for forensic labs. Thus, HCDs may be considered a future smart material to replace tedious and expensive organic compounds for various direct applications.

Author contributions

Dharmendra Kumar and Gaurav Tiwari are responsible for writing the first draft of manuscript. Dr. Mrigendra Dubey **is** responsible for overall supervision, direction and writing final draft of this review manuscript.

Conflicts of interest

“There are no conflicts to declare”.

Data availability

This is a review paper and as such it does not include any primary datasets. No primary research results, software or code have been included and no new data were generated or analysed as part of this review. All the data discussed and analysed within this review are derived from the published studies and literature references in the manuscript. The figures are adopted after copyright permission and cited appropriately.

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