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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, oil–water separation applications, *etc*. This review critically consolidates various preparation methods and fundamentals of hydrophobicity, and provides brief insight into the fluorescence mechanisms of hydrophobic carbon dots. Overall, this 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, lotus leaves exhibit remarkable hydrophobic properties, enabling them to repel water and prevent dust or microbial growth, ensuring self-cleaning and long-lasting vitality. In fact, these natural hydrophobic properties derive from micro/nanostructures present in natural species and make 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, on the journey of materials development from natural plants to engineered carbon dots, hydrophobicity proves to be a powerful and multifunctional property, 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 carbon nanotubes, where they identified 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 materials and termed them ‘‘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 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 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 carbon dots interact with their environment.[7–9] In fact, they can be considered not only as new-generation smart materials, but also as emerging alternatives to multi-step, tedious and over-expensive synthetic organic materials, particularly in applications such as optoelectronics, textiles, bioimaging, oil–water separation, anticounterfeiting inks, *etc*.

The concept of hydrophobicity has evolved over more than two centuries, driven by fundamental theoretical and experimental milestones. The earliest scientific foundation dates back to 1805, when Thomas Young introduced the concept of contact angles in his work ‘Essay on the Cohesion of fluids’,

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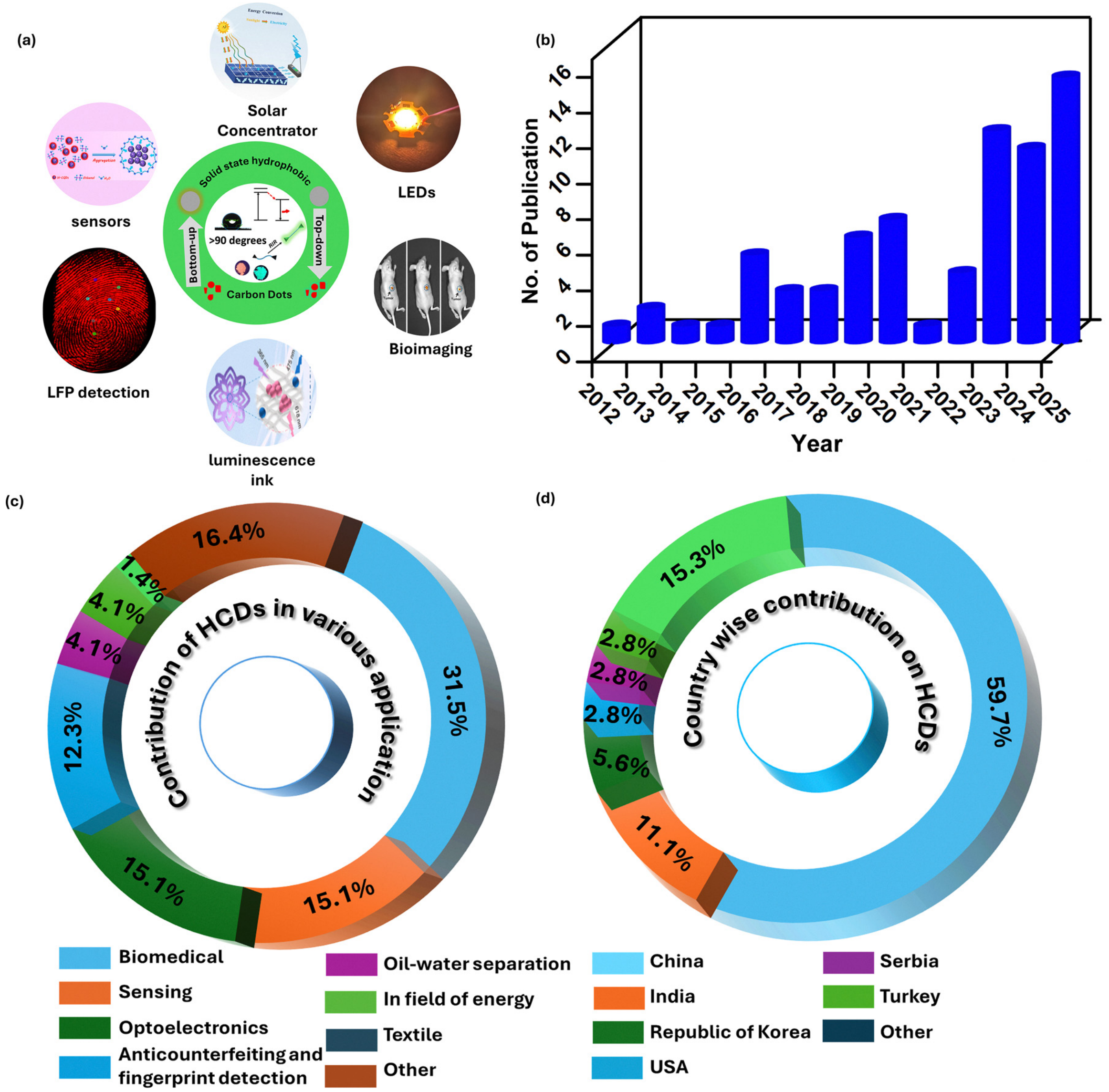

Fig. 1 (a) An overview of hydrophobic carbon dot (HCD) applications (reproduced with permission from ref. 11 © The American Chemical Society 2023, 12 © The American Chemical Society 2023, 13 © The American Chemical Society 2020, and 14 © The Elsevier 2024, respectively).[11–14] (b)–(d) The general progress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

providing the first physical explanation of wetting and non-wetting behaviour at solid-state interfaces, which later became the basis for the concept of hydrophobicity.[10] Furthermore, in 1936, 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 plant leaf surfaces with hierarchical micro–nano structures due to the superhydrophobic behaviour of plant leaves.[17]

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

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 overview of the world of versatile hydrophobic carbon dots. We explored the methods used to synthesise them, the reason behind the hydrophobicity of CDs, the fluorescence mechanisms of HCDs, and their

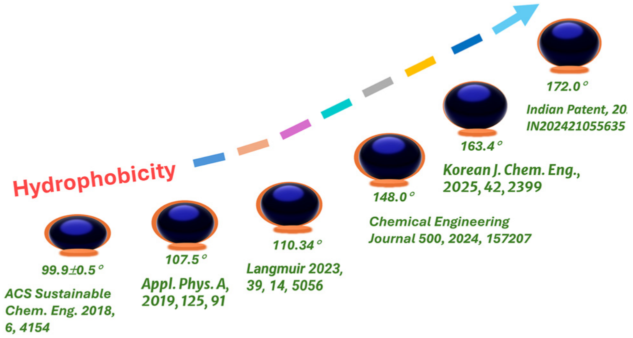

Fig. 2 A general trend of progress in HCDs with contact angle demonstration from hydrophobicity to superhydrophobicity.[11,18–22]

applications across various fields. By bringing these insights together, the current review serves as a valuable guide for anyone looking to harness the unique capabilities of HCDs. Moving forward, the key to unlocking their full potential lies in developing synthesis methods, perfecting their surface design, and focusing on their application because a single type of HCD may show potential across different fields, as discussed in the application section.

2. Classification of CDs

Carbon dots exist in two states: either a dispersive state or a 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 an aqueous medium, because CDs have abundant polar or charged groups (*e.g.*, –OH, –COOH, $-NH_2$, *etc.*) towards the outer surface which facilitate strong binding interactions with water molecules (hydrogen bonding, ion–dipole interaction, *etc*). These hydrophilic moieties make CDs remain colloidally stable in water, exhibit good biocompatibility, and easily adapt to biological environments. For instance, C. Dai *et al.* synthesised high-hydrophilic carbon dots from petroleum coke, which showed potential for enhanced oil recovery applications due to their strong 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 a rational design strategy makes CDs insoluble in water but stable in organic media or in dry states.[4,24,28,29] Scientists also utilized the hydrophobicity concept to achieve aggregation based on the principle that molecules present in CDs organize themselves into hydrophobic and hydrophilic arrays to minimize the contact with water, leading to the formation of 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 methods, mechanisms, and applications, 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 terms of contact angle classification; 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 can be 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, in the case of the water-loving variants of amphiphilic CDs. Another, a significant problem arises, when these dots are packed closely together, they have a natural tendency to aggregate and stack on top of each other, often causing 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, converting carbon dots into solid-state, water-repellent forms by attaching specific organic chains to their surfaces. These dots can be easily dispersed in organic solvents, which makes them highly compatible with 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 HCDs in solid-state applications, such as electronic screens and advanced sensors, as well as their characterization 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 LEGO® 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 a 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 hydrophobic carbon dots is the solvothermal approach, wherein, solution of precursor materials is placed inside a sealed container under high pressure and temperature conditions. The main difference between the two methods lies in the solvent used; if water is used, it is called a hydrothermal process, whereas when an organic solvent is used, the process is referred to as a solvothermal approach. By precisely controlling the reaction 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 application. Another advantage of this technique is, it is considered 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 Fig. 3.[48] Similarly, X. Zhao *et al.* further synthesized red, yellow, and green AIE HCDs by using a 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 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 Fig. 3. This approach makes CDs more environmentally friendly by avoiding acidic media.[50] Furthermore, hydrophobic carbon dots exhibiting superhydrophobicity were reported by M. Ali *et al.*, they made two different combinations of solid-state fluorescence HCDs using DTSA and melamine. To get superhydrophobicity in CDs, they increased the carbon chain length by using hexane. Hexane is a nonpolar solvent, and is used as a suitable candidate to increase hydrophobicity.[21] Using an 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 has 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 developed, such as chemical oxidation, microwave synthesis, pyrolysis, solid-state oven heating, *etc*. A brief discussion of these methods is provided 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 rapid carbonisation, thereby producing CDs. Researchers are attracted towards the method due to its high efficiency and simplicity. For example, A. Goswami *et al.* for the first time reported a facile synthesis of luminescent HCDs using a household microwave oven. Their group used poloxamer Pluronic F-68 and *o*-phosphoric acid at an optimum power of 450 W for 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 the 1,3-dibutylimidazolium dicyanamide ionic liquid and ethyl alcohol *via* the same method at 1300 W for 5 min. Cationic moieties having N-heteroaromatic rings or derivatives of ammonium groups 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 carbon dots using pheophytin and DMF by irradiating microwaves at 150 °C and 400 W for 30 min.[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 such as solvothermal/hydrothermal. Pyrolysis does not require solvents, avoiding dangers of high pressure, while offering advantages such as economic viability, ease of operation, scalability and quick reaction time. Thus, it has become a

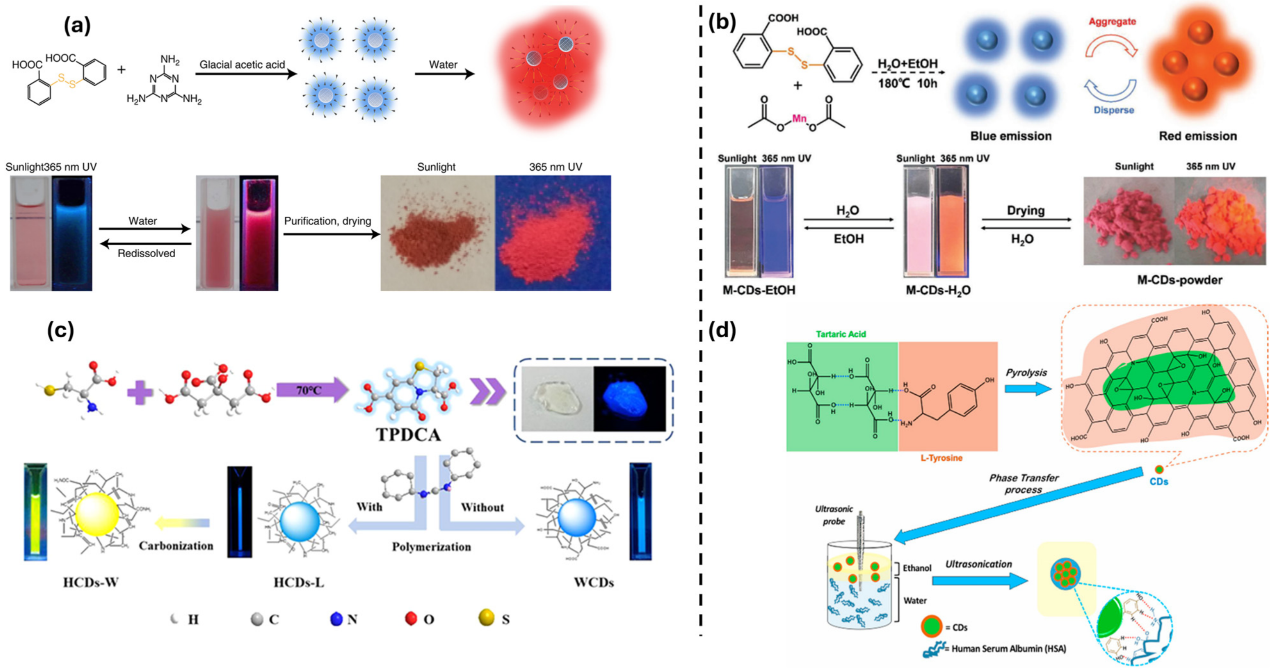

Fig. 3 Preparation route for HCDs through solvothermal (a) dual-mode luminescence solid-state HCDs (reproduced with permission from ref. 48 © The Springer Nature 2019).[48] (b) Synthesis of metal-doped dual-mode switchable fluorescent HCDs (reproduced with permission from ref. 50 © The Wiley 2024).[50]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

preferable choice for researchers.[63,64] To prepare pyrolytic HCDs, a lot of polymeric feedstocks have been explored as demonstrated in Fig. 3. For example, M. Bodik *et al.* synthesised hydrophobic CDs below a size of 5 nm by heating a mixture of the polyoxyethylene–polyoxypropylene–polyoxyethylene triblock copolymer (Pluronic PF-68) and phosphoric acid at 250 °C under 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 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 dots were synthesized using a homogeneous mixture of *o*-phenylenediamine and pyridinedicrboxylic acid (mole ratio, 1 : 1) in a ceramic crucible by heating in an oven at 180 °C for 4 hours.[67] M.Z. Fahmi *et al.* reported carbon dots formed by pyrolyzing tartaric acid and L-tyrosine at 220 °C for 30 min, as demonstrated in Fig. 4.[68] In subsequent years, P. Purkayastha *et al.* synthesised 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 Fig. 3. These HCDs were employed as fluorescent lipid raft markers.[69]

3.4. Solid phase synthesis

Another approach that avoids the use of organic solvents and strong acids/bases is solid-phase synthesis, making it more eco-friendly than 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 dots demonstrated outstanding fluorescence properties and were 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 an industrial scale, in which the organic raw materials are dissolved in a chemical oxidant solution to initiate 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 the chemical oxidation method for the synthesis of HCDs by mixing triolein with concentrated sulphuric acid. After liquid–liquid extraction, precipitation was observed and the organic phase was 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 times (28–180 h). They demonstrated that higher yield of HCDs can be obtained by increasing the reaction time and the concentration of NaOH. In 2019, this group reported the same HCDs

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

S.no.	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–Polyoxyethylene (Pluronic 68)	Stirrer heating	Filtration	Blue	Antibacterial activity	99.9°, 98.9°		18
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	110°	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	Fluorescence 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

again and used them 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 1 M HCl solution.[72]

The solvothermal method is considered the best among the 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, the solvothermal process ensures high-quality, tunable, and stable fluorescence, while also facilitating 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, which are costly and 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 properties of luminescence of these CDs cannot be easily transferred to their solid-state counterpart, due to the severe quenching effect, named aggregation caused quenching (ACQ). With time, luminescence properties of most aqueous or solvent-dispersed carbon dots diminish, thus, to retain the fluorescence properties, an extra matrix of polymer is needed. But usually, a 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 chains or matrices is that 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 fields, where water free conditions are strictly required. Therefore, to address these limitations, a new class of CDs, known as solid state emissive carbon dots, has emerged as a promising approach.[73,74]

Nowadays, unlike conventional carbon dots, hydrophobic carbon dots have attracted researchers' attention because of their ability to exhibit strong fluorescence even in the solid

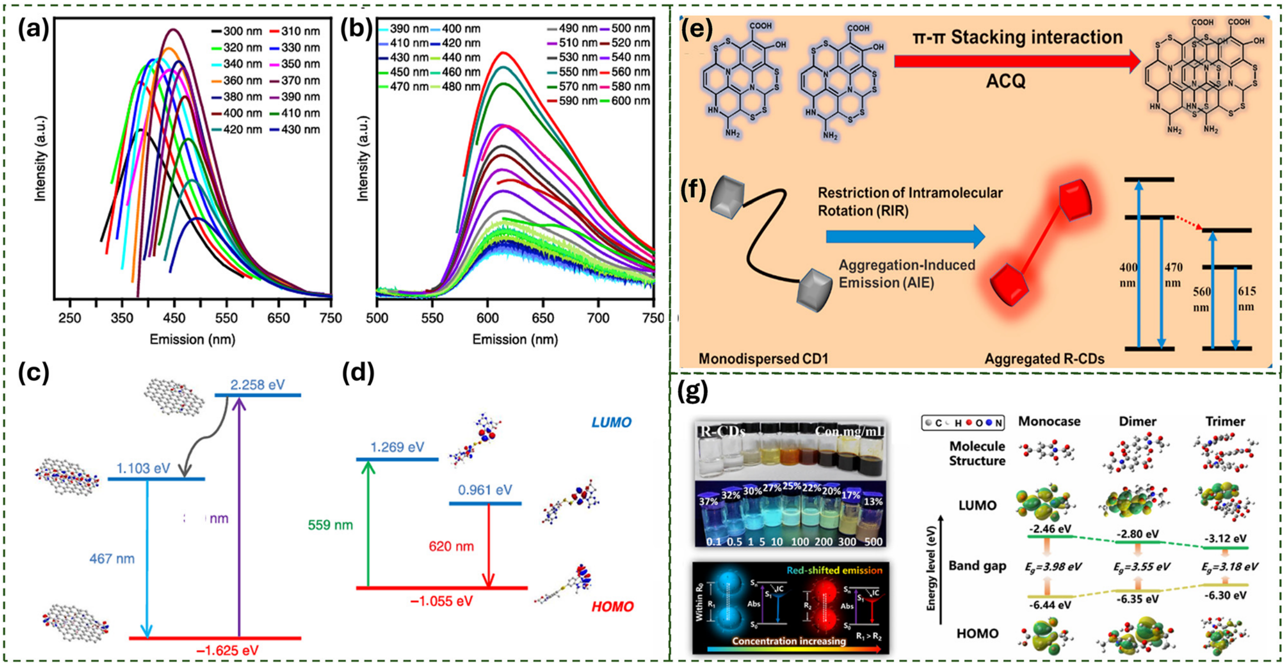

Fig. 4 (a) and (b) PL emission spectra with different excitation wavelengths of H-CD powder and the as-prepared solution. (c) and (d) Energy level diagram of the proposed H-CDs' graphitizing core, and the surface symmetrical heterocycle's molecular orbital (reproduced with permission from ref. 48 © The 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]

state. HCDs are useful for real practical application purposes, such as bioimaging, sensing, optoelectronics, anticounterfeiting, hydrophobic coatings for textiles, oil–water separation, corrosion protection of metals, *etc*. The origin of hydrophobicity in CDs is of considerable interest and a subject of curiosity; thus, based on the literature available and principles of chemistry, a critical overview of the factors responsible for the hydrophobicity of HCDs has 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 are hydrophilic in nature, and some functional groups do not interact with water, as a result, they show hydrophobic behaviour. Typically, polar groups like hydroxyl (–OH), carboxyl (–COOH), and amino (–NH_2) on the surface of CDs readily form hydrogen bonds with water molecules, resulting in excellent aqueous dispersibility and biocompatibility of CDs.

In contrast, hydrophobic carbon dots have a slightly different architecture from that of 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 CDs revealed the absence of NH_2 and OH peaks, because of the reduction of hydrophilic moieties, which makes carbon dots hydrophobic in nature.[75] Similarly, hydrophobic carbon dots reported by J. Hao *et al.*, using an aliphatic chain as one terminal functional group, 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, and –NH_2), which are otherwise responsible for hydrophilicity in conventional CDs. A 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, and their hydrophobicity has typically been evaluated with contact angle measurements, while some researchers have assessed 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 has been revealed in most of the reported results.[12,48,51]

Another surface study of hydrophobic carbon dots was reported by J. Wang *et al.*, and 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 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. Effects of precursors and solvents

The nature of the precursors and solvents is also a significant factor for 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 of the role of DTSA in the generation of hydrophobic carbon dots becomes important. The DTSA has an aromatic benzene ring and a sulphur-rich molecular structure, which strongly influences the surface properties, thus the 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 for 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 a solvent and a catalyst, promoting amidation and controlled carbonisation while limiting the formation of oxygenated polar groups. This results in carbon dots with low surface polarity, with a 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 superhydrophobicity, but by using slightly different solvent conditions. 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 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 hydrophobicity to superhydrophobicity, the nonpolar solvent system promotes dense alkyl chain grafting and nanoscale roughness, thereby reducing surface energy and trapping air pockets at the solid–liquid interface. The resulting contact angle of $\sim 163^\circ$ confirms superhydrophobicity. Thus, the transition from a 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*). The aggregation effect is not only responsible for AIE but also for 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 nonradiative 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 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 water interacts with it, eliminating the need for an expensive apparatus. The hydrophobic nature of grass can be observed in the morning when dew forms droplets on its blades. Similarly, the 'arbi' leaves (Taro leaf or Colocasia leaf) are an excellent example of hydrophobicity. Although hydrophobicity can be observed with the naked eye, it must be quantified scientifically using measurable parameters to enable a reliable comparison between hydrophobic and superhydrophobic materials. The contact angle measurement of water on hydrophobic surfaces has been widely accepted to prove hydrophobicity. From a fundamental perspective, CDs have been consistently characterized by multiple characterisation techniques, such as FTIR, XPS, NMR, TEM, and XRD studies, indicating that CDs possess small crystalline sp^2 carbon cores surrounded by disordered shells enriched with 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 characterization. Contact angle measurement is the most effective technique for assessing the hydrophobicity of carbon dots. However, the observed contact angle is an intrinsic property of the material but it can vary depending on sample preparation methods such as pressed powders, thin films and 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.* reported 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.* obtained contact angles of left 109.56° and right 110.34°, demonstrating good hydrophobic behaviour of CDs made using organic pollutant Rhodamine B, as shown in Fig. 2.[11] M. Ali *et al.* transformed hydrophobic into superhydrophobic CDs, proved through contact angles from 122.33° to 163° by varying the concentration of oleic acid ($10\ \mathrm{g\ L^{-1}}$ to $30\ \mathrm{g\ L^{-1}}$). The mechanism of hydrophobicity has also been explored: melamine and 2,2-dithiodibenzoic acid trigger the formation of a 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 angles are tabulated in Table 1.

5. Fluorescence 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 the ACQ effect when they go from a liquid to solid state, and their hydrophobicity can be either reduced or enhanced, it is 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 HCDs is their dual-emission capability, which is directly linked to their state of aggregation. Generally, DTSA-based CDs show a dispersed state (blue emission), but when HCDs are dissolved in suitable organic solvents (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 sp^2-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 aggregation into a 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)

The AIE phenomenon in CDs is governed by the principle of restriction of intramolecular rotation. The mechanism relies on the specific chemical groups attached to 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. Fig. 4 shows a clear visual representation of the photoluminescence behavior and the underlying 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). 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 the DFT model for geometry optimisation and investigated the energy gap between the HOMO and the LUMO (Fig. 4). By adjusting the proportion of citric acid and urea to 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 blue, green, yellow, and red fluorescence, 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 CD 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 the CD structure plays a crucial role in elucidation of the 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 of the current applications in various fields.

6.1. Anti-counterfeiting and information encryption

The most prominent application of these H-CDs is in 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]

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 their dual nature reversibility depends on the solvent. They showed the blue color in acetic acid, and in contrast, when water was sprayed, they showed the pink colour in the aggregated state under a 365 nm UV lamp on paper, while under visible light, there is no appearance of color as demonstrated in Fig. 5.[12] Similar effects with hydrophobic behaviour are reported by X. Zhao *et al.*, they prepared HCDs at different temperatures, and as a result, they discovered carbon dot powder with blue to red aggregation-induced emission. These HCDs can be used for anti-counterfeiting applications.[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 a 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 for latent fingerprint detection on different material surfaces such as an auto fluorescent lid, floppy disc, glass slide, leather keychain, magazine paper, optical disc, compact disc, aluminium surface and journal cover page. The subpart of a finger impression is shown in Fig. 5.[67] Recently, M. Dubey *et al.* reported superhydrophobic AI-enabled solid-state carbon dots with green, red, orange and yellow fluorescence with an exceptional contact angle of 172°. In this patent, a Python program was used for fingerprint detection, and the sample score matched 90%, which shows a significant similarity to the

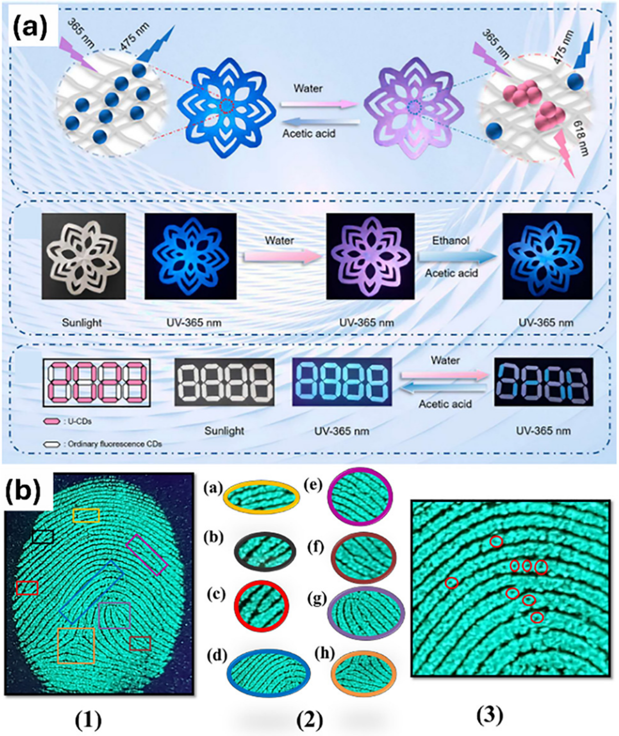

Fig. 5 (a) Dual switch luminescence and information encryption (reproduced with permission ref. 12 © The American Chemical Society 2023).[12] (b) Latent fingerprint detection (reproduced with permission from ref. 67 © The Elsevier 2020).[67]

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 HCDs and non-HCDs for this application.[3,73,75]

6.3. Hydrophobic coatings on textiles

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 achieving advanced functionality. In these types of applications, CDs offer certain characteristic properties because of their 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 angles of 161° and 158°, respectively, as reported in their work as patent. Another researcher, M. Ali's group, gave a clear demonstration of a superhydrophobic contact angle of 163.4° on the textile sample.[21,22]

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 a polymer matrix. LED devices are typically classified by two mechanisms: electroluminescence and 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 with the appropriate polymer matrix, and this matrix is coated onto a UV or blue chip. When the voltage is applied to the UV chip, the coated materials absorbed this radiation and emitted the light in the visible region. This type of fabrication is very easy and useful because carbon dots show properties in a polymer matrix, and then after coating, the result will always be in favour. Furthermore, an advantage is that there is no need to change the band gap of the semiconductor to produce light of different wavelengths. These properties were utilised by X. Zhao *et al.*, who mixed HCD powder in epoxy resin and reported red, green, and yellow LEDs. After that, with the help of these emission properties, a 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 a single study, other researchers have also been studying HCDs, and by using different precursors and solvents they have been regularly reporting many other multicoloured LEDs.[50,51] Here, an example is shown in Fig. 5 with corresponding plots of luminescent properties and CIE coordinates reported by X. Lin's group. They produced HCDs on a 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 a very high CRI (96.6) value.[79,85]

6.5. Solar cells

Carbon dots (CDs) have emerged as promising materials 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

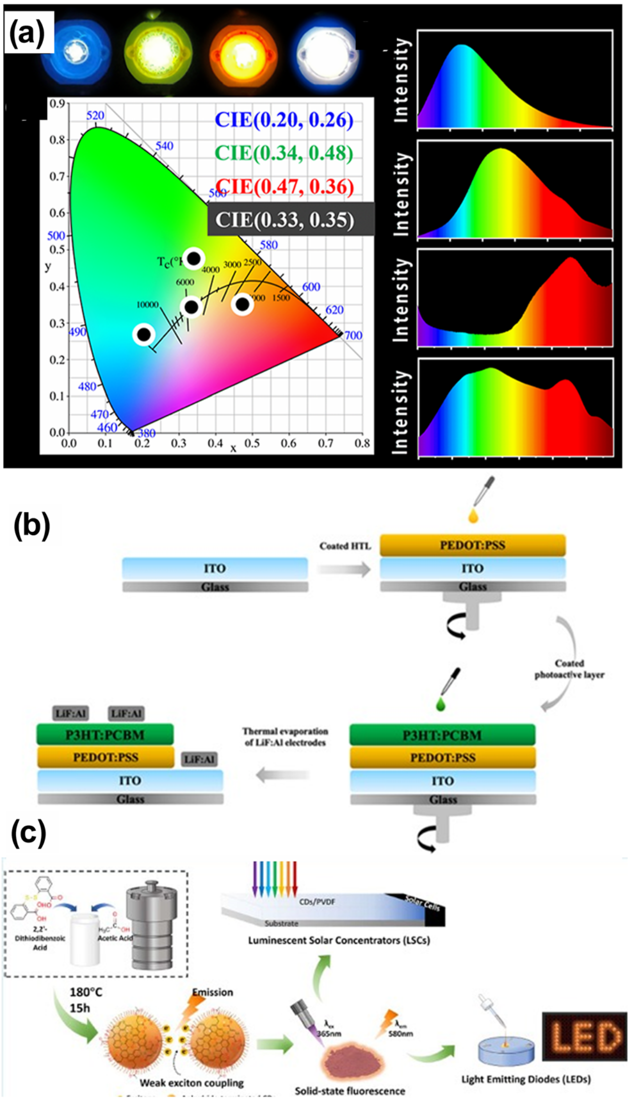

Fig. 6 HCDs used in optoelectronic application, (a) preparation of different color LEDs with CIE coordinated demonstration (reproduced with permission from ref. 79 © The American Chemical Society 2024).[79] (b) Photovoltaic application (reproduced with permission from ref. 30 © The American Chemical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

dots for solar cell fabrication, combining green chemistry with advanced photovoltaic performance.[95] Moreover, very little work is conducted on photovoltaic applications of HCDs, but these types of materials may also be used in solar cell applications. With the advantage of hydrophobicity, 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 retain hydrophobic nature. The device fabrication is shown in Fig. 6. In addition, the productivity performance of all parameters in the device improved after incorporating the CP-DOTs into the photoactive layer. In doped OSCs, the FF increased up to 52.15% compared to the undoped device's FF, which was 48.51% and the short circuit current density increased from 7.842 to 9.113 mA cm^{-2} after introducing 3 vol% carbon dots. CD 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 the 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 anhydride-terminated CDs with solid state emission in the orange region (580 nm) with a good photoluminescence quantum yield (PLQY) of 28%. For the fabrication of a 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 a gel that was drop cast 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 Fig. 6.[89]

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 material applications, the principles of controlling fluorescence through molecular interactions are also relevant to bioimaging.[98,99] The *in vivo* biodistribution of 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

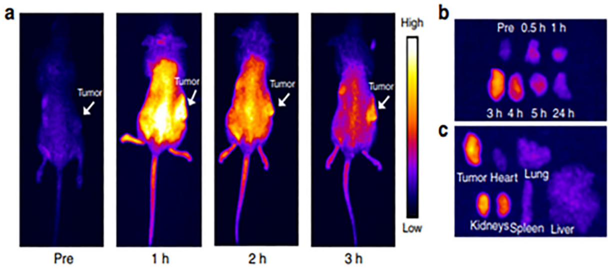

Fig. 7 Tumour cell imaging (reproduced with permission from ref. 97 © The Springer Nature 2018).[97]

quantum dots, displayed bright blue fluorescence. Similarly, the HCDs synthesized using paraplast as a precursor *via* the 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 their 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 in 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 $\lambda_{\text{em}} = 495$ nm ($\lambda_{\text{ex}} = 410$ 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 the inner filter effect. In addition, they also prepared portable fluorescent test strips that showed a gradual decrease of the intensity of fluorescence under 365 nm UV-light, as TNP concentration increased from 2 to 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. Using this procedure, HCDs, exhibiting bright red emission, were synthesised *via* a hydrothermal method using *o*-phenylenediamine, *p*-aminobenzoic acid, manganese chloride, and hydrochloric acid. The authors visualized that the addition of ultrapure 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 conditions 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 Fig. 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, a linear correlation was established 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. Furthermore, some other observations, like interesting reproducibility with standard deviation <6.6% and high recovery rates (96.5–104.5%), validated its effectiveness.[14]

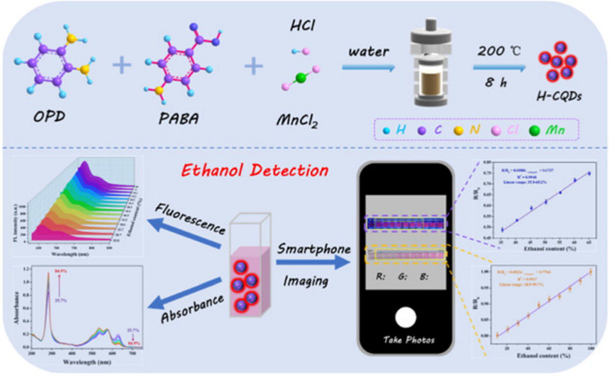

Fig. 8 Schematic illustration of the synthesis process and ethanol detection of HCQDs (reproduced with permission from ref. 14 © The Elsevier 2024).[14]

6.8.2. Physical sensing. Earlier, a few research groups synthesized fluorescent gels and made an effort to enhance their conductive properties by incorporating metal ions or metal nanoparticles with the objective to utilize them 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 fluorescence properties of gels are imparted by CDs that enable their use in physical sensing.[108–114] These nanoparticle-immobilized gels are 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, which bring changes in the electrical signal. The signal is recorded using an instrument, and meaningful data can be used 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 handwriting sensing, and different letter patterns written on the hydrogel generated specific electrical signals, so the fabricated system can be used as a writing pattern decoder as shown in Fig. 9. In addition, the hydrogel sensor was evaluated through a vocal cord monitoring experiment.

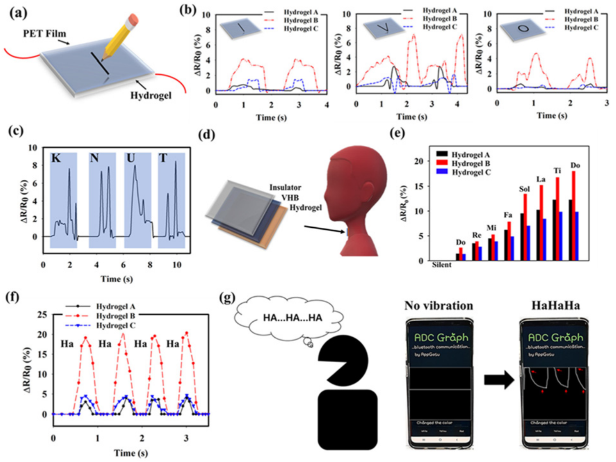

Fig. 9 (a) Illustration of the handwriting analysis using a fabricated hydrogel. (b) Relative resistance response during handwriting analysis of I, V, and O. (c) K-N-U-T resistance signal gain when writing on the surface of hydrogel B. (d) Schematic of the vocal cord vibration using a different PVA/C-Chitosan/f-CD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

The hydrogel sensor was employed to monitor the vibration and transient air gaps generated during the pronunciation of the syllable "Ha", as illustrated in Fig. 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.* have 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]

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. Free oil–water mixtures showed excellent separation efficiency (~98–99%). In contrast, Span 80 stabilised water-in-oil emulsions were more difficult to separate because of their higher stability. Nevertheless, effective separation was achieved, as confirmed by efficient droplet removal and emulsion clarification. They synthesised hydrophobic C_{18}-CQDs using citric acid and octadecylamine *via* a 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 oil–water separation and saltwater desalination simultaneously, showing excellent separation efficiency, a high salinity rejection rate (~99%), and strong antifouling and recyclability, maintaining a high flux (~145 kg m^{-2} h^{-1}). Antifouling properties of C_{18}-CQDs were analysed by measuring the contact angles during the cycles of the separation process. The membrane showed no significant 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).

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 increased 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

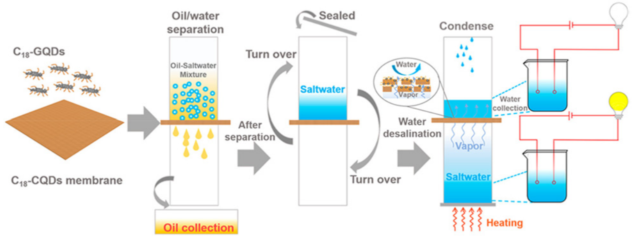

Fig. 10 Separation and saltwater desalination (reproduced with permission from ref. 116 ©, The Americal Chemical Society 2019).[116]

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 exhibited 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 rate 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 (13 500 $\text{L m}^{-2}\ \text{h}^{-1}$), 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, HCDs have demonstrated potential for applications in various fields, such as dual-mode anticounterfeiting inks, latent fingerprint detection and information encryption. The stability of CDs has also been optimised for forensic purposes because of their potential for application in the identification of criminals and secure message transfer technology.

Hereby, researchers are continuously exploring ways to enhance the hydrophobicity of textiles. The optimum range of contact angles, varying between 90° and 163°, has been explored. Further optimisation is required to improve 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 coordinates and fluorescence efficiency have been utilized for LED device 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. Different coloured LEDs can be fabricated easily by using cost effective UV chips and a polymer–HCDs matrix.

In the field of bioimaging, HCDs are very promising materials, and many researchers have reported their research work at a lab scale and demonstrated successful results in tumour detection. Despite their vast potential applications, the development of HCDs on a large-scale remains 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 dopants or incorporated into composites 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 discussed various preparation methods of hydrophobic carbon dots, introducing their promising electronic, optical and environmentally friendly properties. In addition, we primarily summarized the factors responsible for hydrophobicity in HCDs. The properties of HCDs are strongly correlated with preparation methods, solvents, temperature, reactants, *etc.* The unique luminescence and surface properties of HCDs enable their use in various applications. The optical properties are suitable for optoelectronic 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, HCDs are extensively used as physical sensors in motion sensing and chemosensors, for example for detecting ethanol, 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 inks, which are crucial for forensic labs. Thus, HCDs may be considered future smart materials 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 the 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 the literature cited within the manuscript. The figures are adopted after copyright permission and cited appropriately.

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