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Preparation, Structural Regulation and Applications of Petroleum-Based Carbon Quantum Dots

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Abstract. Carbon quantum dots (CQDs) are a type of zero-dimensional carbon nanomaterial with excellent comprehensive properties, which have been widely employed in fluorescence detection, photocatalysis, energy devices, biomedicine and other fields. Petroleum by-products including petroleum coke, needle coke and petroleum asphalt exhibit outstanding advantages such as abundant reserves, low cost and high carbon content. Using these petroleum-derived by-products as precursors to fabricate CQDs is a promising strategy to achieve high-value utilization of petroleum resources and facilitate the green and low-carbon transformation of the petrochemical industry. This paper systematically summarizes recent studies on petroleum-derived CQDs. It clarifies the structural characteristics and pretreatment strategiess of petroleum precursors, and sorts out current progress in preparation technologies, structural regulation and functional applications The existing key challenges in this field are highlighted, and future research directions are prospected. This review can provide a reliable reference for fundamental research and industrial application of petroleum-based CQDs.

Keywords: Petroleum-based carbon quantum dots, Petroleum precursors, Preparation technology, Structural regulation, Functional application

1. Introduction

Carbon quantum dots (CQDs) were first discovered in 2006. Benefiting from their low fabrication cost, negligible biological toxicity, excellent fluorescence stability and facile surface functionalization, CQDs have gradually replaced traditional heavy-metal quantum dots and become a research hotspot in the field of nanomaterials. They have been extensively applied in photocatalysis, sensing detection, photoelectric conversion, biological imaging and other research areas [1, 2]. Massive by-products such as petroleum coke and asphalt are generated during petroleum refining and deep processing. These by-products possess high carbon content but low additional value, and are mostly disposed of by low-cost incineration or extensive utilization, resulting in severe resource waste and environmental burden. Adopting petroleum by-products as precursors for CQD synthesis can diversify raw material sources, lower industrial production costs and improve petroleum resource utilization efficiency. This approach also supports the green and sustainable development of the petrochemical industry with prominent economic and ecological advantages [1, 2].

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Petroleum-based precursors have emerged as a highly competitive and promising direction for CQD preparation due to their high carbon content, low cost and easy accessibility. Foreign studies mainly focus on green fabrication routes and high-end functional applications, while domestic research concentrates on precursor pretreatment optimization and industry-specific applications in the petroleum sector. Both domestic and international studies have achieved preliminary progress. Nevertheless, several bottlenecks still remain: immature pretreatment techniques for raw precursors, insufficient breakthroughs in large-scale production, low precision in structural modulation, limited application scenarios, and the absence of a systematic theoretical and industrial technical framework. Accordingly, this review centers on three core research contents of petroleum-derived CQDs: synthesis methods, structural regulation and functional applications. It summarizes key scientific and technical problems, aiming to provide theoretical guidance for subsequent basic research and industrial transformation.

2. Characteristics and pretreatment of petroleum-based carbon precursors

2.1. Main precursor types and structural characteristics

The performance of petroleum-based CQDs is strongly dependent on the composition and microstructure of precursors. The most commonly used raw precursors include petroleum coke, needle coke and petroleum asphalt. These three materials are produced via different refining processes and differ significantly in microstructure and application suitability.

Petroleum coke is a dominant by-product of delayed coking, with a carbon content of over 90% and extremely low contents of heteroatoms (N, O, S). It is mainly composed of graphite-like microcrystals with a compact microstructure, high graphitization degree and superior chemical stability. After purification and deashing, petroleum coke becomes a high-quality precursor for making highly conductive CQDs. These are well-suited for things like electrochemical energy storage, photoelectric detection, and other areas that need good conductivity [1, 2].

Needle coke is a high-crystallinity special petroleum coke with a carbon content higher than 95%. It exhibits a much higher graphitization degree than conventional petroleum coke, with regularly arranged aromatic structures and excellent electrical conductivity, thermal conductivity and mechanical properties. It is an ideal precursor for high-performance CQDs. The as-prepared CQDs present superior photoelectric conversion efficiency and fluorescence stability, and are suitable for the development of high-end optoelectronic devices [1, 2].

Petroleum asphalt refers to the residual component after petroleum distillation, with carbon content fluctuating between 85% and 95%. It is rich in polycyclic aromatic hydrocarbons and contains relatively high heteroatom species. Its surface has plenty of hydroxyl and carboxyl groups, which give it high chemical reactivity, and it's also very cheap. So asphalt is the go-to raw material for making CQDs on a large scale. However, asphalt contains substantial small-molecule impurities and ash residues. Without appropriate pretreatment, the purity and fluorescence performance of the obtained CQDs will be severely deteriorated. Therefore, pretreatment is an indispensable procedure prior to CQD synthesis [3-5].

2.2. Main pretreatment methods

The core purposes of pretreatment include impurity removal, carbon skeleton activation and enhancement of chemical reactivity, which lay a solid foundation for subsequent carbonization and

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material assembly. The mainstream pretreatment approaches are classified into four categories as follows.

Solvent extraction removes small-molecule organics, ash residues, heavy metals and other impurities based on solubility differences. Common extraction solvents include toluene, ethanol, acetone and N-methylpyrrolidone. This method features simple operation, low cost and no secondary pollution, and is applicable to the preliminary purification of various petroleum-based precursors [3, 4].

Oxidation treatment adopts strong oxidants such as concentrated sulfuric acid, nitric acid, hydrogen peroxide and potassium permanganate to etch the dense carbon skeleton and introduce hydrophilic functional groups (hydroxyl and carboxyl). It comes in two forms: liquid-phase and gas-phase. Liquid-phase oxidation is mild and easy to control, while gas-phase oxidation works faster but uses more energy. The oxidant dosage must be strictly controlled to avoid over-oxidation and destruction of the intrinsic carbon skeleton [3-5].

Activation treatment is adopted to increase the specific surface area and optimize pore structure for improving carbon conversion efficiency, which is generally divided into physical activation and chemical activation. In practical operation, the activator dosage and reaction temperature should be precisely regulated to guarantee satisfactory activation effect and structural integrity of carbon materials [1].

Combined pretreatment integrates two or more individual pretreatment methods to simultaneously achieve purification and activation. Such compound modification has been extensively applied to produce high-quality CQDs from modified petroleum precursors [1, 5].

The carbon content, heteroatom ratio and graphitization degree of precursors directly determine the conversion efficiency and final performance of CQDs. High carbon content contributes to improved fluorescence and electrochemical properties; moderate heteroatoms can act as in-situ dopants to optimize optical performance, while excessive heteroatoms will reduce the purity and structural stability of CQDs. Precursors with high graphitization possess superior electrical conductivity, while those with loose and high-activity structures are more favorable for large-scale CQD fabrication.

3. Preparation methods of petroleum-based carbon quantum dots

3.1. Mainstream preparation technologies

Hydrothermal/solvothermal method is the most widely used route for CQD synthesis in laboratories. The precursor is mixed with a specific solvent and transferred into a high-temperature high-pressure autoclave. CQDs with uniform particle size are formed via carbonization and molecular polymerization. Water serves as the reaction medium in hydrothermal systems. This approach is low-cost and environmentally safe, producing water-soluble CQDs with rich surface functional groups. The solvothermal method employs ethylene glycol, dimethylformamide and other organic solvents to regulate the oil solubility and surface physicochemical properties of CQDs. This route yields CQDs with high purity and excellent fluorescence performance, whereas it suffers from long reaction duration and difficulty in large-scale production, making it more suitable for small-batch preparation of high-performance samples [2-4].

Pyrolysis is conducted at 300–800 °C under an inert atmosphere (nitrogen or argon). This method requires simple equipment with low cost and allows continuous production. The obtained CQDs possess high graphitization degree and excellent electrical conductivity, which are suitable for electrochemical energy storage and photoelectric detection. However, pyrolyzed CQDs usually have

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relatively low purity, broad particle size distribution, scarce surface functional groups and ordinary fluorescence performance, and thus subsequent purification and surface modification are usually required [1, 2].

Chemical oxidation method utilizes strong oxidants for oxidative depolymerization of carbon precursors. After carbon skeleton cleavage, subsequent neutralization, filtration, centrifugation and dialysis purification can harvest high-purity CQDs. This method is easy to operate, cost-effective and high-yield, supporting large-scale industrial production. Sulfuric acid oxidation achieves high product yield and stable fluorescence performance, while its strong corrosiveness brings hidden safety dangers and environmental pressure. Hydrogen peroxide oxidation is eco-friendly and produces water-soluble CQDs, but the reaction efficiency is relatively low. Therefore, precise regulation of oxidation degree is the key to this method [3-5].

3.2. Method comparison and development trend

Hydrothermal/solvothermal, microwave, pyrolysis and chemical oxidation methods differ greatly in reaction conditions, product quality and application scope. The hydrothermal/solvothermal method produces CQDs with uniform size and excellent fluorescence performance, making it suitable for high-value laboratory research. The microwave method features fast reaction rate and simple operation for rapid small-batch synthesis. Pyrolysis allows continuous production with low cost and is suitable for large-scale industrial manufacturing. Chemical oxidation has a simple process and high conversion rate, which is favorable for industrial amplification. Each preparation method has its unique merits, while common limitations remain in product uniformity, environmental compatibility and long-term stability. In the future, the preparation of petroleum-based CQDs will develop toward green chemistry, mild reaction conditions, continuous production and low cost. The coupling of multiple synthesis routes will further enhance product quality and production efficiency, promoting the industrialization of petroleum-based CQDs.

4. Structural regulation and performance optimization

4.1. Regulation of process parameters

Reaction temperature, time, pH value, precursor concentration and oxidant dosage play q key role in the microstructure and performance of CQDs. Rational optimization of these parameters can significantly enhance the comprehensive performance of as-prepared products.

Reaction temperature determines the carbonization degree, particle size and fluorescence behavior. Insufficient temperature leads to incomplete carbonization, while excessively high temperature induces particle agglomeration and fluorescence quenching. Hence, each preparation method corresponds to an optimal temperature window [2-4].

Reaction time affects carbonization completeness and product uniformity. Too short reaction periods produce impure CQDs with unstable properties, whereas prolonged treatment brings particle aggregation, weakened fluorescence and extra energy waste. Operators need to set proper reaction duration based on practical reaction systems [3, 4].

pH value modulates the surface functional groups and surface charge state, thereby altering the intrinsic properties of CQDs. Acidic conditions facilitate the formation of carboxyl and sulfonic groups, improving water solubility and fluorescence intensity; alkaline conditions easily trigger particle agglomeration and reduce fluorescence stability. Consequently, most reaction systems adopt weakly acidic or neutral environments [2, 3].

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Precursor concentration and oxidant dosage also play vital roles in controlling product quality. Excessively high precursor concentration brings severe particle aggregation, and too sparse raw material cuts down production yield. Inadequate oxidant cannot fully crack carbon precursors, and redundant oxidant ruins intrinsic carbon frameworks and worsens product performance. Therefore, parameter balancing and optimization are indispensable [3-5].

4.2. Doping and modification strategies

Heteroatom doping, surface functionalization and composite modification are effective strategies to optimize the performance and broaden the application scope of petroleum-based CQDs, realizing the directional modulation of electronic structure and surface properties.

The commonly used doping elements are N, S and P. N doping effectively improves fluorescence intensity, water solubility and structural stability; S doping enhances photocatalytic activity and electrochemical performance; P doping optimizes electron transport and photoelectric conversion capability. In-situ doping is widely adopted due to its simple operation and stable modulation effect [2-4]. Surface functionalization introduces hydrophilic, hydrophobic or targeting groups via chemical modification, which meets the diverse requirements of aqueous sensing, oil-phase photocatalysis and biological targeted imaging, and further expands the application scenarios of CQDs [2, 3]. Composite modification integrates CQDs with functional materials such as TiO_2, ZnO, graphene, carbon nanotubes and metal nanoparticles. Benefiting from synergistic interactions between different substances, final composites gain better photocatalytic, electrochemical and fluorescence sensing capacity and make up for the defects of pure CQD samples [2-4]. The structure–activity relationship of CQDs is well established: small particle size induces remarkable quantum size effect and high fluorescence quantum yield; nearly spherical morphology ensures good dispersibility and fluorescence stability; high graphitization structure improves electrical conductivity and photocatalytic activity; amorphous carbon structure is beneficial to fluorescence emission; heteroatom doping can precisely modulate the band structure and surface states to achieve synergistic optimization of multiple properties.

5. Applications and development challenges of petroleum-based carbon quantum dots

5.1. Application progress

The most targeted and industrial promising applications of petroleum-based CQDs are concentrated in the petroleum industry. These materials can be applied in oilfield produced wastewater treatment for efficient degradation of phenolic and aromatic pollutants. They enable rapid fluorescence detection of oil and gas components with fast response and ultra-low detection limit. Moreover, they can serve as corrosion inhibitors for oil and gas pipelines, with an inhibition efficiency of over 85%, which prolongs pipeline service life and improves the safety of oil and gas storage and transportation [1, 3, 6]. Producing CQDs from waste petroleum by-products and recycling such materials back into petrochemical processing builds a "resource-material-application" closed cycle. In the field of photocatalysis, petroleum-based CQDs can efficiently degrade organic pollutants, realize antibacterial activity and photocatalytic hydrogen production, with pollutant degradation and antibacterial efficiency exceeding 90%. In optoelectronic devices, they can be applied to fabricate solar cells and LED devices, reducing raw material cost while maintaining device stability and photoelectric conversion efficiency [2-4].

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5.2. Core challenges and future perspectives

At present, the development of petroleum-based CQDs faces five major bottlenecks: low efficiency and poor environmental compatibility of precursor pretreatment; immature large-scale preparation technology and difficulty in maintaining product uniformity; unclear structural regulation mechanism limiting targeted property control; relatively single application scenarios and insufficient exploration of high-end applications; lack of systematic industrial framework and unified quality standards that slow down industrial implementation [1, 3, 4].

Future research will focus on five directions: (1) developing high-efficiency, eco-friendly and low-cost precursor pretreatment technologies to improve raw material utilization and product purity; (2) exploring green, continuous and large-scale fabrication routes to reduce industrial production cost; (3) systematically investigating the structure–activity relationship and regulation mechanism to realize directional performance optimization; (4) exploiting high-end applications such as biomedicine and advanced optoelectronics, while strengthening characteristic applications in the petroleum industry; (5) establishing a complete industrial system and unified quality standards to promote the industrialization of petroleum-based CQDs [1, 3, 4, 6].

6. Conclusions

Petroleum heavy by-products (petroleum coke, needle coke and petroleum asphalt) possess high carbon content, abundant reserves and low price, which are ideal low-cost carbon precursors for CQD synthesis. These materials integrate the intrinsic optical properties of CQDs with the high conductivity and conjugated structure of petroleum-based carbon materials, showing broad application prospects in photocatalysis, fluorescence sensing, optoelectronic devices and energy and environmental fields. This is of great significance for realizing the high-value recycling of petroleum by-products and promoting the green and low-carbon development of the petrochemical industry. This review systematically summarizes the precursor characteristics, pretreatment methods, preparation technologies, structural regulation strategies and application advances of petroleum-based CQDs. Meanwhile, this paper clarifies current research limitations in green pretreatment technology, large-scale production stability, structure–activity relationship analysis and targeted functional design. Overall, petroleum-based CQDs provide a novel route for the resource utilization of heavy petroleum by-products and offer raw material support for the low-cost industrialization of CQDs. Future research should prioritize the development of green and efficient pretreatment, controllable synthesis, precise structural regulation and petroleum-oriented functional materials. Further improvement of theoretical systems and technical standards will facilitate the transformation of petroleum-based CQDs from laboratory research to practical engineering applications, and provide strong support for the high-quality and sustainable development of the petrochemical industry.

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