

Microfluidic Synthesis of Carbon-Based Nanomaterials



Beyza Nur Küçük, Yusuf Aslan, Garbis Atam Akceoglu, and Fatih Inci

Abstract Carbon stands out as an exceptionally versatile element, manifesting in various forms such as diamond, graphite, carbon fiber, fullerene, carbon nanodots, and so on. Leveraging its exceptional mechanical strength, thermal, optical, and density properties, carbon-based materials find widespread applications in industries, spanning from health, electronics, and defense to nanotechnology. The fabrication of diverse carbon materials can be achieved through a range of methods, including wet spinning, top-down and bottom-up approaches like chemical vapor deposition (CVD) and conventional sol-gel processes. In the last decades, the application of microfluidics in synthesizing carbon materials has gained prominence, owing to its superior heat and mass transfer capabilities, minimal chemical requirements, and the ability to create multiplexed and adaptable structures. This chapter provides an in-depth exploration of both conventional and microfluidic fabrication techniques for various carbon-based materials. Herein, an emphasis is placed on elucidating the advantages and limitations faced by these methods, providing a comprehensive overview of the evolving landscape in carbon material synthesis.

Keywords Carbon-based materials · Microfluidic synthesis · Nanomaterial synthesis

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1 Introduction

Nanomaterials are structures with at least one dimension, within a range of 1–100 nm, which are employed in applications such as diagnosis, therapeutics, sensors, and drug delivery [1, 2]. As researchers realized that the properties of materials change as their size decreases to nano sizes, they exhibit distinct chemical and physical properties depending on their different crystal structures, and therefore, their production requires absolute and high control of both thermodynamic and kinetic reactions. In general, nanomaterials are produced using the bottom-up methods, by batch chemical synthesis, template synthesis, or catalytic growth methods, including condensation, chemical precipitation, and hydrothermal synthesis [3, 4]. Despite the use of such different methods, strict control has never been achieved in experimental methods and has resulted in the production of irregular nanoparticles of different sizes. Additionally, most of these methods are complex and require very large devices with trained personnel to use them, and large production and storage areas are needed for the operation [5, 6]. This situation increases the overall cost, wastes resources, and creates safety concerns as well [7]. Another drawback is that these devices are time-consuming and potential contaminations during the synthesis process can occur [6] that may affect the final product homogeneity and size distribution [8]. It is very thought-provoking that such techniques release toxic/dangerous substances that threaten environmental health [5, 8], and act as polluting sources [9]. Choosing a suitable production method that provides proper and effective control over the reaction conditions mentioned above is essential to deliver high-quality products. In this respect, a promising new technology has emerged: microfluidics [10].

Examining the evolutionary trajectory of microfluidic systems reveals their roots in the inception of widely employed analytical tools like capillary electrophoresis. Subsequent years witnessed a confluence of technological advancements, refined production techniques, and progress in material science, culminating in the fabrication of intricate channel structures adaptable for diverse applications. The incorporation of such channel structures into microfluidic systems marked the establishment of an autonomous research domain [11]. Notably, the pioneering work of Manz in the field stands out, particularly in the development of mass chemical analysis systems. These groundbreaking contributions represent the nascent instances of microfluidic technologies, laying the foundation for subsequent advancements in the field [12].

Microfluidic systems exhibit remarkable versatility, accommodating a broad volume spectrum ranging from microliters to picoliters. Their widespread adoption across various industrial domains is attributed to their capacity for delivering heightened efficiency, secure operation, and precise control over reactions [13]. Manipulation of time-control parameters, such as flow rates and the sequence of reagent addition, empowers the regulation of reactions across different phases. This affords adaptive kinetic control and facilitates reactor design tailored to the nuances of the specific reaction mechanism. Notably, the microfluidic system's channels

enable the flow of liquids in the laminar regime, where fluid behavior is predominantly governed by viscosity rather than inertia [14]. The laminar flow characteristic ensures that mass transfer occurs exclusively through diffusion—free from the turbulence inherent in macro-scale liquid flow. Additionally, the distribution of fluid velocity within channels, propelled by pressure-driven flow, adheres to boundary conditions, resulting in a distinctive parabolic velocity profile.

In microfluidic systems, acoustic-, electrokinetic-, magnetic-, and optical-based active approaches are used as mixing mechanisms, as well as passive approaches such as lamination and serpentine channels Tesla microstructures [15, 16]. Microchannels have a high surface-to-volume ratio, allowing the heat produced because of reactions to dissipate quickly and creating more active sites in the reactions [17]. The influence of surface forces (e.g., surface tension), electrical effects, van der Waals interactions, and surface roughness of channel sidewalls becomes more important and often dominates particle motion, especially for nanometer-sized materials (proteins, DNA, etc.), nanocrystals, and so on [18]. These unique properties make microfluidic systems effective tools for applications, such as biological sample preparation, examination of reaction kinetics, chemical synthesis, and biological analyte detection [19]. Moreover, the ability to work with real-time monitoring and high controllability under the laminar regime in small volumes has become indispensable, especially to produce nanomaterials that require precise production techniques. The primary advantage and, concomitantly, the principal challenge inherent in the utilization of microfluidic systems lie in the potential for innovative manipulation of diverse effects and reaction parameters to create tailor-made nanomaterials. Basically, microfluidic systems are used in the control of structures such as shape, size, and structure of nanomaterials, process development of high-efficiency tools, and scale-up through continuous flow processes.

2 Conventional Methods for Producing Carbon-Based Materials

Carbon, among the myriad chemical elements, stands out as one of the most abundant and versatile elements in the universe, playing a distinct and vital role. Its allotropic nature contributes to the foundation of life in nature, as carbon compounds exhibit vastly different properties based on the arrangement of adjacent carbon atoms. Illustratively, diamond and graphite exemplify this phenomenon. Diamond, recognized for its extreme hardness, contrasts sharply with the fragility and brittleness of graphite [20, 21]. Beyond these characteristics, carbon demonstrates the capacity to give rise to a diverse array of nanostructures, encompassing graphene single sheets, mono- and multiwalled carbon nanotubes, carbon fibers, fullerenes, onions, and nanodiamonds. Notably, carbon exhibits a unique capability to form bonds with nearly all chemical elements, resulting in an extensive range of molecules and compounds [22] characterized by diverse chemical and physical

properties. Additionally, graphite is acknowledged for its exceptional thermodynamic stability and high conductivity, rendering it well-suited for applications in electronic devices such as batteries, electrodes, solar panels, and more.

Carbon nanomaterials represent a highly promising avenue in nanotechnology due to their exceptional properties, making them a focal point for extensive research across diverse fields. In the realm of biomedical applications, they introduce novel functionalities, particularly in areas such as sensing and the controlled release of pharmaceuticals and drugs [23, 24]. In electronics, their molecular-sized diameter, combined with a microscopic-scale length and outstanding optical and electrical properties [25], paves the way for the innovation of advanced electronic devices. Utilizing composites derived from carbon nanomaterials proves advantageous, as they exhibit favorable mechanical properties and electrical and thermal conductivities, rendering them valuable in various applications. These materials also showcase desirable attributes for corrosion protection, positioning them as ideal reinforcing elements, as well as protective materials [26], in the development of new engineering products [27]. Furthermore, the applications of microfluidic systems extend to environmental domains, making notable contributions to environmental remediation, separation processes, and water treatment [28–31].

Nanomaterial production can be realized through two fundamental approaches: the top-down and bottom-up methods [32]. The top-down method entails the reduction of larger structures through mechanical force to achieve industrial-scale dimensions. However, this approach lacks precise control over particle growth and demands costly equipment and substantial energy input. Commonly utilized techniques involve mild electric current, pressure, microwave, heat, and ultrasound, often necessitating multistep processes with extended application times. Prominent methods within this category include electrodeposition, solvo-microwave, hydrothermal, solvo-exfoliation, and mechanical exfoliation [33]. Notably, mechanical extraction, exemplified by the tape-peeling technology pioneered by Geim and colleagues, represents a straightforward method for separating graphene layers by overcoming Van der Waals attraction [34].

In contrast, the bottom-up approach requires self-assembly of atomic or molecular precursors to produce nanomaterials. Although this method often tends to form agglomeration and can produce amorphous particles with increased solubility and bioavailability, it is used to produce many nanomaterials. The most used methods are self-assembly, sol-gel, coprecipitation, microemulsion, template synthesis, sonochemical, chemical vapor deposition (CVD), magnetron, Evaporation, and ion beam sputtering [35–38]. Among these, CVD is one of the most frequently used methods in the production of carbon nanomaterials, such as carbon nanotubes and graphene [39]. In CVD, gaseous reactants are applied on a suitable substrate at relatively high temperatures (600–2300 °C), and this application causes the products to accumulate layer by layer on the suitable substrate [40].

The methodological constraints inherent in traditional production methods hinder the seamless transition of nanomaterial production into practical applications across diverse fields, such as electronics, biomedicine, green synthesis, drug delivery, and biosensors. These limitations impede the progress of research endeavors

within these areas [41, 42]. As a result, there is an urgent need for an easy and effective production technique to produce high-quality nanomaterials.

3 Microfluidic-Based Carbon Material Synthesis

The term *miniaturized total chemical analysis systems* was first utilized by Andreas Manz in 1990, and this technology became the father of current microfluidics [12]. By the definition of George Whitesides, microfluidics alludes to micron-sized channels, including systems that can manipulate 10^{-9} to 10^{-18} L fluid [13]. Recent developments in micro- and nanofluidics help to make rapid analysis with less volume of samples in several areas, such as biology, chemistry, and nanotechnology [43–45]. At micro- and nanoscale flows, with the effects of decreased flow rate and increased surface-to-volume ratio, interfacial and viscous forces become more important than gravitational and inertial forces [46]. These characteristics of microfluidic systems provide them certain advantages, such as enhanced heat and mass transfer and laminar flow, in nanomaterial synthesis compared with the conventional top-down and bottom-up methods, which are generally problematic in terms of reaction control, scalability, harsh chemical utilization, and time-consuming multistep pre- and posttreatments [47, 48]. A significant obstacle in bulk synthesis is nonuniform mixing of chemicals, which causes diminished uniformity in particle distribution. By altering the geometry and size of the channels, flow direction and rate can be controlled precisely. Considering these manners, micromixer-like structures can be utilized for the process of homogenous mixing and reacting within the microchannels, resulting in monodispersed and controllable shaped materials [49]. Moreover, several channels can be run in one chip at the same time continuously and rapidly, which is promising for large-scale synthesis, and eventually, it decreases batch-to-batch differences.

Depending on how the flow pattern is altered, there are two primary categories of microfluidics, which are single-phase (continuous-flow microfluidics) and multiphase flow (droplet-based microfluidics). Continuous flow synthesis is typically carried out in laminar flow with a smaller Reynolds number [50]. The majority of microfluidic fabrications include single-phase flow, which is quite suitable for nanoprecipitation techniques [51]. Moreover, it is an alternative method for graphene-based fiber and graphene oxide sheet fabrication, instead of wet-spinning, solution-spinning, and coaxial spinning [52]. Previous studies have demonstrated that the strong π -interaction during the conventional synthesis process causes significant reassembling of graphene oxide nanosheets, destroying the material's excellent conductive properties and severely limiting its applications [16]. In addition, graphene-utilized fiber-shaped super micro conductors are commonly used materials for wearable fabric electronics considering their flexible and light structure, but to preserve their superior properties, the correct fabrication method must be chosen [53]. Recent reports point out that microfluidic spinning is an emerging technology to address the challenges in the existing fabrication methods (Fig. 1a)

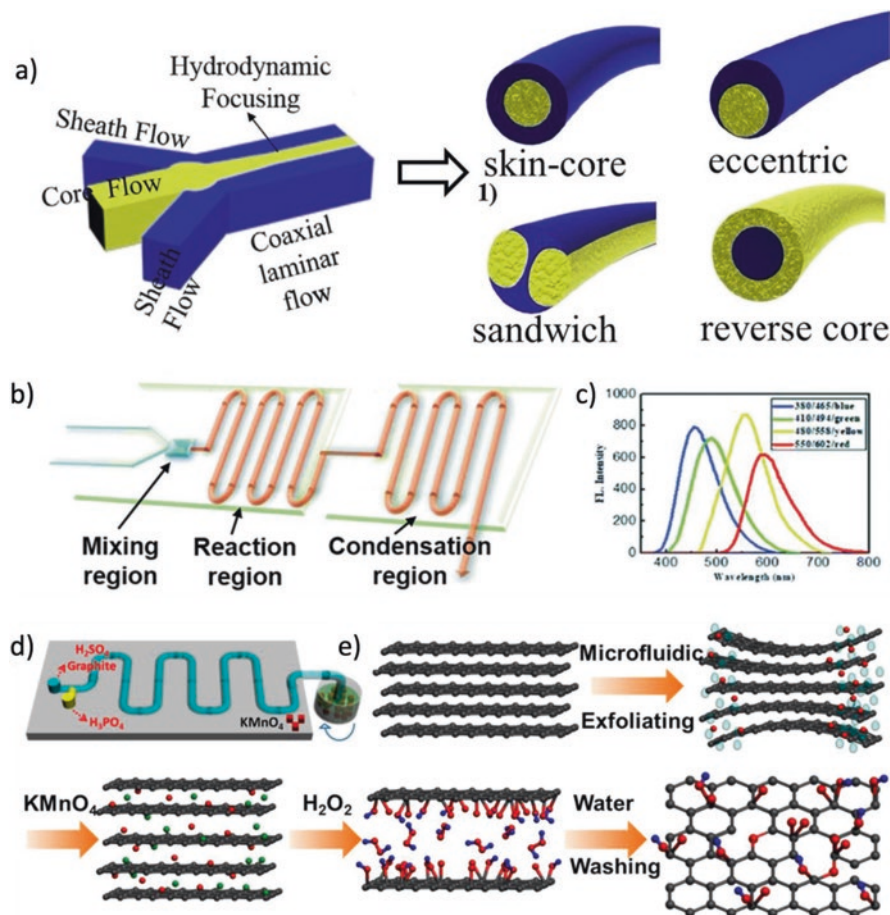


Fig. 1 Schematics of (a) shape tunable composite carbon fiber fabrication. (Reproduced with permission from Hu et al. [54], Copyright 2019, Elsevier); (b) three-stepped synthesis of carbon dots and (c) full spectrum of synthesized CDs. (Reproduced from Shao et al. [55], Copyright 2019, Royal Society of Chemistry). (d) microdroplet formation and (e) graphene oxide exfoliation in microdroplet microreactors. (Reproduced the permission from Qiu et al. [56], Copyright 2020, American Chemical Society)

[54]. Homogenous porous structures created on the fibers with precise control during the microfluidic fabrication would increase the surface area, which provides enhanced areal-specific capacitance and areal energy density. For instance, N-doped graphene fibers with uniformly distributed porous structures fabricated with a microfluidic system exhibit a promising areal energy density of $95.7 \mu\text{Whcm}^{-2}$ [57]. The second flow pattern is multiphased microfluidics, which involves more than one immiscible phase, which could be liquid-liquid or gas-liquid. Herein, by applying multiple reactants from separated channels at a controllable rate, the mixing ratio can be arranged and materials with diverse morphology and hybrid characteristics

can be fabricated, such as hollow, core-shell, or composite fibers and Janus particles [58]. Droplet-based microfluidics focuses on the creation and manipulation of micron- to nano-sized separate droplets within the channels. Droplet production can be precisely monitored in real time and controlled over material fabrication processes by adjusting the device geometry, channel dimensions, and flow rates of individual fluids [59]. It is a commonly employed method for uniformly distributed nanoparticle synthesis, such as carbon dots (CDs) (Fig. 1b, c) [60], quantum dots (QDs) [61], and porous carbon nanospheres [62]. Through microfluidic systems, nanomaterials can be fabricated and modified with various materials in a single step [63]. Furthermore, each droplet can be used as a separate microreactor for multiplexed carbon material synthesis (Fig. 1d, e) [56]. Regarding these, microfluidic fabrication offers an alternative green synthesis technique with a micro- and nano-scaled control on the material.

Even though microfluidics has superior features over conventional methods, including high reproducibility, controllable particle shape and tunable particle size, less reagent consumption, and advanced heat and mass transfer, they also have restraints. First and most commonly, micron-sized channels are prone to clogging. Aggregated particles in the channels, for instance, cause obstacles in the chip that may affect the mixing rate of reagents and cause the reaction to fail. Material compatibility and enhancement could be another problem because, generally, materials are selected for conventional laboratory methods; however, microfluidics may require extreme properties. Moreover, the fabrication of microfluidic devices itself can be intricate and may need specialized expertise and equipment. In view of these, there are still many features that can be improved for microfluidics.

4 Synthesis of Carbon-Based Materials On-Chip

Carbon is destined to play a highly significant role in the nanotechnology age due to its small size and bond hybridization. These properties enable carbon to form numerous structures with versatile dimensions, such as zero-dimensional carbon dots (CDs) and fullerenes, one-dimensional carbon nanotubes (CNTs), and two-dimensional graphene sheets. Carbon-based materials offer a broad spectrum of applications in numerous scientific fields, including biomedical imaging [64], drug delivery systems [65], organic electronics [66], in vivo and in vitro biomedical applications [67], and biosensors [53].

Conventionally, the synthesis of carbon-based materials includes top-down methods, such as laser ablation [68], electron beam ablation [69], and electric arc discharge [70], as well as there are strategies on bottom-up chemical methods, such as chemical process methods [71]. Further, carbon nanomaterials require sorting and isolation for achieving monodispersed size and structure. This is highly vital since the spectral properties of carbon materials are dependent on their size. The polydispersity results in low control over the physical and electronic structure of the material [7]. The conventional methods lack control over structural parameters, and

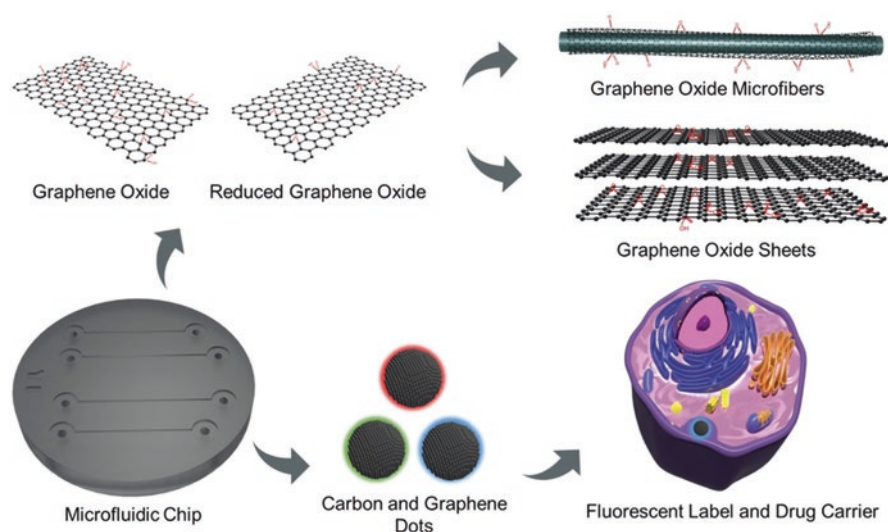


Fig. 2 Schematic representation of the microfluidic synthesis of carbon-based nanomaterials

reproducible large-scale fabrication is not established for such methods. However, the utility of microfluidics enables well-manipulations over the structural parameters by providing a controllable and continuous process. Today, many carbon-based materials have been synthesized via microfluidic techniques, including carbon and graphene quantum dots, graphene, graphene oxide, reduced graphene oxide, and carbon spheres. Current microfluidic methods include either direct or hybrid synthesis [72]. Direct synthesis utilizes microfluidic methods to fabricate carbon nanomaterials with desired properties, whereas hybrid synthesis is the integration of carbon nanomaterials with others using microfluidic techniques as a tool. This section explains the microfluidic synthesis of carbon-based nanomaterials, specifically examining carbon dots, graphene dots, and integrated hybrid nanomaterials, which include graphene oxide (GO) and reduced graphene oxide (rGO) (Fig. 2).

4.1 Synthesis of Carbon and Graphene Dots On-Chip

Right after the discovery through the isolation of single-walled carbon nanotubes (SWCNTs), carbon dots (CDs) became the central part of the carbon-based nanomaterials [73], which was then awarded the Nobel Prize for Chemistry in 2023 [74]. Carbon nanodots have tunable photoluminescence, high quantum yield, and biocompatibility properties. In addition, they utilize cost-effective precursors for the synthesis, resulting in high quantum yield [75]. They are utilized in numerous applications encompassing biosensors [76], bio-imaging [77], theranostics [78], catalysis [79], optoelectronics [80], and photonic crystals [60]. CDs are frequently

generated using laser ablation technique, wherein a carbon substrate is subjected to an intense laser beam in an atmosphere of water vapor, while argon is utilized as the carrier gas. However, this strategy and many other conventional methods have confines over the control over size and structure control, which impede the scalability of the process. Microfluidic synthesis inside microreactors provides a facile and rapid optimization process with enhanced heat- and mass-transfer rates [81]. Teflon capillaries [82], PTFE tubes [83], stainless steel tubes [83] (Fig. 3a), foamy copper microchannels [84] (Fig. 3b), and ceramic microfluidics [86] are the preferred materials for the synthesis of carbon materials on a chip. These materials are chosen for either their high thermal conductive (i.e., stainless steel and copper) or inert and chemical-resistant (i.e., ceramic, PTFE, and Teflon) properties. For the generation of CDs, continuous laminar flow microfluidics with linear-like, snake-like, and spiral mixing modules [87] and droplet-based microreactors with x-, t-, and combined junctions [60] are utilized. Citric acid, ascorbic acid, ethylenediamine, diethylenetriamine, L-cysteine, ethanolamine, and phosphoric acid are some of the examples of carbon sources [81]. Handling of different types of solvents is also an essential step for the process optimization step. By adjusting these process parameters and the microfluidic platform, the photoluminescence properties of CDs can be tuned in a controlled manner. In conclusion, the utilization of microfluidics accelerates the optimization and enhanced processes with remarkable heat- and mass-transfer rates, compared to other large-scale synthesis techniques, such as hydrothermal, microwave-assisted, and magnetic hyperthermia methods [81].

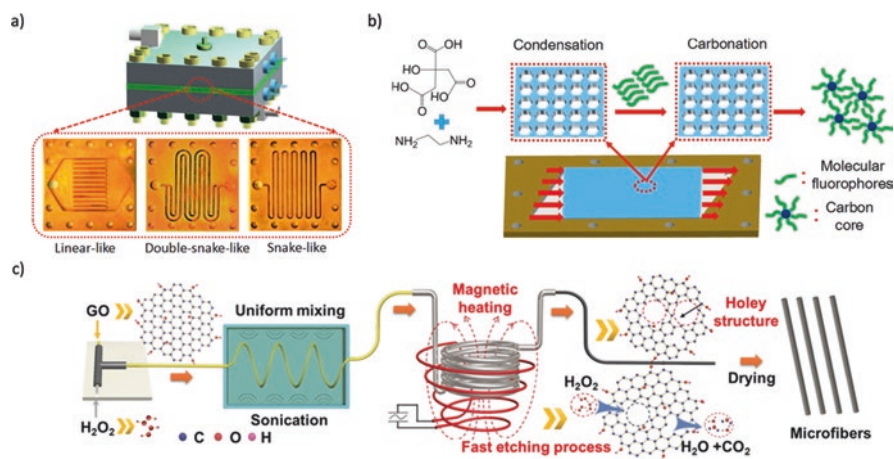


Fig. 3 Some examples of microfluidic-based synthesis of carbon-based nanomaterials. (a) Interior of a stainless steel microreactor for efficient CD synthesis with three different mixing module designs [83], Copyright 2017, Elsevier. (b) Fabrication of nitrogen-doped CDs using microreactor, including foamy copper [84], Copyright 2018, Elsevier. (c) Fabrication steps of GO integrated and magnetothermal microfluidic-assisted core-shell microfibers [85], Copyright 2020, Wiley

4.2 *Synthesis of Graphene Oxide (GO) and Reduced Graphene Oxide (rGO) Integrated Hybrid Nanomaterials On-Chip*

Graphene, a two-dimensional sheet composed single-atom-thick carbon-carbon structure, is firstly fabricated from a block of graphite [88]. Contrary to its single-atom thickness, graphene is much stronger than diamond and has higher electrical conductivity than copper. Furthermore, this material has great charge mobility and almost perfect optical transparency [89]. Graphene is synthesized using both bottom-up and top-down techniques. The most utilized and practical technique is mechanical exfoliation. Many methods provide sufficient large-scale fabrication of graphene; yet, the control over the size remained as the major drawback. Graphene oxide (GO) is obtained by layering both sides of a graphene sheet with functional oxygen groups [90] and conventionally fabricated by the oxidation of graphite and further exfoliated into GO sheets [91]. In addition, GO is synthesized by chemical [92] and electrochemical methods [93]. However, the introduction of oxygen, including functional groups ($=O$, $-OH$, $-O-$, $-COOH$), forms a large amount of crystal defects, which deteriorates the optical and conductive properties compared to graphene. Further, GO exhibits hydrophilic and electrical insulator properties, which are utilized in various applications, including drug delivery [94], biosensing [53], water purification [95], ion-selective membranes [96], and building materials [97]. Reduction of GO into rGO provides the reacquisition of the graphene-like properties of good electrical conductivity and strong optical absorption through reductive treatments [98].

Microfluidics are utilized to integrate graphene, GO, and rGO into hybrid nanomaterials by providing efficient and rapid optimization methods over the structure and size of the hybrid nanomaterials. Flexible and high-conductive fibers [56, 57], core-shell microfibers [85] (Fig. 3c), and spherical microspheres [99] are some of the hybrid material examples, which are synthesized using microfluidic techniques. These obtained hybrid materials possess outstanding properties that can be applied to cutting-edge technologies, such as wearable electronics [56], supercapacitors [85], and dye removal [100].

5 Controlling Material Production

A controlled environment provided by microfluidic devices enhances the reproducibility of monodisperse carbon material synthesis, leading to products with tailored properties, including specific morphologies and well-defined structures [101]. The origin of these parameters can be explained by dividing the synthesis process into temporal steps, including mixing and reaction, nucleation, growth, and termination [102]. Mixing and reaction occurs in the mixing regime of the microfluidic platform and ensures the efficient diffusion of precursors for the synthesis of monodisperse carbon-based materials. After mixing, the solute concentration increases to a critical

concentration where a super saturation limit is established. Supersaturation of monomers initiates the nucleation process and following sequential addition of monomers provides growth until the depletion of reagents terminates the growth process. These temporal synthesis steps can be manipulated in a desired manner by optimizing the design of the microfluidic platform. Adjusting the kinetic parameters [59], such as precursor residence time and concentration, diffusion rate, and mass- and heat-transfer rate, in a microfluidic synthesis process, the properties of carbon-based materials can be tuned accordingly. One of the most important parameters in microfluidic fabrication is the reaction volume, and this volume in microfluidics is adjusted via the diameter and length of microchannels or the size of generated droplets. As the volume inside the microchannel is reduced, the heat- and mass-transfer rates are improved [103]. Further, the diameter of the mixing channels affects the heat and mass transfer and flow rate of the system, and literature shows smaller diameters present better heat and mass transfer of the introduced raw materials from different inlets [55]. Microfluidic fabrication of CDs has also benefited from enhanced heat and mass transfer rate. CDs nucleation process is complex, and a model explaining the process is not found in literature yet [104]. CDs are synthesized from the thermal decomposition of carbon precursors. In this process, the heat transfer rate and residence time are crucial for adjusting the size of CDs which will significantly affect their emission spectrum and quantum yields [105]. Moreover, the design of the microfluidic system is another factor that enhances material synthesis. As mentioned, homogenous mixing of the initially introduced precursor and curing material has crucial importance for uniformly distributed material synthesis. Especially for the carbon-based nanoparticles, the mixing rate, mixing time, and precipitation time affect the size and size distribution and consequently polydispersity index (PDI) of the particles [106]. Because the solvents and antisolvents are mixed in a time period that is shorter than the time needed for precipitation, smaller nanoparticles are formed with a lower PDI when the duration of mixing is less than the duration of precipitation [107]. In bottom-up approaches, the synthesis of nanoparticles takes place in comparatively bigger-scale environments, such as a millimeter or centimeter scale. This leads to localized variations in concentration, which in turn causes the particles to have a big size and a wide variety of sizes due to the greater mixing time than the precipitation time. Mixing in microfluidics happens on a very small scale, often in the range of micrometers. To achieve high-quality nanoparticles and efficient mixing, different shaped microfluidics are introduced in literature, such as spiral [108] and serpentine [109].

6 Conclusion

Carbon-stemmed materials exhibit remarkable electrical, optical, mechanical, and thermal properties, predominantly influenced by size and crystal structure. These properties pave the way in presenting diverse opportunities across various research realms. Conventional fabrication methods however suffer from polydispersity and a

paucity of control over material growth, resulting in the reduced yield of carbon materials with desired features. Moreover, the need for additional sorting and isolation steps prolongs the process time and increases costs. On the other hand, microfluidic synthesis shows an innovative approach that allows for precise manipulation of the growth process of carbon materials. This fine-tuning capability over material properties is achieved through adjustments to kinetic parameters and microfluidic design. This chapter elucidates the principles underlying microfluidic synthesis for carbon-based materials, delving into the fundamentals of carbon-based materials, microfluidic systems tailored for their synthesis, and the pertinent production parameters. The exploration of microfluidic synthesis in this context promises enhanced control, efficiency, and customization in the production of carbon-based materials, addressing key challenges associated with traditional fabrication methods.

The design of the mixing regime within a microfluidic platform plays a pivotal role in shaping the trajectory of a reaction, exerting control over the diffusion rate and residence time of precursors. Through meticulous adjustment of the microfluidic chip design and kinetic parameters, one can precisely tune the morphology and size of carbon-based materials. A defining advantage of employing microfluidic technology in the synthesis of high-performance carbon-based materials lies in its potential for large-scale production, characterized by consistency, devoid of batch-to-batch variations, and amenability to real-time characterization. In particular, single-phase microfluidic platforms stand out for providing a homogeneous mixing regime, ensuring reproducible size and morphology in the synthesized materials. On the other hand, multi-phase platforms offer the additional benefit of automated production, further enhancing efficiency and scalability in the synthesis process. The nuanced control afforded by microfluidic design not only optimizes the material properties but also streamlines the production process, marking a significant advancement in the synthesis of carbon-based materials.

Today, there is a significant emphasis on utilizing microfluidic fabrication techniques for producing carbon and graphene dots, owing to their wide range of applications. By precisely controlling the growth process of a carbon dot with a microfluidic design, its fluorescence spectrum and quantum yield can be adjusted accordingly. A noteworthy trend in microfluidic technology is directed towards the fabrication of near-infrared carbon dots, strategically designed to achieve low-absorption coefficients, thereby enhancing their efficacy in both *in vivo* and *in vitro* bioimaging applications. Furthermore, graphene undergoes oxidation and subsequent reduction processes, resulting in the formation of graphene oxide and reduced graphene oxide sheets. These versatile materials serve as building blocks for various hybrids, including fibers, membranes, and flexible patches. The applications of these hybrid materials span diverse fields, such as wearable electronics, supercapacitors, biosensors, and water purification. It is important to state that currently, direct microfluidic synthesis of both graphene and carbon nanotubes (CNT) from carbon sources is not prevalent in the literature. This scarcity can be attributed to concerns related to fluid interactions potentially introducing crystal defects during the synthesis process. Despite this limitation, the exploration of microfluidic techniques in the fabrication of carbon and graphene-based materials underscores their

significance in advancing the field and addressing specific challenges in material synthesis.

The usage of microfluidics for carbon-based materials also faces many challenges, such as (i) the blockage of microfluidic channels during the growth process, (ii) potential contamination from microchannel walls, and (iii) low throughput, which poses significant hurdles, particularly in meeting the demands of commercial and industrial-scale production. Additionally, there exists a knowledge gap in the understanding of fluid dynamics and growth processes in the microfluidic synthesis of carbon-based materials, necessitating further research. Looking ahead, we anticipate that future studies in the microfluidic synthesis of carbon-based materials will trend toward addressing these challenges. The trajectory of research is expected to focus on advancements in green synthesis techniques, aiming for environmentally friendly processes. Moreover, we foresee a notable integration of machine learning-assisted chip design, leveraging artificial intelligence to optimize and enhance microfluidic systems for improved performance and efficiency. By addressing these challenges and leveraging emerging technologies, the field of microfluidic-based synthesis, especially for carbon-based materials, is poised for continued growth and innovation.

7 Multiple Choice Questions

1. What differentiates microfluidic synthesis of carbon-based materials from conventional methods?
 - A. Polydisperse nature of size and morphology.
 - B. Low heat and mass transfer rate.
 - C. Monodisperse nature of size and morphology.
 - D. Expensive and time-consuming optimization.
2. Which of the following carbon allotrope is not directly fabricated in microfluidics from its carbon sources?
 - A. Carbon nanotubes.
 - B. Graphene oxide.
 - C. Reduced graphene oxide.
 - D. Carbon dots.
3. Which of the following features about graphene oxide is true?
 - A. GO has higher charge mobility and conductivity than graphene and rGO.
 - B. GO and graphene have identical properties.
 - C. GO has lower surface area than graphene and rGO.
 - D. GO contains crystal defects due to oxygen-functionalized groups.

4. It is known that microfluidics has superior features that are advantageous in carbon material synthesis. Which of the following describes the behavior of the fluids in a micro-scale structure?
- Provides improved heat and mass transfer with a high Reynolds number.
 - Provides chaotic flow and better mixing with a high Reynolds number.
 - Provides improved heat and mass transfer with a low Reynolds number.
 - Provides better mixing because gravitational and inertial forces become more dominant over interfacial and viscous forces.

5. (I) Top-down synthesis of carbon nanomaterials can be controlled precisely at the nanoscale.

(II) Graphene fibers, which are a commonly used material in wearable electronics, can be fabricated through continuous-flow microfluidic systems.

(III) Uniformly distributed micron- to nano-sized droplets that can be utilized as a particle or microreactor can be produced via multi-phase microfluidic systems.

Which of the following is the answer when the given sentences are evaluated as “true” or “false”?

- T-T-F.
- F-T-T.
- F-F-T.
- F-F-T.

6. Which of the following is true when the blanks are filled in appropriately?

- In , gaseous reactants are applied on a suitable substrate at relatively high temperatures (600–2300 °C), and this application causes the products to accumulate layer by layer on the suitable substrate.
- can be utilized for the process of homogenous mixing and reacting within the microchannels, resulting in monodispersed and controllable shaped materials.
- Conventionally, the synthesis of carbon-based materials includes methods, such as laser ablation, electron beam ablation, and electric arc discharge, as well as there are strategies on methods such as chemical process methods.

- Chemical vapor deposition—continuous flow—bottom-up—top-down.
- Chemical vapor deposition—micromixers—top-down—bottom-up.
- Thermogravimetric analysis—serpentine channels—bottom-up—top-down.
- Coprecipitation—micromixers—top-down—bottom-up.

8 Answers

1. C
2. A
3. D
4. C
5. B
6. B

References

1. Ece E, Hacısmanoğlu N, Inci F. Microfluidics as a ray of hope for microplastic pollution. *Biosensors*. 2023;13:332.
2. Erdem Ö, Eş I, Saylan Y, et al. In situ synthesis and dynamic simulation of molecularly imprinted polymeric nanoparticles on a micro-reactor system. *Nat Commun*. 2023;14:4840. <https://doi.org/10.1038/s41467-023-40413-8>.
3. Aljuhani E, Al-Ahmed ZA. Evaluation of the physical parameters of nano-sized tetrachlorosilane as an inorganic material a mixed solvent using fuoss-shedlovsky and fuoss-hsia-fernández-prini techniques. *Biointerface Res Appl Chem*. 2020;10:5741–6. <https://doi.org/10.33263/BRIAC104.741746>.
4. Saleh TA. Nanomaterials: classification, properties, and environmental toxicities. *Environ Technol Innov*. 2020;20:101067. <https://doi.org/10.1016/j.eti.2020.101067>.
5. Chatterjee A, Kwatra N, Abraham J, et al. Chapter 11 – Multimodal applications of phytonanoparticles. In: Thajuddin N, Mathew SBT-P, editors. *Micro and nano technologies*. Elsevier; 2020. p. 195–219.
6. Shrimal P, Jadeja G, Patel S. A review on novel methodologies for drug nanoparticle preparation: microfluidic approach. *Chem Eng Res Des*. 2020;153:728–56. <https://doi.org/10.1016/j.cherd.2019.11.031>.
7. Liu Y, Jiang X. Why microfluidics? Merits and trends in chemical synthesis. *Lab Chip*. 2017;17:3960–78. <https://doi.org/10.1039/C7LC00627F>.
8. Chatterjee A, Kwatra N, Abraham J. Chapter 8 – Nanoparticles fabrication by plant extracts. In: Mathew SBT-P, editor. *Thajuddin N. Micro and nano technologies*: Elsevier; 2020. p. 143–57.
9. Fajar MN, Endarko E, Rubiyanto A, et al. A green deposition method of silver nanoparticles on textiles and their antifungal activity. *Biointerface Res Appl Chem*. 2020;10:4902–7. <https://doi.org/10.33263/BRIAC101.902907>.
10. Zhao X, Bian F, Sun L, et al. Microfluidic generation of nanomaterials for biomedical applications. *Small*. 2020;16:1901943. <https://doi.org/10.1002/smll.201901943>.
11. deMello AJ. Control and detection of chemical reactions in microfluidic systems. *Nature*. 2006;442:394–402. <https://doi.org/10.1038/nature05062>.
12. Manz A, Graber N, Widmer HM. Miniaturized total chemical analysis systems: a novel concept for chemical sensing. *Sensors Actuators B Chem*. 1990;1:244–8. [https://doi.org/10.1016/0925-4005\(90\)80209-I](https://doi.org/10.1016/0925-4005(90)80209-I).
13. Whitesides GM. The origins and the future of microfluidics. *Nature*. 2006;442:368–73. <https://doi.org/10.1038/nature05058>.
14. Zhang J, Yan S, Yuan D, et al. Fundamentals and applications of inertial microfluidics: a review. *Lab Chip*. 2016;16:10–34. <https://doi.org/10.1039/C5LC01159K>.

15. Ahmed D, Peng X, Ozcelik A, et al. Acousto-plasmo-fluidics: acoustic modulation of surface plasmon resonance in microfluidic systems. *AIP Adv.* 2015;5:97161. <https://doi.org/10.1063/1.4931641>.
16. Luo Y, Zhang Y, Zhao Y, et al. Aligned carbon nanotube/molybdenum disulfide hybrids for effective fibrous supercapacitors and lithium ion batteries. *J Mater Chem A.* 2015;3:17553–7. <https://doi.org/10.1039/C5TA04457J>.
17. Ehrfeld W, Hessol V, Löwe H. *Microreactors: new technology for modern chemistry.* Wiley; 2000.
18. Huang Y, Duan X, Wei Q, Lieber CM. Directed assembly of one-dimensional nanostructures into functional networks. *Science.* 2001;291:630–3. <https://doi.org/10.1126/science.291.5504.630>.
19. Kovarik ML, Gach PC, Orloff DM, et al. Micro total analysis systems for cell biology and biochemical assays. *Anal Chem.* 2012;84:516–40. <https://doi.org/10.1021/ac202611x>.
20. Koziol K, Boskovic BO, Yahya N. Synthesis of carbon nanostructures by CVD method. *Adv Struct Mater.* 2011;5:23–49. https://doi.org/10.1007/8611_2010_12.
21. Shenderova OA, Zhirnov VV, Brenner DW. Carbon nanostructures. *Crit Rev Solid State Mater Sci.* 2002;27:227–356. <https://doi.org/10.1080/10408430208500497>.
22. Hirsch A. The era of carbon allotropes. *Nat Mater.* 2010;9:868–71. <https://doi.org/10.1038/nmat2885>.
23. Akceoglu GA, Saylan Y, Inci F. A snapshot of microfluidics in point-of-care diagnostics: multifaceted integrity with materials and sensors. *Adv Mater Technol.* 2021;6:2100049. <https://doi.org/10.1002/admt.202100049>.
24. Inci F. Benchmarking a microfluidic-based filtration for isolating biological particles. *Langmuir.* 2022;38:1897–909. https://doi.org/10.1021/ACS.LANGMUIR.1C03119/ASSET/IMAGES/MEDIUM/LA1C03119_0010.GIF.
25. Ghosh D, Sarkar K, Devi P, et al. Current and future perspectives of carbon and graphene quantum dots: from synthesis to strategy for building optoelectronic and energy devices. *Renew Sust Energ Rev.* 2021;135:110391. <https://doi.org/10.1016/j.rser.2020.110391>.
26. Lu Y, Huang G, Wang S, et al. A review on diamond-like carbon films grown by pulsed laser deposition. *Appl Surf Sci.* 2021;541:148573. <https://doi.org/10.1016/j.apsusc.2020.148573>.
27. Page AJ, Ding F, Irle S, Morokuma K. Insights into carbon nanotube and graphene formation mechanisms from molecular simulations: a review. *Rep Prog Phys.* 2015;78:36501. <https://doi.org/10.1088/0034-4885/78/3/036501>.
28. Kanao E, Kubo T, Otsuka K. Carbon-based nanomaterials for separation media. *Bull Chem Soc Jpn.* 2020;93:482–9. <https://doi.org/10.1246/bcsj.20190372>.
29. Wang R, Chen D, Wang Q, et al. Recent advances in applications of carbon nanotubes for desalination: a review. *Nanomater (Basel, Switzerland).* 2020;10:1203. <https://doi.org/10.3390/nano10061203>.
30. Wu W, Shi Y, Liu G, et al. Recent development of graphene oxide based forward osmosis membrane for water treatment: a critical review. *Desalination.* 2020;491:114452. <https://doi.org/10.1016/j.desal.2020.114452>.
31. Yildiz UH, Inci F, Wang S, et al. Recent advances in micro/nanotechnologies for global control of hepatitis B infection. *Biotechnol Adv.* 2015;33:178–90. <https://doi.org/10.1016/j.biotechadv.2014.11.003>.
32. Hamdallah SI, Zoqlam R, Erfle P, et al. Microfluidics for pharmaceutical nanoparticle fabrication: the truth and the myth. *Int J Pharm.* 2020;584:119408. <https://doi.org/10.1016/j.ijpharm.2020.119408>.
33. Liu Y-P, Liu Q-L, Zhang X-L, et al. Bioactive monoterpene indole alkaloids from *Nauclea officinalis*. *Bioorg Chem.* 2019;83:1–5. <https://doi.org/10.1016/j.bioorg.2018.10.013>.
34. Novoselov KS, Geim AK, Morozov SV, et al. Electric field in atomically thin carbon films. *Science (80-).* 2004;306:666–9. <https://doi.org/10.1126/science.1102896>.

35. Garbacz H, Wieceński P, Adamczyk-Cieślak B, et al. Studies of aluminium coatings deposited by vacuum evaporation and magnetron sputtering. *J Microsc.* 2010;237:475–80. <https://doi.org/10.1111/j.1365-2818.2009.03297.x>.
36. Kong J, Cassell AM, Dai H. Chemical vapor deposition of methane for single-walled carbon nanotubes. *Chem Phys Lett.* 1998;292:567–74. [https://doi.org/10.1016/S0009-2614\(98\)00745-3](https://doi.org/10.1016/S0009-2614(98)00745-3).
37. López-Quintela MA. Synthesis of nanomaterials in microemulsions: formation mechanisms and growth control. *Curr Opin Colloid Interface Sci.* 2003;8:137–44. [https://doi.org/10.1016/S1359-0294\(03\)00019-0](https://doi.org/10.1016/S1359-0294(03)00019-0).
38. Wu J-J, Lu T-R, Wu C-T, et al. Nano-carbon nitride synthesis from a bio-molecular target for ion beam sputtering at low temperature. *Diam Relat Mater.* 1999;8:605–9. [https://doi.org/10.1016/s0925-9635\(98\)00283-0](https://doi.org/10.1016/s0925-9635(98)00283-0).
39. Li Y, Gao T, Yao Y, et al. In situ “Chainmail Catalyst” assembly in low-tortuosity, hierarchical carbon frameworks for efficient and stable hydrogen generation. *Adv Energy Mater.* 2018;8:1801289. <https://doi.org/10.1002/aenm.201801289>.
40. Addou R, Dahal A, Sutter P, Batzill M. Monolayer graphene growth on Ni(111) by low temperature chemical vapor deposition. *Appl Phys Lett* 2012;100. <https://doi.org/10.1063/1.3675481>.
41. Ahmed BS, Mostafa AA, Darwesh OM, Abdel-Rahim EA. Development of specific nano-antibody for application in selective and rapid environmental diagnoses of salmonella arizonae. *Biointerface Res Appl Chem.* 2020;10:7198–208. <https://doi.org/10.33263/BRIAC106.71987208>.
42. Monajjemi M, Naghsh F, Mollaamin F. Bio-lipid nano capacitors: resonance with helical myeline proteins. *Biointerface Res Appl Chem.* 2020;10:6695–705. <https://doi.org/10.33263/BRIAC106.66956705>.
43. Inci F, Ozen MO, Saylan Y, et al. A novel on-chip method for differential extraction of sperm in forensic cases. *Adv Sci.* 2018;5:1800121. <https://doi.org/10.1002/advs.201800121>.
44. Wang S, Inci F, De Libero G, et al. Point-of-care assays for tuberculosis: Role of nanotechnology/microfluidics. *Biotechnol Adv.* 2013;31:438–49. <https://doi.org/10.1016/j.biotechadv.2013.01.006>.
45. Wang SQ, Inci F, Chaunzwa TL, et al. Portable microfluidic chip for detection of Escherichia coli in produce and blood. *Int J Nanomedicine.* 2012;7:2591–600. <https://doi.org/10.2147/IJN.S29629>.
46. Wibowo D, Zhao C-X, He Y. Chapter 2 - Fluid properties and hydrodynamics of microfluidic systems. In: Santos HA, Liu D, Zhang H, editors. *Micro and nano technologies*. William Andrew Publishing; 2019. p. 37–77.
47. Baki A, Löwa N, Remmo A, et al. Micromixer synthesis platform for a tuneable production of magnetic single-core iron oxide nanoparticles. *Nanomaterials.* 2020;10:1845. <https://doi.org/10.3390/nano10091845>.
48. Tokel O, Yildiz UH, Inci F, et al. Portable microfluidic integrated plasmonic platform for pathogen detection. *Sci Rep.* 2015;5:9152. <https://doi.org/10.1038/srep09152>.
49. Moragues T, Mitchell S, Akl DF, et al. Droplet-based microfluidics platform for the synthesis of single-atom heterogeneous catalysts. *Small Struct.* 2023;4:2200284. <https://doi.org/10.1002/SSTR.202200284>.
50. Hessel V, Löwe H, Schönfeld F. Micromixers—a review on passive and active mixing principles. *Chem Eng Sci.* 2005;60:2479–501. <https://doi.org/10.1016/j.ces.2004.11.033>.
51. Nie G, Huang Z, Ding S, et al. SnCo nanoalloy/graphene anode constructed by microfluidic-assisted nanoprecipitation for potassium-ion batteries. *ACS Appl Nano Mater.* 2022;5:2616–25. <https://doi.org/10.1021/acsanm.1c04285>.
52. Jia Y, Ahmed A, Jiang X, et al. Microfluidic fabrication of hierarchically porous superconductive carbon black/graphene hybrid fibers for wearable supercapacitor with high specific capacitance. *Electrochim Acta.* 2020;354:136731. <https://doi.org/10.1016/j.electacta.2020.136731>.

53. Erdem Ö, Derin E, Zeibi Shirejini S, et al. Carbon-based nanomaterials and sensing tools for wearable health monitoring devices. *Adv Mater Technol.* 2022;7:1–25. <https://doi.org/10.1002/admt.202100572>.
54. Hu X, Tian M, Pan N, et al. Structure-tunable graphene oxide fibers via microfluidic spinning route for multifunctional textiles. *Carbon.* 2019;152:106–13. <https://doi.org/10.1016/j.carbon.2019.06.010>.
55. Shao M, Yu Q, Jing N, et al. Continuous synthesis of carbon dots with full spectrum fluorescence and the mechanism of their multiple color emission. *Lab Chip.* 2019;19:3974–8. <https://doi.org/10.1039/C9LC00683D>.
56. Qiu H, Wu X, Hong R, et al. Microfluidic-oriented synthesis of graphene oxide nanosheets toward high energy density supercapacitors. *Energy Fuel.* 2020;34:11519–26. <https://doi.org/10.1021/acs.energyfuels.0c02143>.
57. Wu G, Tan P, Wu X, et al. High-performance wearable micro-supercapacitors based on microfluidic-directed nitrogen-doped graphene fiber electrodes. *Adv Funct Mater.* 2017;27:1702493. <https://doi.org/10.1002/ADFM.201702493>.
58. Kong B, Liu R, Guo J, et al. Tailoring micro/nano-fibers for biomedical applications. *Bioact Mater.* 2023;19:328–47. <https://doi.org/10.1016/j.bioactmat.2022.04.016>.
59. Niculescu A-G, Chircov C, Bîrcă AC, Grumezescu AM. Nanomaterials synthesis through microfluidic methods: an updated overview. *Nanomaterials.* 2021;11. <https://doi.org/10.3390/nano11040864>.
60. Li G, Cheng R, Cheng H, et al. Microfluidic synthesis of robust carbon dots-functionalized photonic crystals. *Chem Eng J.* 2021;405:126539. <https://doi.org/10.1016/J.CEJ.2020.126539>.
61. Li GX, Li Q, Cheng R, Chen S. Synthesis of quantum dots based on microfluidic technology. *Curr Opin Chem Eng.* 2020;29:34–41. <https://doi.org/10.1016/j.coche.2020.02.005>.
62. Jin Z, Jiang X, Dai Z, et al. Continuous synthesis of nanodroplet-templated, N-doped microporous carbon spheres in microfluidic system for CO₂ capture. *ACS Appl Mater Interfaces.* 2020;12:52571–80. <https://doi.org/10.1021/acsami.0c14044>.
63. Guo J, Yu Y, Sun L, et al. Bio-inspired multicomponent carbon nanotube microfibers from microfluidics for supercapacitor. *Chem Eng J.* 2020;397:125517. <https://doi.org/10.1016/j.cej.2020.125517>.
64. Mansuriya BD, Altintas Z. Carbon dots: classification, properties, synthesis, characterization, and applications in health care—an updated review (2018–2021). *Nanomaterials.* 2021;11:2525. <https://doi.org/10.3390/NANO11102525>.
65. Liu Z, Robinson JT, Tabakman SM, et al. Carbon materials for drug delivery & cancer therapy. *Mater Today.* 2011;14:316–23. [https://doi.org/10.1016/S1369-7021\(11\)70161-4](https://doi.org/10.1016/S1369-7021(11)70161-4).
66. Guo Y, Wei X, Gao S, et al. Recent advances in carbon material-based multifunctional sensors and their applications in electronic skin systems. *Adv Funct Mater.* 2021;31:2104288. <https://doi.org/10.1002/ADFM.202104288>.
67. Zhang J, Shi J, Zhang H, et al. Localized fluorescent imaging of multiple proteins on individual extracellular vesicles using rolling circle amplification for cancer diagnosis. *J Extracell Vesicles.* 2020;10:e12025.
68. Castro HPS, Souza VS, Scholten JD, et al. Synthesis and characterisation of fluorescent carbon nanodots produced in ionic liquids by laser ablation. *Chemistry.* 2016;22:138–43. <https://doi.org/10.1002/CHEM.201503286>.
69. Kamura Y, Imura K. Space-selective fabrication of light-emitting carbon dots in polymer films using electron-beam-induced chemical reactions. *ACS Omega.* 2019;4:3380–4. https://doi.org/10.1021/ACSOMEGA.9B00085/ASSET/IMAGES/LARGE/AO-2019-00085U_0006.JPEG.
70. Xu J, Sahu S, Cao L, et al. Carbon nanoparticles as chromophores for photon harvesting and photoconversion. *ChemPhysChem.* 2011;12:3604–8. <https://doi.org/10.1002/CPHC.201100640>.

71. Zhu W, Feng X, Zhao M, et al. Scalable and atom economic preparation of red-near-infrared emitted N-doped graphene quantum dots with a high quantum yield. *Diam Relat Mater.* 2021;116:108395. <https://doi.org/10.1016/J.DIAMOND.2021.108395>.
72. Kang K-K, Lee B, Lee C-S. Recent progress in the synthesis of inorganic particulate materials using microfluidics. *J Taiwan Inst Chem Eng.* 2019;98:2–19. <https://doi.org/10.1016/j.jtice.2018.08.027>.
73. Sun YP, Zhou B, Lin Y, et al. Quantum-sized carbon dots for bright and colorful photoluminescence. *J Am Chem Soc.* 2006;128:7756–7. https://doi.org/10.1021/JA062677D/SUPPL_FILE/JA062677DSI20060417_112943.PDF.
74. Murray CB, Norris DJ, Bawendi MG. Synthesis and characterization of nearly monodisperse CdE (E = S, Se, Te) semiconductor nanocrystallites. *J Am Chem Soc.* 1993;115:8706–15. https://doi.org/10.1021/JA00072A025/ASSET/JA00072A025.FP.PNG_V03.
75. Yuan F, Li S, Fan Z, et al. Shining carbon dots: synthesis and biomedical and optoelectronic applications. *Nano Today.* 2016;11:565–86. <https://doi.org/10.1016/J.NANTOD.2016.08.006>.
76. Sagdic K, Eş I, Sitti M, Inci F. Smart materials: rational design in biosystems via artificial intelligence. *Trends Biotechnol.* 2022;40:987–1003. <https://doi.org/10.1016/j.tibtech.2022.01.005>.
77. Li H, Yan X, Kong D, et al. Recent advances in carbon dots for bioimaging applications. *Nanoscale Horizons.* 2020;5:218–34. <https://doi.org/10.1039/C9NH00476A>.
78. Mohebbichamkhorami F, Faizi M, Mahmoudifard M, et al. Microfluidic synthesis of ultrasmall chitosan/graphene quantum dots particles for intranasal delivery in Alzheimer's disease treatment. *Small.* 2023;19:2207626. <https://doi.org/10.1002/SMLL.202207626>.
79. Feng T, Tao S, Yue D, et al. Recent advances in energy conversion applications of carbon dots: from optoelectronic devices to electrocatalysis. *Small.* 2020;16:2001295. <https://doi.org/10.1002/SMLL.202001295>.
80. Stepanidenko EA, Ushakova EV, Fedorov AV, Rogach AL. Applications of carbon dots in optoelectronics. *Nanomaterials.* 2021;11:364. <https://doi.org/10.3390/NANO11020364>.
81. Chen L, Wang CF, Liu C, Chen S. Facile access to fabricate carbon dots and perspective of large-scale applications. *Small.* 2023;19:1–24. <https://doi.org/10.1002/sml.202206671>.
82. Lu Y, Zhang L, Lin H. The use of a microreactor for rapid screening of the reaction conditions and investigation of the photoluminescence mechanism of carbon dots. *Chem—Eur J.* 2014;20:4246–50. <https://doi.org/10.1002/CHEM.201304358>.
83. Rao L, Tang Y, Li Z, et al. Efficient synthesis of highly fluorescent carbon dots by microreactor method and their application in Fe³⁺ ion detection. *Mater Sci Eng C.* 2017;81:213–23. <https://doi.org/10.1016/J.MSEC.2017.07.046>.
84. Tang Y, Rao L, Li Z, et al. Rapid synthesis of highly photoluminescent nitrogen-doped carbon quantum dots via a microreactor with foamy copper for the detection of Hg²⁺ ions. *Sensors Actuators B Chem.* 2018;258:637–47. <https://doi.org/10.1016/J.SNB.2017.11.140>.
85. Qiu H, Cheng H, Meng J, et al. Magnetothermal microfluidic-assisted hierarchical microfibers for ultrahigh-energy-density supercapacitors. *Angew Chemie - Int Ed.* 2020;59:7934–43. <https://doi.org/10.1002/anie.202000951>.
86. De Pedro SG, Salinas-Castillo A, Ariza-Avidad M, et al. Microsystem-assisted synthesis of carbon dots with fluorescent and colorimetric properties for pH detection. *Nanoscale.* 2014;6:6018–24. <https://doi.org/10.1039/C4NR00573B>.
87. Li G, Liu C, Zhang X, et al. Low temperature synthesis of carbon dots in microfluidic chip and their application for sensing cefquinome residues in milk. *Biosens Bioelectron.* 2023;228:115187. <https://doi.org/10.1016/j.bios.2023.115187>.
88. Gerstner E. Nobel Prize 2010: Andre Geim & Konstantin Novoselov. *Nat Phys.* 2010;6:836. <https://doi.org/10.1038/nphys1836>.
89. Craciun MF, Russo S, Yamamoto M, Tarucha S. Tuneable electronic properties in graphene. *Nano Today.* 2011;6:42–60. <https://doi.org/10.1016/J.NANTOD.2010.12.001>.
90. Morales-Torres S, Jiříčková A, Jankovský O, et al. Synthesis and applications of graphene oxide. *Materials.* 2022;15:920. <https://doi.org/10.3390/MA15030920>.

91. Tarcan R, Todor-Boer O, Petrovai I, et al. Reduced graphene oxide today. *J Mater Chem C*. 2020;8:1198–224. <https://doi.org/10.1039/c9tc04916a>.
92. Hummers WS, Offeman RE. Preparation of graphitic oxide. *J Am Chem Soc*. 1958;80:1339. https://doi.org/10.1021/JA01539A017/ASSET/JA01539A017.FP.PNG_V03.
93. Cao J, He P, Mohammed MA, et al. Two-step electrochemical intercalation and oxidation of graphite for the mass production of graphene oxide. *J Am Chem Soc*. 2017;139:17446–56. https://doi.org/10.1021/JACS.7B08515/ASSET/IMAGES/LARGE/JA-2017-08515D_0008.JPEG.
94. Jafari Z, Rad AS, Baharfar R, et al. Synthesis and application of chitosan/tripolyphosphate/graphene oxide hydrogel as a new drug delivery system for Sumatriptan Succinate. *J Mol Liq*. 2020;315:113835. <https://doi.org/10.1016/J.MOLLIQ.2020.113835>.
95. Qi Y, Yang M, Xu W, et al. Natural polysaccharides-modified graphene oxide for adsorption of organic dyes from aqueous solutions. *J Colloid Interface Sci*. 2017;486:84–96. <https://doi.org/10.1016/J.JCIS.2016.09.058>.
96. Sun P, Zhu M, Wang K, et al. Selective ion penetration of graphene oxide membranes. *ACS Nano*. 2013;7:428–37. https://doi.org/10.1021/NN304471W/SUPPL_FILE/NN304471W_SI_001.PDF.
97. Gong K, Pan Z, Korayem AH, et al. Reinforcing effects of graphene oxide on portland cement paste. *J Mater Civ Eng*. 2014;27:A4014010. [https://doi.org/10.1061/\(ASCE\)MT.1943-5533.0001125](https://doi.org/10.1061/(ASCE)MT.1943-5533.0001125).
98. Lundie M, Šljivančanin Ž, Tomić S. Electronic and optical properties of reduced graphene oxide. *J Mater Chem C*. 2015;3:7632–41. <https://doi.org/10.1039/C5TC00437C>.
99. Zhao C, Fan J, Chen D, et al. Microfluidics-generated graphene oxide microspheres and their application to removal of perfluorooctane sulfonate from polluted water. 2016. <https://doi.org/10.1007/s12274-015-0968-7>.
100. Wang R, Guo W, Li X, et al. Microfluidic generation of 3D graphene microspheres for high-efficiency adsorption. *J Mater Sci*. 2017;52:13930–9. <https://doi.org/10.1007/S10853-017-1487-6/FIGURES/7>.
101. Abedini-Nassab R, Pouryosef Miandoab M, Şaşmaz M. Microfluidic synthesis, control, and sensing of magnetic nanoparticles: a review. *Micromachines*. 2021;12:768. <https://doi.org/10.3390/mi12070768>.
102. Wang J, Song Y. Microfluidic synthesis of nanohybrids. *Small*. 2017;13:1604084. <https://doi.org/10.1002/SMLL.201604084>.
103. Manay E, Sahin B. The effect of microchannel height on performance of nanofluids. *Int J Heat Mass Transf*. 2016;95:307–20. <https://doi.org/10.1016/J.IJHEATMASSTRANSFER.2015.12.015>.
104. Xia C, Zhu S, Feng T, et al. Evolution and synthesis of carbon dots: from carbon dots to carbonized polymer dots. *Adv Sci*. 2019;6:1901316. <https://doi.org/10.1002/ADVS.201901316>.
105. Wojnicki M, Hessel V. Quantum materials made in microfluidics - critical review and perspective. *Chem Eng J*. 2022;438:135616. <https://doi.org/10.1016/j.cej.2022.135616>.
106. Ge H, Xu H, Lu T, et al. Microfluidic production of porous carbon spheres with tunable size and pores. *J Colloid Interface Sci*. 2016;461:168–72. <https://doi.org/10.1016/j.jcis.2015.09.018>.
107. Agha A, Waheed W, Stiharu I, et al. A review on microfluidic-assisted nanoparticle synthesis, and their applications using multiscale simulation methods. *Discov Nano*. 2023;18:18. <https://doi.org/10.1186/s11671-023-03792-x>.
108. Zhao X, Chen H, Xiao Y, et al. Sharp-edge-driven spiral acoustic micromixers for functional nanoarray engineering. *Mater Today Nano*. 2023;22:100338. <https://doi.org/10.1016/j.mtnano.2023.100338>.
109. Hong SO, Park K-S, Kim D-Y, et al. Gear-shaped micromixer for synthesis of silica particles utilizing inertio-elastic flow instability. *Lab Chip*. 2021;21:513–20. <https://doi.org/10.1039/D0LC00834F>.