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Microfluidics-enabled intelligent manufacturing of metal halide perovskite nanocrystals

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ABSTRACT

Large-scale and controllable fabrication is an indispensable step for the industrialization and commercialization of halide perovskite nanocrystals, which are new-generation semiconductor materials for optoelectronic applications. Microfluidics, which provides continuous and precise synthesis, has been considered as a promising technique to fulfill this aspect. The research studies over the past decades have witnessed the advancement of microfluidics as a powerful tool in the fabrication of halide perovskite nanocrystals. In this Perspective, the state-of-the-art research based on microfluidics is introduced initially, including the synthesis of functional structures and materials, devices, as well as the interdisciplinary interactions between microfluidics and artificial intelligence and machine learning, etc. We then detail the issues and challenges in hindering progress in the above areas. Finally, we provide future directions and trends for the technology to achieve its full potential. This Perspective is expected to benefit the collective efforts between the field of nanomaterials and microfluidics in advanced manufacturing.

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INTRODUCTION

As new-generation semiconductor materials, metal halide perovskite nanocrystals (PeNCs), which are also referred to as quantum dots, with the characteristic dimension being commensurate with (or smaller than) the Bohr radius of exciton of the counterpart bulk materials have attracted worldwide attention over the past decades. PeNCs with a chemical formula of ABX_3 , where A can be methylammonium (MA: $CH_3NH_3^+$), formamidinium [FA: $CH_3(NH_2)_2^+$], inorganic cations (e.g., Cs^+), and B and X represent bivalent metal ions (e.g., Pb^{2+} and Sn^{2+}) and halide ions (Cl^- , Br^- , and I^-), have manifested massively promising applications in optoelectronics, especially including lighting and display (Fig. 1),¹ owing to their overwhelming merits over conventional semiconductor materials. The optical properties such as photoluminescence quantum yields (PLQY) of conventional group III–V nanocrystals (NCs) (e.g., InP and InAs) and I–III–VI NCs (e.g., $CuInS_2$) are far behind the standard for commercial use. The vacuum condition and high temperature required in the chemical reaction render a high cost in the manufacturing of III–V NCs. Although group II–VI NCs (e.g., CdSe, CdS, and CdTe) possess high PLQYs suitable for commercialization, the expensive and high-cost

stringent conditions for the synthesis of materials with core-shell structures hinder their progress. On top of that, the toxic cadmium (Cd)-based NCs jeopardize the ecological environment and human health, further fading their competition in the world of NCs. In contrast, PeNCs are more mitigatory over conventional NCs in environmental impact. The amount of Cd is restricted to 100 ppm in electronic products while 1000 ppm for Pb, following the laws of restriction of hazardous substances (RoHS) enacted by the European Union.¹ On the other hand, the facile tunability of the size of PeNCs enables a wide range of emission/absorption bands covering entire visible spectra, which is beneficial for the development of optical-electronic-conversion devices. In addition, high color purity and light absorption coefficient with high defect tolerance engender high brightness with a narrow full width at half maximum (FWHM) around 12–40 nm.^{2,3} Moreover, solution processability at low temperatures contributes to a cost-effective approach in the synthesis of PeNCs. All these advantages enable PeNCs to be dominant materials in optoelectronic fields including lighting,^{4,5} display,^{1,6} and solar cells.⁷

The manufacturing methods are critically important for the advancements and applications of materials. Up to now, hot

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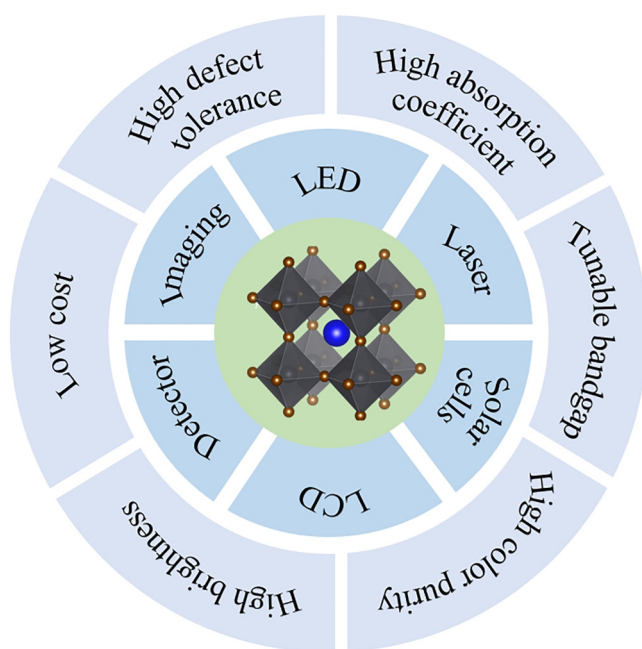


FIG. 1. PeNCs feature low cost, tunable optical, and electrical properties. Their wide applications span energy conversion, illumination, displays, detectors, and imaging.

injection (HI)² and antisolvent⁸ or so-called ligand-assisted reprecipitation (LARP)⁹ are the most common methods used for the preparation of PeNCs. However, the PeNCs synthesized by the two methods are limited to small volumes with a mode of a single batch. Such a batch mode suffers from the irreproducibility and large distribution of sizes for the synthesis of NCs from batch to batch, impeding the advancement of the materials toward advanced manufacturing for industrialization.

Microfluidics with “continuous flow reactors” is a promising pathway to solve these challenges. On the one hand, the continuous flow reactors enable a continuous-massive fabrication of materials, during which the size and morphology of the materials can be tuned through the heaters and flow-control components integrated into a microfluidic system. The rapid mixing of reagents occurring in the microchannels with large surface-area-to-volume ratios results in rapid transfers of heat and mass in fluid. The small-instantaneous fluid volumes in microreactors ensure rapid and efficient chemical reactions that can be controllable precisely, contributing to the continuous formation of NCs with a narrow distribution of sizes.¹⁰ On the other hand, the compatibility between online parametric screening and microchannels allows for a precise feedback system which ensures that the manufacturing process is controllable and efficient.^{11–13} Therefore, microfluidics offers the feasibility of highly controlled batch-to-batch manufacturing on a massive scale. With a rapid development, a growing number of technologies, such as inkjet printing¹⁴ and fiber-spinning chemistry,¹⁵ have been integrated with microfluidics in recent years to achieve new-generation functional

materials. This remarkable progress suggests that a sophisticated and intelligent manufacturing is coming.

In this Perspective, we briefly review recent advances in microfluidics in fabricating PeNCs and then showcase some typical and important applications of microfluidic systems in the interdisciplinary fields between functional materials and advanced manufacturing. Furthermore, we discuss issues that currently hinder progress in microfluidic manufacturing and finally provide an outlook for research opportunities in this technology.

RECENT ADVANCES IN THE FABRICATION OF PeNCs BY MICROFLUIDICS

Microfluidics evolved at the end of 20th century and has been applied in four aspects: microanalysis in chemistry and biochemistry, military defense, molecular biology, and microelectronics.^{16,17} After a rapid development, it has been evolved as a new fabrication technology in biomedicines, chemistry, and materials, especially, nanoparticles and nanocrystals. Recently, PeNCs as new-generation optoelectronic materials for lighting and display have attracted wide attention. Microfluidic technology, offering fabrication approaches with dominant merits over other methods, has been considered as the most promising channel in reaching industrialization and commercialization.

Metal halide perovskite nanocrystals (MHPNs) synthesized by microfluidics were investigated initially by Lignos *et al.*¹⁸ In their research, a microfluidic platform was designed for the synthesis of CsPbX₃ (X = Cl, Br, and I) perovskite NCs, in which an online optical monitoring system was integrated for the real-time characterization. The quality of the NCs characterized by the online monitoring system is used to activate and control the chemical payload (such as the fractions of the lead and cesium sources) for the parameter optimization during the synthesis of the NCs. Three years later, Abdel-Latif *et al.*¹⁹ proposed a microfluidic apparatus as a “quantum dots changer” by anion exchange. Except for the monitoring strategy of absorption and photoluminescence (PL), they constructed a micromixer module to achieve the premixing of precursor streams, which avoided unwanted inhomogeneous reactions in the moving precursor slug [Fig. 2(a)]. The experimental results show that the FWHM of the PL spectra of CsPbBr₃ QDs during the anion exchange experienced a decrease first and reached a plateau late with increasing the flow rate of the precursor solution, indicating a mass-transfer limit for the reactions of anion exchange.¹⁹

Other than the inorganic perovskite CsPbX₃ NCs, hybrid organic perovskite FAPbX₃ (X = Br and I) NCs were also synthesized by microfluidics, as reported by Maceiczky *et al.*¹² Two precursor solutions used to form hybrid organic perovskite nanocrystals were delivered into a microfluidic reactor individually with syringe pumps precisely. The flow rates of the precursor solutions in a capillary were adjusted for the stoichiometric control of the reaction solution. The capillary was set up on a temperature-controlled heating apparatus to allow precise control of the reaction temperature. They found that the PL spectra of the specimens exhibited mono peak characteristics when the ratio of Pb:I was 1:2 while decreasing the FA concentration led to the presence of a shoulder in the PL spectra [Fig. 2(b)]. For the ratio of Pb:I being

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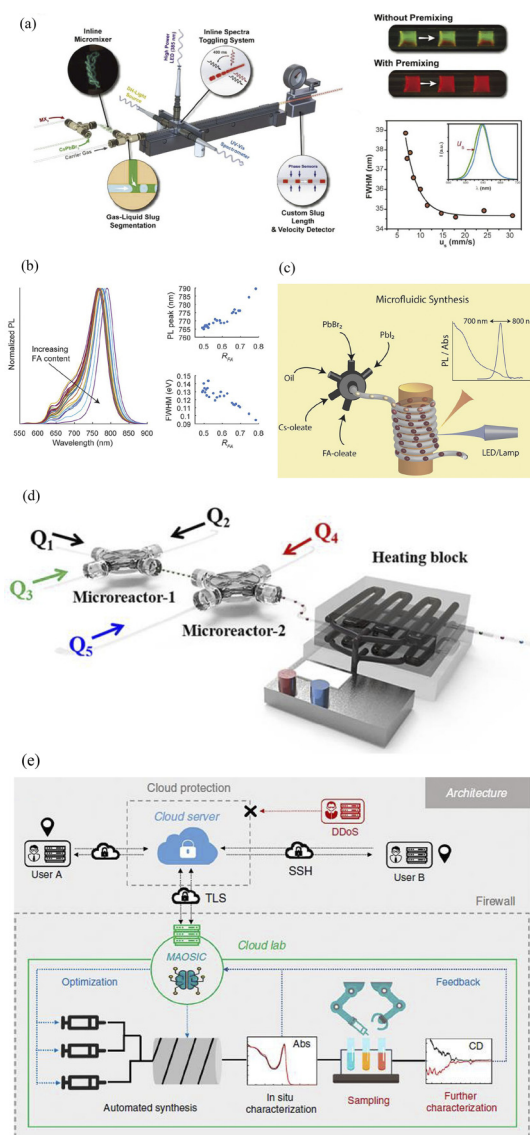


FIG. 2. (a) Microfluidic apparatus as a “quantum dots changer” achieved by anion exchange with a micromixer module for the premixing of the precursor solution. Reproduced with permission from Abdel-Latif *et al.*, *Adv. Funct. Mater.* **29**, 1900712 (2019). Copyright 2019 John Wiley and Sons. (b) FA-content dependent PL spectra of FAPbI₃ NCs with the corresponding PL peaks (upper right) and FWHM (lower right). Reproduced with permission from Maceiczky *et al.*, *Chem. Mater.* **29**, 8433–8439 (2017). Copyright 2017 American Chemical Society. (c) Schematic of microfluidics with a seven-port manifold for the synthesis of the multinary model Cs_xFA_{1-x}Pb(Br_{1-y}I_y)₃ PeNCs. Reproduced with permission from Lignos *et al.*, *ACS Nano* **12**, 5504–5517 (2018). Copyright 2018 American Chemical Society. (d) Double-microreactors platform and heating system for the preparation of CsPbX₃ and CsPb(X/Y)₃ (X = Br, Y = Cl and I) PeNCs. Reproduced with permission from Kang *et al.*, *Chem. Eng. J.* **384**, 123316 (2020). Copyright 2020 Elsevier. (e) Workflow of the cloud lab for discovering new optically active PeNCs. Reproduced with permission from Li *et al.*, *Nat. Commun.* **11**, 2046 (2020). Copyright 2020 Author(s), licensed under the Creative Common Attribution (CC BY) license.

1:1, 3:1, and 1:3, multiple peaks are present in the PL spectra. Several months later, Lignos *et al.*²⁰ reported the formation of hybrid organic perovskite FAPb(Cl_{1-x}Br_x)₃ NCs in a microfluidic system with online parametric screening and offline characterization of microstructures. The NCs would evolve to nanoplates when the ratio of FA:Pb exceeds 1.8 or the ratio of oleic acid (OA):oleylamine (OLA) is 10:1.

With the progress in the microfluidic technology, multinary (mixed-cation and mixed-halide) PeNCs have been successfully synthesized and emerged as novel perovskites with enhanced stability over unmixed A-site mixed-halide.^{21–23} The mixed halides play a role in the extension of the bandgap scale. A typical multinary model Cs_xFA_{1-x}Pb(Br_{1-y}I_y)₃ PeNCs synthesized by microfluidics was reported by Lignos *et al.*¹¹ A seven-port manifold with individual capillary in each port was employed for carrying precursor solutions, which allows for independent variations of precursor molar ratios (e.g., Cs/FA, Cs/Pb, FA/Pb, and Br/I), and the control of growth parameters such as growth time and temperature [Fig. 2(c)]. The setup also enables an interdependent and efficient mixing of precursor solutions, thus ensuring unique characteristics of the as-synthesized specimens.

It is worth mentioning that these online monitoring designs have accelerated the development of integrated microfluidics systems. Kang *et al.*²⁴ developed an integrated double-microreactors platform and a heating system for the preparation of CsPbX₃ and CsPb(X/Y)₃ (X = Br, Y = Cl and I) PeNCs [Fig. 2(d)]. The first microreactor was used to form dispersed emulsion reactors first while the second microreactor served as the channels for the exchange of anion ions. The heating system provided the control of temperature for the synthesis of PeNCs and was used for the nucleation and growth of NCs. Recently, Geng *et al.*²⁵ formed zirconium (Zr)-based lead-free Cs₂ZrX₆ (X = Cl and Br) double perovskite NCs via a microfluidic approach and studied the influences of the flow rate of precursor solutions and ligand ratio on the morphology and optical characteristics of the NCs. Their study showed the possibility of fabricating lead-free double perovskite NCs by microfluidics and a step forward for the development in the fields of lead-free double perovskites and microfluidics.

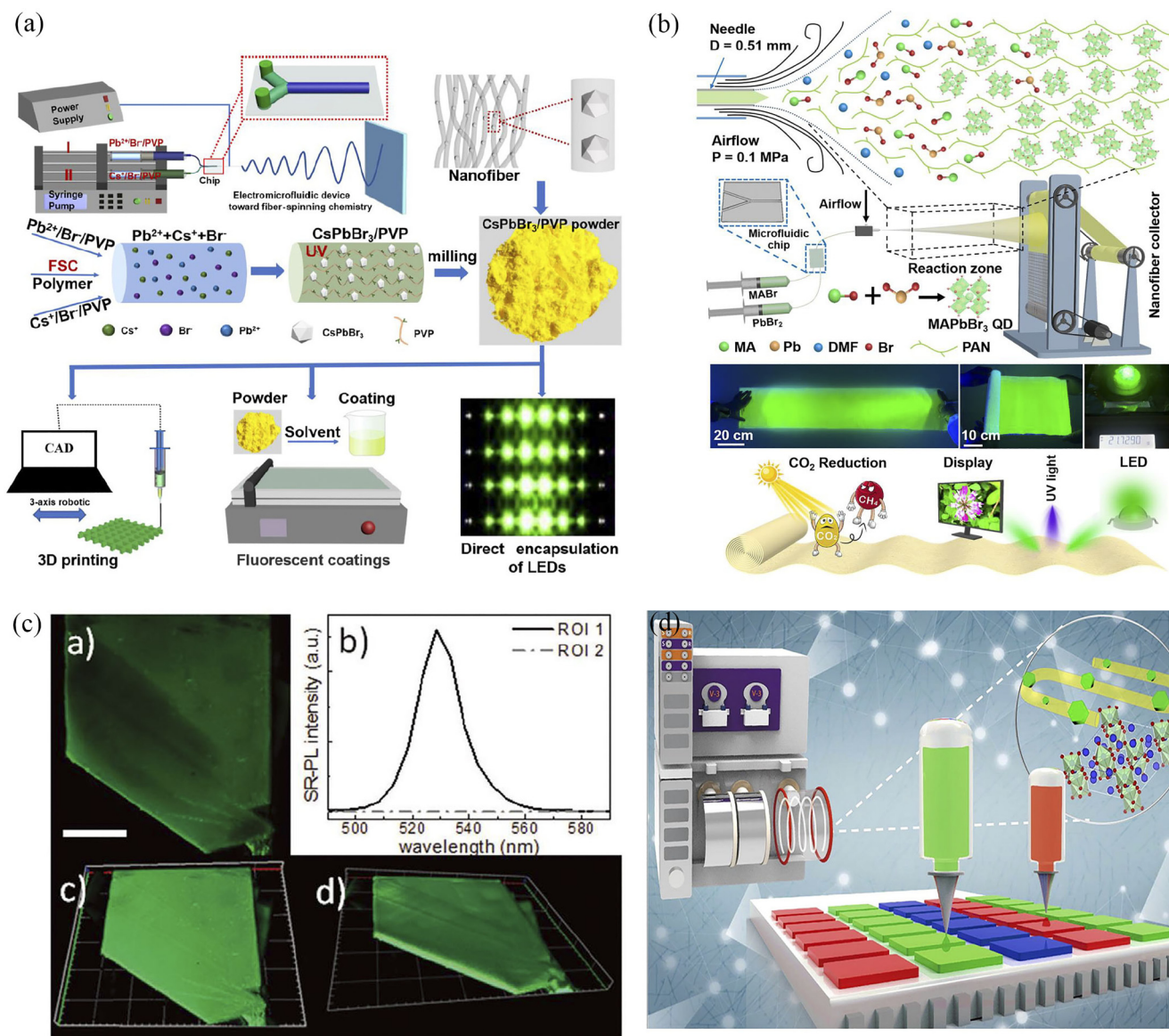
The past years also witnessed a rapid development of artificial intelligence (AI), which provides a powerful and versatile path for high throughput data collection, target prediction, automated operation, etc. It is a foreseen bright manufacturing field linking AI and microfluidics, and researchers have initialized this three years ago. Li *et al.*²⁶ devised an intelligent cloud lab for the chirality detection of CsPbBr₃ perovskites [Fig. 2(e)]. Using this intelligent platform, the synthesis, characterization, and parameter optimization were autonomously obtained, which enables on-demand experimental designs for remote users. The platform also offered a detection component for the investigation of inter-structure and infrastructure-induced mechanisms. This intelligent cloud lab opens an efficient and reliable online channel for the access of global collaborations to extend the scope of materials (e.g., biosensing, chiral catalysis, and chiral photonics) and to discover new materials. Moreover, some other emerging techniques such as machine learning²⁷ and programmable synthesis²⁸ were incorporated with microfluidics as well, which presents proof-of-concept experiments for the integration of automation and microfluidics.

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APPLICATIONS

Given the overwhelming merits of high throughput and reproducibility of using microfluidics in the fabrication of PeNCs, a variety of manufacturing technologies has been incorporated into microfluidics toward advanced applications for industrialization

and commercialization in recent years. As a cutting-edge application, microfluidics can be integrated with a fiber-spinning system. Dong *et al.*¹⁵ reported *in situ* synthesis of polyvinylpyrrolidone-based perovskite nanocrystals by the fiber-spinning chemistry (FSC) method on a microfluidic platform [Fig. 3(a)]. Subsequently,



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FIG. 3. (a) Schematic for the synthesis of CsPbBr_3 powders by the FSC technique for the applications in 3D printing, coatings, and LED. Reproduced with permission from Dong *et al.*, ACS Appl. Mater. Interfaces **13**, 39748–39754 (2021). Copyright 2021 American Chemical Society. (b) Schematic of the reaction mechanism for *in situ* formation of MAPbBr_3 nanocrystals in nanofiber films by microfluidic-blow-spinning method and the potential applications. Reproduced with permission from Cheng *et al.*, Angew. Chem. Int. Ed. **61**, e202204371 (2022). Copyright 2022 John Wiley and Sons. (c) Fluorescent images of MAPbBr_3 single crystals by microfluidics-assisted technique on a patterned ITO substrate. Reproduced with permission from Viola *et al.*, Adv. Mater. Technol. **8**, 2300023 (2023). Copyright 2023 John Wiley and Sons. (d) Microfluidic synthesis of $\text{CsPbBr}_3/\text{Cs}_4\text{PbBr}_6$ nanocrystals for inkjet printing of mini-LEDs. Reproduced with permission from Bao *et al.*, Chem. Eng. J. **426**, 130849 (2021). Copyright 2021 Elsevier.

applications for 3D printing, direct encapsulants for light-emitting diodes, and fluorescent coatings were explored owing to the excellent fluorescent characteristics of the as-prepared specimens. During the 3D printing process, a mixture of CsPbBr₃ and PVP-ethanol was used as an ink, enabling the successful formation of a series of fluorescent patterns and the large-area uniform films with a size of 45 × 18 cm². Cheng *et al.*²⁹ used the same microfluidic-blow-spinning-based FSC technique for the manufacturing of perovskite nanocrystal-based nanofiber films [Fig. 3(b)]. The films, which are composed of MAPbBr₃ nanocrystals and polymer, possess an absolute PLQY of 71% and a green-emissive PL centered at 527 nm with a FWHM of 23 nm. Thanks to the protection of the polymer matrix, the as-obtained fiber films exhibited long-term stability in water over 60 days. The films also had good blue light emission and thermal stabilities. This work foresees the potential applications in a variety of areas, including display and photocatalytic CO₂ reduction.

As a prospective field in biomedicine, microfluidics has never stopped advancing, and its combination with perovskite nanocrystals has been endlessly studied in the related field. Bian *et al.*³⁰ synthesized perovskite nanocrystals-encapsulated multi-colored barcode particles through a microfluidic technique with seven-channel capillary tubes. The barcode particles have potential applications in biomolecular fields of multiplex encoding and assays, such as tumor screening. Sanjayan *et al.*³¹ reported a paper-analytical device from CsPbBr₃ nanocrystals, which was used as fluorometric detection of lung cancer biomarkers concurrently for carcinoembryonic antigen (CEA) and neuron specific enolase (NSE). The operation of such a device was motivated by the high sensitivity and multicolor imaging ability offered by perovskite CsPbBr₃ and CsPbI₃ nanocrystals. Such a device is expected to be a promising diagnostic tool for the detection of other proteins or biomarkers for research in food safety. In addition, microfluidics has been used in the fabrication of single crystals for photodetection [Fig. 3(c)], demonstrating a wide span for materials synthesis and optoelectronic applications.³²

In 2021, Bao *et al.*¹⁴ fabricated CsPbBr₃/Cs₄PbBr₆ NCs via a microfluidic platform for the inkjet printing of mini-LEDs (light-emitting diodes). The NCs were synthesized first and then used as an ink material for the printing. Their work combined the concepts of microfluidics and inkjet printing [Fig. 3(d)]. As a similar approach, Guo *et al.*³³ synthesized CsPbI₃/SiO₂ nanocomposites on a microfluidic platform, which solved the issue of large-scale continuous production of core-shell PeNCs with excellent chemical and thermal stability attributed to the mesoporous silica coated on the surface of the core of CsPbI₃. The work revealed that the water resistance property and thermal stability of the CsPbI₃/SiO₂ specimens exhibited twofold and threefold improvement over the one without the shell layer of mesoporous silica. This also suggests the successful fabrication of core-shell structured NCs via microfluidics. As a result, CsPbI₃/SiO₂ based LEDs exhibited a superior optical stability over the one without the shell layer.

CHALLENGES AND OUTLOOK

Further technological advancement of microfluidics will require continued optimization of multiple aspects, including the

synthesis of PeNCs, microfluidic technique, integration between real-time characterization and microfluidic platforms, and intellectualization of microfluidic platforms.

From the perspective of synthesis, the critically important and fundamental aspects of the devices and the methods for the synthesis of PeNCs in microfluidics are of emergency to be upgraded from the point of sustainability, though tremendous progress has been made in microfluidics for the fabrication of PeNCs in the past few years. Up to now, all the methods for lead-based PeNCs by microfluidics are based on the solutions containing toxic organic solvents,^{12,18,19,34} which pose a threat to the health of human beings and the environment and hinder the progress of industrialization of the materials. The green-route methods are calling for the development of microfluidics in terms of their use in chemistry and materials. On the other hand, it is still challenging for the synthesis of lead-free PeNCs by microfluidics. Tang *et al.*³⁵ demonstrated the green-route synthesis of Sn-based Cs₂SnCl₆ PeNCs through a microfluidic platform. However, microfluidics was used in this method for the first step—the synthesis of Cs₂SnCl₆ powder. The second step for the synthesis of Cs₂SnCl₆ NCs requires further baking and ultrasonication, which is time-consuming and energy-consuming from the point of industrialization. It is worth mentioning that a controlled growth of lead-free cesium zirconium (Zr) halide double PeNCs through a microfluidic channel was reported by Abdel-Latif *et al.* recently.²⁵ Nevertheless, the method is also called hot injection and is similar to the one for the synthesis of group II–VI NCs, which still involves the use of toxic solvents such as ODE (1-octadecene).

It needs to be pointed out that green solvents have been investigated as a hot spot in the conventional antisolvent method in which green solvents are used for the preparation of precursor solutions. For example, Zheng *et al.*³⁶ synthesized CsPbBr₃ PeNCs with water and EtOH. This method is the closest one to a green method but not a green method, because EtOH is not totally called green solvent. Also, this reported method was achieved through a conventional antisolvent method instead of microfluidics. Water is a non-toxic green solvent. However, it remains a challenge to fabricate PeNCs by water on a microfluidic platform. In this regard, the sustainable fabrication of PeNCs by microfluidics needs more efforts to be devoted.

From the perspective of functionalization, analysis channels for the *in situ* characterization of physical and chemical properties of materials on a microfluidic platform are of no doubt for the facilitation of the function of microfluidics.^{11,18,20,37} Such analysis channels including online optical measurement systems (such as photoluminescence and absorption measurements) and online monitoring of reaction parameters (such as the reaction temperatures, and reaction times) offer a rigorous and rapid feedback of the product. According to the feedback, parameters will be adjusted and the optical properties of the product can be improved.^{11,18} Although integration of analysis channels including optical and structural properties have been implemented, these components are so large that are hard to be transferred.^{38,39} Miniature, portable analysis channels can be convenient for the characterization and quality test of materials and can be flexible in measurements to fulfill the potential of lab on a chip for microfluidics. However, the challengeable achievement for this is still under the way.

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On top of that, the limitations of the quality and practicality of PeNCs prepared through microfluidic methods should also be considered. First, the chemical stability of PeNCs is still not good enough for commercial use, which is attributed to the properties of the nanocrystals. Second, PeNCs-based LEDs have short lifetimes. It is a huge challenge to prolong the working lifetime of PeNCs-based LEDs. Third, PeNCs exhibit significant thermal quenching of PL characteristics, which limits the high-temperature applications (usually larger than 100 °C) of PeNCs-based devices and systems.

Certainly, microfluidics provides an efficient path for the large-scale synthesis of PeNCs with high-quality reproducibility, which is a major step to reach industrialization and commercialization. However, it is still challenging in the large-scale synthesis of PeNCs on a microfluidic platform. Currently, the microfluidic platforms used to synthesize PeNCs are based on the principle of either the hot injection or the antisolvent method. During the processing of PeNCs, two different precursor solutions are loaded into two pump systems, in which the precursor solutions will be injected and mixed. Therefore, the temperature rise likely occurs locally during the loading and mixing of the precursor solutions. Also, it is difficult to control the mono-dispersity of PeNCs produced by microfluidics during large-scale production, in which agglomeration of PeNCs becomes a serious issue. These are two major issues/concerns for the manufacturing of PeNCs via microfluidic systems. There are several potential solutions, which may address these issues. For the former, two pump systems connected with fluidic channels can be placed into a temperature chamber to enable reactions at a constant temperature. For the latter, proper surfactants and solvents need to be used to limit the agglomeration for the storage of PeNCs. Under such a scenario, the polarities of solvents are a key parameter determining the applicability of surfactants and PeNCs.

Several techniques have been incorporated into microfluidics to achieve multi-functions of microfluidics. Inkjet printing is of immense promise in the field of display and lighting due to its role in the efficient fabrication of LEDs. Wei *et al.*⁴⁰ recently reported the preparation of PeNCs-based LEDs by inkjet printing. However, there is still some distance for this approach to be industrialized as the ink is limited for the batch-to-batch fabrication of devices. In this case, microfluidics is expected to likely solve this issue and forward the advancement of the industrialization of LEDs by advanced manufacturing.

In terms of continuous manufacturing, another application of microfluidics can be focused on electrospinning or fiber-spinning chemistry. The batch-to-batch precursor solution or so-called ink can guarantee the up-scaling model of the specimen. However, it is difficult to obtain a high-rate throughput of fiber microreactors and the products for microfluidic spinning.²⁹ Hence, more efforts are needed for this aspect. With the development of AI, a highly integrated system for microfluidics is expected to meet the pace and requirement for the manufacturing of nanoparticles and nanocrystals. Therefore, computerized-intelligent synthesis or so-called programmable microfluidics will dominate microfluidic platforms including the fabrication of PeNCs, which are expected to offer a highly efficient and automated approach to fulfill the general and tailored designs.

CONCLUSIONS

The solution-processed feature for the synthesis of NCs offers an opportunity for microfluidics spanning from the biomedical field to chemical materials. PeNCs are considered as new-generation semiconductor materials for lighting and display, which allows microfluidics to be a significant platform for advanced manufacturing of materials and devices. To date, microfluidics has been employed to fabricate a variety of PeNCs and has demonstrated huge potential in continuous batch-to-batch fabrication, which, however, is young in integration although the components including optical and structure characterization have been incorporated into microfluidic platforms. Such advanced integrations include the heating system, synthesis channels, and detection zones, which will enable a portable and efficient tool for the manufacturing of materials. There is a great need calling for the integration of computerized-intelligent and programmable systems in microfluidic systems to provide a further precise control of the synthesis of PeNCs. This can help the development of AI-assisted microfluidic platforms to advance the commercialization and industrialization of microfluidics for the synthesis of PeNCs.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Xiaobing Tang: Conceptualization (supporting); Data curation (lead); Investigation (equal); Methodology (equal); Writing – original draft (lead). **Fuqian Yang:** Conceptualization (lead); Funding acquisition (lead); Investigation (equal); Methodology (equal); Project administration (lead); Resources (lead); Supervision (lead); Validation (equal); Writing – review & editing (lead).

DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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