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Advances in printing techniques for thermoelectric materials and devices

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Abstract

Thermoelectric materials and devices have garnered significant attention in recent years due to their potential for converting waste heat into usable electricity, opening new avenues for sustainable energy harvesting. As research in the field of thermoelectric materials and devices continues to grow, so does the need for efficient and scalable fabrication methods. Among various fabrication techniques, printing methods have emerged as promising approaches to producing thermoelectric materials and devices, offering advantages such as low cost, high throughput, and design flexibility. Here, we overview the recent advances in printing methods for the fabrication of thermoelectric materials and devices. We discuss the key principles, challenges, and opportunities associated with various printing techniques, including screen printing, inkjet printing, and 3D printing, with a focus on their applications in thermoelectric materials and devices. Furthermore, we highlight the progress made in optimizing the printing parameters, ink formulations, and post-processing methods to enhance the thermoelectric performance of printed materials and devices. Finally, we provide insights into the prospects and potential research directions in the field of printing methods for thermoelectric materials and devices. This review aims to provide a comprehensive overview of the state-of-the-art printing techniques for thermoelectric materials and devices and to serve as a reference for researchers and practitioners working in this rapidly growing field.

Keywords: Flexible thermoelectric generators, large-scale fabrication, printing technology, transport of charge carriers, phonon scatterings

INTRODUCTION

Thermoelectric materials, which can directly convert heat energy into electricity, have gained significant attention because of their potential for sustainable energy harvesting from waste heat sources[1-3]. With increasing concerns on energy efficiency and renewable energy sources, thermoelectric materials and devices have been explored for a wide range of applications, including waste heat recovery in industrial processes, automotive waste heat recovery, and powering wearable devices[4-6]. The performance of thermoelectric materials is determined by the dimensionless figure of merit, zT[7-9], expressed by $zT = S^2\sigma T/\kappa$, in which S is the Seebeck coefficient, σ is the electrical conductivity, and κ is the thermal conductivity [comprising electron (κ_e) and lattice (κ_l) thermal conductivities][10-12].

Significant progress has been achieved to enhance zT through increasing $S^2\sigma$ and decreasing κ_l. Since S, σ, and κ_e are coupled via the carrier concentration (n_H), optimising n_H is the prerequisite to maximise zT[13]. Electrically, to increase $S^2\sigma$, band engineering to align multiple band valleys has been established to ensure a large band degeneracy (N_v)[14,15]. Improving the carrier mobility (μ_H) is also a method to increase $S^2\sigma$[3,16-18]. Thus, engineering light bands to reduce the band effective mass can effectively enhance thermoelectric performance. Other strategies that have also been proven to elevate $S^2\sigma$ include introducing resonant energy levels into the density of states (DOS), minority charge carrier filtering, and quantum confinement[19-21]. Thermally, κ_l can be decreased by enhancing phonon scatterings[22]. This includes strengthening inherent phonon-phonon interactions and introducing external scattering centres[23-26]. To identify the intrinsically strong phonon-phonon interactions, the acoustic and optical phonon branches should be overlapped in frequency to allow sufficient scattering channels[27-29]. The slope of acoustic branches should be as low as possible to ensure a small phonon group velocity[28,30,31]. Further reinforcing phonon scatterings is realised by introducing various types of nanostructures and lattice imperfections[32-34].

Additionally, the fabrication technique used to create thermoelectric materials and devices plays a crucial role in determining their performance[35-38]. Bulk processing techniques involve the synthesis of thermoelectric materials in a solid-state form, such as solid-state reaction, ball milling, or melt processing, followed by shaping and consolidation processes to obtain bulk materials with desired properties[39-42]. Vacuum deposition methods, such as sputtering and thermal evaporation, realize the deposition of thin films of thermoelectric materials onto substrates using vacuum chambers and specialized equipment[43-47].

While these traditional fabrication methods have been widely used in the field of thermoelectrics and have resulted in significant advances, they also have limitations[48]. For instance, bulk processing methods may have limitations in terms of achieving high-resolution patterning or complex geometries, and vacuum deposition methods may be costly and complex for large-scale production[49]. Additionally, these methods may require high processing temperatures or harsh conditions, which can limit the selection of materials and substrates[36].

To harness the full potential of thermoelectric materials and devices, efficient and scalable fabrication methods are crucial[50]. Among various fabrication techniques, printing methods have emerged as promising approaches for the production of thermoelectric materials and devices, offering advantages such as low cost, high throughput, and design flexibility[51-53].

Printing methods for thermoelectric materials and devices can be categorized into different types [Table 1], such as screen printing[54-57], extrusion printing[58-61], inkjet printing[62], aerosol jet printing[63], drop casting[64],

Table 1. Typical flexible thermoelectric materials synthesized by printing methods

Materials	Method	Additive	Treatment T (°C)	S (mV·K^{-1})	(S·cm^{-1})	S^2 (mW·m^{-1}·K^{-2})	Ref.
Sb_2Te_3	Screen printing	-	500 °C	98	1,500	1,441	[54]
Bi_2Te_3/PEDOT	Screen printing	dimethyl sulfoxide	450 °C	-138	73	138.6	[55]
$Sb_{1.6}Bi_{0.4}Te_3$/Te	Screen printing	α-Terpineol + Disperbyk-110	450 °C	204	720	3,000	[56]
$Bi_2Te_{2.8}Se_{0.2}$	Screen printing	α-Terpineol + Disperbyk-110 + glass frits	430 °C	-126	310	490	[57]
Bi_2Te_3/epoxy	Extrusion printing	epoxy resin + anhydride -based hardener	250 °C	-157	61	150	[58]
Bi_2Te_3/Se/epoxy	Extrusion printing	epoxy resin	350 °C	-170	96	277	[59]
Sb_2Te_3/epoxy	Extrusion printing	2-butoxy ethanol + dibutyl phthalate	250 °C	160	63	160	[60]
$Bi_{0.5}Sb_{1.5}Te_3$	Extrusion printing	glycerol,	450 °C	165	554	1,508	[61]
TiS_2(HA)x	Inkjet printing	N-Methylformamide	110 °C	-70	430	211	[62]
Sb_2Te_3/Te	Aerosol jet printing	ethylene glycol, + glycerol, + ethanol	400 °C	198	560	2,200	[63]
SnSe/PEDOT	Drop casting	-	328 °C	110	320	390	[64]
CNT/P3HT	Spray printing	-	-	97	345	325	[65]

and spray printing[65]. These printing techniques have been extensively explored in recent years, and significant progress has been made in optimizing the printing parameters, ink formulations, and post-processing methods to enhance the thermoelectric performance of printed materials and devices[66]. Furthermore, printed thermoelectric materials and devices have found applications in energy harvesting, waste heat recovery, wearable electronics, and flexible electronics[67-70].

Given the significant advances and potential applications of printing methods for thermoelectric materials and devices, it is timely to review the recent progress in this field. This review aims to provide a comprehensive overview of the recent advances in printing methods for thermoelectric materials and devices, covering the key principles, challenges, and opportunities associated with various printing techniques. The review highlights the progress made in optimizing the printing parameters, ink formulations, and post-processing methods to improve the thermoelectric performance of printed materials and devices. Furthermore, their applications in energy harvesting, waste heat recovery, wearable electronics, and flexible electronics, are discussed, providing insights into the current state-of-the-art and future directions of this promising field.

SCREEN PRINTING

Screen printing is a versatile printing technique used for a wide range of applications, including textiles, graphics, electronics, and more recently, thermoelectric materials and devices[71-74]. The process involves several steps, as depicted in Figure 1A[75]. First, pastes or inks are added to a screen, which is a mesh-like stencil made of fabric or other materials. The paste is then smeared across the screen surface using a blade, spreading it evenly over the openings in the screen. Next, a squeegee is pressed against the screen with pressure, driving the paste through the holes in the screen. As the squeegee passes across the screen, it leaves a thin layer of paste on the substrate in the desired pattern. Ultimately, the deposited pattern is formed on the substrate after removing the screen.

Figure 1B depicts flexible thermoelectric generators (TEGs) that has been screen printed onto polyimide substrates[76]. The thermoelectric legs were n-type $Bi_2Te_{2.7}Se_{0.3}$ and p-type $Bi_{0.5}Sb_{1.5}Te_3$, which were mixed with epoxy resin and then screen printed onto the substrates. The formed radial structured TEGs with five

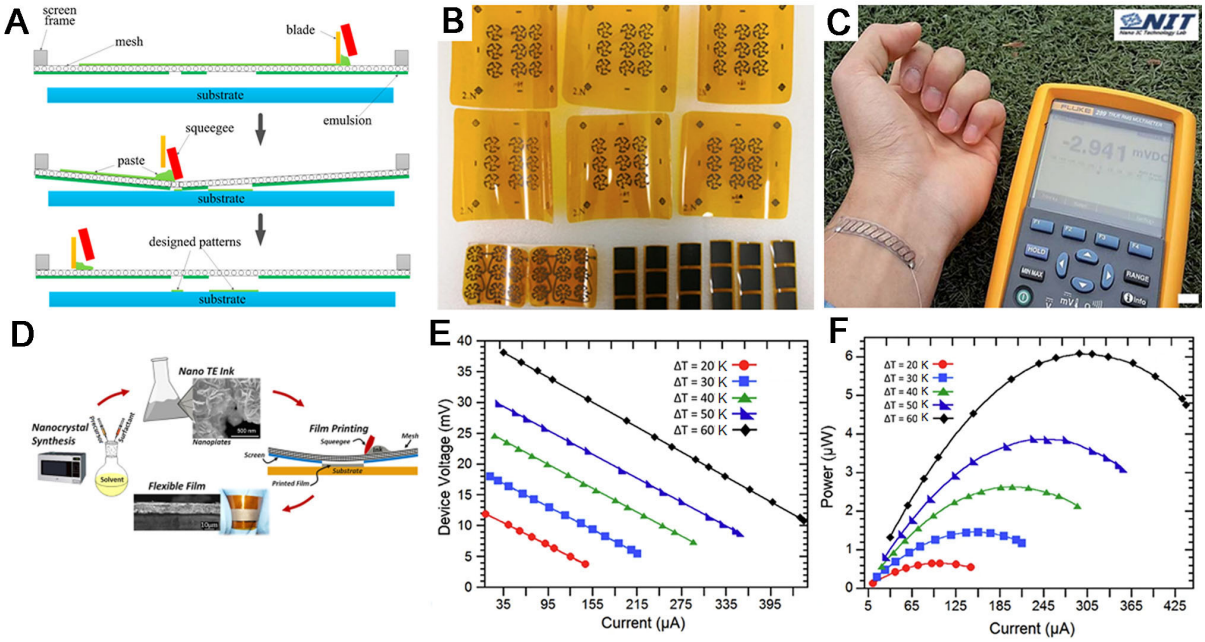

Figure 1. (A) Schematic diagram showing the pattern deposition by screen printing. Reproduced with permission[75]; (B) Screen-printed radial structured TEG with n-type $Bi_2Te_{2.7}Se_{0.3}$ and p-type $Bi_{0.5}Sb_{1.5}Te_3$ on polyimide substrates. Reproduced with permission[76]; (C) Screen-printed TEG on flexible glass fabric substrate and the voltage generated using human body heat[54]; (D) Fabrication process for the screen-printed flexible thermoelectric films. Measured output (E) voltage and (F) power of the thermoelectric device as a function of current. Reproduced with permission[57]. TEG: Thermoelectric generator.

thermoelectric couples employed a heat source in the center and generated an open-circuit voltage of 68.41 mV and an output power of 5.81 μW.

Another screen-printed prototype TEG shown in Figure 1C was composed of eight thermocouples made of Bi_2Te_3 and Sb_2Te_3 thick films with dimensions of 15 × 20 × 0.5 mm. The substrate was Al_2O_3[54]. When subjected to a temperature difference (ΔT) of 50 K, this TEG produced an open-circuit voltage of 90 mV. The Al_2O_3 substrate acted as a heat sink, which increased the power density to be 3.8 mW·cm^{-2} with a ΔT of 50 K. It was also discovered that the devices were lightweight, with an overall density of 0.13 g·cm^{-2}, and yield a high specific output power of 28 mW·g^{-1} when a ΔT was 50 K. With this TEG, a wearable device was built to illustrate the likelihood for power generation using the human body heat, as shown in Figure 1C. Figure 1D shows the fabricating process of flexible thermoelectric films using a screen-printing technique with colloidal inks composed of bismuth telluride-based nanoplates[57]. These nanoplates were synthesized by a microwave-heated solution method, which was likely to scaleup the material synthesis. Figure 1E and F show the measured output voltage and power of this TEG at different ΔT between the hot side and cold side[57]. The maximal output voltage reached 38 mV with a ΔT of 60 K. Moreover, the maximum power was up to 6.1 μW. This typical TEG highlights the potential of screen printing as the highly scalable and low-cost method for fabricating flexible TEGs. The results open promising opportunities for advancing thermoelectric energy harvesting and cooling applications[77-79].

INKJET PRINTING

Inkjet printing has become a promising technique for producing thermoelectric materials and devices due to its ability to precisely deposit materials with high resolution, which enables the fabrication of complex thermoelectric structures and patterns. Inkjet printing thermoelectric materials involves using specialized inks that contain thermoelectric materials[80,82].

Figure 2A shows the typical process of employing inkjet printing to prepare flexible thermoelectrics[83]. First, nanowires of metal chalcogenides were synthesized via a chemical method using the template of tellurium nanowires. Then, the collected nanowires were used as inks to print thermoelectric films and devices. Figure 2B is a typical TEG that is comprised of 20 inkjet-printed graphene legs connected by silver[84]. The device is printed on a flexible Kapton polyimide substrate, making it suitable for conforming to non-planar surfaces or for energy harvesting from body heat in wearable applications. The as-prepared large-area flexible graphene thin films exhibited remarkable thermoelectric properties. Due to the phonon-glass electron-crystal behavior, the all-graphene films exhibit a high room-temperature $S^2\sigma$ of 18.7 $\mu W \cdot m^{-1} \cdot K^{-2}$, representing a significant improvement of over threefold compared to previous solution-processed all-graphene structures.

The post-treatment of inkjet-printed nanowires by hot press sintering is used as a method to further enhance thermoelectric performance of the printed nanowires. During the sintering process, glass fiber membranes are employed to protect the inkjet-printed nanowires from separating from the substrate due to the high temperature and pressure applied. Figure 2C is the scanning electron microscope (SEM) image of Ag_xTe film sintered at 673 K, in which the original nanowire morphology is well-preserved[83]. Figure 2D and e show the σ and S of printed Ag_xTe films with different Ag contents sintered at 673 K[83]. As can be seen, the σ increases significantly with increasing Ag content, from 185.8 $S \cdot cm^{-1}$ for $Ag_{1.9}Te$ to 523.3 $S \cdot cm^{-1}$ for $Ag_{2.1}Te$ at room temperature. The corresponding absolute value of S decreases from 80.4 to 65.2 $\mu V \cdot K^{-1}$. The remarkably enhanced thermoelectric performance resulted in highly efficient TEGs. Figure 2F exhibits the measured output voltage and power of a TEG made of four $Ag_{2.1}Te$ thermoelectric legs function of the output current at different temperature gradients[83]. As a result, the maximum output power of 101.3 nW is achieved at a ΔT of 30 K. The developed $Ag_{2.1}Te$ based TEG was used to generate electricity using the human body. Figure 2G shows a prototype wearable thermoelectric generator. Figure 2H is the corresponding temperature and output power. A stable voltage of 2 mV can be obtained from the ΔT between the wrist skin and the external environment.

The successful demonstration of inkjet-printed thermoelectric devices highlights the potential for flexible, scalable, and low-cost thermoelectric applications. This includes the possibility of harvesting energy from body heat in wearable applications, where the flexibility, conformability, and cost-effectiveness of the inkjet-printed graphene-based thermoelectric films can be leveraged. These findings pave the way for advancements in the field of flexible thermoelectric materials and devices, with potential applications in wearable electronics, energy harvesting, and other related fields[85,86].

AEROSOL JET PRINTING

Aerosol jet printing is a technique that can be used for printing thermoelectric materials to fabricate thermoelectric devices. It is a form of additive manufacturing where a fine aerosolized mist of material is jetted through a small nozzle and deposited onto a substrate to create a pattern or structure[18,87].

Figure 3A is a typical aerosol jet printing nozzle, allowing for the direct printing of thermoelectric devices with high spatial resolution on both 2D planar and 3D curved substrates[88]. One of the key advantages of aerosol jet printing is its ability to achieve sub-micron thickness control of the deposited material, which is critical for optimizing the performance of thermoelectric devices. Furthermore, aerosol jet printing can utilize colloidal nanoparticle inks with a wide range of viscosities. This flexibility enables the printing of inks composed of different thermoelectric materials, dopants, or additives, allowing for tailoring of the

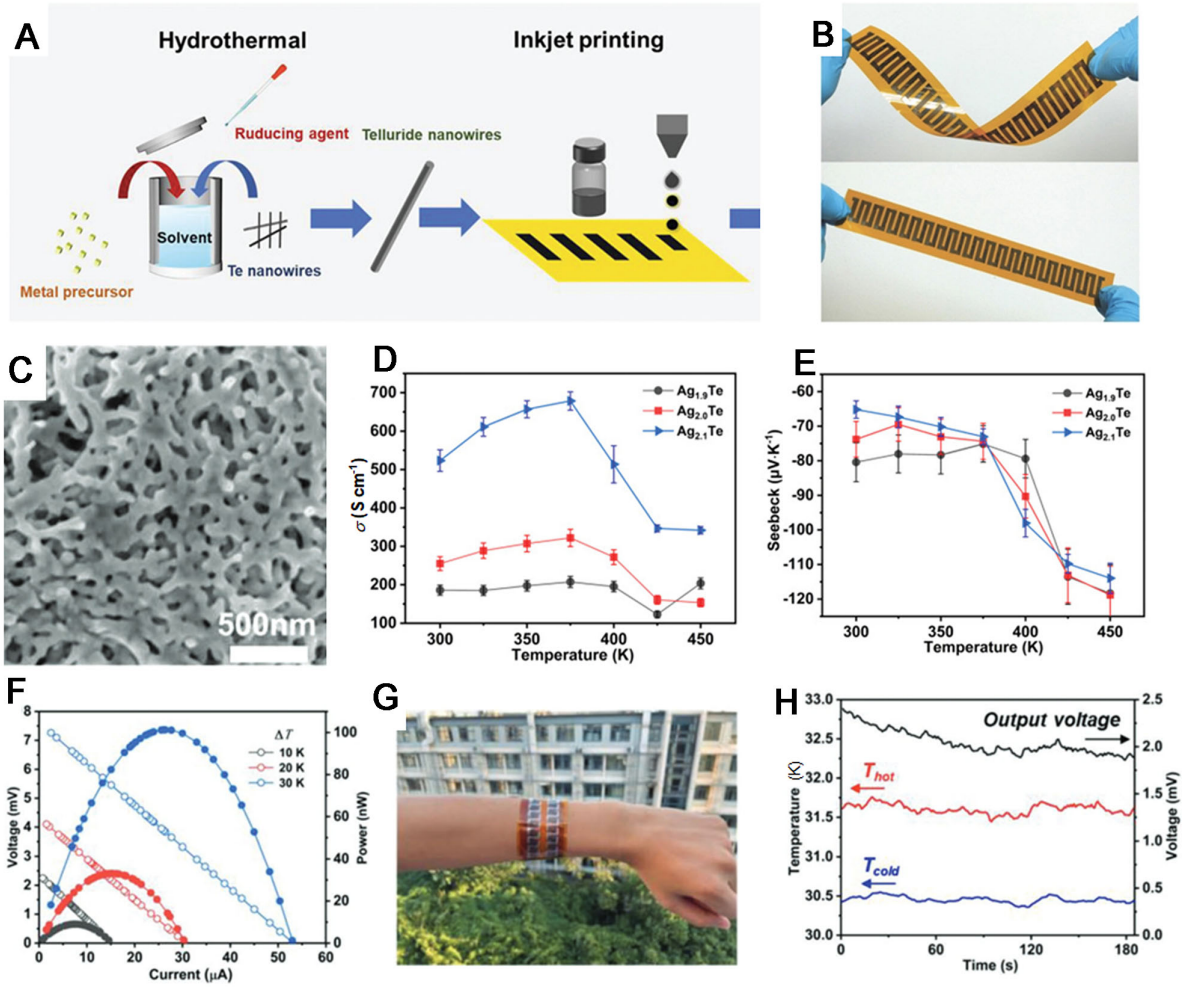

Figure 2. (A) Fabrication process of inkjet printing to produce TEGs[83]; (B) Photograph of an inkjet-printed thermoelectric device consisting of 20 silver and graphene legs[84]; (C) SEM images of Ag_2Te films sintered at 673 K; Temperature-dependent (D) σ and (E) S of printed Ag_xTe films with different Ag ratios sintered at 673 K[83]; (F) Measured power and voltage as a function of the current at different T; (G) Photograph of a wearable flexible TEG fabricated by inkjet printing; (H) Measured temperature and voltage when wearing on the wrist[83]. SEM: Scanning electron microscope; TEG: thermoelectric generator.

properties of the final thermoelectric device. This ability to handle a variety of inks with different material properties makes aerosol jet printing a versatile technique for optimizing the design and performance of thermoelectric devices, making it suitable for a wide range of applications.

Using the colloidal nanocrystal ink, the thermoelectric films with virtually any patterns can be fabricated by aerosol jet printing method onto 2D flexible substrate (e.g., polyimide, Figure 3B) and 3D curved substrate (e.g., glass tube, Figure 3C)[88]. A flexible TEG with aerosol jet printed Sb_2Te_3-Te films and Ag electrodes was fabricated to demonstrate the printed TEG for energy harvesting[63]. Figure 3D and E show the measured voltage and power as a function of electrical current tested at different ΔT. A maximum power output of 1.15 μW was obtained with a ΔT of 60 K. Figure 3F plots the power density. With increasing ΔT, the power density reaches the maximal value of 7.65 mW·cm^{-2}[63]. Such a high power density achieved by aerosol jet printed flexible TEG indicates that even small-sized TEG can provide sufficient power to drive typical Internet of Things (IoT) devices and sensors. This demonstrates the potential of aerosol jet printing as a scalable and efficient method for producing compact and high-performance flexible TEGs that can be used in various IoT and sensor applications where size and power density are critical factors.

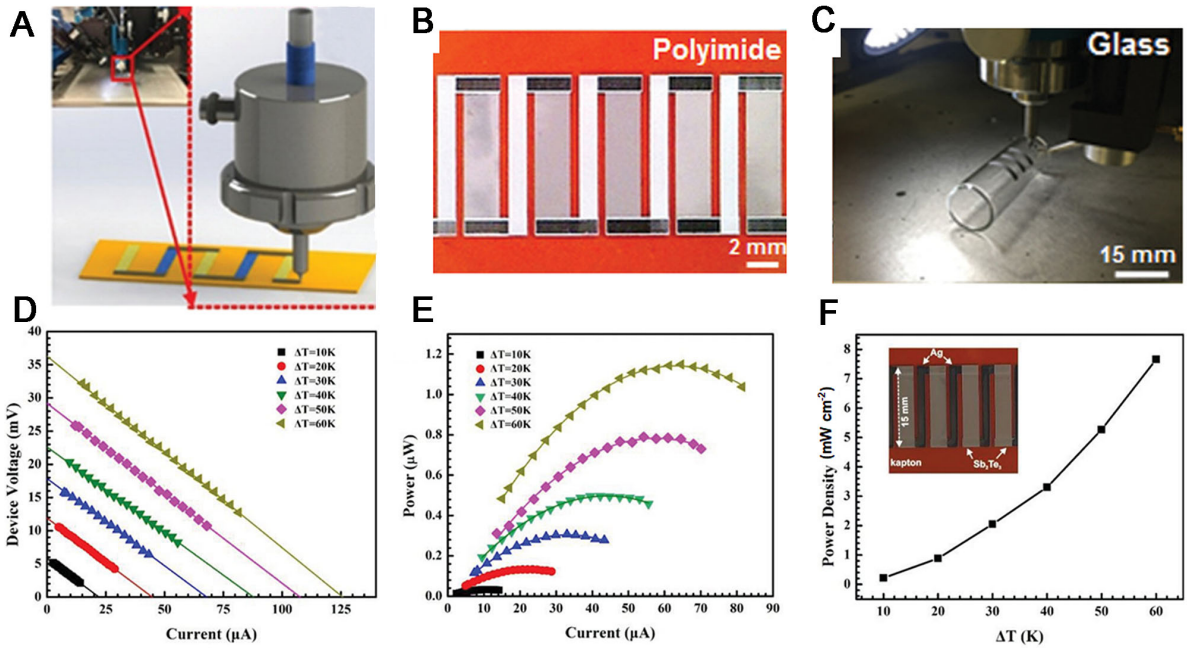

Figure 3. (A) Aerosol jet printing nozzle. The aerosol-printed films with n-type $Bi_2Te_{2.7}Se_{0.3}$ legs on (B) flat polyimide substrate and (C) glass tube[88]; As for an aerosol-printed thermoelectric devices with p-type Sb_2Te_3 legs connected by Ag, measured output (D) voltage and (E) power as a function of current at various ΔT; (F) Measured power density tested at various ΔT[63].

3D PRINTING

Three-dimensional (3D) printing has emerged as a promising technique for fabricating thermoelectric materials and devices. 3D printing also enables the fabrication of thermoelectric devices on flexible and curved substrates, making them suitable for wearable and conformal applications. This allows for the integration of thermoelectric generators into various wearable devices, sensors, and IoT applications, where flexibility and shape adaptability are critical.

Figure 4A-C illustrates the use of extrusion-based 3D printing to fabricate thermoelectric materials with defined shapes using all-inorganic inks[61]. One of the main challenges in this approach is the development of all-inorganic inks that do not contain any organic binders. This requires tailoring the rheology of the ink to ensure reliable flow through fine nozzles during printing, while maintaining the structural integrity of the printed shape[61].

Inorganic ChaM (chalcogenide-based metal) ions have been reported as surface ligands for nano- and microscale particles, providing stability to the particles in solution through electrostatic interactions. In the case of Sb_2Te_3 ChaM ions in concentrated colloid inks, they effectively hold Bi_2Te_3-based thermoelectric particles together in an electrostatic manner. This enhances the colloidal stability of the thermoelectric inks and impacts the rheological properties of the medium in the static state[61].

The use of inorganic ChaM ions in the inks allows for improved stability during 3D printing, enabling reliable extrusion through fine nozzles and maintaining the structural integrity of the printed shapes. This is crucial for achieving high-quality, well-defined thermoelectric materials with desired shapes using 3D

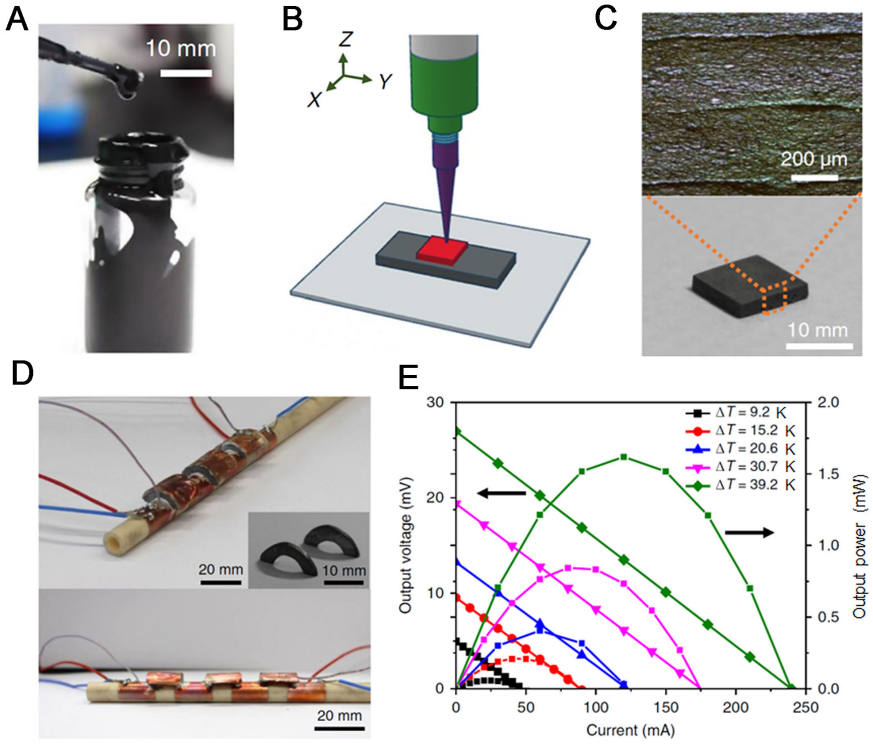

Figure 4. (A) Photograph displaying all-inorganic thermoelectric ink with viscoelastic feature; (B) Depiction of a 3D printing process; (C) Optical microscopy image, as well as a photograph, of the thermoelectric materials printed through 3D printing; (D) Photographs of the fabricated half-ring-based conformal TEG; (E) Measured voltage and power of the TEGs at different T[61]. TEG: Thermoelectric generator.

printing techniques. By overcoming the challenge of ink stability and rheological properties, all-inorganic thermoelectric inks with tailored ChaM ions hold significant promise for advancing the field of 3D printing of thermoelectric materials with defined shapes and enhanced performance.

Figure 4D shows a TEG fabricated by 3D printing method, which was used to harvest waste heat from hot water[61]. Figure 4E plots the measured voltage and power of the TEG. With increasing ΔT, the 3D printed TEG exhibited a linear increase in output voltage and a quadratic increase in output power. The maximum output voltage achieved was 27.0 mV and the maximum power reached 1.62 mW at a ΔT of 39 K, leading to an output power density of 1.42 mW·cm^{-2}. This performance was comparable to that of previously reported TEGs using composite inks consisting of Bi_2Te_3-based powders with Sb_2Te_3 ChaM[61]. These results highlight the potential of 3D printed thermoelectric materials for achieving high output voltage, power, and power density suitable for practical energy harvesting applications.

Based on the solution synthesized materials, more TEGs were fabricated via 3D printing[89]. Figure 5A is the schematic diagram of the 3D printing process, which was conducted for 25 times to increase the thickness of films. The infrared thermal image shown in Figure 5B provides a visual representation of the temperature distribution in the TEG. The uniform heat flow along the thermoelectric leg indicates that consistent thermal conductance is achieved in the layers. This uniformity in thermal conductance is essential for efficient energy harvesting and indicates the potential of 3D printing for precise control over the thermoelectric performance of the fabricated devices[89]. Figure 5C shows the measurement setup. Figure 5D plots the measured voltage and power as a function of current at a ΔT of 32 K. This TEG provides an output

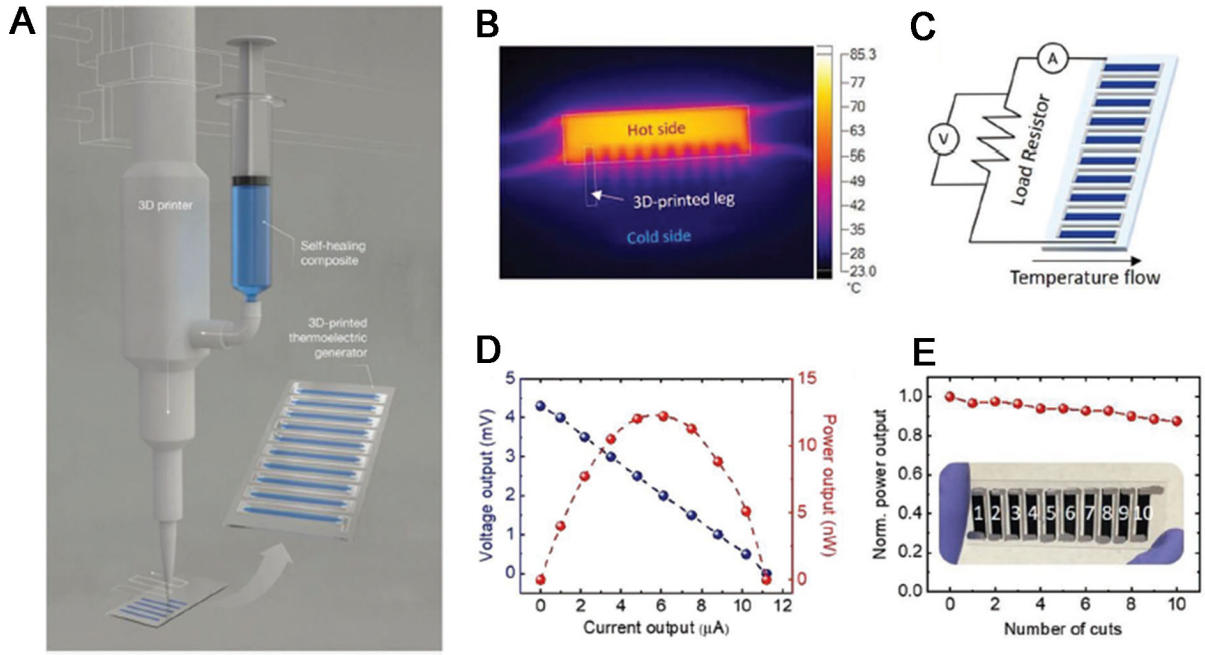

Figure 5. (A) 3D printing process; (B) An infrared thermal image that illustrates the thermal gradient within the 3D-printed TEG sample; (C) Performance evaluation of TEGs. The current and voltage were measured using a variable load resistor; (D) Measured current-dependent voltage and power with ΔT of 32 K; (E) Normalized output power in the cases of different number of cuts[89]. TEG: Thermoelectric generator.

voltage of 4.3 mV[89]. As a result, this TEG yielded a peak power of 12.2 nW. The achieved output voltage and power are sufficient to operate electronic devices that require low power consumption[89].

Impacts on maximum power output were monitored while cutting each leg to test the self-healing property of 3D-printed TEGs upon physical damages by cutting [Figure 5E]. Despite an increase in the number of cuts, the power output remained relatively constant, with over 85% of its initial value retained. As can be seen, the self-healing feature of the thermoelectric composite enables the fabrication of 3D-printed TEGs with excellent mechanical integrity, making them robust and durable for practical applications.

ROLL-TO-ROLL PRINTING

Roll-to-roll (R2R) printing is a high-throughput manufacturing technique that involves continuously feeding a flexible substrate material, such as plastic or metal foil, through a series of processing stations. In the context of thermoelectric materials, R2R printing has emerged as a promising fabrication method for producing large-area, flexible, and cost-effective thermoelectric devices. The R2R printing process typically involves depositing or printing multiple layers of thermoelectric materials, such as p-type and n-type semiconductors, along with metal contacts and insulating layers, onto a flexible substrate[90].

Figure 6A shows the process of R2R printing for PEDOT:PSS thermoelectric patterns[91]. The copper roller has convex stamping features that are made by mechanical machining. The features are treated with 2.5 M NaOH and 0.13 M $(NH_4)_2S_2O_8$ solutions to achieve the desired ink wettability. The substrate is initially hydrophobic, with a water contact angle of over 90°. Different chemical solutions are used to modify the ink wettability. Figure 6B and C are the photographs of the printed thermoelectric devices. To improve the ink wettability, the plastic films were oxygen plasma treated for one hour, which resulted in an ink contact angle of around 20°.

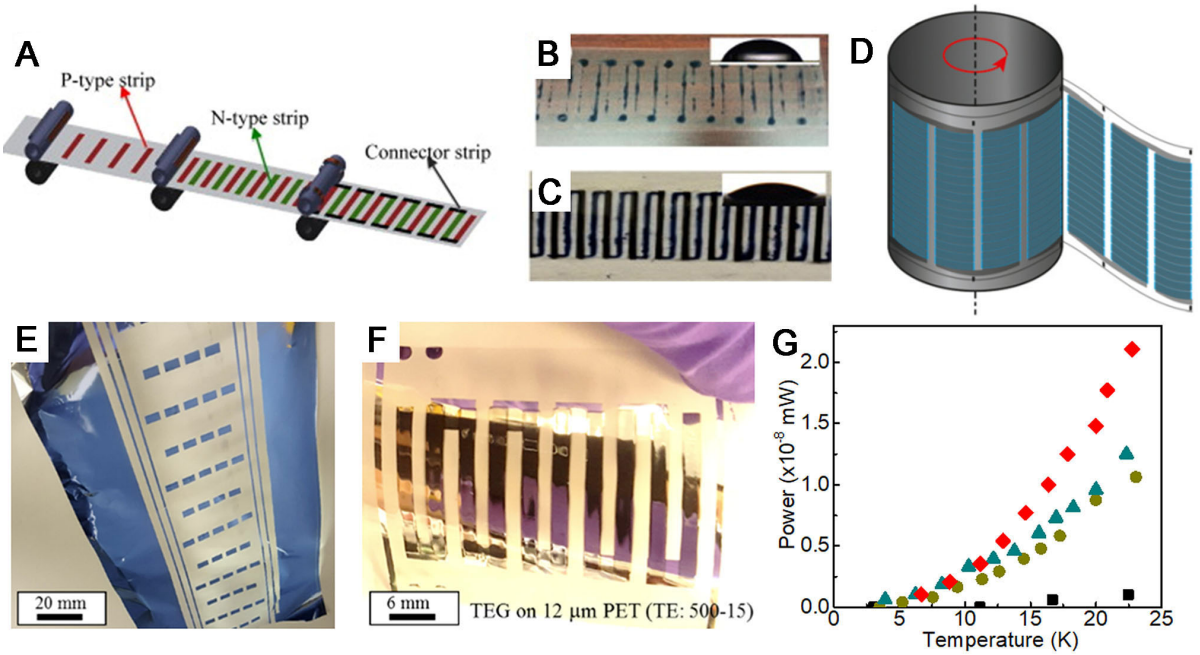

Figure 6. (A) Illustration of R2R printing of flexible thermoelectric devices. R2R printed thermoelectric films on (B) UV-treated plastic substrate and (C) plasma-treated plastic substrate. The inset images show the contact angle between substrate and the inks[91]; (D) Illustration of the rolled-up final device[92]; (E) Printed electrode cleaned by electron beam; (F) Complete TEG on pristine electrodes; (G) generated output power as a function of temperature difference[93]. R2R: Roll-to-roll; TEG: thermoelectric generator; UV: ultraviolet.

After printing, the thermoelectric junctions was divided into smaller stretches and rolled up on an aluminum cylinder [Figure 6D][92]. This work also implemented low-energy electron beam (LEB) to clean surface. This technique employs flexographic oil masking onto flexible substrates to achieve continuous R2R patterning. After material deposition, the oil masking is removed, resulting in the patterned deposited material being left behind.

The fabricated TEGs are used to demonstrate the viability of using LEB as a surface cleaning technique in R2R. The fabrication steps involve thermally evaporating aluminum electrodes using R2R flexographic oil masking. The substrate used is a 12 μm polyethylene terephthalate (PET). The Al electrode patterned substrate is then cleaned via LEB for 0, 3, 5 or 15 min.

Four pairs of thermoelectric legs are made using a shadow mask and then form TEGs, as shown in Figure 6E and F. The TEG shows high output power. Figure 6G plots the measured output power as a function of ΔT. As can be seen, a maximal power of 2.1×10^{-11} W was achieved at a ΔT of 22 K.

SPRAY PRINTING

Spray-printing is a novel fabrication method that has been explored for fabricating thermoelectric materials and devices. In this method, thermoelectric materials are atomized into a spray, which is then deposited onto a substrate to form a thin film. The thin film can be patterned into different shapes and sizes to create thermoelectric devices with specific geometries.

Figure 7A schematically shows the fabrication process of spray-printing, associated with the chemical structure, and the fabricated PEDOT:PSS TEGs on a flexible substrate. This study utilized glass and polyethylene naphthalate (PEN) substrates and applied an ultraviolet (UV) ozone treatment to produce

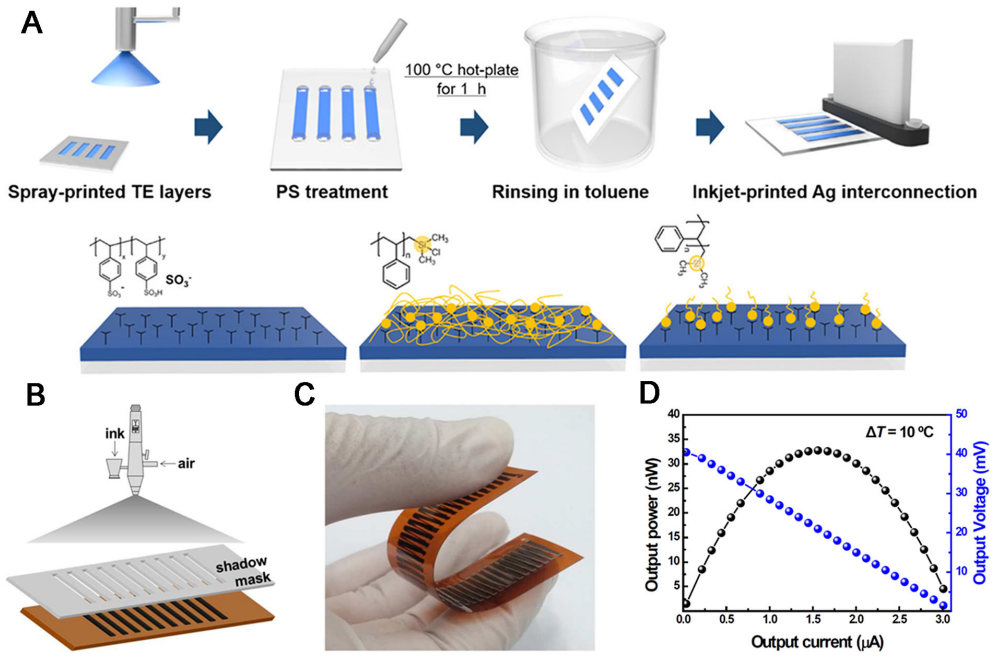

Figure 7. (A) Schematic diagram showing the process of fabricating PEDOT:PSS TEGs via spray printing[94]; (B) Schematic drawing of the spray-printing process of a flexible CNT/P3HT TEG; (C) Photograph of the CNT/P3HT TEG demonstrating flexibility; (D) Measured output power and voltage as a function of current[65]. TEG: Thermoelectric generator.

hydrophilic surface properties, which enhance ink wetting. Dimethyl sulfoxide (DMSO) with a concentration of 5 vol% was added to PEDOT:PSS to increase the σ of PEDOT:PSS thermoelectric legs. The solution was then deposited on substrates by a customized spray printing system using a patterned mask. The printing conditions were optimized to achieve highly uniform PEDOT:PSS layers with a thickness of 1 μm. For selective treatment at the contact region, a 0.4 wt% dimethyl chlorosilane-terminated polymer solution dissolved in toluene was applied, followed by annealing on a 373 K hot plate for 1 h. Upon rinsing in toluene to eliminate any remaining uncoupled residues, the series-connected thermoelectric legs were furnished with highly conductive interconnects by printing nanoparticle-type Ag ink using an inkjet printer. To ensure the quality of the interconnects, the Ag electrodes were then subjected to annealing at 423 K for 30 min. A flexible organic TEG consisting of only p-type carbon nanotube/poly(3-hexylthiophene) (CNT/P3HT) was fabricated by spray printing[65]. Figure 7B schematically shows the spray printing process, in which the CNT/P3HT patterns were printed on a polyimide substrate. Figure 7C is the photo of the CNT/P3HT-based TEG. Excellent flexibility of TEG can be seen. Figure 7D plots the measured output power and voltage as a function of current for the spray-printed flexible CNT/P3HT TEG[65]. A maximum open circuit voltage of 40 mV and a maximum output power of 32.7 nW was achieved at a T of 10 K. This study successfully demonstrated the electrical power generation of the spray-printed TEG.

CONCLUSION AND OUTLOOK

The review provides a comprehensive overview of different printing techniques, including screen printing, inkjet printing, aerosol jet printing, 3D printing, R2R printing, and spray printing for fabricating thermoelectric materials and devices. The materials used, the process of each printing technique, and the potential for enhancing thermoelectric performance through post-treatment methods are discussed in detail. The advantages and applications of each technique are highlighted, demonstrating their potential for enabling scalable and cost-effective fabrication of thermoelectric devices. With continued efforts in

materials design, process optimization, and device integration, thermoelectric printing has the potential to revolutionize the field of thermoelectric materials and devices, offering sustainable and efficient solutions for energy conversion and harvesting applications.

The field of thermoelectric printing has seen significant advancements in recent years, offering promising prospects for the fabrication of thermoelectric materials and devices using printing techniques. These techniques offer several advantages, including high scalability, flexibility in device design, and potential for integration into various applications, such as wearable devices, energy harvesting systems, and smart textiles.

However, there are still challenges and limitations that need to be addressed to fully realize the potential of thermoelectric printing. These include the need for further optimization of printing parameters, materials design, and post-treatment methods to achieve improved thermoelectric performance. Additionally, standardized characterization methods and performance metrics for printed thermoelectric devices need to be established for accurate comparison and evaluation.

Future research and development in the field of thermoelectric printing could focus on the exploration of novel thermoelectric materials specifically designed for printing techniques, the advancement of printing methods for complex device architectures, and the integration of printed thermoelectric devices into practical applications. Further advancements in post-treatment methods, device characterization, and understanding of the fundamental transport mechanisms in printed thermoelectric materials are also crucial for advancing the field. Specifically, the future directions include:

(1) Further enhancing thermoelectric performance. Strategies for optimizing material design, device structure, and printing parameters will be developed to result in improved thermoelectric properties of printed materials. This, combined with advancements in printing precision and efficiency, will contribute to increased power densities and enhanced practical application value of flexible thermoelectric devices.

(2) Ensuring reliability and stability. Ongoing research will focus on improving the reliability and stability of printed thermoelectric devices. By optimizing material formulations and device architectures, as well as understanding degradation mechanisms, longer lifespans and better performance in practical applications can be achieved.

(3) Addressing the limitation of in-plane device. Improving heat transfer within the thin film structure is crucial for enhancing the performance of in-plane devices. Strategies such as incorporating high thermal conductivity layers or engineering thermal interfaces can enhance heat dissipation and improve the temperature gradient across the device, resulting in improved thermoelectric efficiency.

(4) Realizing cost reduction and environmental sustainability. With the expansion of production scale, process optimization, and equipment improvements, the cost of printed thermoelectric devices is expected to decrease. This will make them more commercially viable and accessible for a wide range of applications. The development of environmentally friendly printing materials and processes will be a significant focus. Exploring thermoelectric materials that are abundant, non-toxic, and compatible with printing techniques, as well as eco-friendly inks and post-treatment methods, will help reduce the environmental impact of thermoelectric printing.

DECLARATIONS

Authors' contributions

Initiated the idea: Hong M, Chen ZG, Li M, Shi XL, Liu W
Conducted the literature review: Hong M, Sun S, Lyu W
Drafted the manuscript: Hong M
Reviewed and revised the manuscript: Hong M, Chen ZG, Shi XL, Liu W, Li M
All authors have read the manuscript and approved the final version.

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Conflicts of interest

Not applicable.

Ethical approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

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