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REVIEW

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Electrohydrodynamic (EHD) printing of nanomaterial composite inks and their applications

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Abstract

The utilization of high-resolution printed flexible electronic devices is prevalent in various fields, including energy storage, intelligent healthcare monitoring, soft robotics, and intelligent human–machine interaction, owing to its compact nature and mechanical flexibility. The EHD jet printing technology has the potential to develop the field of printing industry through its ability to fabricate high-resolution, flexible, stretchable, and 3D structures for electronic applications such as displays, sensors, and transistors. The EHD jet printing technology involves the use of solution-based inks made of diverse functional materials to print a wide range of structures. Consequently, it is imperative to have a comprehensive understanding of nanomaterial composites that are printed using EHD jet printing technology. This review provides a thorough overview of nanomaterial composite inks printed for electronic devices using EHD jet printing technology. In particular, a comprehensive overview has been provided about the utilization of EHD jet printing for nanomaterial composites in several domains, including flexible electrodes, flexible displays, transistors, energy harvesting, sensors, and biomedical applications. Moreover, this analysis presents a concise overview of the limitations and prospective future directions for nanomaterial composites fabricated by EHD jet printing.

Keywords Electrohydrodynamic, Nanomaterial composite, Electronics applications

Introduction

The rapid advancement of high-tech devices needs the use of high-resolution and low-cost manufacturing techniques. Printing technologies such as screen printing [1], transfer printing [2], inkjet printing [3], and EHD jet printing [4] have recently piqued the curiosity of scholars globally for printed electronics. Particularly, EHD jet printing, in which the electric field plays an important role in printing is a topic of paramount importance because of its variety of printed electronic applications. The EHD jet printing technology has several notable features, including high resolution, accurate pattern precision, noncontact printing, driven by a high-voltage electric field, cost-efficiency, and the ability to jet solutions even with high viscosity. The utilization of EHD jet printing technology facilitates high-resolution printing of flexible electronic devices. This is accomplished through the generation of a Taylor cone of ink, which is facilitated by the usage of an electric field between the nozzle's tip and the substrate [5]. When the Coulombic force within the Taylor cone exceeds the surface tension of the functional ink, high-resolution continuous jet lines or droplets are printed on a substrate by EHD jet printing.

Various functional materials (i.e. metal materials [6–8], conducting polymers [9, 10], and carbon-based materials [11]) have been used to fabricate high-resolution flexible electronic devices through the utilization of EHD jet printing technology. In recent years, several review works have summarized the EHD printing of conducting polymers, metal materials, and carbon-based materials

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[12–17] but there is no detailed review focusing on nanomaterial composites printed by EHD. Nanomaterial composites have frequently been used for various engineering applications due to their superior electrical, mechanical, optical, electrochemical, and biological properties. The physical and chemical characteristics of nanomaterial composites have a significant impact on both the print quality of functional inks on substrates and the overall operation of printed electronic devices [18]. While traditional inkjet printing or microfabrication technologies are limited in the printing or pattern resolution, EHD jet printing of nanomaterial composites can offer higher patterning resolution than can lead to performance enhancement. Therefore, the choice of suitable nanomaterial composite inks for the EHD jet printing technology is of utmost importance in ensuring the sensitivity, electrical conductivity, and mechanical stability of devices printed with high-resolution.

This short review focuses on the developments made in the field of EHD jet printing of nanomaterial composites and their subsequent applications. Initially, we provided a concise overview of EHD jet printing technology and outlined the several types of materials, namely insulating, semiconducting, conductive, and biomaterials, which have the potential to serve as functional inks in the context of EHD jet printing technology. Further, we reviewed nanomaterial composites printed by EHD and their applications in the areas of flexible displays, transistors, electrodes, energy harvesting, storage devices, sensors, and medical devices [19–24]. Finally, we discussed the future prospects and constraints of printed electronic device printing with next-generation EHD jet technology.

EHD jet printing technology

Brief overview

Before exploring the applications of nanomaterial composites printed by EHD jet printing, it is necessary to elucidate the fundamental concept of EHD jet printing in order to present a concise overview of the underlying technology. The origins of EHD jet printing are attributed to the examination of droplet dynamics in the existence of an electric field, resulting in the formation of a droplet with a near-conical shape. The operation of EHD jet printing differs from conventional inkjet printers in that it enables direct deposition of materials through specific modes of droplet generation, such as micro dripping, dripping, and cone jet, which is illustrated in Fig. 1a and b [25]. The EHD jet printing process primarily relies on the formation of a Taylor-cone under the electric field effect, as depicted in Fig. 1c [26]. This approach enables the production of intricate nano/micrometer-sized designs that surpass the capabilities of regular inkjet printers.

EHD jet printing of materials

Insulating materials

The use of functional ink, which plays a key role in determining the ultimate characteristics of the electronic device, stands as a pivotal facet within the realm of printed electronics technology. Various materials have been printed using diverse printing processes, however, our primary emphasis lies on materials printed through the utilization of EHD printing technology (Fig. 2). The investigation of EHD jet printing of insulating materials has received limited attention because of the inherent difficulties associated with achieving uniform patterning

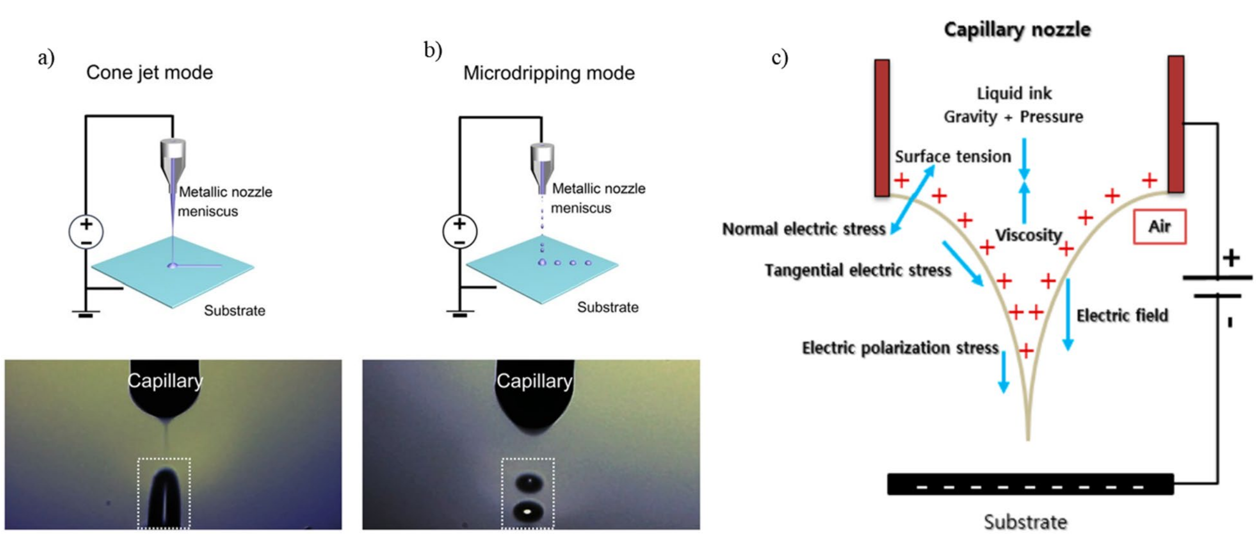

Fig. 1 Schematic and physics of EHD jet printing. **a** and **b** Cone jet mode and micro dripping mode, reprinted from reference [19]. **c** Forces acting on a capillary tip to form a Taylor cone, reprinted from reference [20]

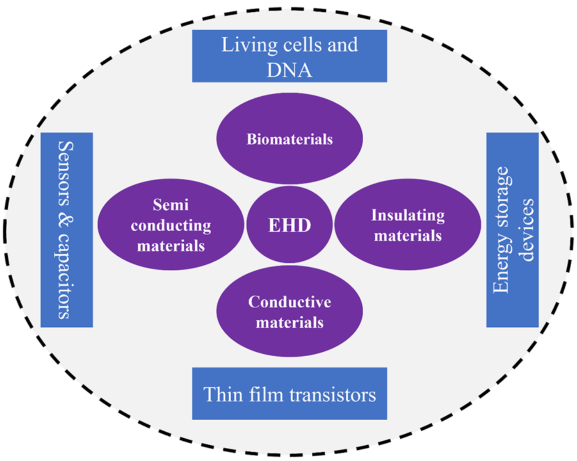

Fig. 2 Schematic representation of materials printed by EHD and their applications

and smooth surfaces [25]. However, researchers have attempted EHD jet printing to create direct patterns of insulating materials and then used them to create electronic devices like gate electrodes and thin film transistors (TFTs) applications [23, 27, 28]. A comprehensive investigation by Tang et al. was conducted to examine the efficacy of EHD jetting with insulating polymers, specifically focusing on the electrostatic-force-assisted dispensing mode to print gate insulators. Once the EHD-printed layer completely covered the bottom gate electrode (BGL), the top electrode was subsequently deposited onto the gate insulator (GI) layer. This process resulted in the formation of a capacitor, as depicted in Fig. 3a and b. Compared to the conventional dispensing process, these designs printed by EHD had more regular patterns even when printing for extended periods of time [29].

Semi-conducting materials

Both organic and inorganic semiconductors have been developed and used extensively for electronic applications. Along with other printing technologies, the EHD jet printing method also enabled semiconductors to be applied to intricate and integrated high-resolution devices [30, 31]. EHD printing has been utilized to pattern high-resolution zinc–tin oxide (ZTO) semiconductors for TFT applications [32] (Fig. 3c). EHD jet-printed structures outperformed spin-coated and inkjet technologies in terms of positive bias stability and hysteresis behavior. In another study, EHD jet printing was applied

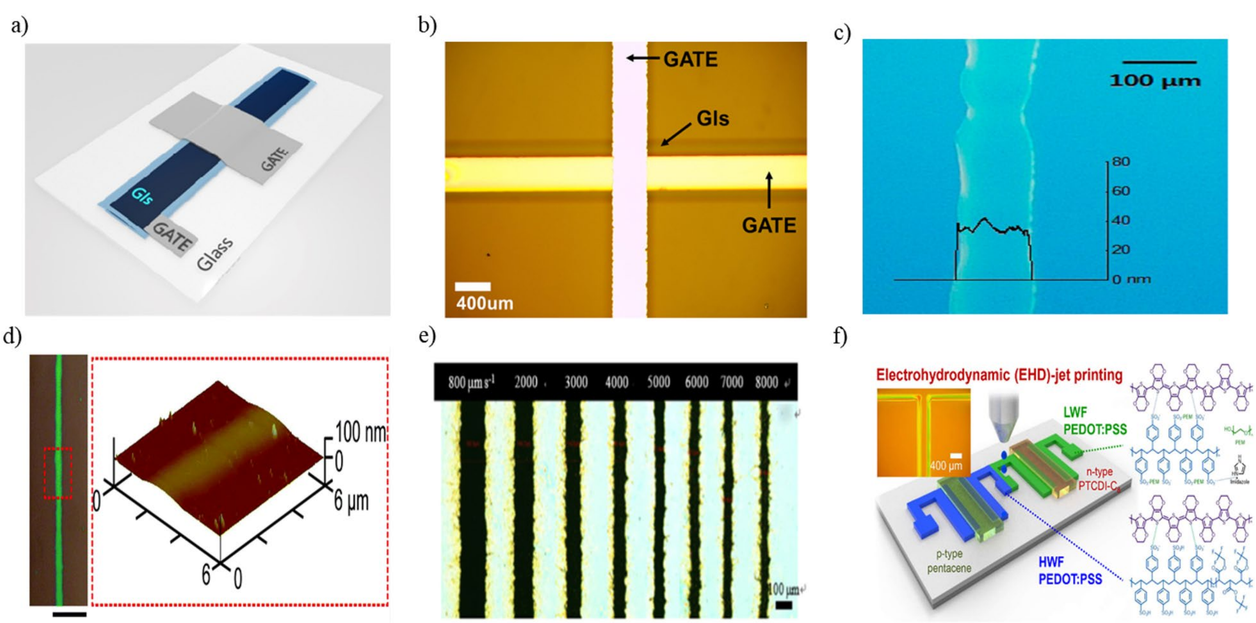

Fig. 3 EHD printing of different materials. **a** and **b** Schematic illustration of insulating material printed by EHD and Optical microscopy (OM) images of EHD-printed GI layers of the metal–insulator-metal (MIM) capacitor with insulating material, reprinted from reference [24], Copyright (2021) American Chemical Society. **c** OM images of ZTO printed by EHD (semiconducting material) active pattern, reprinted from reference [27], Copyright (2014) American Chemical Society. **d** EHD-printed droplets of PMMA solution (semi conducting material) w. r. t. applied voltage, reprinted from reference [28]. **e** OM of Cu lines printed by EHD (conductive material) on ZTO with different stage speeds of 1000 to 8000 μm/s, reprinted from reference [29]. **f** Schematic of fabrication process using PEDOT:PSS for complementary NOT gate (conductive material) electrodes with various work functions (WFs), and EHD jet printed OM images of high work function (HWF) and low work function (LWF) of PEDOT:PSS electrodes on the substrate (Si/SiO2), reprinted from reference [33], Copyright (2020) American Chemical Society

to print indium oxide (In_2O_3) structures to fabricate TFTs with the improvement in device stability having good electrical characteristics. TFTs were successfully fabricated on plastic substrates, which showed an excellent mobility of ~230 cm^2/V.s with the use of high-k dielectrics (Fig. 3d), indicating that EHD jet printed In_2O_3 is a potential material to use in the various printed electronics [33].

Conductive materials

The literature has presented a wide range of alternative materials that can be used for the printing process with conductive properties, including various metals, i.e., nickel, gold, copper, silver, and palladium. Additionally, carbon-based materials like graphene, carbon nanotubes, and carbon black have been explored as potential options for the printing process. Furthermore, conductive polymers such as polyacrylic acid (PAA), polycaprolactone (PCL), and poly (3,4-ethylenedioxythiophene): polystyrene sulfonate (PEDOT: PSS) have also been investigated [34–38]. Thuy et al., primarily investigated copper (Cu) electrode patterns printed by EHD having a high resolution of 40 µm for TFT application as depicted in Fig. 3e [34]. In another study, the fabrication of contact organic thin-film transistors (OTFTs) with a high resolution of 5 µm was achieved through the utilization of the EHD jet printing of silver (Ag) electrodes [35]. The OTFTs were able to produce a high current output due to the extremely reproducible nature of the printing process and the small differences between individual devices. The printing of source and drain electrodes for OTFTs involved the utilization of PEDOT: PSS electrodes which were recognized as one of the most conductive polymers. By adjusting the work function of the electrodes, specifically by achieving a high work function for one electrode and a low work function for the other, the resulting OTFTs exhibited effective charge transport and enabled the development of logic circuits [38]. These modified work functions of PEDOT: PSS electrodes were used to create superior electrical characteristics in OTFTs (Fig. 3f).

Biomaterials

Numerous biological applications exist for conventional inkjet printing, but when printing biomaterials, the resolution is poor, which is a drawback. Therefore, high-resolution patterns of various biomaterials, including hydrogels, proteins, and DNA, are attempted by jet printing techniques. The utilization of the EHD jet printing technique has the potential to enhance the resolution of printed biomaterials by increasing the density of dots within a given unit area [39]. Researchers applied EHD printing to create DNA probes with nanoliter-scale droplets on the surfaces of DNA microarrays [40]. A wide range of complicated single and double-stranded DNA configurations were printed to develop aptamer-based biosensors. This biosensor was designed to facilitate the detection of adenosine molecules by fluorescence analysis [41]. The EHD jet printing technology enabled the deposition of living cell suspensions onto surfaces while preserving the integrity and functionality of the cells [42]. In another study, a wide range of complicated single- and double-stranded DNA patterns were created for simple aptamer-based biosensors to detect adenosine molecules' fluorescence. Kim et al. also studied the EHD jet printing technology to deposit living cell suspensions on surfaces without affecting the structure and functions of cells. The EHD direct printing can create 3D structures of cell-adhesive biomaterial such as collagen, as well as patterns of live bacteria [43]. Different biomaterials, mixing living cells with hydrogels [44–46], polyurethane and poly(methylsilsesquioxane) polymers [47], and polydiacetylene-embedded polystyrene nanofiber [48] have also been reported in the literature.

EHD jet printing of nanomaterial composites and applications

Transistors and displays

Previously we discussed different materials printed by EHD printing technology and their applications in different fields. In this section, we will overview different nanomaterial composites printed by EHD printing technology and their applications in various fields. EHD jet printing method helped the precise printing of metal oxide semiconductors, resulting in its effective implementation for the production of transistor arrays and integrated circuits. Zinc oxide (ZnO) nanorods exhibited notable electron mobility and catalytic efficacy, making them a highly prospective substance for augmenting the electrical characteristics of functional devices. The patterning technique of the substrate with ZnO was described by Zhang et al. where they utilized EHD jet printing to deposit composite filaments consisting of polyethylene oxide-zinc nitrate (PEO)-Zn $(NO_3)_2$. Subsequently, the substrate was subjected to a heating process, resulting in the decomposition of (PEO)-Zn $(NO_3)_2$ and the formation of ZnO nanorods [49], as depicted in Fig. 4a. In another study, EHD printing was applied to print electrodes in p-type OTFTs, where the printed lines of multiwalled carbon nanotubes/polystyrene sulfonate (MWCNT/PSS) showed remarkable electrical properties, as depicted in Fig. 4b. Conversely, the MWCNT/Triton X-100 (TX-100) lines had exceptional electrical characteristics, as illustrated in Fig. 4c [50]. The EHD printing process was also studied to shape straight lines

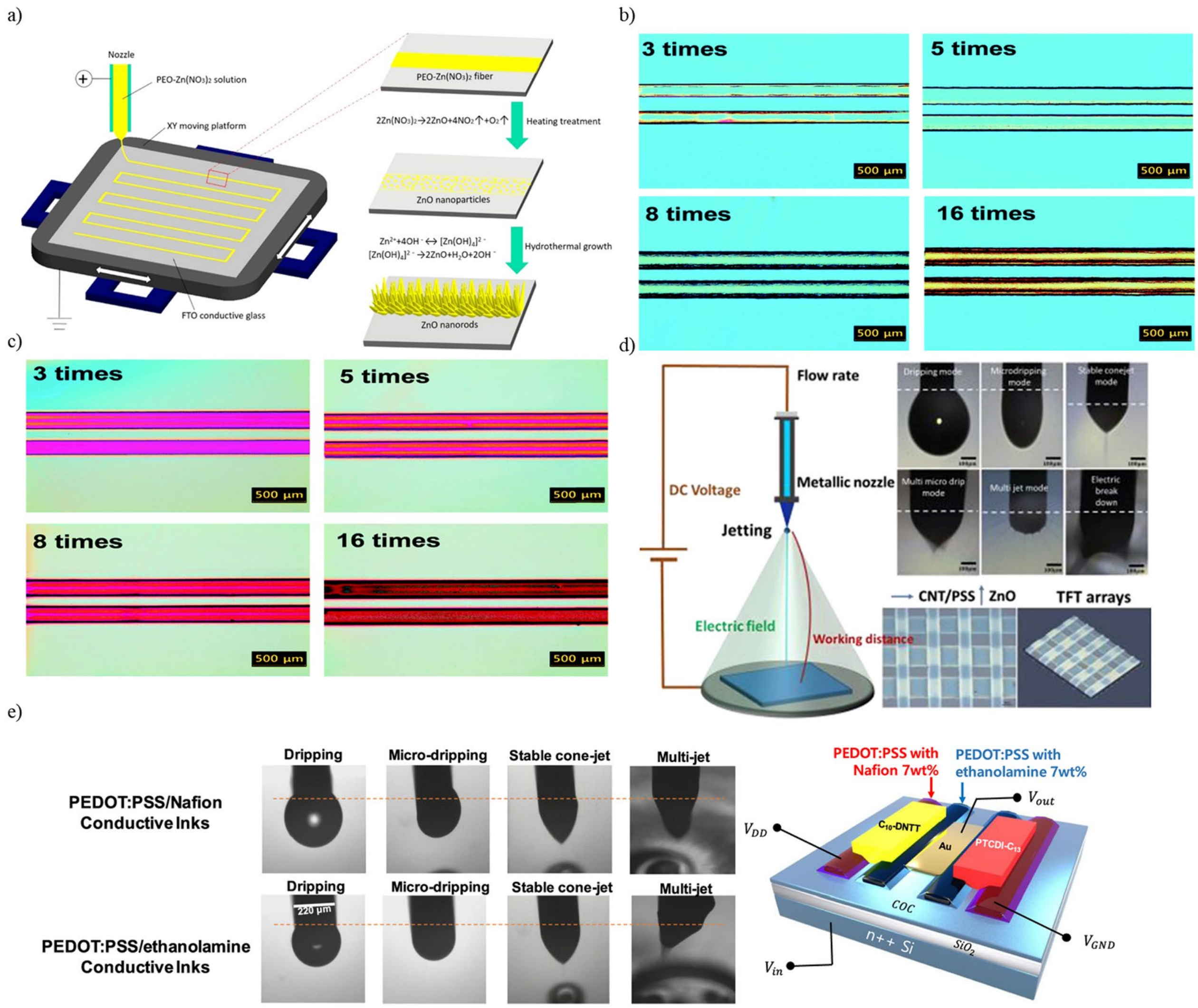

Fig. 4 Nanomaterial composites fabricated by EHD printing technology for transistors. **a** Schematic of ZnO nanorods patterned by the EHD printing process, reprinted from reference [44] b and **c** MWCNT/TX100 and MWCNT/PSS lines printed by EHD, reprinted from reference [45]. **d** Schematic of printing process and EHD-printed OM images of ZnO: Three types of S/D electrodes with EHD-printed in TFT arrays by MWCNT/PSS and Ag nanoparticles, with conductive PEDOT:PSS, reprinted from reference [46]. **e** OM images of conductive inks for PEDOT:PSS/Nafion and PEDOT:PSS/ethanolamine for four different EHD printing modes along with organic complementary inverter composed of an n-type and a p-type electrodes, reprinted from reference [47], Copyright (2020) American Chemical Society

of ZnO semiconductors, which were derived from zinc acrylate. These straight lines were then utilized as the active layers of TFTs with satisfactory outcomes [51]. The devices that are produced have TFT characteristics, and the use of a minor quantity of indium doping has the potential to enhance their overall performance. In addition, the study involved the printing of three various conducting materials onto pre-patterned ZnO substrates. This was done to create ZnO thin-film transistor (TFT) arrays that incorporate semiconductors and source/drain (S/D) electrodes that are directly drawn onto the substrate, as depicted in Fig. 4d.

The EHD printing was also employed for the fabrication of source and drain electrodes in PEDOT: PSS-based composites, catering to both p-type and n-type OFETs [52]. Nafion and ethanolamine were employed as modifiers to enhance the functionality of PEDOT: PSS. The resulting composites, namely PEDOT: PSS/Nafion and PEDOT: PSS/ethanolamine, were then printed using the EHD-jet printing technique. This allowed for the creation of asymmetric source and drain electrodes in OFETs, as depicted in Fig. 4e. The printed asymmetric electrodes, consisting of both p-type and n-type materials, showed excellent performance in terms of inverters and displayed favorable voltage transfer characteristics. Liu et al. employed the EHD jet printing technique to fabricate high-resolution micropatterns consisting of a composite material composed of Cesium Lead Bromide ($CsPbBr_3$) quantum dots (QDs) and PCL. This composite material was intended for applications in light-emitting optical fiber and flexible display technology [24]. The use of the EHD printing process enabled the production of stretchable nanofibers that produce green light. These nanofibers on flexible PDMS substrates were composed of hybrid inorganic bases and were found to exhibit robust green luminescence and water stability when encapsulated within a PCL polymer matrix, as depicted in Fig. 5a and b. In another research, researchers employed EHD jet-printing to create composite patterns of methylammonium lead bromide/polyacrylonitrile $MAPbBr_3$/PAN that were translucent, flexible, and stable. The resulting patterns exhibited a high resolution of around 10 µm, as demonstrated in Fig. 5c and d [53]. Moreover, the researchers demonstrated that the nanomorphology of the pattern surfaces could be controlled. In addition, the processing conditions and printing inks were further optimized to ensure the exceptional properties of optoelectronic of the $MAPbBr_3$/PAN composite patterns for flexible display applications, while also achieving a stable EHD jet printing process.

Energy harvesting and storage

EHD printing has been extensively used to fabricate devices such as capacitors and batteries for energy-harvesting applications. In one study, EHD jet printing was employed in the fabrication of electrochemical energy storage devices to control the porosity of a composite material consisting of microscale carbon and nickel (C-Ni), as depicted in Fig. 6 [19]. A flexible zinc ion battery (ZIB) system of manganese selenide (MnSe)/reduced graphene oxide (rGO) was developed by EHD, which was connected to a touch-controlled light-emitting diode (LED) [54]. In this study, the researchers used a high-precision EHD jet printing technique to fabricate flexible miniaturized energy storage devices for MnSe/rGO. These devices were then coupled with an LED array

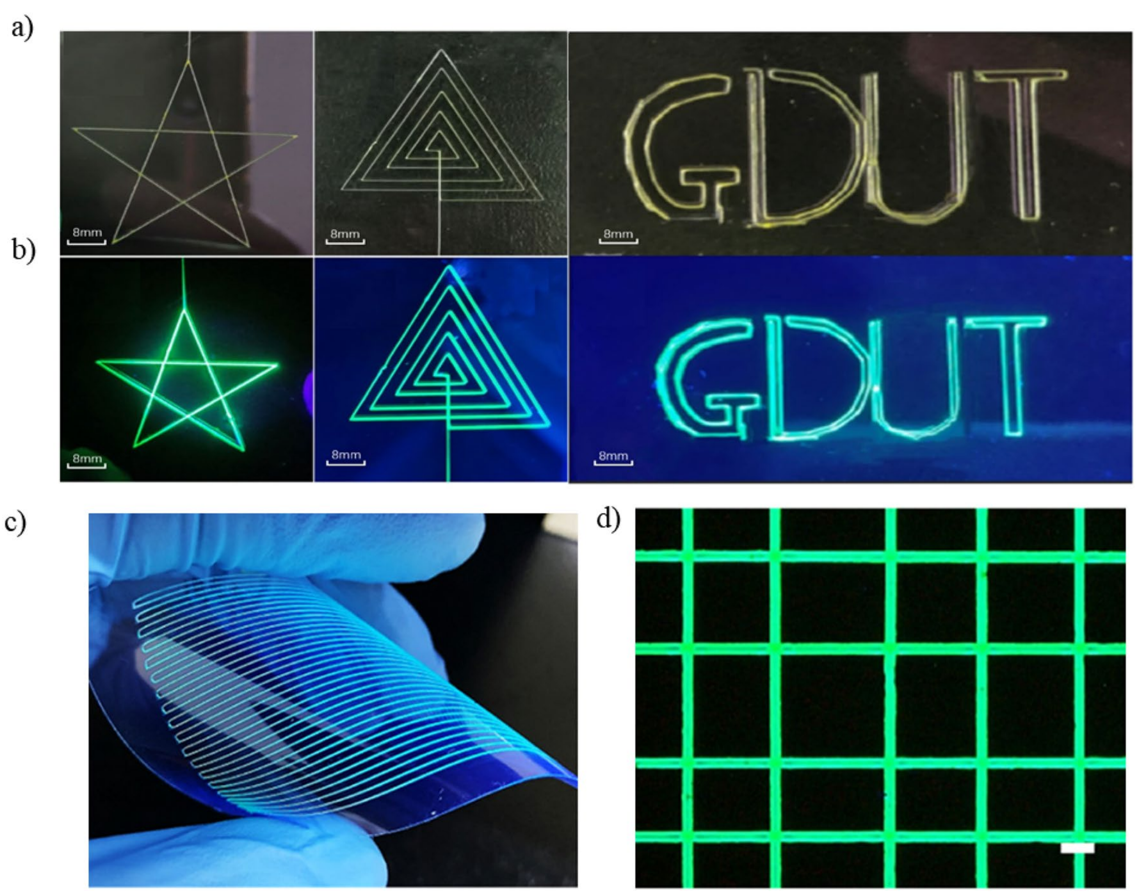

Fig. 5 Nanomaterial composites fabricated by EHD printing technology for displays. **a** Printed high-resolution patterns (stacking 15 layers of fibers). **b** Pattern printed under ultraviolet light, reprinted from reference [48]. **c** Flexible and transparent micropatterning of MAPbBr3/PAN composite ink on a PET substrate. **d** Under UV light, the emission of mesh and line patterns, reprinted from reference [49]

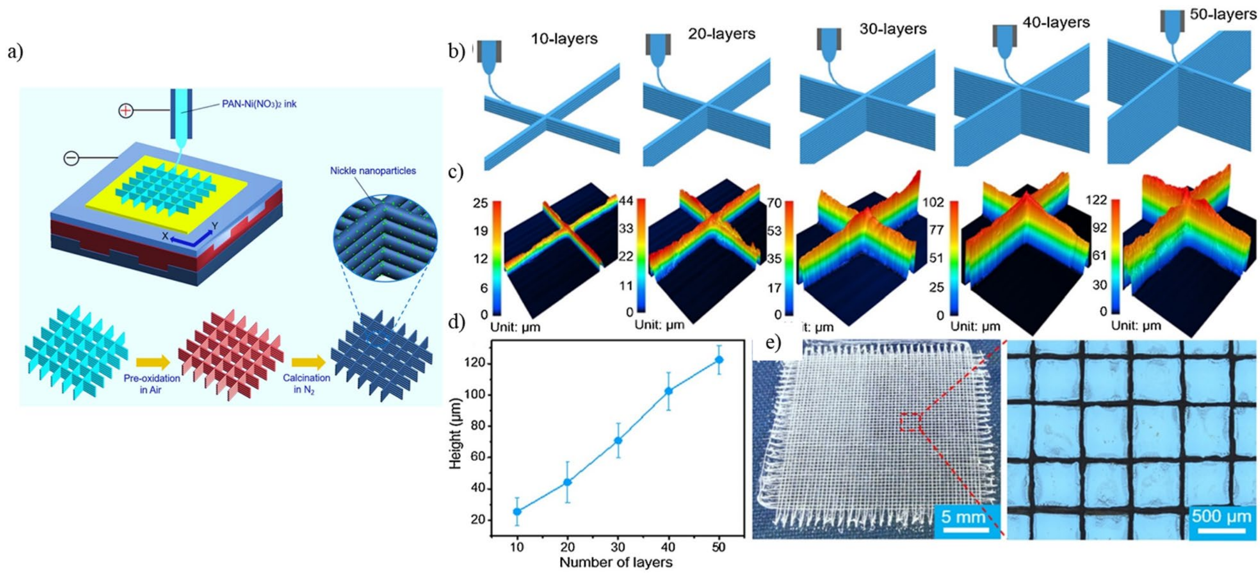

Fig. 6 Filter's components fabrication on a substrate. **a** EHD 3D printed schematic illustration of C-Ni composite electrodes. **b** EHD-printed 3D structures schematic diagram with 10 to 50 layers. **c** Laser confocal scanning 3D profiles of printed structures. **d** The structural height w. r. t. the no. of layers. **e** OM of 50-layers lattice structure having a space of 500 μm gap, reprinted from reference [50]

system to highlight the potential application of flexible EHD jet-printed micro-batteries, as depicted in Fig. 7a. The rGO's mechanical stability further ensured that flexible ZIBs could be printed using an EHD jet to create a flexible, integrated functional system. Molybdenum disulfide (MoS_2), an excellent two-dimensional building part, is a good candidate for lithium-ion battery (LIB) anode in complete cell LIB configurations. Wei et al., chemically exfoliated MoS_2 powders in bulk into ce-MoS_2 and then re-dispersed in a solution for EHD jet printing [55]. A functional layer of an anode was printed by EHD jet printing for electrolyte-supported solid oxide fuel cells. An electrolyte plate was used to print ink containing 10% scandia-stabilized zirconia (10% ScSZ) and nickel oxide for solid oxide fuel cell (SOFC) [56]. In addition, a comparison was made between the performance of the EHD jet-printed cell and the screen-printed cell. Surprisingly, the EHD jet printed cell showed superior performance despite the significantly thinner thickness of the EHD jet printing, which was found to be 7 to 10 times thinner than that of screen printing. Biohybrid photo-electro protein micro-capacitors were fabricated on a non-conductive PET film substrate through the utilization of EHD printing. These micro-capacitors

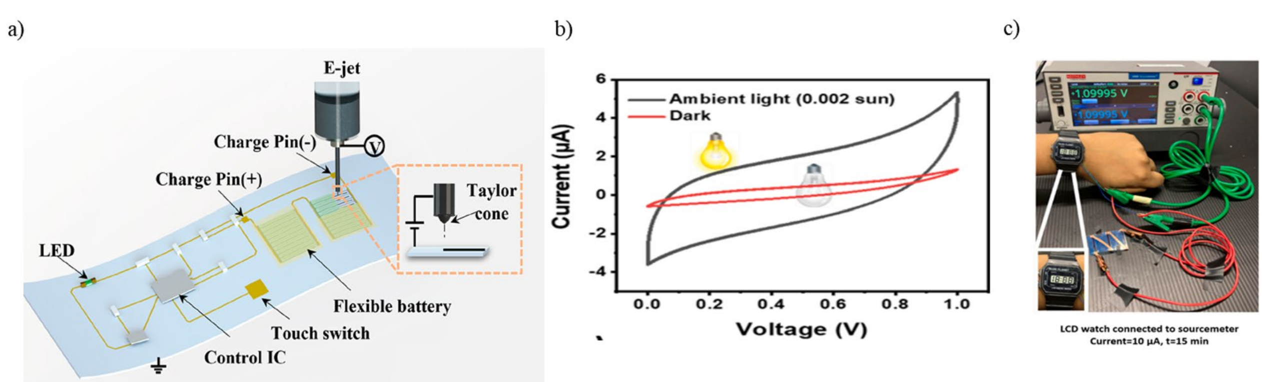

Fig. 7 Nanomaterial composites fabricated by EHD printing technology for energy harvesting and storage. **a** EHD jet printing process schematic for a flexible ZIB device, reprinted from reference [51], Copyright (2023) American Chemical Society. **b** and **c** CV profiles of a standard MC under in the dark and indoor light and external charges injected by applying constant current of 10 μA connected to LCD watch, reprinted from reference [54]

exhibited exceptional uniformity in performance and demonstrated remarkable operational stability [57]. The biohybrid micro-capacitors were subjected to optimization procedures to achieve a targeted specific capacitance of 110 mF/g and a scan rate of 10 mV/s. The efficacy of these micro-capacitors was determined to be satisfactory for supplying power to a nano/microelectronic device, as depicted in Fig. 7b and c. In this way, EHD-printed solid-state devices can be employed within the domain of portable, flexible, and wearable electrochemical energy storage systems.

Electrodes and sensors

EHD printing was also applied to fabricate different electrodes and sensors for various applications. In one study, EHD jet printing was applied to print methyl-red/graphene composite sheets with a thickness of 300 nm, which have been deposited on silver electrodes [20] (Fig. 8a). In another study, the composite electrodes were successfully employed as a humidity sensor by quantifying the change in electrical resistance, which demonstrated an inverse correlation with relative humidity. The great sensitivity (96.36% resistive and 2869500% capacitive sensitivity against humidity) of the sensor was achieved by depositing a composite layer of methyl-red/graphene onto silver electrodes. The fabrication of graphene/Pt (G/Pt) composite microelectrodes involved the utilization of the drop-on-demand (DoD) EHD printing technique. This method enabled the printing of graphene lines with a thickness of 5 nm onto Pt microelectrodes. It was observed that the resistance of 4.2 mΩ cm [58] was achieved for each printed line. According to the electrochemical test, the peak current of microelectrodes of G/Pt composite was found to be more than twice as large compared to bare Pt microelectrodes. The microelectrodes of G/Pt significantly enhanced sensing sensitivity, making them suitable for high-performance electrochemical sensors, as shown in Fig. 8b.

The EHD jet printing was also employed to fabricate self-healing electrodes suitable for dielectric elastomer actuators using a gelatin-based composite that incorporates conductive ions and hydrogen bonds [21]. The

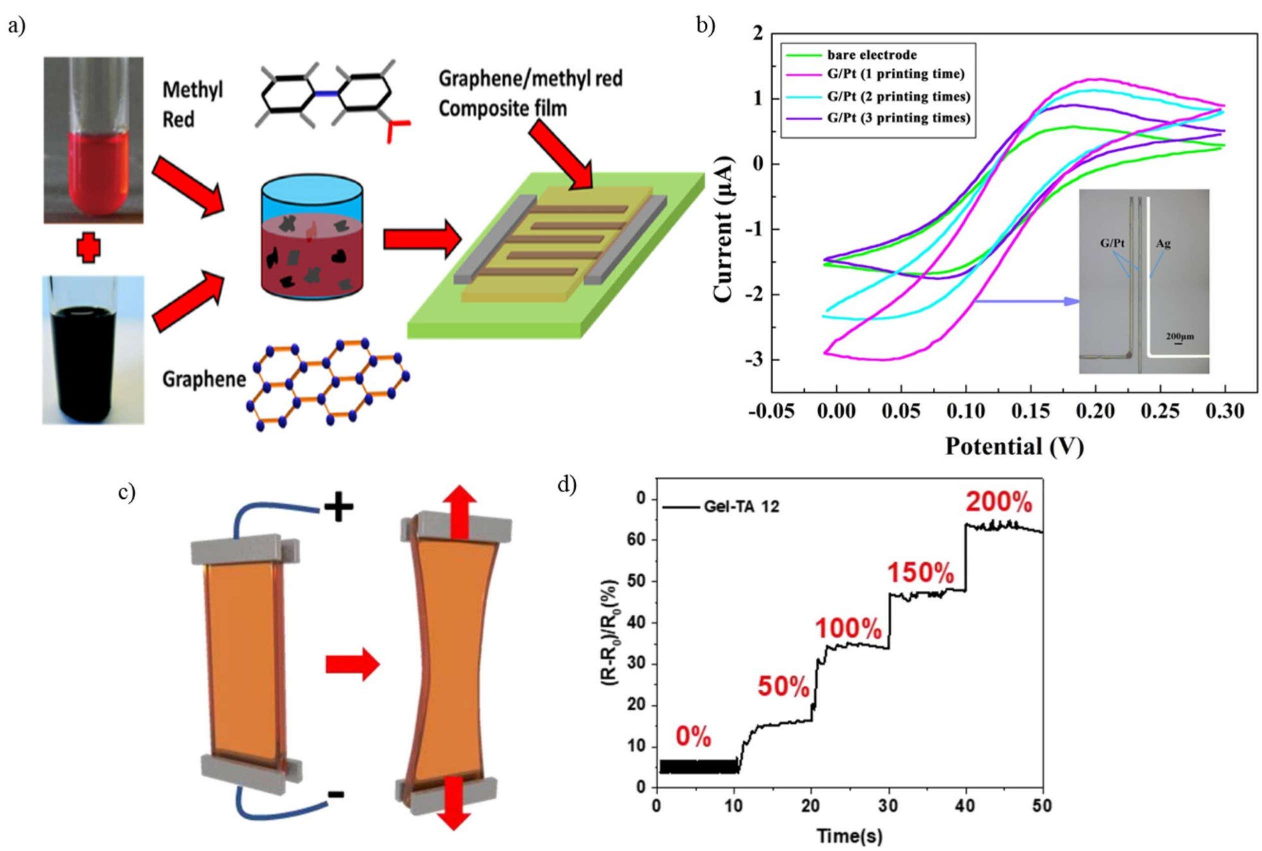

Fig. 8 Nanomaterial composites fabricated by EHD printing technology for electrodes and sensors. **a** Graphene/methyl-red composite-based humidity sensor schematic representation, reprinted from reference [55]. **b** Cyclic voltammetry curves of the G/Pt microelectrodes, reprinted from reference [56]. **c** Strain sensor schematic illustration. **d** Depending on the tensile strain, the plot of the changes in resistance of the strain sensor Gel-TA12, reprinted from reference [57]

process of fabricating dielectric elastomer actuators involved the application of gelatin-based electrodes onto elastomeric substrates through the EHD printing technique. The actuators exhibited satisfactory performance and showed a remarkable ability to regain their functionality, achieving an efficiency of up to 96.8% despite sustaining damage to the gelatin-based electrodes. In addition, the potential suitability of the gelatin-based electrode as a strain sensor was also examined, wherein a tensile stimulus was converted into an electrical resistance signal, as depicted in Fig. 8c and d. EHD printing was employed as a technique for fabricating cost-effective foldable electronics with environmentally benign attributes, which involved the printing of circuits composed of a composite material consisting of PEDOT: PSS and graphene onto films made of polyvinyl alcohol (PVA) [59]. Another study investigated the influence of different weight ratios of graphene and PEDOT: PSS inks on the morphology and electrical characteristics of printed patterns, employing diverse printing circumstances. Moreover, a resistive temperature sensor was examined in the form of a printed PEDOT: PSS/graphene circuit, which was used to monitor body temperature and respiratory behavior. The application of EHD jet printing was employed to print anisotropic conductive films (ACFs) that are used for bonding in communication devices and displays. The final results of this process were shown to be significantly affected by the characteristics of the ink and the conditions under which the printing was conducted [60]. Hence, to assess the viability of utilizing EHD printing technology for the production of ACFs, polymer inks suitable for EHD ink jet printing were formulated by incorporating conductive particles as the primary constituent. The findings showed a satisfactory level of electrical conductivity in the ink formulation having conductive particles.

Biomedical applications

The applications of EHD printed nanomaterial composites encompass a wide range of fields, including electronic devices, as well as bio and health-related devices such as living tissues, rug delivery, and health monitoring [22, 57, 61]. To generate microscale living tissue patterns, a combination of RGD (GGGGRGDSP)-functionalized alginate and fibrin system (RAF) was employed alongside PEDOT: PSS to create an electro-conductive hydrogel as shown in Fig. 9 [61]. Three-dimensional (3D) EHD printing was employed in the fabrication of a composite membrane that incorporates both hydrophilic and hydrophobic molecules. This flexible multi-drug composite membrane consisted of two distinct sections, namely cellulose acetate-ibuprofen and cellulose acetate-paracetamol, with an intermediate folding component composed of PCL [22]. To help the process of swallowing, the composite membranes possess the capability to be folded and conveniently housed into commercially available capsules. The composite membrane exhibited biocompatibility and flexibility, hence enabling its utilization in diverse applications such as drug combination therapy and personalized medicine.

The EHD jet printing technology was used to create organized patterns and intricate patterns by employing two polymeric materials, namely polyurethane (PU) and poly (methylsilsesquioxane) (PMSQ), and coarse processing needles. The results underscore the promise of this approach for the direct 3D fabrication of biological polymers and composites, with implications for a diverse array of biomedical applications [47]. A melt-based EHD printing method was developed to fabricate microfibrous scaffolds composed of PCL and MWCNTs [62]. The incorporation of MWCNT into PCL did not significantly affect the stability of melt-based EHD printing, however, it lowered the impedance of fibrous scaffolds. The mechanical characteristics of microfiber PCL/MWCNTs composite scaffolds printed by EHD printing were found to be similar to those of pure PCL scaffolds. Moreover, these composite scaffolds exhibited favorable cytocompatibility, as evidenced by their ability to support cellular spreading and proliferation in laboratory settings. The technique that has been revealed exhibits significant potential in the uniform integration of functional bionanomaterials into EHD-printed microfibrous architectures, intending to achieve specific biological functionalities. In another study, to achieve a more accurate replication of collagen fibers and hydroxyapatite nanocrystals found in actual bones, composite scaffolds consisting of microscale PCL and hydroxyapatite nanoparticles were fabricated by EHD 3D printing [63], where the composite scaffolds were biocompatible and aided cell alignment and proliferation in vitro. This approach had the potential to effectively regulate the cellular microenvironment across various sizes and materials, hence enhancing tissue regeneration.

Others

To achieve the desired characteristics of fast response, effective control, and safe interactions between humans and machines in low-strength magnetic fields, the researchers utilized an advanced four-dimensional EHD printing technology to order and deposit soft magnetic composites [64]. The EHD-printed memristor was constructed using a composite of graphene quantum dots (GQDs) and poly 4-vinylphenol (PVP) to attain a state of high stability and linearity, where the dielectric layer of the capacitor as depicted in Fig. 10 [65]. The EHD printing method offered a convenient and effective method

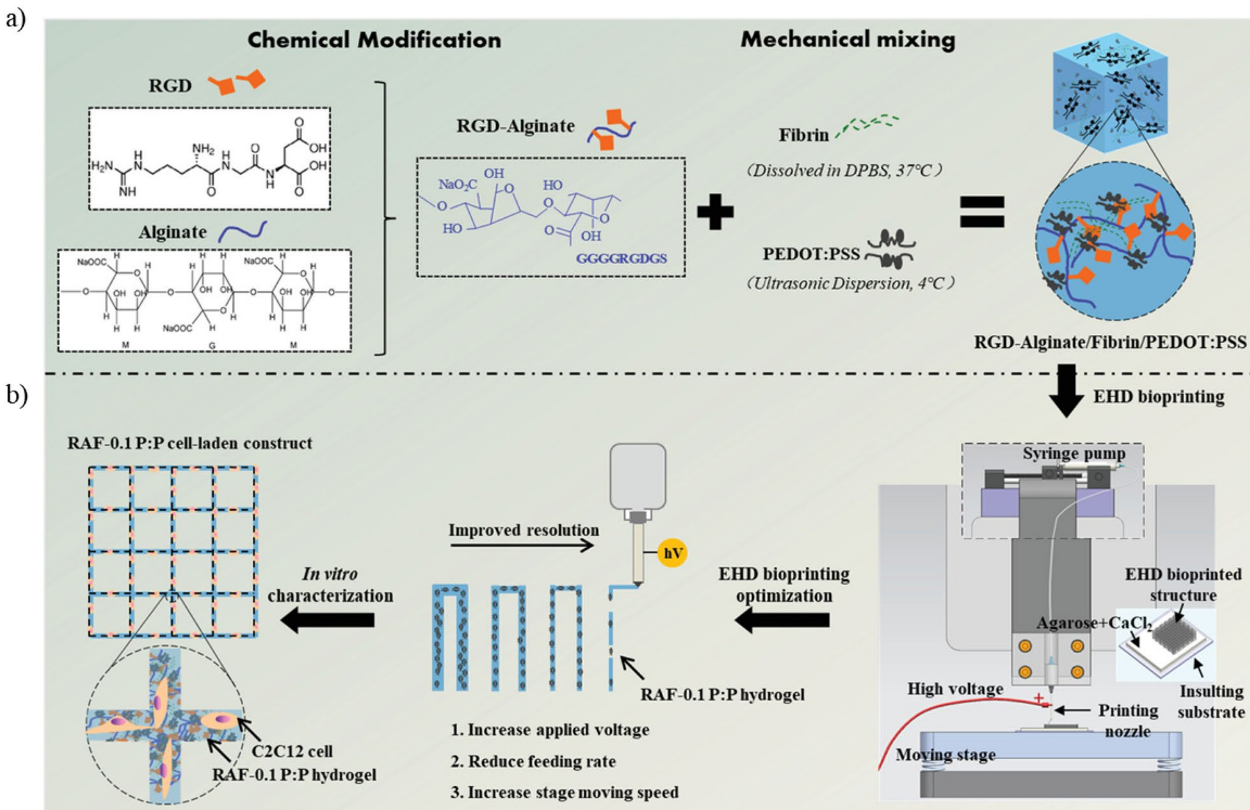

Fig. 9 Nanomaterial composites fabricated by EHD printing technology for biomedical applications. **a** Electro-conductive bioink synthesized schematic. **b** EHD-bioprinting schematic of electro-conductive bioink, reprinted from reference [60]

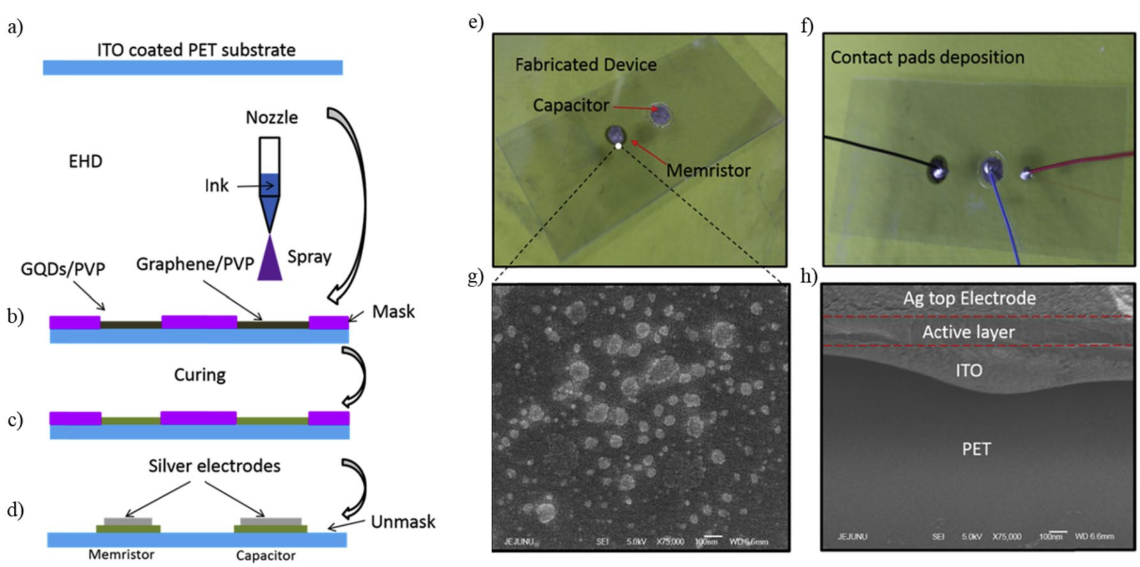

Fig. 10 Filter's components, fabrication steps on a substrate. **a** Substrates of ITO coated PET. **b** Memristor and capacitor deposition of sandwich layers on the substrate of ITO coated PET. **c** Curing condition of sample. **d** Depositing top electrodes as well as unmasking. **e** Capacitor and memristor fabrication. **f** Proposed device along with contact pads. **g** Memristor (GQDs/PVP) active layer SEM image. **h** memristor cross-sectional view showing three layers, reprinted from reference [64]

for fabricating soft magnetic composites, hence enabling their use in several prospective applications, including actuators. A hybrid conductive ink with excellent printability and stability was synthesized through the incorporation of polyaniline (PANI) nanoparticles into a composite of silver flake and thermoplastic polyurethane (TPU) using an EHD jet printing technique [66]. The flexible heaters printed by EHD demonstrated exceptional flexibility and endurance, boasting a remarkable resolution of 45 µm. The heater's resistance exhibited remarkable mechanical stability after undergoing outer bending cycles of 3000 with a 0.5 mm radius. The heater can also be affixed to an individual's body, showing the potential for novel wearable electrical applications.

In another work, the process of EHD printing was employed to fabricate a hybrid structural adjustable lens, which was afterward powered by dielectric elastomer actuators (DEA) during the printing process [67]. In order to fabricate the dielectric elastomers (DEs) driven component, a highly dielectric silicone rubber (SR) based ink was employed to form the encapsulating layer of the lens, which was loaded with copper phthalocyanine (CuPc). The tunable lens that has been produced exhibited the ability to automatically change its focal length in a manner like to the human eye. This innovation has significant promise for a range of applications including imaging, information storage, beam steering, and bifocal technology. The successful preparation of lead zircon titanate (PZT) composite films using EHD printing where another PZT thin films with varying thicknesses of 362, 725, and 1092 nm were fabricated on a Ti/Pt bottom electrode via the sol–gel technique. PZT composite films were subsequently fabricated by employing an EHD printing technique to deposit PZT thick films over the pre-existing PZT thin films [68]. The hybrid method possesses considerable potential for widespread application in the fabrication of PZT composite sheets intended for utilization in micro-nano devices. Nanomaterial composites printed by EHD printing technology have also been extensively studied in the literature (Table 1).

Conclusion and future directions

The present work conducted an extensive overview of the underlying principle of EHD jet printing and its applications in different fields, with a particular focus on the prospective utilization of nanomaterial composites as functional inks. The application of EHD jet printing allows for the production of complex patterns

Table 1 Nanomaterial composites printed by EHD jet printing technology

No	Composites	Applications	References
1	ZnO/MWCNT/PSS	TFTs arrays	[51]
2	MWCNT/PSS	OFETs	[69]
3	Cu/PEO	OFETs	[70]
4	CB/TX-100	OFETs	[71]
5	AgNWs/PEO	OFETs	[72]
6	PEDOT: PSS/CNT	OTFTs	[73]
7	CNT/graphene	OFETs	[74]
8	Carbon/Nickel	Supercapacitor	[19]
9	Graphene/PVP	Capacitor	[65]
10	PANI/carbon nanofiber	Hybrid capacitors	[75]
11	MoS_2 & PEO	Humidity sensors	[76]
12	PVDF/SWCNTs	Pressure sensor	[77]
13	PCL/graphene	Sensor	[78]
14	PVP/PEO	Anti-microbial drug	[79]
15	PEDOT: PSS/GR/SWCNTs	Health monitoring	[10]
16	PEDOT: PSS/RGD	Living tisue	[61]
17	PCL/PEG/ROX*	Bone tissue engineering	[80]
18	PCL/PEO/Fe_3O_4 *	Fibers in wound dressings	[81]
19	PCL/PEO/Graphene	Nerve restoration and regeneration	[82]
20	$Ni_3Al–Cr_3C_2$	Solar films	[83]
21	Al_2O_3/ITO	IoT devices	[84]

* PEG (polyethylene glycol)

* ROX (roxithromycin)

* Fe_3O_4 (iron oxide)

using different materials as well as nanomaterial composites at the micro to nanoscale, hence enabling the manufacturing of devices with high precision, wide coverage, and efficient production ability.

The commercialization of printed electronic devices in various industrial areas is now hindered by several drawbacks (external and internal) of the EHD jet printing technology. The external factors that affect the EHD jet printing technology are humidity and temperature which adversely disturb the ejection procedure and performance of flexible electronic devices. Moreover, the internal factors that influence the EHD printing technology are uniform electric field, functional ink properties, and handling of the system. Ensuring repeatability is a significant challenge in the realm of commercialization, as it becomes difficult to adequately control the mixability and precise proportions of various ingredients to make nanomaterial composite inks in EHD printing technology. Another issue for the commercialization is the viability of high-resolution flexible electronic devices produced by EHD jet technology which are constrained in certain domains such as soft robotics, intelligent healthcare monitoring, and human–machine interaction mostly due to their inadequate repeatability and sensing stability. Hence, a significant task and potential for high-resolution flexible electronics lies in the development of distinctive materials, techniques, and technology for EHD jet printing, with the aim of creating reliable and expensive devices. However, the EHD printing technology exhibits considerable appeal from multiple perspectives and is currently garnering significant attention. Therefore, it is expected that the advancements achieved in several facets of EHD jet printing technology will expedite the commercialization of printed devices in the future.

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Data Availability
Availability of data and materials is not applicable for this article since this is a review paper.

Declarations

Competing interests
The authors declare that they have no competing interests.

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