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Recent Progress of Printing Memristors: Material, Method, and Performance

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Abstract. This review article explores recent advancements in printable memristors (pMRs), focusing on materials, fabrication methods, and performance metrics. As the demand for high-efficiency computing grows in fields such as big data and artificial intelligence, conventional computing architectures face limitations due to transistor miniaturization and the von Neumann bottleneck. Memristors, serving as fundamental components of neuromorphic computing (NMC), have gained significant attention due to their simple two-terminal structure, low power usage and simple manufacturing. The article provides a comprehensive discussion on materials for printed electrodes (e.g., metal inks, carbon-based inks, conjugated polymers, and conductive metal oxides) and dielectric layers, highlighting their impact on device performance. Furthermore, various printing techniques, covering inkjet printing, screen - type printing, aerosol jet - based printing, electrohydrodynamic (EHD) jet printing, 3D printing, spin coating, gravure printing, flexographic printing, and doctor blade coating, are examined in terms of their principles, advantages, and applications in pMR manufacturing. This study is intended to act as a meaningful reference resource for scholars engaged in memristor-related and neuromorphic computing research domains.

Keywords: Printable Memristors, Printing Techniques, Electrode and Dielectric Materials

1. Introduction

Today, high-efficiency computing is becoming more and more prominent in fields, such as big data and artificial intelligence. It helps improve technology products with faster processing speed and better computing performance. However, the demand for big-data in human practice is also increasing, and conventional computing architecture cannot completely satisfy all the requirements. Specifically, from the device level, the size of the transistors in the chip now is close to the physical limitation. It becomes difficult to cram more transistors into a chip to achieve better performance. Since the 1990s, the downsizing of devices has persisted steadily, yet following trajectories that grew more and more divergent from Dennard scaling principles [1] At the system level, the constraint posed by the von Neumann architecture has highlighted the restriction on data processing rates between a computer's central processing unit and its memory. Neuromorphic computing (NMC) has come forth as a solution to tackle these limitations. At its core lies the memristor (MR), a device that mimics both volatile memory functionality and the computational behavior of biological neurons—serving as a foundational building block for NMC systems. Recently, printed

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memristors (pMRs) have emerged as a focal point in the field, owing to their simplified two-terminal design, facile fabrication processes, and low power requirements [2]. this review consolidates the latest advancements in pMRs. It encompasses materials for printed electrodes and dielectric layers, printing methodologies, and resultant device performance. We hope this review could provide useful guidance to the MR and NMC field.

2. Different materials of the pMR

Typically, selecting materials for both electrodes and dielectric layers in pMRs exerts a pivotal influence on defining memristor behavior, switching dynamics, and overall device functionality. In this section, we summarize the pMR from the printing electrode and dielectric layer separately.

2.1. Technology printing electrode

1)Metal Inks: Metal-based inks are a cornerstone of pMR electrode fabrication. Silver (Ag) inks, available as nano-dispersions for droplet-based techniques or pastes for screen printing, are widely used [3]. For example, Kosmala et al. (2011) developed a Ag nanoparticle ink via silver ion reduction with hydrazine in the presence of sodium bis(2-ethylhexyl) sulfosuccinate (AOT), followed by dispersion in water using a PEO–PPO–PEO triblock copolymer and high-intensity focused ultrasound treatment [4]. Noh and colleagues (2012) delved deeper into the compatibility of silver pastes, examining electrochemical migration (ECM) phenomena on silicon substrates featuring screen-printed silver-epoxy circuits under a range of sintering parameters [5]. Copper (Cu) inks, with lower sintering temperatures (<100°C), provide an economically viable option; Duraisamy and colleagues (2012) demonstrated Cu electrode fabrication using electro-hydrodynamic printing [6].

2)Carbon-Based Inks: Carbon-based conductive inks comprise graphite, carbon black, and emerging carbon allotropes like graphene and carbon nanotubes [7]. In 2018, Islam's research team showcased a straightforward, scalable, and cost - effective approach to fabricate functional carbon black (CB) ink. The method involves dry and wet medium blending of burned charcoals. This process cuts down on both the processing time and the need for sophisticated manufacturing instruments [8].

3)Conjugated Polymers: Conjugated polymers comprise unsaturated moieties (e.g., benzene rings or acetylene units) linked by single bonds with partial π -bond character, forming extensive domains of delocalized, polarizable π -electrons. These materials have been integrated into polymer-based electronics, including field-effect transistors and rechargeable batteries [9]. In 2015, Worfolk and colleagues achieved the large-scale production of highly conductive PEDOT-polystyrene sulfonate thin layers through solution shearing methods. This approach yielded an impressive electrical conductance of 4,600 ± 100 siemens per centimeter, with the films maintaining strong light transmittance [10].

4) Conductive Metal Oxide Inks: Owing to their superior electrical conductivity relative to other ink types, metallic oxide nanoparticle inks are frequently employed for creating conductive circuits and patterns [11]. In 2020, McGhee and their research group engineered and produced capacitive humidity sensors for printed electronics. They utilized specially developed conductive indium tin oxide inks and dielectric aluminum oxide inks tailored for screen printing [12].

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2.2. Printing dielectric layer

1) Metal Oxides: Metal oxides are of immense technological value in electronics, as they can produce charge carriers when activated by a suitable energy source [13]. On the other hand, Gale and team members reviewed Resistive Random Access Memory (ReRAM) with a focus on titanium dioxide in 2014. They identified two primary mechanisms underlying the resistance switching and memristance in TiO_2-based devices: ionic and thermal processes. The ionic mechanism relies on ion migration—such as oxygen ion movement—to render TiO_2-based ReRAM conductive. The thermal mechanism achieves the same conductive state through Joule heating: upon the application of an electric field, an electric current is generated, which heats the material and induces structural changes [14].

2) 2D Materials: 2D materials have ultrahigh surface area-to-volume ratios and have anisotropic properties, resulting in unique material properties due to the quantum confinement they experience. When combined with the distinct family - associated properties, 2D materials further expand and diversify their characteristics, for instance, transforming into semiconductors [15]. They offer great flexibility in terms of tuning their electronic properties [16]. Based on these unique physical and chemical properties, 2D materials are critical in printed dielectric layers of pMRs, with broad application implications. In 2012, Sun et al. first synthesized freestanding, three-atom-thick SnS_2 monolayers via scalable exfoliation [17]. The same year, their team fabricated large-area, freestanding, four-atom-thick monolayers of non-layered ZnSe using a lamellar hybrid intermediate strategy. Under 300 W Xe lamp irradiation, it shows a photocurrent density of 2.14 mA/cm^2 at 0.72 V, 195-fold higher than bulk Ag/AgCl [18].

3) Organic Materials: Organic materials are an essential kind of material frequently used in field of display and electronic applications. In the past few years, they have been widely studied to improve its processability and tunability focusing on organic electroluminescence [19]. Additionally, in 2012, Roar and colleagues provided an overview of the Roll - to - Roll (R2R) techniques capable of producing organic thin films and indicated that certain issues, like improved registration control, remain to be addressed [20].

4) Hybrid Materials: As a rapidly emerging class of materials at the forefront of technological innovation, hybrid materials generate unique material properties through the synergistic integration of inorganic and organic components at the molecular level. The predominant types of organic - inorganic materials are generally categorized into three groups: intercalation compounds, organic derivatives of inorganic solids, and sol - gel hybrid materials [21]. For instance, leveraging the distinctive geometric nanostructure and electrical characteristics of carbon nanotubes (CNTs), when electroactive metal oxides were deposited onto CNTs, CNTs can not only facilitate the dispersion of oxide nanoparticles but also offer direct electron - conducting pathways [22]. The presence of the nanoparticle's aids charge transfer through trapping and hopping mechanisms [23].

4) Nanoparticles: Many physical phenomena in organic and inorganic materials involve inherent length scales of 1–100 nm (equivalent to 10^2–10^7 atoms). Owing to their nanometer - scale dimensions and finite size, nanoparticles exhibit unique electronic and optical properties for future applications. In 2000, Murray's group reported solution - phase synthesis and size - selective separation methods for semiconductor/metal nanocrystals, with sizes tunable from 1–20 nm and monodispersity $\leq$ 5%. These samples span molecular - to - bulk nanometer regimes, enabling systematic characterization of structural, electronic, and optical properties [24]. In 2004, Jaffar et al. demonstrated a technique to manipulate quantum dot (QD) surface chemistry and direct spatial organization for integration. They used charge - driven polyelectrolyte coating of carboxylated ZnS -

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capped CdSe QDs in aqueous solution to fabricate functional nanocrystals. Results showed individual polyelectrolyte shell coating of QDs [25].

3. Different printing methods of the pMR

3.1. Inkjet printing

Inkjet printing, a non-contact, digitally controlled technique, was gained traction for pMR fabrication due to its simplicity, low material waste, and versatility across industries—from textiles to biomedicine [26]. Two primary variants dominate: continuous inkjet (CIJ) and drop-on-demand (DoD). CIJ systems eject droplets continuously via an applied electric field and charging electrode, enabling high-speed printing but with lower resolution. DoD systems, in contrast, generate droplets through voltage waveforms (e.g., thermal or piezoelectric actuation), offering higher precision and compatibility with nanoparticle inks [27,28]. The CIJ system permits the collection and reuse of unprinted droplets. Owing to the high drop speed (around 25 m/s), rapid solvent evaporation, and the use of inks with volatile solvents, there is a relatively large gap between the printhead and the substrate. This also helps with adhesion on numerous substrates. It also comes with drawbacks: relatively low printing resolution, extremely high maintenance requirements, negative environmental impacts, and restrictions on the printed fluid, which must be chargeable. The DoD system, conversely, has distinct characteristics depending on its subcategories. Thermal inkjet printing offers advantages such as the ability to handle potentially small drop sizes and a high nozzle density. However, the fluid it uses must be able to vaporize and endure extremely high local temperatures. Similarly, piezoelectric inkjet printing achieves higher resolution but can only use conductive fluids, and the cost of implementing this technology is relatively high [33]. Most inkjet cartridges feature nozzles that are tens of microns in size. To avoid nozzle clogging, dispersed particles must be kept within a range below approximately ≈ 200nm [28].

3.2. Screen printing

Screen printing, which emerged in China during the Song dynasty, has undergone rapid and dynamic development [29]. Now, as one of the most popular printing technologies in the world, electrodes manufactured using it are widely used to produce printed electrodes, transistors, supercapacitors, anti-reflection coating (ARC) and batteries. Screen printing forms thin layers with custom 2D patterns using a mesh screen, where ink is evenly distributed via a squeegee. For manual operations, the mesh is first positioned at a precise distance above the substrate—misalignment at this stage can degrade pattern quality. Next, the requisite ink or paste gets applied onto the mesh and then spread uniformly via the squeegee; a reverse - direction second pass removes excess ink to regulate layer thickness. An improper distance might impair the printed pattern's quality. Next, the suitable ink or paste is pressed onto the mesh. With a squeegee, the ink should distribute evenly across the screen, and an opposite - direction second stroke eliminates surplus ink to achieve the right thickness. After this, the ink or paste transfers through the mesh's open regions, forming the desired pattern on the substrate. Finally, the screen must be cleaned to avoid ghosting and haze formation on images. However, robotics' rapid advancement has enabled machines to perform industrial screen printing, following methodology largely identical to manual processes. Screen printing can produce films with layer thicknesses ranging from 10–100μm REF _Ref205593323 \r \h [30].

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3.3. Aerosol Jet Printing

Aerosol Jet Printing (AJP), a printing method originally developed for electronic circuit fabrication, is now being explored for multiple uses: active and passive electronic parts, actuators, sensors, and various targeted chemical/biological reactions. AJP represents a contactless direct - writing method designed to produce high - resolution features on diverse substrates. Unlike traditional manufacturing or direct - writing techniques, AJP allows researchers to fabricate devices with greater geometric complexity, leveraging freeform deposition and a relatively large standoff distance [31]. It employs printable inks made of solutions or nanoparticle suspensions, containing metals, alloys, ceramics, polymers, adhesives, and/or biomaterials [32].. For the inks that AJP consumed during the printing process, they are required to have viscosity in the range of 1–1000mPas, particle size less than one micron and solvents with boiling points above [34].

3.4. Electrohydrodynamic (EHD) jet printing

Electro-hydrodynamic (EHD) jet printing leverages solution - based inks formulated with diverse functional materials to fabricate various structures. Owing to its compact form factor and mechanical adaptability, high - resolution printed flexible electronic devices enabled by EHD jet printing are widely applied in fields such as energy storage, intelligent healthcare monitoring, soft robotics, and intelligent human - machine interaction. Unlike traditional inkjet printers, EHD jet printing allows direct material deposition via specialized droplet - generation modes, including micro - dripping and cone - jet configurations. It boasts key advantages: high resolution, precise pattern fidelity, cost - effectiveness, and the capability to dispense high - viscosity solutions [35]. For the EHD jet printing process, it involves viscous spraying in the cone - jet mode that uses electrical forces to pull liquids from the nozzle tip, rather than applying thermal energy to push liquid from a fine capillary. The electric field induces charge accumulation at the liquid - cone surface, where the cone shape is determined by the combined force of the electric field and surface tension. As the electric field strength increases, electric stress at the cone tip surpasses surface tension, causing a liquid droplet or jet to be ejected toward the grounded substrate [36].

3.5. 3D printing

Designed to meet diverse product specifications and manufacturing demands, they differ from traditional subtractive printing methods. Instead, 3D printers adopt an additive process for pMR production. Fused Deposition Modeling (FDM) and Stereolithography (SLA) stand as two leading consumer - grade additive manufacturing processes. SLA leverages an ultraviolet light source to selectively cure resin, whereas FDM dispenses semi - liquid plastic in a predefined pattern to form objects [37]. F For conventional 3D printing, it relies on data formatted as STL, AMF, or other types to sequentially fuse or deposit thin material layers. Each layer is outlined by a series of closed contours that map the outer surface of the item being manufactured at the corresponding layer. 3D printers construct each layer by filling the region bounded by these contours with a material of a specified thickness, say 1mm [38]. A recent study by Bisht and team (2025) illustrated the promise of 3D printing for pMR arrays via fused filament fabrication (FFF). They used copper - polycaprolactone (Cu-PCL) filaments for electrodes and copper - polylactic acid (Cu-PLA) for the active layer, realizing bipolar switching with set/reset voltages of 16V and -8V, respectively. Notably, these devices showed an on/off ratio of 100, underscoring FFF's potential for scalable

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neuromorphic hardware—though issues like layer adhesion and device uniformity still warrant further improvement [39].

3.6. Spin coating

Spin coating serves as the dominant approach for preparing uniform thin films of photosensitive materials, where thicknesses span from micrometers down to nanometers [40]. Within the pMR manufacturing workflow, spin coating acts as an alternative approach for attaching the dielectric layer. The fundamental operations of the spin coating process can be illustrated by breaking down the entire process into four stages: transition, spin - up, spin - off, and solvent evaporation. First, the substrate is positioned on the spin coater, and the material to be coated — in solution form — is dispensed onto the substrate surface. TVia either static or dynamic deposition, the material being coated spreads due to centrifugal force, reducing its height to a critical level. Second, following material deposition, the substrate starts to rotate until it attains a specific speed. The centrifugal force arises from the acceleration and angular speed imparted to the substrate. Consequently, fluid flows outward across the substrate surface. Third, solution thinning — also referred to as surface solution thinning — is regulated by liquid viscous forces as soon as the wafer begins spinning at a constant rate. Fourth, the substrate is spun at an event and the coating thinning conduct is dominated by solvent evaporation [41].

3.7. Gravure printing

Printing methods are classified into groups using two core dimensions: throughput and resolution. A trade - off commonly exists between these aspects; high - speed processes usually limit pattern sizes. Non - digital techniques like screen printing and gravure printing are better suited for large - scale, high - throughput manufacturing. As a key technological enabler, gravure - printed transistors can fabricate high - speed transistors with sub - 5μm channel lengths and sub - 5V operation. Unlike screen printing, gravure printing uses an inked roller to transfer patterned ink onto substrates. In the detailed gravure printing workflow, Initially, the roller rotates through a large ink reservoir, filling the cells. Then, optimally, the cells are fully inked (no trapped air), and excess ink is wiped off by a doctor blade. Next, the roller contacts the substrate. Finally, ink spreads on the substrate to fill gaps between cells, forming the final pattern. For patterns printed with this technology, they typically break pixels into individual square cells, with feature sizes far below 1μm—all while maintaining high - speed printing (≈1m/s) [42]. In 2005, Yudas et al. developed a rotogravure printing approach for creating conductive lines on paper and plastic films. The inks had silver particles in an organic medium, cured at 70–120°C. These produced conductor lines with printed resistance as low as 50 m/mm and ≈4–7μm thickness [43].

3.8. Flexographic printing

Flexographic printing is valued for its versatility, cost-efficiency, high throughput, superior image quality, and eco-friendliness [44]. Its uses extend to packaging, publishing, and—more recently—printed electronics, where its capacity to work with various substrates (e.g., plastics, paper) offers an advantage. In flexographic printing, there are three options available: one for straightforward operation and double-sided substrate printing, another for precise registration, and a third for printing on heavier materials like corrugated boards. This method closely resembles letterpress printing, which transfers images from raised elements on a plate cylinder to nearly all substrate

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types [45]. Print quality with this method is typically characterized by line width, thickness, and ink transfer. A narrower line width and thinner layer indicate the ability to print finer details. In flexographic printing for pMR production, ink is first delivered to the anilox—either via a chamber or a fountain roll. It is then regulated by a doctor blade before being transferred to the printing plate. This plate features a flexible surface with raised patterns in image areas. Thanks to the anilox's engraved structure, which ensures consistent ink supply, the plate makes contact with the target substrate at the impression cylinder, transferring the image in the process [46]. Many researchers have attempted to utilize this printing technique to produce electronic equipment. Deganello et al. has conducted experiments that employed flexography printing to print micro-scale conductive networks. Results have demonstrated that conductive tracks within networks of 74.6 μm width and 0.74 μm height has a sheet resistance of 1.26 Ω/m at 18.6% coverage [47].

3.9. Doctor blade coating

Doctor blade coating, boasting merits like simplicity, versatility, cost-effectiveness, and compatibility with roll-to-roll processes, is extensively studied for depositing thin layers in diverse film applications. The “doctor blade” coating technique traces back to 1940, initially recognized for producing thin piezoelectric material sheets and capacitors. The core operating principle of this printing method hinges on continuous relative motion between the blade and substrate, achieving near-100% utilization efficiency—outperforming spin coating. This process enables computer-controlled operation, facilitating seamless integration into roll-to-roll workflows and large-scale manufacturing. The step-by-step printing workflow using this technology is as follows. First, a sharp-edged blade is positioned at one end of the substrate to initiate coating. Next, the balde is checked that it can move at a constant speed and the ink with the proper viscosity is placed in front of the blade. Then, the ink can get spread uniformly on the substrate surface. The surface wetting of the deposited layer can be affected by the distance between the blade and substrate, ink viscosity and surface tension of the precursor used. However, there also exists some limitations for the doctor blade coating. Due to the open ink system, doctor blade coating should be carried out with fixed knife and the balde durability [48]. In industrial use, doctor blade coating finds application in various fields. Xu et al. fabricated β-phase transistors composed of small printed molecular materials via doctor blade coating. Specifically, they leveraged a new soluble phthalocyanine, tetra-n-butyl peripheral substituted copper (II) phthalocyanine (CuBuPc), to form a 3D network of 1D nanofibers. They also used the doctor blade method to minimize material waste. The resulting transistor showed an on/off ratio of 10^3, charge mobility of 0.030 $cm^2/(V \cdot s)$, and a threshold voltage of - 8.5 V. Its charge-carrier mobility is 50 times higher than that produced by drop-casting [49].

4. Conclusion

Printable memristors (pMRs) exhibit significant potential for neuromorphic computing and next-generation electronics, owing to their unique properties and manufacturing versatility. This review systematically summarizes the material selection, fabrication techniques, and performance characteristics of pMRs, demonstrating their capability to address the limitations of traditional computing architectures. By optimizing electrode and dielectric materials, pMR performance has been substantially enhanced. Additionally, diverse printing technologies offer scalable and cost-effective solutions for pMR production. However, challenges such as endurance, uniformity, and integration remain critical barriers to widespread adoption. To drive pMR commercialization, future work should prioritize three areas: developing novel materials (e.g.,2D-organic hybrid dielectrics),

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optimizing printing processes for uniformity (e.g., tuning sheath gas flow in AJP), and establishing industry-wide performance metrics (e.g., switching endurance >10^5 cycles). This review synthesizes material-process-performance correlations, offering targeted guidance for advancing neuromorphic computing hardware.

Declaration

This manuscript has undergone language refinement with the assistance of AI tools to improve clarity and readability. The authors have meticulously reviewed and edited the AI-refined text to ensure that it accurately conveys the scientific content and intended meaning. All elements of the work have been carefully verified, and the authors assume full responsibility for the integrity and accuracy of the final manuscript.

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