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CRITICAL REVIEW

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Direct writing of graphene electrodes for point-of-care electrochemical sensing applications

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Electrochemical sensors are increasingly garnering attention as valuable tools for point-of-care (POC) testing due to their low costs, high sensitivities, and ease of miniaturization. Graphene-based materials, renowned for their tunable electrical conductivity, high specific surface areas, versatile functionality, and biocompatibility; are highly suited for the fabrication of electrochemical sensors with heightened sensitivities. Non-contact, maskless, direct writing methods allow the rapid, large-scale production of graphene electrodes with high design flexibility. Researchers globally are advancing graphene electrode production, aiming for smaller, faster, and more efficient sensors. This review provides a comprehensive overview of recent advances on the direct writing of graphene electrodes for electrochemical sensing applications. It covers the basics of direct writing techniques, the advancements in graphene ink/precursor preparation, structural design, and device integration, with a focus on POC platforms.

1 Introduction

The escalating significance of healthcare, food safety control, and environmental preservation underscores the pressing need for precise disease diagnostics and accurate contaminant detection. Established methodologies such as chromatography, mass spectrometry, spectroscopy, polymerase chain reaction, and enzyme-linked immunosorbent assays have conventionally addressed these exigencies.[1–3] Nevertheless, their protracted procedures and intricate protocols necessitate specialized personnel and sophisticated equipment, thereby presenting substantial challenges, particularly in resource-limited settings. Electrochemical sensors have emerged as a compelling

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alternative owing to their cost-effectiveness, high sensitivity/selectivity, facile miniaturization, and user-friendly operation.[4–7] They facilitate the transformation of physical, biological, and chemical information into easily measurable electrical signals, which can be recorded using simple, portable setups. Consequently, electrochemical platforms hold great promise for revolutionizing point-of-care (POC) devices. In order to be considered true POC diagnostic devices, they should comply with the WHO's REASSURED criteria, which states that tests should have real time connectivity, be environmentally friendly, affordable, sensitive, specific, user-friendly, rapid, equipment free and deliverable.[8]

Graphene derivatives are excellent candidates for electrode fabrication in electrochemical sensing applications, owing to their remarkable attributes such as high surface area, tunable conductivity, and good biocompatibility.[9–11] Graphene is ideally a single carbon atom-thick sheet consisting of a hexagonal network with no defect sites.[12] It shows remarkable conductivity (200 000 cm^2 V^{-1} s^{-1}) with a theoretical specific surface area as high as 2630 cm^2 g^{-1}. Low-defect graphene is expensive to produce and tends to aggregate in most solvents, due to π–π stacking and van der Waals interactions, making it difficult to process.[13] Graphene oxide (GO) retains the high surface area and flexibility of graphene, while the hydrophilic oxygen-containing groups (hydroxyl, carbonyl, carboxyl and epoxy groups) make it soluble and easy to functionalize in aqueous solutions.[14] The functional groups change the carbon atoms from sp^2 to sp^3, which disrupts the graphene lattice, making GO thermally insulating and electrically resistive. The poor conductivity leads to an unfavorable electrochemical interface for sensing. To solve this problem, the oxygen-containing groups can be partially removed, resulting in reduced graphene oxide (rGO) with improved conductivity and fewer defect sites.[15]

Despite recent advances in graphene production, the fabrication of graphene electrodes remains slow and costly. Photolithography patterns graphene with high precision/resolution,[16] but is limited by the need for expensive equipment and clean room facilities. Screen printing,[17] gravure printing,[18] and contact-transfer printing[19] are promising alternatives, but require pre-patterned templates, which does not allow the rapid and facile iteration of electrode designs as well as being wasteful and expensive.

Recently, direct writing methods, also known as "digital writing" or "digital printing", have been the subject of much research in the field of graphene electrode production.[20,21] Direct writing encompasses a set of maskless techniques for material patterning based on computer-aided digital designs; they show great potential for the simple, rapid, and scalable patterning of electrodes with high design flexibility.[22,23] Various direct writing systems have been employed to fabricate graphene electrodes, including direct ink writing,[24–26] 3D printing,[27] and direct laser writing.[22] These techniques allow customization of graphene electrode design, morphology, chemical composition, and conductivity. As such they can be used to make electrodes smaller, faster, and improve the sensing performance of the resulting electrodes. In this review, we present a comprehensive overview of the recent literature related to graphene electrode fabrication *via* direct writing, specifically focusing on designs and methods that have been used for electrochemical sensing. The review covers the fundamentals of direct writing techniques, advancements in graphene-based inks/precursors, electrode designs/structures, and their electrochemical sensing applications. The combination of different direct writing methods, low-cost tools, and integrated sensing platforms for smart sensing applications are also discussed. Notably, emphasis is placed on the creation of POC platforms.

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Table 1 Notable graphene inks from the literature

Ink	Filler	Solvent	Printing technique	Ref.
Gr/PANI	—	Water/ethanol	IJP	31
MoS_2/Gr	EC	Cyclohexanone/terpineol, NMP, or IPA	IJP	32
Gr	EC	Ethylene glycol mixes with ethanol, DMF or NMP	IJP	33
Gr	Pyrene sulfonic acid sodium	Water	IJP	34
GO	—	Water	IJP	35
Gr/Ag nanoparticle	EC	Terpineol/cyclohexanone	IJP	36
Gr Ag@Au	—	Water	IJP	37
PANI/Gr	SDBS	Water	IJP	38
Gr	—	NMP	IJP	39
Gr	—	Ethanol	IJP	40
Gr	—	IPA/PVA	IJP	41
GO	Sodium dodecyl sulfate	EG/water	IJP	42
GO		Water/ethanol/EG	IJP	43
GO/Ti_3C_2	Nafion	Water	IJP	44
Gr, Gr/AgNPs, Gr/PEDOT: PSS	—	IPA/EG	IJP	45
Gr/polyurethane composite hydrogel	—	Organic solvent mixture	EXP	46
Gr/CNT	Ethyl cellulose	Ethanol/terpineol	EXP	47
GO	—	Water	EXP	48
GO/PANI/PEDOT:PSS, rGO/PEDOT: PSS	—	Water	EXP	49
Gr	Poly(ionic liquid)	Water	EXP	50
Gr/PDMS	—	—	EXP	51
GNPs/MWCNT/polyethylene oxide	—	Water	EXP	52
(PANI/GO) gel	—	*N*-methyl-2-pyrrolidinone/water	EXP	53
Gr nanoplatelets/PVB	—	Ethanol	EXP	54
GO mix with polymers, ceramics, or steel	—	Water	EXP	55
rGO/Pluronic F127	—	Water	EXP	56
GO/MWCNTs/Nafion	—	Water	EXP	57
Chitosan/ /rGO	—	Water	EXP	58
Fe_2O_3/Gr/Ag	PVDF	Nmp	EXP	59
Lignin/GO	—	Water	EXP	60
GO	Branched copolymer/glucono-δ-lactone	Water	EXP	61
GO	Hydroxypropyl methylcellulose	Water	EXP	62
Gr	Nitrocellulose/ethylene glycol	Glycerol/ethyl lactate	EXP	63
MOF/CNT/GO	—	Water	EXP	64
GO/ZnV_2O_6@$Co_3V_2O_8$, GO/$Co_3V_2O_8$, GO/VN	—	Water	EXP	65
Gr	PLG	DCM/EGBE/DBP	EXP	66
Gr/PDMS	—	—	EXP	67
Gr	EC	Terpineol/cyclohexanone	EHDP	68, 69
			AJP	70
GO	—	Water	EHDP	71
rGO	—	NMF	EHDP	72
Gr	BSA	Water	EHDP	73, 74
PEDOT:PSS/Gr/SWCNTs	Polyethylene oxide	Water	EHDP	75
Gr	Perylene	Dichloromethane	EHDP	76
MXene/rGO	PVDF	DMF/acetone	EHDP	77
Gr	Nitrocellulose	Dibutyl phthalate/ethyl lactate	AJP	78–80
Electrochemically exfoliated graphene	—	Water	AJP	81
Gr	Graphene quantum dot	Terpineol/cyclohexanone	AJP	82
Gr	EC	Ethanol/ethyl lactate	AJP	83
rGO/CNT/PEDOT:PSS	—	DMSO/water/methanol	AJP	84
Gr	1-Pyrenesulfonic acid sodium salt	Water	AJP	85
Gr/Ag NPs	EC	Ethanol/terpineol/water	AJP	86

Abbreviations: AJP, aerosol jet printing; EHDP, electrohydrodynamic printing; EXP, extrusion printing; Gr, graphene; PANI, polyaniline; PEDOT: PSS, poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate); MWCNT, multi-walled carbon nano-tube; PDMS, polydimethylsiloxane; EC, ethyl cellulose; SDBS, sodium dodecyl benzene sulfonate; PVDF, polyvinylidene difluoride; NMP, *N*-methylpyrrolidone; EG, ethylene glycol; PVP, polyvinyl pyrrolidone; SDS, sodium dodecyl sulfate.

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2 Direct writing of graphene

2.1 Direct ink writing

Direct ink writing techniques allow the direct, controlled, maskless and contactless deposition of materials onto rigid, flexible, and even deformable substrates.[20,26,28] They do not require complicated and expensive exposure and etching processes, and have the advantage of working with a wide range of materials. They are easy-to-use, cheap, and produce less material waste than other printing techniques. Common examples of this technique include inkjet printing, aerosol jet printing, electrohydrodynamic printing, and extrusion printing. They have been used to prepare graphene micro/nano-structures for different applications. Moreover, the morphology and properties of these structures can be meticulously adjusted through modifications in ink composition and printing parameters.

2.1.1 Graphene inks. Printed electronic sensors can make POC devices more accessible. The core technology at the heart of printed electronics is the conductive ink. Conventionally, metallic nanoparticle/metal precursor inks are mainly being used,[29] however, newer inks have been developed containing carbon, two-dimensional materials, and polymers.[30] Among these, graphene-based inks stand out due to their remarkable electrical conductivity, biocompatibility, and scalability in manufacturing. The progress of graphene ink formulations has been particularly notable in direct writing techniques. A list of graphene inks and their compositions is reported in Table 1.

The choice of solvent plays an important role in creating stable and well-performing graphene dispersions, targeting the characteristics of each printing/writing method. A solvent is required that readily disperses the graphenic material, that does not damage the substrate or cause issues downstream.[25] A number of solvents or solvent mixtures have been used for graphene ink preparation, such as *N*-methyl-2-pyrrolidone, *N*,*N*-dimethylformamide and dimethyl sulfoxide, terpineol, ethanol, isopropanol, ethylene glycol, glycerol, ethylene acetate and deionized water.[41,87–89] Additives are frequently used to adjust the viscosity and surface tension of the inks, or to stabilize graphene flakes *via* π–π stacking, van der Waals forces, hydrogen bonding or electrostatic interactions.[90,91]

Pure graphene-based inks suffer from poor conductivity mainly due to the deposition of disconnected flakes and making difficult the creation of a relatively direct percolation path, which limits their applicability.[92] Composites of graphene and metallic nanoparticles[93] or conductive polymers[94] is often used to address this. The integration of such additives not only hinders aggregation, improving the stability of the ink, but also increases the conductivity of the printed devices.

As an electrode material, graphene is known for its good electrochemical, biological, and thermal stability, making it ideal as a material choice for POC diagnostic device development. However, the stability and selectivity of any biosensor towards the analyte largely depends on the bioreceptor (enzyme/antibody/aptamer/molecular imprinted polymer)[4,7,95] making a summary of these performance factors from the literature superfluous to the scope of this review.

2.1.2 Inkjet printing. Inkjet printing (IJP) is a well-established standard in the field of printed and flexible electronics. Ink droplets are ejected from nozzles by the mechanical force generated by a transducer (Fig. 1a).[26,91] The transducer can either be a piezoelectric crystal, which undergoes a physical deformation when an electrical signal is applied, or an electrical heater that generates air micro-bubbles rapidly vaporizing the ink in close proximity of the resistor. To achieve a stable printing process (free from satellite drops and nozzle clogging), the rheological properties of the inks must be tuned to obtain a Laplace number (Z) between 1 and 10.[96] Typically, graphene flakes with average sizes <0.02 times the nozzle diameter, are dispersed in low viscosity (4–30 cP) solutions, to formulate inks with surface tensions between 20–50 mN m^{-1}.[97] It is important to note that while these parameters are recommended, practical printing often relies on a trial-and-error approach to identify the best combination of printers, inks, and substrates.[98,99] With $Z \approx 19$, uniform features still can be printed on SiO_2, glass and paper without satellite drops or nozzle blocking, yielding a high electrical conductivity of 3.91×10^4 S m^{-1} after annealing at 300 °C for 1 h (Fig. 1a).[34]

2.1.3 Aerosol jet printing. During aerosol jet printing (AJP), an ink is atomized into micrometer-sized aerosol droplets through pneumatic or ultrasonic methods; droplets are then transported into a deposition nozzle and directed onto a substrate using a sheath gas (Fig. 1b).[102] AJP demonstrates the capability to print inks with viscosities from 1 to 1000 cp, at resolutions down to a few micrometers, on both planar and non-planar surfaces.[79,103] To obtain consistent outcomes, relatively high volume of ink is required and various parameters such as atomizer power, atomizer/sheath gas flow rates, substrate temperature, and printing speed must be optimized.[70,104]

AJP can produce sophisticated 3D structures due to its compatibility with highly viscous inks. For example, a water-based graphene ink was used to construct graphene pillars at different angles with respect to the substrate without using supporting fillers (Fig. 1b).[100] By setting the plate temperature to 120 °C, the graphene microstructures obtained were conductive immediately after printing.

2.1.4 Electrohydrodynamic printing. In electrohydrodynamic printing (EHDP), an electric field is applied between the nozzle and the substrate to deposit continuous lines (or droplets) onto the substrate, forming 2D/3D structures (Fig. 1c).[24] EHDP exhibits excellent compatibility with a variety of materials, accommodating a wide range of ink viscosities from 0.1 to 10 000 cp, realizing the large-scale, high-resolution (down to nanometer) fabrication of electronics[105,106] . For example, graphene drops

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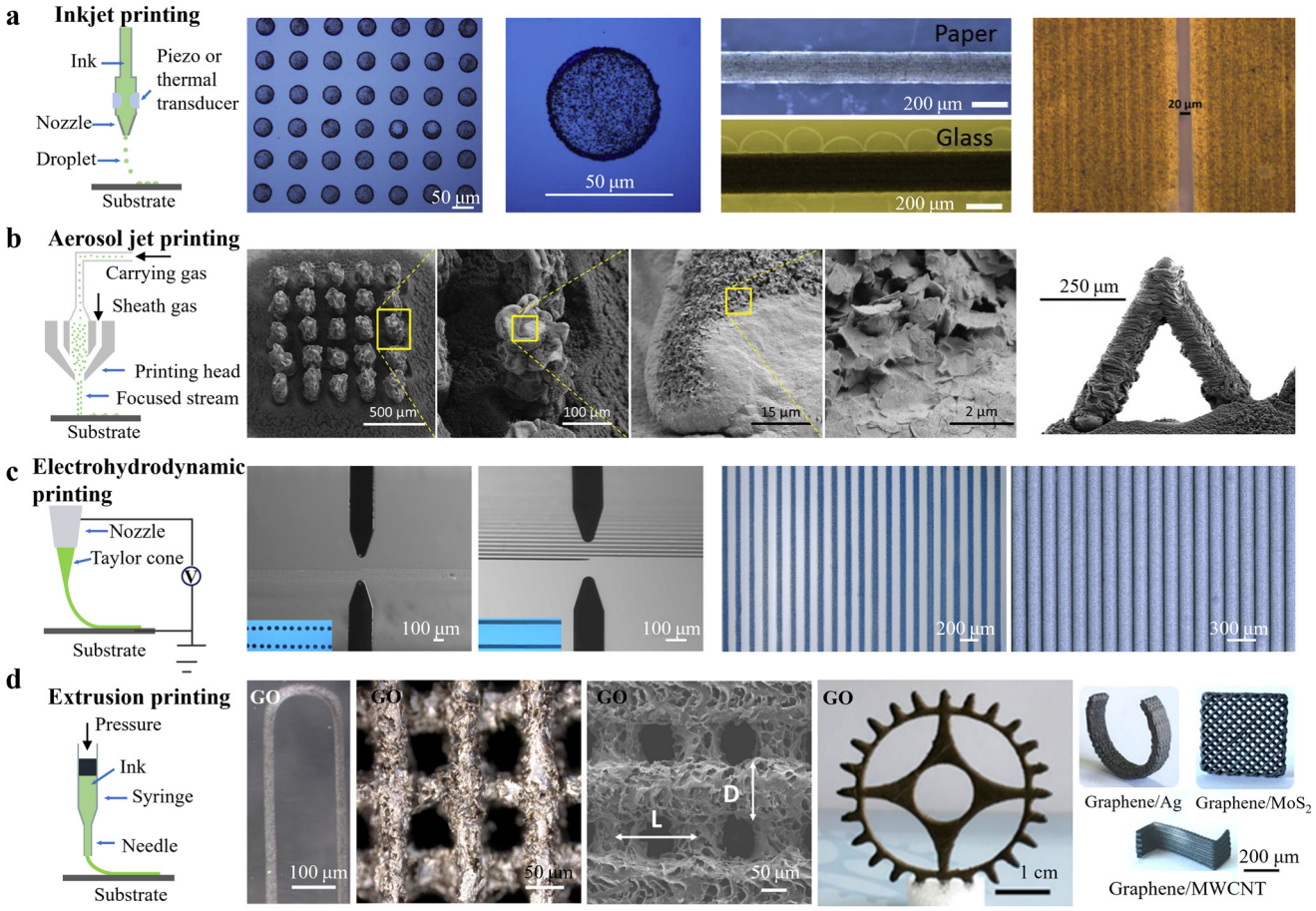

Fig. 1 Direct ink writing of graphene. From left to right: (a) inkjet printed graphene drops to show the drop size and uniformity, inkjet printed graphene lines on different substrates, and two printed graphene lines on paper separated by a gap of ~20 μm. Reproduced from ref. 34 with permission from Elsevier, copyright 2019. (b) Aerosol jet printed graphene vertical and tilted micropillars. Reproduced from ref. 100 with permission from John Wiley and Sons, copyright 2023. (c) Electrohydrodynamic printed graphene traces with high accuracy and uniformity. Reproduced from ref. 69 with permission from IOP Publishing Ltd, copyright 2020. (d) Graphene-based 3D structures constructed by extrusion printing. Reproduced from ref. 101 with permission from American Chemical Society, copyright 2018.

and lines were printed, obtaining features with standard deviations down to 1.9 μm (Fig. 1c).[69] The strong electric field generates droplets far smaller than the nozzle diameter, avoiding nozzle clogging. Despite all these advantages, only a few works have been found that demonstrate graphene electrode production by EHDP, potentially due to the complexity of the technique.[72,77,107]

2.1.5 Extrusion printing. In extrusion printing (EXP), a pneumatic gas pump or a motor-driven screw pushes viscoelastic inks through a fine nozzle; instead of individual droplets, the ink emerges as a continuous flow that can be layered to construct complex 3D structures (Fig. 1d).[108] The wide range of ink viscosities, spanning from 0.5 to 1 000 000 cp, compatible with EXP means that it has been used to print: liquids, colloidal suspensions, hydrogels, and composites.[109] Printing parameters that must be tuned to the rheological properties of the ink include: the pressure applied, nozzle size, and printing speed.[110,111]

Using EXP, highly viscoelastic graphene inks have been used to produce self-supporting 3D graphene-based architectures, with high specific surface areas, good electrical conductivity, and excellent electrochemical properties.[110,112] Novel printable graphene inks can be made by tuning the graphene preparation,[113] condensation,[114] ion linking[111] and polymer stabilization.[60] For example, negatively charged 0D, 1D and 2D materials were integrated into GO gels, yielding uniform graphene-based inks, in which aggregation was prevented by robust electrostatic repulsion between the GO and the other composite materials.[101] Complex architectures fashioned from these mixed-dimensional materials were printed on various substrates (Fig. 1d). Graphene aerogel electrodes were also printed by this method and exhibited excellent electrochemical performance owing to their efficient ion- and electron-transport.

Overall, the quality of the printed patterns is influenced by the rheological properties of the inks, surface properties of the substrates and sintering methods. These factors dramatically influence the properties of the printed devices, including their resolution, conductivity, adhesion, and structural integrity. Graphene printed from low-viscosity inks

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has lower contact resistances[98] and a strong affinity to the substrate after sintering.[94,115] This is highly favorable for low-cost, flexible, thin film-based electrochemical devices. However, the low graphene content within the ink, typically 0.002–1 wt%, requires extensive layer printing.[42] Additives such as ethyl cellulose and surfactants are helpful, but their subsequent removal through an annealing process is required (usually at temperatures ≥300 °C);[47] which limits the choice of substrates to heat-resistant materials. Complex 3D structures have been printed with highly viscous-elastic graphene inks and applied in energy devices and strain sensors.[27,113,116] However, their applications as electrochemical sensors are rarely reported, potentially due to the hydrophobicity and structural fragility of the 3D porous graphene matrix.[58] As well as 0D, 1D and 2D printing, EXP can also be used to 3D print devices.

2.2 3D printing

3D printing is a group of computer-controlled techniques for creating three-dimensional objects by depositing material layer by layer along the *Z*-axis.[26–28,117] Common techniques include fused deposition modelling (FDM), stereolithography (SLA), and powder bed fusion (PBF).

FDM can be adapted to 3D print graphene-based electronics. Graphene-doped organic filaments are thermally melted, extruded through a nozzle, and deposited layer by layer on the support, obtaining free standing frameworks after solidification by cooling (Fig. 2a).[118] It has the advantage of being low cost, easy to use, and fast; drawbacks include low resolution, potential nozzle clogging, and electrical anisotropy of the final devices. Acrylonitrile butadiene styrene copolymers (ABS) and polylactic acid (PLA) are commonly utilized filaments for graphene electrode production.[119,120] For example, a PLA-based composite with a high content of oriented colloidal graphite and graphene (15–25 vol%) was used to print complex electrodes (Fig. 2a).[121] The structural anisotropy was tuned by changing the printing direction, showing conductivity as high as 22 S m^{-1}. Mechanical, chemical, electrochemical, or thermal treatments are generally necessary to expose and activate the graphene encapsulated within the polymer matrix.[122–124]

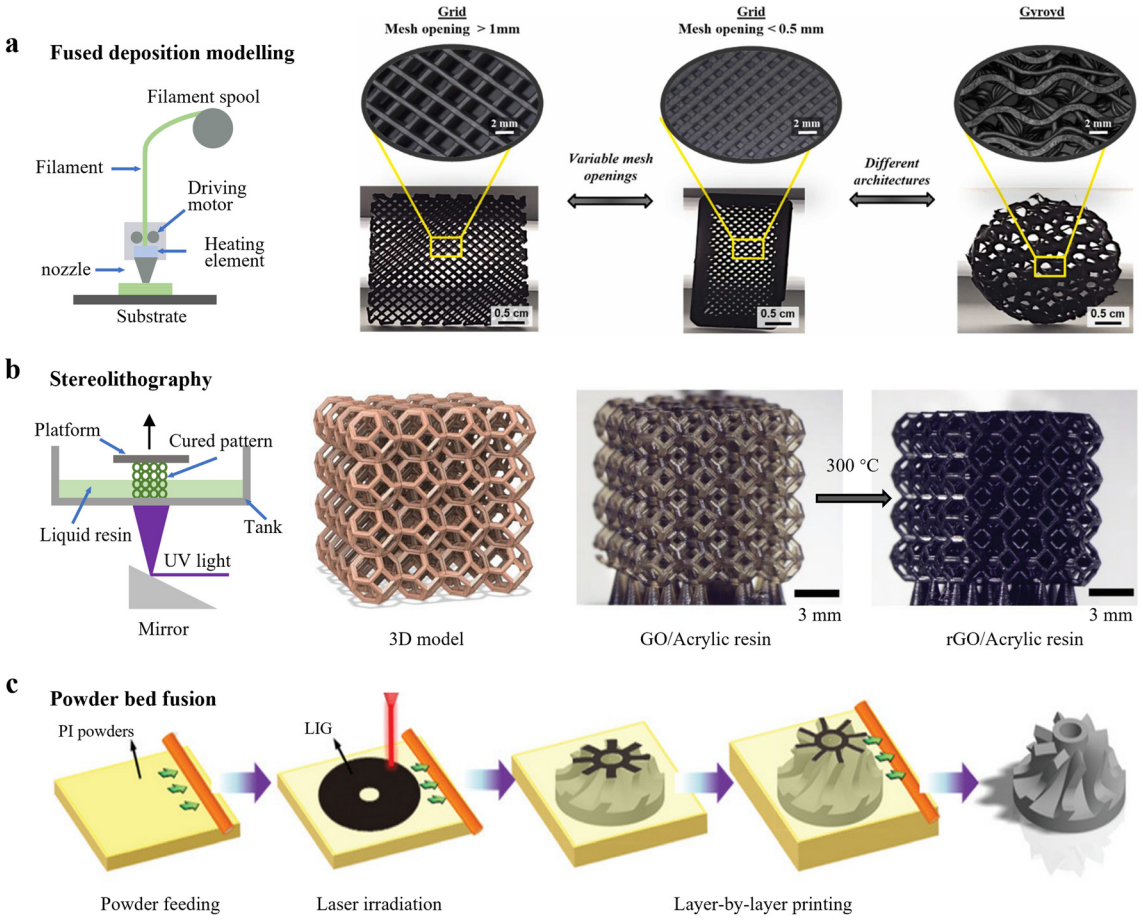

Fig. 2 3D printing of graphene. (a) Different architectures by fused deposition modelling. The printing direction affects graphene's properties. Reproduced from ref. 121 under creative commons CC-BY license. (b) Graphene cell printed by stereolithography. Reproduced from ref. 125 with permission from Wiley-VCH GmbH, copyright 2023. (c) Sophisticated graphene devices printed with a modified powder bed fusion method. Reproduced from ref. 126 under creative commons CC-BY license.

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SLA employs radiation (often UV light lasers) to selectively solidify liquid polymers, achieving fast 3D printing with high spatial resolution (10–150 μm), high throughput, and smooth surfaces (Fig. 2b).[127] Patterning of graphene structures has been demonstrated by UV polymerization of monolayer GO blended acrylic resin (Fig. 2b).[125] Thermal treatment at 300 °C allows for the *in situ* thermal reduction of GO to rGO, while the resin remained unchanged, resulting in conductivities of up to 0.012 S m^{-1}. Concentrated carbon-based materials tend to absorb and scatter light, which hinders the photopolymerization process, making it difficult to obtain graphene electrodes with high electrical conductivity by SLA.[128,129]

Graphene can be 3D printed by a modified FDM method. (Fig. 3e).[126] The strategy is based on selective laser sintering, in which CO_2 laser irradiates polyimide powder-bed, realizing both particle-sintering and graphene-converting processes layer-by-layer by adjusting the laser power and layer thickness. This unique strategy could assemble various types of graphene architectures including identical-section, variable-section, and graphene/PI hybrid structures, forming bulk 3D graphene with freeform structures without introducing extra binders, templates, and catalysts. Direct conversion of liquid organic precursor into versatile 3D graphene could be done with a similar process.[130]

2.3 Direct laser writing

Direct laser writing (DLW) is an easy, efficient, low-cost, eco-friendly and maskless method to simultaneously produce and pattern porous graphene electrodes without any catalysts or harmful solvents.[137–139] In the DLW process, a laser is used to irradiate the samples, inducing photochemical and/or photothermal reactions, resulting in laser induced graphene (LIG). LIGs are highly porous 3D networks with large active surface areas, good chemical/thermal stability, and fast electron transfer rates, making them ideal electrode materials for electrical/electrochemical sensing. Intensive studies have been conducted with different laser sources (ultraviolet–visible, infrared, and ultra-short pulse lasers) and precursors; as a result, it is now possible to tune the LIG morphology and composition, and thereby their physical, chemical and electrical properties.[137,138]

Polyimide has quickly become the most popular precursor for the production of LIG since it was first reported by Lin *et al.* in 2014.[131,140] While it is debatable whether or not this is truly graphene, it possesses many of the properties of layered, highly crystalline graphene. This method allows different patterns to be obtained with porous structures abundant in five- and seven-membered rings, as opposed to the conventional hexagonal lattice of graphene (Fig. 3a). LIG can be used to prepare graphene with a variety of sophisticated structures by changing the laser parameters, these structures could be designed such that their resistances changed upon structure deformation (Fig. 3b)[132] GO is another commonly used precursor to produce LIG with a low laser power.[138] Low cost, high performance, porous graphene was obtained with a LightScribe DVD writer (788 nm, 5 mW); which was capable of reducing and patterning GO with a 20 μm resolution.[141] Spatially shaped femtosecond lasers (SSFL)

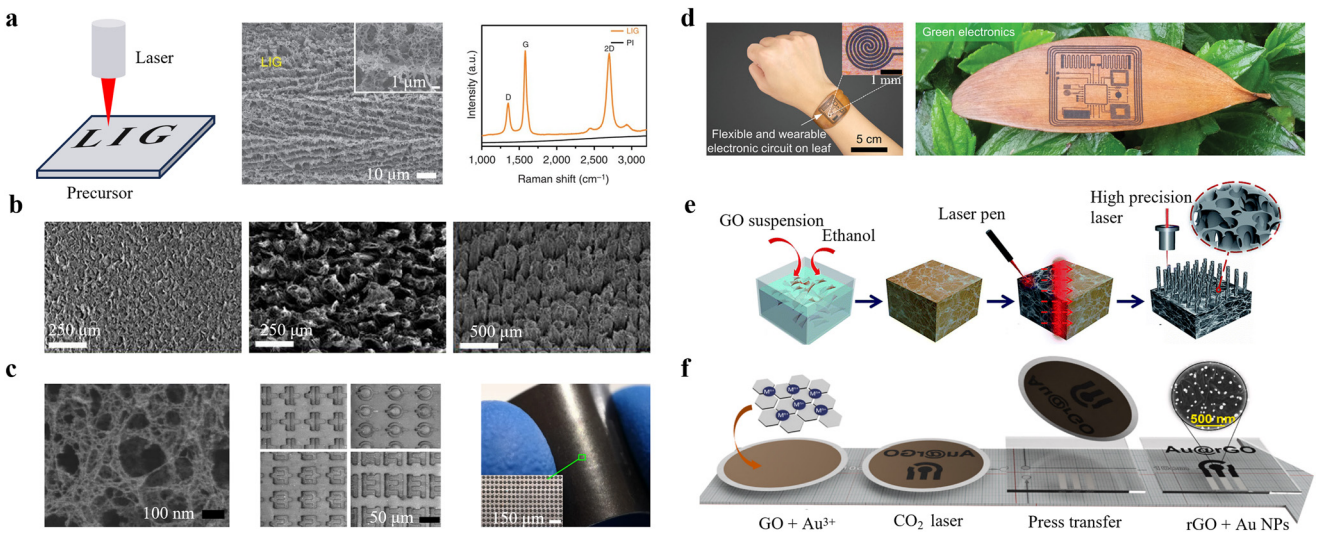

Fig. 3 Laser induced graphene (LIG). (a) LIG from polyimide, showing porous structures with typical Raman signals of graphene. Reproduced from ref. 131 under creative commons CC-BY license. (b) Morphology variation led by different laser parameters. Reproduced from ref. 132 with permission from, American Chemical Society, copyright 2023. (c) High density, high resolution LIG/MnO_2 patterns fabricated on an industrial scale. Reproduced from ref. 133 under creative commons CC-BY license. (d) Flexible, wearable electronics made from plants. Reproduced from ref. 134 with permission from Wiley-VCH Verlag GmbH & co. KGaA, Weinheim, copyright 2019. (e) Highly vertically ordered pillar array of graphene framework constructed with a two-step lasering process. Reproduced from ref. 135 with permission from Royal Society of Chemistry, copyright 2018. (f) Selective transfer of laser reduced graphene composite by a press transfer process. Reproduced from ref. 136 under creative commons CC-BY license.

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can produce graphene electrodes from GO on an industrial scale. In SSFL, the shape of the laser beam is modulated to allow it to pattern the graphene surface, this allows the laser to rapidly stamp the surface in a manner compatible with large scale fabrication. Using SSFL, over 30 000 graphene devices with dimensions as Small as 10 μm, separated by 500 nm could be produced in 10 minutes (Fig. 3c).[133] Renewable materials have proven to be great precursors to produce LIG.[142,143] Cellulose, hemicellulose, and lignin can be converted into graphene like materials using a UV femtosecond laser in ambient air; this greatly improves the sustainability of electronics manufacturing, and has been shown capable of producing electrical connections, flexible temperature sensors and supercapacitors (Fig. 3d).[134] A versatile method has also been developed to produce LIG from cross-linked polystyrene, epoxy resins, and cellulose.[144] A first laser irradiation step converts the substrates into amorphous carbon, and a second laser step transforms the amorphous carbon into graphene. The resulting LIG displayed a low sheet resistance of around 5 Ω sq^{-1} and was used to create supercapacitors on the surface of a coconut. The method shows that any precursor that can be converted into amorphous carbon can be transformed into graphene.

Although DLW is a 2D patterning process, there have been attempts to use it for 3D device fabrication.[126,130,145] For example, a highly vertically ordered pillar array of graphene framework was produced from GO. GO hydrogel was transformed into rGO framework by laser irradiation, and then shaped into desired structures with macroporous networks using a highly precise laser (Fig. 3e).[135]

Typically, LIG remains on the surface where it was generated. However, the successful transfer of LIG onto various substrates has recently been achieved by mechanical pressing,[146] elastomer embedding,[147,148] and hydrogel[149]/adhesive tape[150]/solvent[151] assisted transfer. Stamp transfer methods have been proposed to transfer laser reduced graphene electrodes on a wide variety of substrates including PET, paper, nitrocellulose, glass, fabric, or silicon.[152–154] In this process, rGO formed by the laser reduction of GO is selectively transferred to other substrates with a mechanical press (Fig. 3f).[136]

2.4 Low-cost direct writers

The accessibility of affordable direct writing tools holds immense significance, as it empowers the creation of personalized sensing devices within laboratories with limited resources and even by non-professionals at home. Consumer-grade inkjet printers show distinct advantages over their high-end research-grade counterparts in terms of affordability, accessibility, printing speed, and the number of available ink channels.[26,155,156] For instance, AgNP and PEDOT: PSS inks were deposited on poly(ethylene terephthalate) substrate with a printer cost of only 60 $.[157] One can find commercial desktop 3D printers or laser engravers on the market for less than 100 $ easily. Furthermore, several teams have created cost-effective extrusion printers (under 300 $) using open-source software and commercial materials.[158,159]

2.5 Additional smart strategies

2.5.1 Multi-material deposition. The ability to print multiple materials simultaneously allows for the fabrication of more complex structures. One noteworthy example of this is the work published by Zeng *et al.*, in which a combinatorial aerosol jet printing approach was used to create patterns with compositional gradients with a microscale spatial resolution (Fig. 4a).[148] Two inks were atomized into aerosols containing microscale droplets. The combined ink streams were then mixed within a single nozzle and deposited onto the substrate using a sheath gas. This method facilitated *in situ* mixing ratio adjustments, leading to combinatorial doping, functional grading, and chemical reactions within versatile material libraries, including 0D, 1D, and 2D nanomaterials, and even seemingly incompatible materials like MXene and Sb_2Te_3. In another work, mono/co-axial microfibers were created using an extrusion printing process with GO and $Ti_3C_2T_x$ dispersions with mono/coaxial needles (Fig. 4b).[160]

2.5.2 Fully written devices. Nowadays, there is a growing need for portable, wearable, miniaturized sensing devices. This requires the incorporation of various functional components, including electrodes, electrical circuits, electrochemical cells, microfluidics, and data processors/transmitters.[163,164] Direct writing methods are inherently suited for this purpose owing to the digital, maskless nature of fabrication with high design freedom. For example, a fully 3D printed portable analytical setup was described comprising a batch injection analysis cell and an electrochemical platform with eight sensing electrodes (Fig. 4c).[161] The device can be printed within 3.4 h using a multi-material printer equipped with insulating and conductive filaments at a cost of *ca.* ~1.2 $ per unit. Direct laser writing exhibits notable advantages in the manufacturing process of multilayered, integrated devices. Utilizing a laser engraver, various components such as LIG electrode arrays, microfluidic channels, and adhesive layers can be precisely patterned. This approach yields to cost-effective, disposable, and wearable epidermal patches capable of simultaneous sweat sampling and signal monitoring (Fig. 4d).[162] The combination of different direct writing techniques can overcome their constraints, structures with higher resolution and special functions. For example, 3D needle-like electrodes and planar connections were printed by AJP and IJP techniques, displaying excellent performance for recording extracellular electrophysiological signals from living cells.[165]

2.6 Comparison with other patterning methods

While printing has many advantages as a method of ink deposition and patterning, there are alternative methods;

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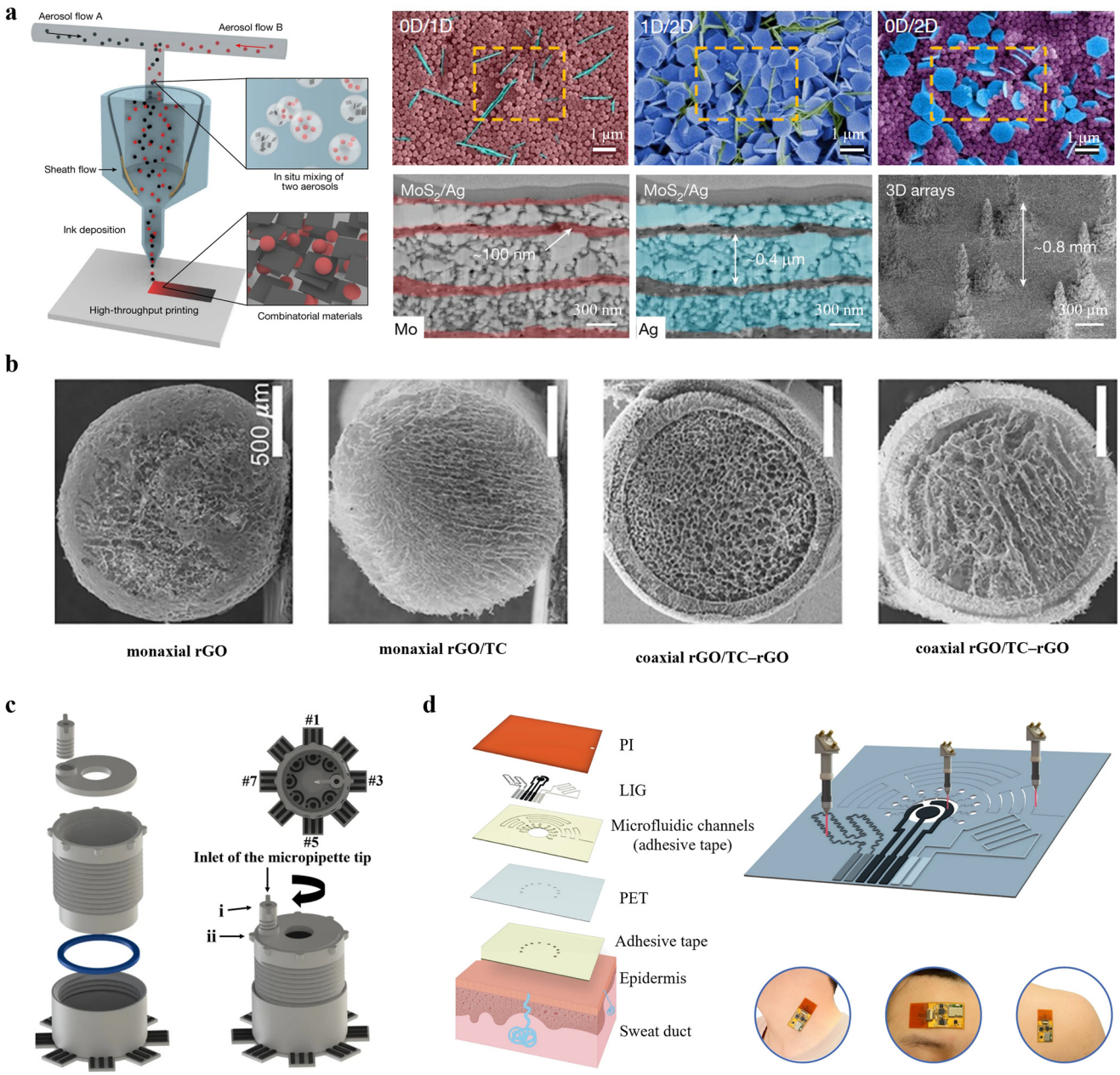

Fig. 4 (a) Simultaneous multi-material printing at various dimensions by AJP. Reproduced from ref. 148 under creative commons CC-BY license. (b) rGO/ $Ti_3C_2T_x$ (TC) monaxial and coaxial fibers produced by EXP. Reproduced from ref. 160 with permission from American Chemical Society, copyright 2023. (c) A fully 3D printed electrochemical cell with 8 integrated electrodes. Reproduced from ref. 161 with permission from Springer Nature, copyright 2022. (d) A laser engraved wearable electrochemical sensor for simultaneous determination of various signals. Reproduced from ref. 162 with permission from Springer Nature, copyright 2019.

these include screen printing and photolithography. Screen printing typically has similar or lower resolution (μm)[166] to direct writing methods, requires pre-fabricated masks that are expensive and time consuming to iterate, also a lot of the ink is wasted when printing by this method, making the cost per device typically higher than those produced by direct writing. Photolithography can also be used to pattern electronic devices, however this requires a large upfront capital investment in the equipment and facilities.[16] Photolithography is not inherently iterative, requiring complex optimization to produce working devices, however once the device fabrication has been optimized, it is inherently scalable and the costs of mass-produced devices can be low. Of all these techniques photolithography has the highest resolution/ precision, achieving low nm in three dimensions (depending on many factors).[167] Direct writing methods are advantageous over both of these alternatives since they are versatile, iterative and can produce small or large batches of devices at a low cost with minimal material waste. The drawbacks of direct written include the low

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resolution, the reproducibility of fabricated devices, and the typical need to include ligands and stabilizers in the inks that need to be removed after fabrication.

3 Electrochemical sensing applications of direct-written graphene electrodes

Electrochemical sensors enable the real-time analysis of analytes in diverse matrices, making them important tools in healthcare, environmental monitoring, and food safety applications. Direct writing facilitates the customization of graphene electrodes, alternating their shape, morphology, conductivity and electrochemical properties. This means that graphene electrodes possess the versatility required for detecting a range of targets including pH, ions, molecules, proteins, and pathogens (Table 2).

The early detection of health-related biomarkers greatly aids disease prevention and enhances the efficiency of medical treatments. As well as traditional POC sensors, wearable biosensors are a hot topic for active real-time monitoring of health conditions.[193] Inkjet printing can use a wide variety of inks and design flexibility to fabricate wearable sensors for multiplexed biosensing (Fig. 5a).[194] The fully-printed system was capable of detecting glucose, alcohol, pH, and skin temperature in sweat. The influence of pH and temperature variations were monitored to allow the stable monitoring of glucose and alcohol for more than 30 h.

The electrochemical properties of graphene composites are well-suited to detecting biomarkers *in vivo*. For example, Li *et al.* used direct laser scribing of metal-complexed polyimide to fabricate stretchable, tissue-resembling electrodes, consisting of interconnected graphene/metal oxide nanoparticle/elastomer networks (Fig. 5b).[195] These electrodes were used for continuous, real-time, monoamine sensing in the brain, and serotonin detection in the gut without disturbing peristalsis. All the work was done in a living mouse *via* fast-scan cyclic voltammetry with a portable potentiostat.

3D printed graphene electrodes have been used for the detection of COVID-19, proving it could be a promising method of decentralized and low-cost manufacturing of POC diagnostic tools. Competitive immunosensors were developed to monitor COVID-19 in both buffered and diluted serological samples, realizing trace level response in 20 min (Fig. 5c).[124] Similarly, infectious pathogens were detected in spiked buffer and artificial urine with LIG-based immunosensors, exhibiting a wide dynamic range (917–2.1 × 10^7 CFU mL^{-1}) with a low LOD (283 CFU mL^{-1}) (Fig. 5d).[152] POC detection was demonstrated by incorporating a portable wireless system controlled and monitored by a smartphone.

Environmental monitoring and food safety have direct implications for both human and animal welfare. Both require POC analytical methods to accurately monitor relevant hazards. A recent work by Chen *et al.* has shown that four different targets could be detected by integrating microfluidics and electrochemical cells through the direct laser writing of PI (Fig. 5e).[178] By tuning the laser parameters, the electrical conductivity, surface morphology, and wettability of the LIG were adjusted, meaning that both the microfluidics and electrodes could be fabricated by the laser on the same substrate. The developed ion-selective and enzymatic electrodes, capable of detecting K^+, NO_3^-, NH_4^+, and parathion simultaneously. An aerosol jet-printed high-resolution interdigitated electrode with a line width of around 40 μm was developed for the detection of histamine using electrochemical impedance spectroscopy.[78] The functionalization of the electrode with monoclonal antibody resulted in a wide histamine sensing range of 56.25 μM–1.8 mM and a low detection limit of 30.7 μM, in tuna broth samples.

4 Conclusions, challenges and perspectives

Direct writing methods have been developed for the rapid prototyping of graphene electrodes using various graphene inks, filaments, or precursors. The choice of direct writing techniques, their parameters, and the chemistry of the materials being used, can be tuned to obtain graphene electrodes with different morphologies, geometries, and electrochemical performances. The advantages of direct writing, such as affordability, potential for mass production, ease of modification and integration, combined with the remarkable properties of graphene, have led to significant progresses in electrochemical sensing. This progress is driving a shift from traditional bulky electrochemical setups to flexible, portable, cost-effective, miniaturized POC devices. Despite the recent advancements in the literature, graphene-based devices for POC testing in real-life scenarios still encounter challenges in fabrication, operation, and data analysis.

The ongoing development of graphene inks faces challenges associated with non-uniform particle sizes and structures. While various methods exist for producing graphene-based materials, the industrial scale production has yet to accomplish. Achieving a balance between ideal printing and an electrode with good functional performance is difficult, as both depends factors such as the surface tension and viscosity of the ink, the substrate surface energy, and sintering conditions. This can potentially be overcome by preparing novel inks, pre-functionalizing the substrates, and post printing treatments. For 3D printing, the availability of graphene-based fillers is limited. The presence of polymer fillers can significantly diminish the advantages of graphene, such as its large specific surface area and conductivity. The underlying mechanism for LIG production is not yet fully understood, making it difficult to precisely tune the properties of LIG with high reproducibility. Nevertheless, doping with heteroatoms or nanoparticles can improve its performance.[196] Mechanical stability is also a concern, as the

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Table 2 Representatives of electrochemical sensors based on direct written graphene electrodes

Precursors	Patterning methods	Sensing platform	Targets	Dynamic range	Limit of detection	Ref.
Gr dispersion	AJP	Gr/antibody	Histamine	56.25 μM–1.8 mM	30.7 μM	78
Gr dispersion	AJP	Gr/antibody	Interferon gamma	0.1–5 ng ml^{-1}	25 pg ml^{-1}	79
			Interleukin 10	0.1–2 ng ml^{-1}	46 pg ml^{-1}	
Gr dispersion	AJP	Gr/antibody	Spike RBD	1–1000 ng ml^{-1}	22.91 ± 4.72 pg ml^{-1}	168
			Spike s1 (COVID - 19)		110.38 ± 9.00 pg ml^{-1}	
Paperboard	DLW	LIG/aluminosilicate particle	Ascorbic acid	2.0–5.0 mM	—	169
			Caffeic acid	0.91–2.86 mM		
			Picric acid	0.48–2.0 mM		
GO/noble metal ions	DLW	rGO/Au	Caffeic acid	0.5–100 μM	50 nM	170
		rGO/Au	NO_2^-	1–100 μM	120 μM	
		Pt@rGO	H_2O_2	5–2000 μM	0.6 μM	
PI	DLW	LIG/PEDOT/MIP	Chloramphen	1 nM–10 mM	0.62 nM	171
PI	DLW	LIG	Clothianidin	10–40 μM	823 nM	172
			Imidacloprid		384 nM	
			Thiamethoxam		338 nM	
			Dinotefuran		682 nM	
PI	DLW	LI/PPPA/anti-cortisol	Cortisol	0.43–50.2 ng ml^{-1}	0.08 ng ml^{-1}	173
PI	DLW	LIG/magmatic bead/DNA	*Cryptosporidium parvum*	0.1 pM–25 nM	3 pM (ECL) 47 pM (EC)	174
GO	DLW	rGO/anti-*E. coli*	*E. coli* O157:H7	917–2.1 × 10^7 cfu ml^{-1}	283 cfu ml^{-1}	152
GO	DLW	rGO/Cu NPs	Glucose	1 μM–4.54 mM	0. 35 μM	175
GO	DLW	rGO/Ag NPs	H_2O_2	0. 1–10 mM	7. 9 μM	176
PI	DLW	LIG/AuNS/aptamer	Human epidermal growth factor receptor 2	0.1–200 ng ml^{-1}	0.008 ng ml^{-1}	177
PI	DLW	LIG/ion selective membrane	K^+	10^{-6}–10^{-2} M	$10^{-5.01}$ M	178
			NO^{3-}		$10^{-5.07}$ M	
		LIG/enzyme	NH^{4+}	40–120 pM	$10^{-4.89}$ M	
			Parathion		15.4 pM	
PI	DLW	LIG/anti-Salmonella	*Salmonella enterica*	25–10^5 cfu ml^{-1}	13 ± 7 cfu ml^{-1}	179
PI	DLW	LIG	Uric acid	20–80 μM	0.74 μM	162
			Tyrosine	50–200 μM	3.6 μM	
PI	DLW	LIG	*Trans*-resveratrol	0.2–50 μM	0.16 μM	180
GO/MWCNTs/Nafion	EXP	rGO/MWCNTs/Nafion aerogel	Acetone vapor	0–100 ppm	5 ppm	57
			Dopamine	0.1–10 μM		
CNT/GO	EXP	Field effect transistor, with CNT/rGO electrodes, PEDOT: PSS channel	Dopamine	1 μM–10 mM	6 μM	181
Chitosan/rGO	EXP	Chitosan/rGO	Glucose	0.5–4 mM	0.45 mM	58
Gr/PLA	FDM	rGO-PLA/tyrosinase	Catechol	30–700 μM	0.26 μM	182
			Serotonin	0.3–10 μM	0.032 μM	
Gr/PLA	FDM	Gr/PLA/Bi NPs	Cd^{2+}	100–500 nM	82 nM	183
		Gr/PLA/Bi NPs	Pb^{2+}	80–500 nM	11 nM	
		Gr/PLA	Hg^{2+}	20–100 nM	6.1 nM	
Gr/PLA	FDM	Gr/PLA	Cocaine	20–100 μM	6 μM	184
Gr/PLA	FDM	Gr/PLA/Au NPs/DNA	COVID-19	1.0–50.0 μM	0.30 μM	185
		Gr/PLA/Au NPs	Creatinine	0.050–3.2 m M	0.02 mM	
Gr/PLA	FDM	Gr/PLA/GOx	Glucose	0.5–6 mM	15 μM	186
		Gr/PLA	Uric acid	0.5–250 μM	0.02 μM	
		Gr/PLA	Nitrite	0.5–250 μM	0.03 μM	
Gr/PLA	FDM	Gr/PLA/Au NPs/HRP	H_2O_2	150–600 μM	9.1 μM	187
Gr/PLA	FDM	Gr/PLA	L-methionine	5.0–3000 μM	1.39 μM	188
Gr dispersion	IJP	Gr/Au NPs/anti-cortisol	Cortisol	0.2–1.0 mM	10 μM	189
		Gr/GOx/PB	Glucose	10 pm–100 nM	10 pm	
Gr dispersion	IJP	Gr/aminated montmorillonite clay mineral	Gentisic acid	1–21 μM	0.33 μM	190
Gr dispersion	IJP	Gr gate electrode/GOx	Glucose	30–5000 μM	100 nM	191
		Gr gate electrode/lactate oxidase	Lactate	2–30 mM	100 nM	
GO dispersion	IJP	rGO/anti-HT-2	Ht-2 mycotoxin	6.3–100 ng ml^{-1}	1.6 ng ml^{-1}	42

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Table 2 *(continued)*

Precursors	Patterning methods	Sensing platform	Targets	Dynamic range	Limit of detection	Ref.
FeO NPs/rGO/graphite dispersion	IJP	FeO NPs/rGO/graphite	Lurasidone	50–2150 ng ml^{-1}	15.64 ng ml^{-1}	192

Abbreviations: AJP, aerosol jet printing; EHDP, electrohydrodynamic printing; EXP, extrusion printing; FDM, used deposition modelling; DLW, direct laser writing; Gr, graphene; MWCNTs, multi-walled carbon nanotube; CNT, carbon nanotube; PPPA, poly (pyrrole propionic acid); Gox, glucose oxidase; NPs, nanoparticles.

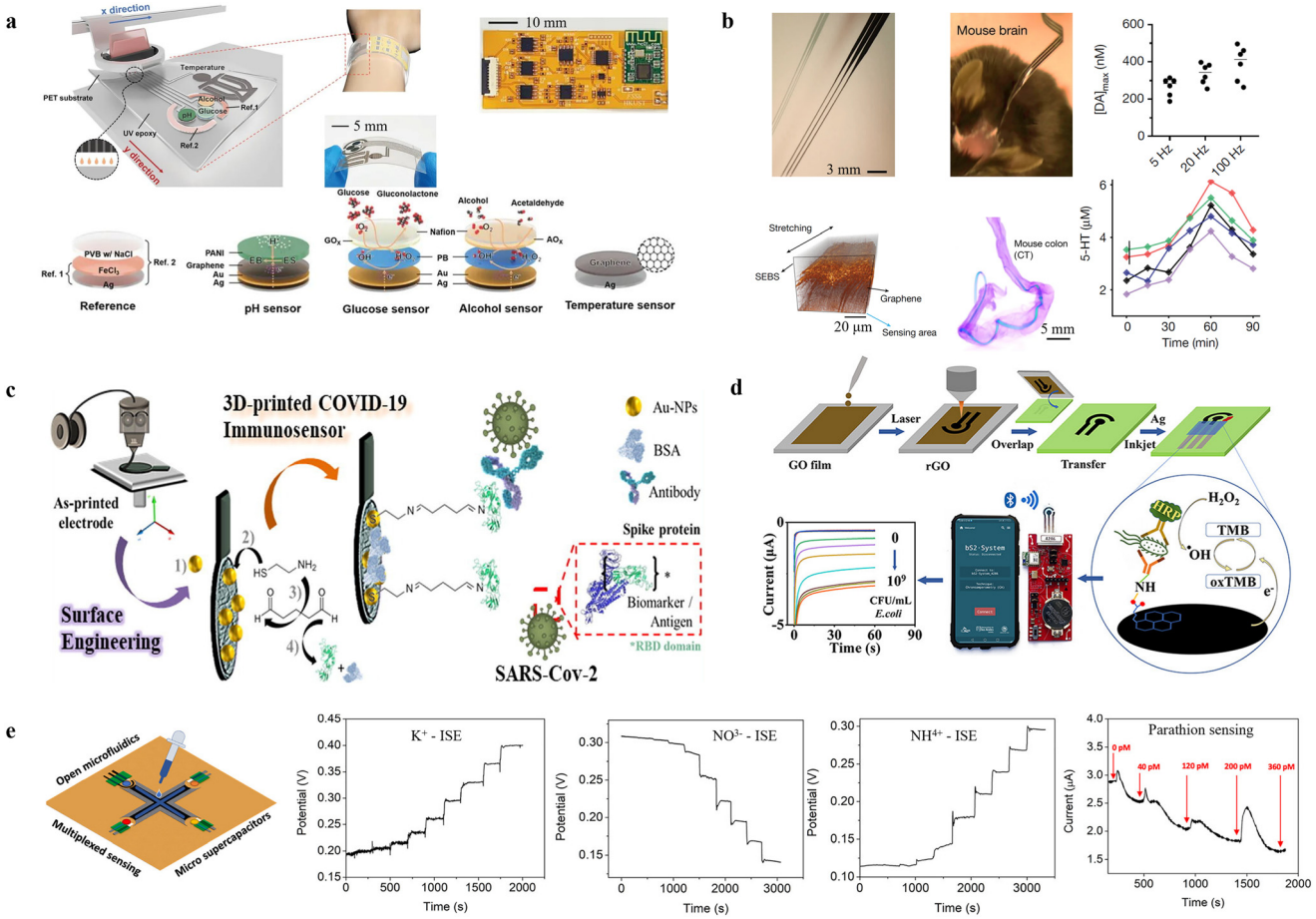

Fig. 5 Direct written graphene-based sensors for electrochemical sensing. (a) A fully inkjet printed wearable system for multiplexed epidermal sweat analysis. Reproduced from ref. 194 under creative commons CC-BY license. (b) Stretchable, implantable LIG-based neurochemical sensors, for the detection of dopamine (DA) in the brain and serotonin in the gut. Reproduced from ref. 195 with permission from Springer Nature, copyright 2022. (c) 3D printed graphene electrodes for the POC detection of COVID-19. Reproduced from ref. 124 with permission from Elsevier, copyright 2021. (d) A laser-reduced graphene oxide electrode-based portable platform for *E. coli* detection. Reproduced from ref. 152 under creative commons CC-BY license. (e) LIG-based open microfluidic system for multiplexed environmental biosensing. Reproduced from ref. 178 with permission from American Chemical Society, copyright 2022.

delicate porous structure of LIG may degrade over long-term exposure to the atmosphere. This becomes more pressing when flexible/portable or wearable sensors are highly desired these days. Combining with elastomers[195] and hydrogels[197] can be promising to obtaining robust, flexible, stretchable graphene electrodes.

There are still difficulties for the commercialization of electrochemical sensors, so as to graphene-based ones. One of the main reasons is the fouling of sensors due to the complexity of the biomatrix. Nanoengineered structures and coatings can be a promising avenue for antifouling.[198] Notably, most biosensing processes necessitate multiple washing steps, which are both time-consuming and relying on trained personnel, thereby rendering them less user-friendly for POC testing. Label-free sensing strategies[199] and automatic sample processing setups[200] present a potential

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solution. However, it is crucial to thoughtfully select stable, specific bioreceptors with strong affinity towards the targets.

Machine learning could guide the entire design-to-fabrication process, eliminating the intricate and time-consuming optimization of the printing process.[201] It can also aid in eliminating signals stemming from contaminants and facilitates the interpretation of extensive data, thereby achieving heightened sensitivity, accuracy, and a broader response range.[202]

Miniaturized, integrated, multifunctional devices are highly desired for electrochemical sensing, but significant challenges persist.[203] Such devices require sampling and sensing structures, software, data transmission/analysis, and power sources. New functional inks and improved printing resolutions, could help to enhance the detection sensitivity and reduce the device size. Moreover, the ability to direct write many of other components, such as: substrates, electronics, microfluidics and batteries[204–206] means that we are heading towards a truly whole printed system.

With all the fabrication techniques described in this review, there is a constant need to improve their respective devices compliance with the REASSURED criteria established by the WHO. All devices and fabrication methods can be improved to reduce their costs, make them more environmentally friendly, more sensitive, specific, and deliverable. User friendliness can be improved by incorporating with sister technological developments such as smartphones. Direct writing methods are inherently suited to these requirements, since the devices can be quickly fabricated at the point of use at a low cost and typically with less waste than other methods, but there is always scope for improvement in all of these aspects of the device performance.

Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

Conflicts of interest

There are no conflicts to declare.

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