Different approaches for fabrication of low-cost electrochemical sensors

Jéssica Santos Stefano[1], Luiz Otávio Orzari[1], Habdias Araújo Silva-Neto[2], Vanessa Neiva de Ataíde[3], Letícia Francine Mendes[3], Wendell Karlos Tomazelli Coltro[2], Thiago Regis Longo Cesar da Paixão[3], Bruno Campos Janegitz[1,*]

[1]Department of Nature Sciences, Mathematics, and Education, Federal University of São Carlos, Araras, SP 13600-970, Brazil.

[2] Institute of Chemistry, Federal University of Goiás, Goiânia, GO 74690-900, Brazil.

[3] Department of Fundamental Chemistry, Institute of Chemistry, São Paulo University-USP, São Paulo, SP 05508-900, Brazil.

***** Corresponding author: Prof. Bruno C. Janegitz

E-mail: brunocj@ufscar.br

Abstract

Electrochemistry combined with economical and sustainable platforms (such as paper) provides portable, affordable, robust, and user-friendly devices. In general, techniques such as photolithography and sputtering are excellent alternatives for producing these platforms. However, due to the requirement of expensive and sophisticated instrumentation as well as cleanroom facilities, these techniques have limited access. Thus, the search for easy to use and produce approaches have been reported, employing consumables, including adhesives, carbon ink, graphite, pencil, office paper, paperboard, among others. In this sense, in this mini-review, we discuss various strategies explored to fabricate low-cost electrochemical sensors, including its main applications. Different manufacturing methods, such as screen and stencil printing, laser-scribing, and pencil drawing, will be discussed here, emphasizing the performance of the obtained devices, in addition to their advantages and disadvantages.

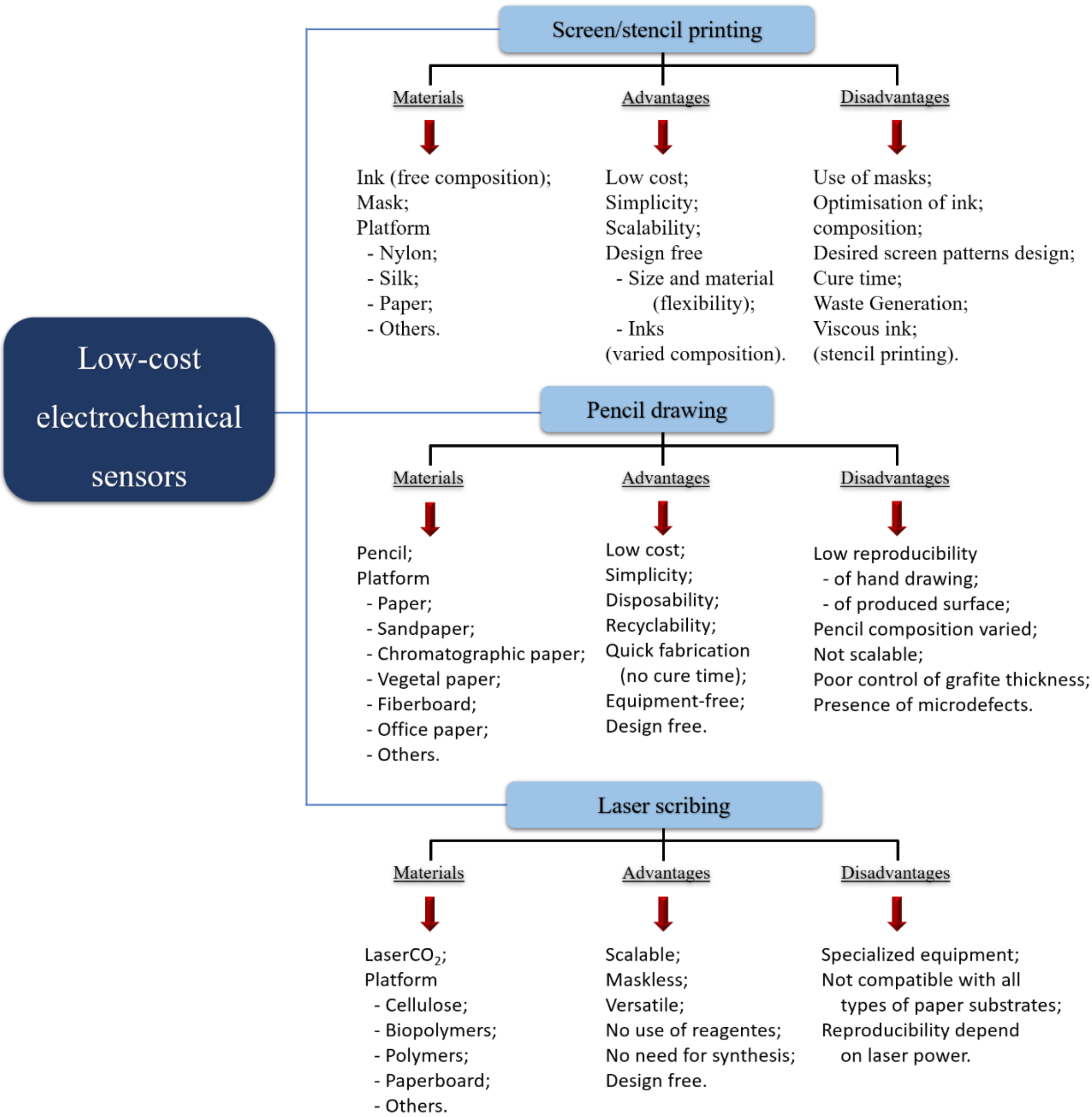
Keywords: screen-printing, pencil drawing, laser scribing, manufacture of devices, and low-cost analytical devices

Introduction

Disposable electrochemical sensors manufactured on low-cost and eco-friendly platforms (such as nylon, silk, or even paper) provide the possibility to develop portable analytical systems which are more accessible to the academic community, contributing to the design of new analytical tools for sensing a variety of compounds, and enabling the development of new point-of-care devices. The extensive use of paper for the development of electrochemical sensors, after the creation of Henry et al. 12 years ago [1], shows how important and attractive the use of cheap and readily available materials is for the fabrication of portable, miniaturized, susceptible, and costless analytical systems [2]. Although paper is a cheap raw material, its use is very employed coupled with expensive and sophisticated instrumentation, such as photolithography or sputtering techniques, to manufacture analytical platforms, restricting access to low-budget laboratories [3,4].

Therefore, the development of homemade disposable sensors based on techniques that do not require elaborated steps and expensive equipment, such as screen- and stencil printing, pencil drawing, laser-scribing, among others, is desirable. Many applications can be found in literature, including areas such as medical, pharmaceutical (drug quality control), industrial (waste monitoring), energy, and environmental (monitoring of potentially harmful substances) [5–11].

These techniques consist of the fabrication of conductive tracks with minimal instrumentation in inert surfaces for electrochemical transduction and allow the use of cheap materials, including pencils, inks, papers, and adhesives, allowing the production of devices with free designs for all kinds of applications [4]. However, some points in the manufacturing of sensors using these techniques have to be highlighted since they can strongly influence the performance of the final device, thus producing sensors with low reproducibility, conductivity, and sensitivity. Scheme 1 presents the main characteristics of these techniques, highlighting their advantages and disadvantages.



Scheme 1. Representative scheme presenting the advantages and disadvantages, and the materials employed for each approach.

Now, we present a brief overview regarding different approaches for the production of low-cost electrochemical sensors using easy-to-find consumables such as adhesives, carbon inks, graphite, pencil, office paper, paperboard, among others, including pencil drawing, laser-scribing, and screen-printing techniques. A brief description of the fabrication protocols including the required materials will be presented along with recent applications, highlighting the characteristics for each method, advantages, and disadvantages, and the advances considered significant in the development of this area.

Manufacturing and Materials

Recent scientific literature presents many alternatives for disposable device production, some aiming for large-scale, others tend towards slower-paced and detailed approaches, with freedom of design and raw materials [12–15]. Among these, screen- and stencil-printing, pencil drawing, and laser-scribing techniques bring an exciting balance of the characteristics mentioned above.

Screen and stencil-printing

The screen-printing technique focused on electrochemical devices consists of applying a conductive ink over an inert surface through a mesh screen that delimitates the electrode design. Commonly, these screens are composed of nylon, silk, or even paper [16–18]. Stencil-printing is a very similar approach, involving a less expensive screen, usually made of cheaper polymers, while also not requiring the coupled equipment for ink immobilization [19,20]. A representative scheme for the fabrication of electrochemical devices using those techniques is summarized in Figure 1.

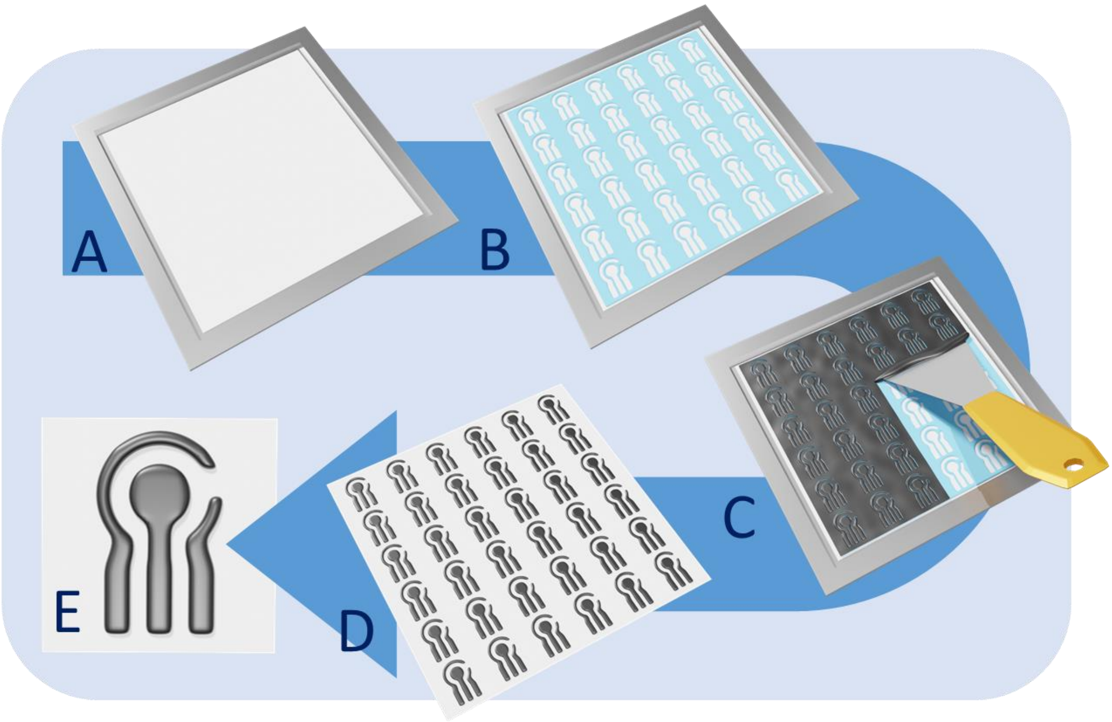


Figure 1. Fabrication scheme of electrochemical systems using a screen or stencil printing. Over the surface of a substrate (A), the screen is placed (B). The electrodes are painted with conductive ink (C), and after the removal of the screen (D), the device is ready to use (E).

The relatively lower cost, scalability, and design freedom are attractive qualities of these procedures [21,22]. Still, it is essential to highlight that some waste is generated and the electrodes may need time to cure, depending on the polymeric vehicle and additives used. While developing conductive inks for such methods, it is essential to pay attention to a few questions: for screen-printing, almost any ink composition could be used, but the stencil-based operation needs more viscous ink compositions to avoid mask leaking and excessive adhesion; the polymeric matrix must be carefully chosen, aiming for higher conductive particle dispersibility, without significant viscosity change. As the final composition must maintain electrical conductivity and still be liquid, the addition of conductive particles must be done carefully. It is common to think that higher amounts of these particles would result in better electrochemical performance (higher peak current, lower peak-to-peak separation, and overall device sensibility). However, after a material threshold, the ink may lose its liquid aspect, or the final device may present a high surface charge, changing the electrochemical behavior. Another essential aspect to consider is the use of ink additive agents, such as stabilizers and solvents. These can be used to avoid composition problems, such as nanoparticle agglomeration or lower or slower material interaction,. Especially the addition of solvents must be considered with care, as small quantities may be sufficient for a considerable fast cure process, cracking the surface of the electrode, making them unusable. Also, the device substrate used must not interfere with the electrochemical processes, but its choice may affect the ink adhesion. Higher exposed functional groups interaction moments between substrate-ink, while lower interaction moments between ink-mask and substrate-mask are preferable [21,22].

The use of ink-based devices brings two main possibilities to the electroanalytical chemistry: the high versatility of the ink composition, as numerous problems could be solved by the addition of a new component in the formulation; and the still possibility of electrode surface modification, now with a substrate of a relatively low-cost.

Screen and stencil-printed electrodes could be modified to perform specific and controlled biological processes [5–7]. In this regard, Orzari et al.[23] reported a self-adhesive electrode preparation. The graphite ink is applied on an adhesive paper sheet, which is cut into a series of electrodes with a cutting printer. After 40 min of drying, electrodes were modified by casting a glucose oxidase dispersion. Succeeding, the modified devices were employed to detect glucose in saline solution and synthetic saliva samples. Also, Srisomwat and collaborators [6] for example, proposed the first pop-up DNA device as an alternative 3D microfluidic paper-based analytical device platform for label-free HBV DNA detection using cellulose paper. The electrodes were manufactured using an in-house screen-printing technique. The pop-up design was devised to allow the control of a fluid path, incubation time, and electrical connectivity by simple folding. In addition, the architecture has allowed the accessible introduction of samples, avoiding contamination and minimizing the exposure of biofluids.

Pencil drawing

Pencil drawing protocol is a faster way to transfer graphite flakes to substrates since it does not require additional liquid binder to incorporate the carbon-based particles and more steps involving heating to evaporate solvents [24,25]. Thus, using graphite pencils and a good area determining the material, drawing any 2D electrode structure of interest over a suitable substrate is possible. The well-known porous analytical platforms successfully deposited conductive graphite particles, including cellulose fiber and polymeric plastics [24–26]. However, it is noteworthy that the pencil grade is crucial, as the quantity of graphite dispensed by friction must be significant for better results. It is of note that a pencil grade of 6B or higher (softer pencils) is preferable [27,28].

When successfully transferred to porous substrates via mechanical friction between the pencil tool and target substrate, the pencil graphite enables the creation of well-defined graphite regions with adequate electrical conductivity. The layers formed or times drawn are also essential factors. Since the process is handmade, it is subject to the strength and pressure applied, thus, providing devices with different electrochemical performances with non-homogeneous graphite distribution. Therefore, it is recommended to re-draw the same device around 10 times, as Foster et al.[26] the painting process of the RE with Ag ink or Ag/AgCl ink can also guarantee a more stable pseudo-reference electrode (p-RE) [28,29]. The delimitation of the geometric area from the obtained devices is the key to fabrication success. Also, higher electrical conductivity can be obtained in sandpaper platforms because this abrasive structure contributes to depositing more graphite flakes and consequently increases the thickness film dimensions [29]. This free-binder fabrication offers a material surface with similar electrical resistivity values (60 to 700 Ω cm) to conventional materials constructed via protocols based on laser scribing, stencil, inject, and screen-printing technologies. One of the most common substrates employed in pencil-drawn devices is cellulose-based ones, such as paper [22] and fiberboard [14]. The attractive features of electrochemical paper-based devices (ePADs), like relatively low-cost, disposability, recyclability, and effective wide potential window are very attractive as well as the creation of flexible and biocompatible devices, with easy integration of the electrochemical system to others, such as microfluidics [13] and several on-skin electronics [30].

The pioneering study involving the manufacturing of pencil-drawn electrodes was proposed by Dossi et al.[31]. A full system was successfully deposited upon cellulose fiber substrate with a geometric area delimited by wax ink in their report, as summarized in Figure 2A. Santhiago et al.[32] manufactured flexible and higher conductively graphite-based films upon cellulose structures (Figure 2B). The reported pencil-drawn electrodes (PDEs) were treated via electrochemical oxidation/reduction to remove excess non-conductive materials impregnated on the surface and to increase sensing performance. Kanaparthi and collaborators [33] reported a wearable device combining pencil graphite, cellulose fiber, and textile mask (Figure 2C) with a cost of ca. US$ 0.03. Koga et al.[8] constructed ZnO nanowires incorporated with PDEs, as illustrated in Figure 2D. The reported results revealed a hybrid surface efficiently bridged network structures with satisfactory capacitive performance of NO_2 gas with detectable levels ranging from 3.9 to 98 ppm.

Other paper substrates, such as filter paper [31], chromatographic paper [25], vegetal paper [9], corrugated fiberboard (CFB) [14], office paper [28] and sandpaper [29] can also be found in the literature as emerging and affordable materials to direct deposited graphite flakes [22,31]. In this sense, Orzari et al.[14] manufactured PDEs upon CFB platform (Figure 2E). Ataide et al.[28] constructed PDEs on office paper and exposed the surface to a CO_2 laser (Figure 2F). This pre-treatment step improved the electrodes' performance and revealed satisfactory LOD values. Rocha et al.[29] reported a combination of PDEs and a 3D-printed holder (Figure 2G) with a cost *ca.* US$ 0.45. This strategy promoted a reusable delimitation of the geometric area from the ePAD, avoiding problems associated with sampling evaporation.

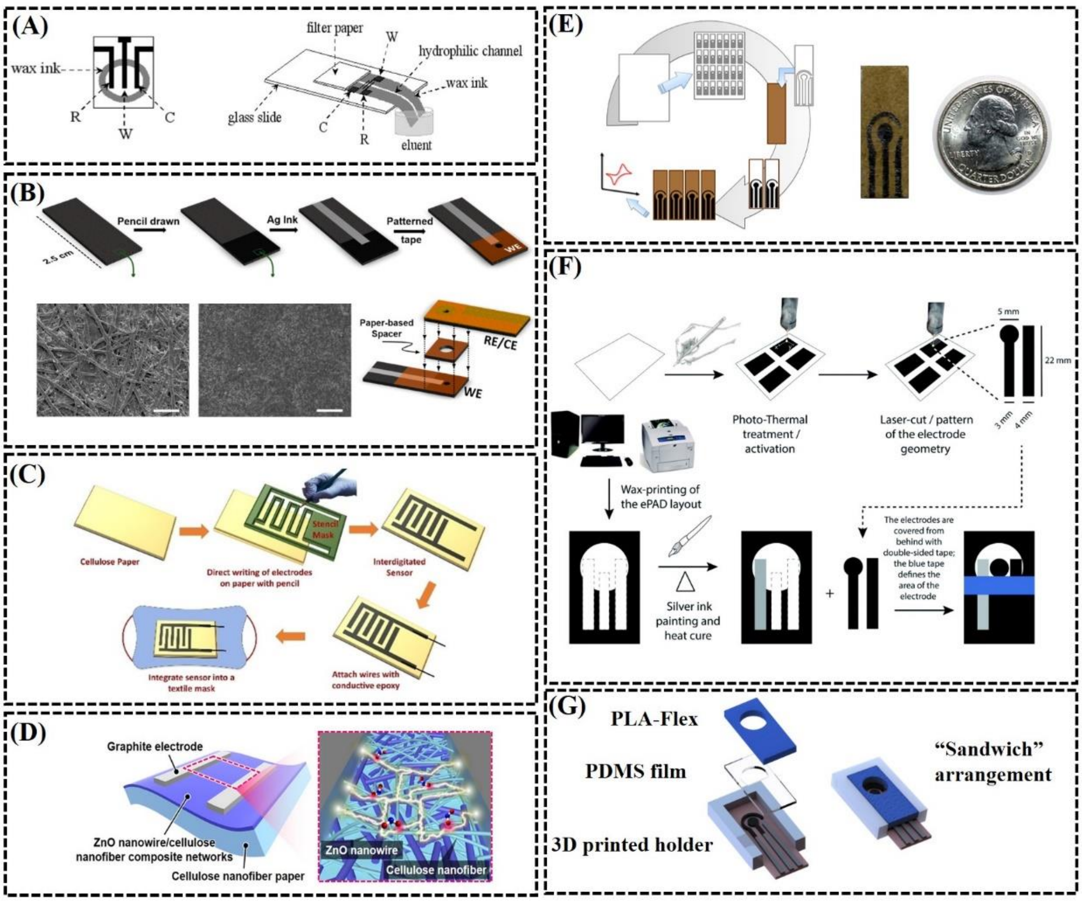


Figure 2. Schematic representation of the pencil drawing process. (A) electrodes fabricated upon cellulose and glass slide reproduced from [31] with permission. (B) steps involving the manufacturing of flexible graphite-based electrodes reproduced from [32] with permission. (C) pencil drawing electrodes integrated on a textile mask reproduced from [33] with permission. (D) nanomaterials of ZnO combined with pencil drawing electrodes reproduced from [8] with permission. (E) multiples electrodes development upon CBF substrate reproduced from [14] with permission. (F) fabrication and pre-treatment steps of the electrodes reproduced from [28] with permission. (G) combination of the electrochemical cell and 3D printing reproduced from [29] with permission.

Laser-scribing

The laser-scribing technique consists of controlled carbonization, using a laser (generally CO_2) to produce a conductive path in materials with insulating characteristics (non-conductive carbon, polymers, cellulose, and biopolymers). This conversion is possible since the pyrolysis of organic compounds, such as cellulose generates carbon-based materials. In this sense, the non-conductive material is turned into graphite-like structures under high temperatures (up to 2000 ºC). The precursor material influences the physical and chemical characteristics of the obtained structures [34].

The laser parameters (such as power, focal distance, pulse density, among others) and carbonization conditions play an essential role in the material's morphological, chemical, and physical characteristics [35]. Furthermore, laser-scribing physical and chemical properties can be modulated according to the atmosphere where the substrate carbonization is carried out [36] or even using treated/modified platform surfaces [37].

Precisely because these parameters are critical for the final obtained materials, their fabrication reproducibility is still challenging.

Nonetheless, this technique has been widely studied and enables sensors and biosensors for the most varied purposes, from pharmaceutical and biological to energy storage, environmental analysis, among others [10]. Nayak et al.[38] fabricated electrodes using direct laser writing on polyimide under ambient conditions (Figure 3A). Furthermore, these developed sensors can be modified to improve sensitivity, detectability, specificity, and other characteristics. In this sense, different materials can be employed, including nanoparticles, polymers, and biological recognition materials (enzymes, antibodies, nucleic acids) [37,39,40].

In 2017, Araujo et al.[3], reported a simple, fast, single-step laser-scribing, green method for scalable fabrication electrochemical sensors on a paperboard platform for the first time. Their material presented randomly oriented porous structures and graphene sheets (Figure 3B). Other precursor materials can also be used to obtain graphene from the laser-scribing technique. In this regard, Zhang and collaborators [41] fabricated for the first time electrochemical devices using phenolic resin as precursor material and an inexpensive 405 nm laser under ambient conditions to fabricate an electrochemical glucose biosensor after the modification of the obtained electrode with Ferroce formic acid (Figure 3C). Also, Beduk and co-workers [11] used polyimide as a precursor material. They combined it with molecularly imprinted polymer to form a biorecognition layer for the sensitive and selective electrochemical detection of Bisphenol A for environmental monitoring waters and plastic samples (Figure 3D).

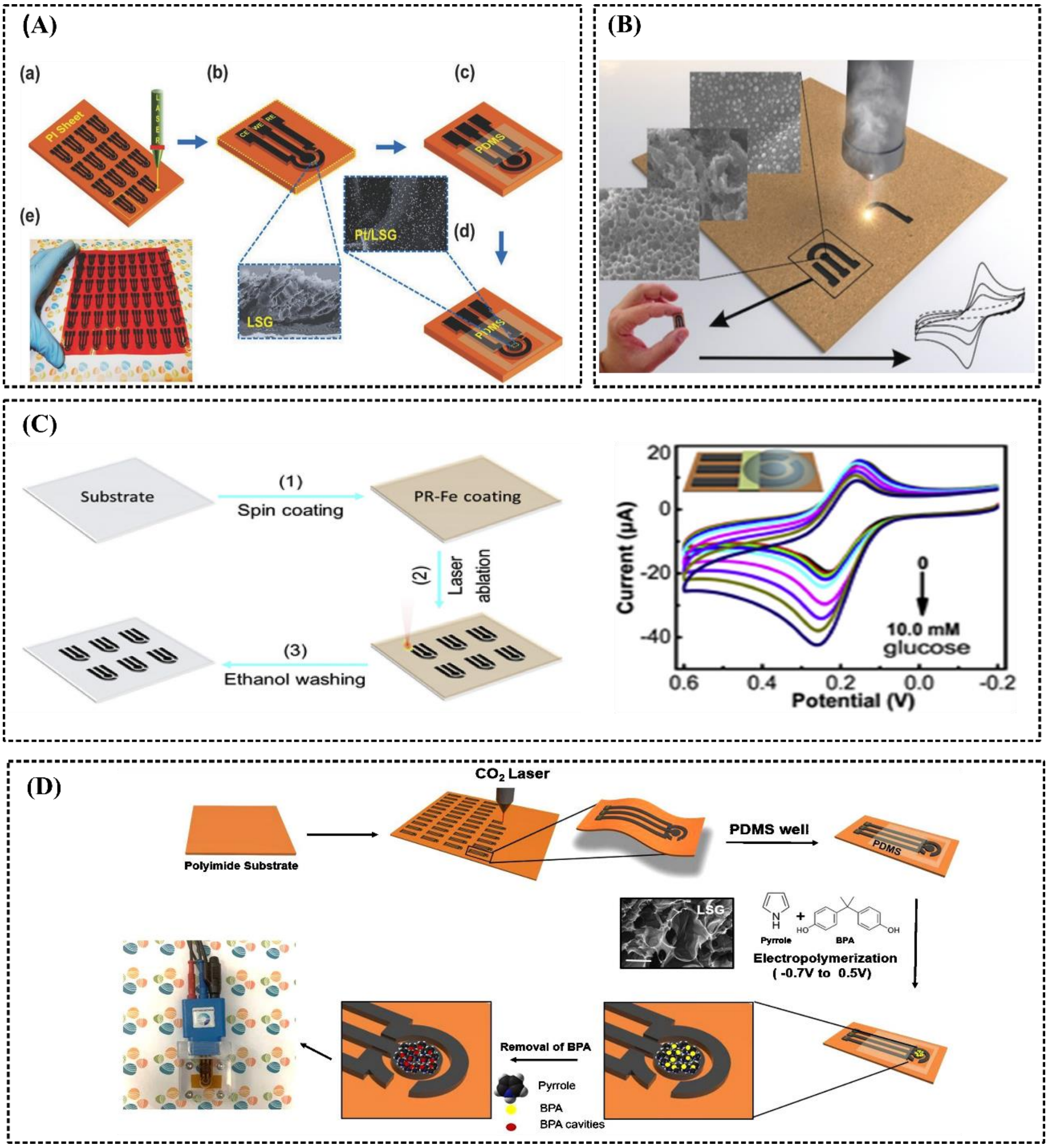


Figure 3. Schematic representations of the manufacturing processes of carbon-based electrochemical devices on different substrates using the laser-scribing technique. (A) Fabrication of electrodes using laser on a polyimide (PI). Reproduced with permission from [38]. (B) Laser-scribing electrochemical paper-based devices (LS-ePADS) from a cardboard substrate using a CO_2 laser. Reproduced with permission from [3]. (C) Fabrication procedures of the graphene-like electrodes and the electrochemical detection of glucose. Adapted with permission from [41]. (D) Fabrication steps of LIG devices

modified with molecularly imprinted polymer (MIP). Reproduced with permission from [11].

As can be seen, the aforementioned techniques are very interesting alternatives for the manufacturing of low-cost electrochemical sensors. The ease obtention of the employed materials makes these approaches very desirable for creating new platforms. However, some disadvantages of each method have to be considered. The need for specialized equipment, required for laser scribing, increases the cost for this technique, nevertheless, the presence of the laser brings as advantage the possibility of a scalable production. Stencil and screen printing techniques also allow a scalable production, simply and with design free, however, the use of masks is required and, in opposition to laser scribing, there is a wasting generation from the ink residues employed. The pencil drawing approach involves the handmade production, thus, the scalable production of pencil-drawn sensors is not possible. Nevertheless, this technique is very simple, there is no waste generation, and no equipment is required for their preparation.

The reproducibility of the obtained sensors in all cases can be a problem, however, the possibility of creating several devices in a fast way, and the disposability of the obtained devices allow the easy replacement of the devices with poor performance.

The use of the aforementioned approaches, though already very explored in the literature, opens up space for new perspectives. The use of different (nano)materials can be explored, as well as the immobilization of different biological materials, increasing the range of application of the obtained sensors. Furthermore, the validation of the obtained sensors, and their application in real samples is of great importance, focusing on the commercialization of these devices, which are a great basis for the production of low-cost point-of-care devices.

Conclusions

The techniques presented here have demonstrated great importance for the advancement in the manufacture of low-cost disposable electrochemical sensors for the possibility of using consumables as a base material for their construction, providing attractive characteristics such as simplicity, sustainability, and applicability in a single device. Therefore, these approaches contribute significantly to reaching simple, inexpensive, robust, and quick analytical systems for on-site application monitoring of compounds potentially harmful to human health and the environment, rapid tests for pathogens, forensic compounds, industrial or food quality control, among others. However, it is worth mentioning that even presenting several qualities as mentioned above, these techniques have disadvantages that require special care, such as reproducibility of the analytical devices, and consequently, systematic errors in the desired applications can be present. Nevertheless, it is possible to circumvent such disadvantages by creating several devices in a row. The techniques allow the manufacture of various devices quickly, easily, and with low consumption of raw materials relatively cheap. Thus, a poor-quality device can easily be replaced and provide satisfactory and reproducible results. Finally, such techniques allow a significant advance in the area of sensors, allowing various academic research groups to have access to alternative means of homemade manufacture electrochemical sensors and biosensors, spreading the access of scientific research to academic/research groups worldwide, thus, producing a significant number of works with relevance.

Conflict of interest statement

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

Papers of particular interest, published within the period of review have been highlighted as:

* of special interest
** of outstanding interest

[1] W. Dungchai, O. Chailapakul, C.S. Henry, Electrochemical detection for paper-based microfluidics, Anal. Chem. 81 (2009) 5821–5826. https://doi.org/10.1021/ac9007573.

[2] A.M. López-Marzo, A. Merkoçi, Paper-based sensors and assays: A success of the engineering design and the convergence of knowledge areas, Lab Chip. 16 (2016) 3150–3176. https://doi.org/10.1039/c6lc00737f.

[3] W.R. de Araujo, C.M.R. Frasson, W.A. Ameku, J.R. Silva, L. Angnes, T.R.L.C. Paixão, Single-step reagentless laser scribing fabrication of electrochemical paper-based analytical devices, Angew. Chemie Int. Ed. 56 (2017) 15113–15117. https://doi.org/10.1002/anie.201708527.

*[4] V.N. Ataide, L.F. Mendes, L.I.L.M. Gama, W.R. de Araujo, T.R.L.C. Paixão, Electrochemical paper-based analytical devices: ten years of development, Anal.

Methods. 12 (2020) 1030–1054. https://doi.org/10.1039/C9AY02350J.

It reports the developments of electrochemical analytical devices based on paper and reports on the manufacturing procedures and applications of the most diverse sensors produced.

**[5] P. Teengam, W. Siangproh, S. Tontisirin, A. Jiraseree-amornkun, N. Chuaypen, P. Tangkijvanich, C.S. Henry, N. Ngamrojanavanich, O. Chailapakul, NFC-enabling smartphone-based portable amperometric immunosensor for hepatitis B virus detection, Sensors Actuators B Chem. 326 (2021) 128825. https://doi.org/10.1016/j.snb.2020.128825.

A smartphone-controlled screen-printed electrochemical sensor operated entirely by Near Field Communication to detect the hepatitis B virus using a low-cost sensor system operated from a smartphone.

[6] C. Srisomwat, P. Teengam, N. Chuaypen, P. Tangkijvanich, T. Vilaivan, O. Chailapakul, Pop-up paper electrochemical device for label-free hepatitis B virus DNA detection, Sensors Actuators B Chem. 316 (2020) 128077. https://doi.org/10.1016/j.snb.2020.128077.

*[7] R.K. Mishra, J.R. Sempionatto, Z. Li, C. Brown, N.M. Galdino, R. Shah, S. Liu, L.J. Hubble, K. Bagot, S. Tapert, J. Wang, Simultaneous detection of salivary Δ9-tetrahydrocannabinol and alcohol using a Wearable Electrochemical Ring Sensor, Talanta. 211 (2020) 120757. https://doi.org/10.1016/j.talanta.2020.120757.

Talanta. 211 (2020) 120757. https://doi.org/10.1016/j.talanta.2020.120757.

A new detection platform based on a wearable electrochemical detection device for the direct and decentralized simultaneous detection of THC and alcohol in saliva. The device is based on a ring containing a voltammetric THC sensor and an amperometric alcohol biosensor, along with the wireless electronic components embedded in the ring box, thus, can allow law enforcement personnel to track drivers in a single traffic stop.

[8] H. Koga, K. Nagashima, Y. Huang, G. Zhang, C. Wang, T. Takahashi, A. Inoue, H. Yan, M. Kanai, Y. He, K. Uetani, M. Nogi, T. Yanagida, Paper-based disposable molecular sensor constructed from oxide nanowires, cellulose nanofibers, and pencil-drawn electrodes, ACS Appl. Mater. Interfaces. 11 (2019) 15044–15050. https://doi.org/10.1021/acsami.9b01287.

[9] A.A. Dias, T.M.G. Cardoso, C.L.S. Chagas, V.X.G. Oliveira, R.A.A. Munoz, C.S. Henry, M.H.P. Santana, T.R.L.C. Paixão, W.K.T. Coltro, Detection of analgesics and sedation drugs in whiskey using electrochemical paper-based analytical devices, Electroanalysis. 30 (2018) 2250–2257. https://doi.org/10.1002/elan.201800308.

*[10] L. Fornasini, S. Scaravonati, G. Magnani, A. Morenghi, M. Sidoli, D. Bersani, G. Bertoni, L. Aversa, R. Verucchi, M. Riccò, P.P. Lottici, D. Pontiroli, In situ decoration of laser-scribed graphene with TiO2 nanoparticles for scalable high-performance micro-supercapacitors, Carbon N. Y. 176 (2021) 296–306. https://doi.org/10.1016/j.carbon.2021.01.129.

This work demonstrates satisfactory results of the use of laser engraved TiO_2-graphene nanostructures as notable candidates in microsupercapacitors for low-cost, large-scale and environmental-friendly applications.

[11] T. Beduk, A. Ait Lahcen, N. Tashkandi, K.N. Salama, One-step electrosynthesized molecularly imprinted polymer on laser scribed graphene bisphenol a sensor, Sensors Actuators, B Chem. 314 (2020) 128026. https://doi.org/10.1016/j.snb.2020.128026.

[12] B. Nagar, M. Jović, V.C. Bassetto, Y. Zhu, H. Pick, P. Gómez-Romero, A.

Merkoçi, H.H. Girault, A. Lesch, Highly Loaded Mildly Edge-Oxidized Graphene Nanosheet Dispersions for Large-Scale Inkjet Printing of Electrochemical Sensors, ChemElectroChem. 7 (2020) 460–468. https://doi.org/10.1002/celc.201901697.

[13] N. Dossi, S. Petrazzi, F. Terzi, R. Toniolo, G. Bontempelli, Electroanalytical cells pencil drawn on PVC supports and their use for the detection in flexible microfluidic devices, Talanta. 199 (2019) 14–20. https://doi.org/10.1016/j.talanta.2019.01.126.

[14] L.O. Orzari, I.A. de Araujo Andreotti, M.F. Bergamini, L.H. Marcolino, B.C. Janegitz, Disposable electrode obtained by pencil drawing on corrugated fiberboard substrate, Sensors Actuators B Chem. 264 (2018) 20–26. https://doi.org/10.1016/j.snb.2018.02.162.

[15] M. Jović, J.C. Hidalgo-Acosta, A. Lesch, V. Costa Bassetto, E. Smirnov, F. Cortés-Salazar, H.H. Girault, Large-scale layer-by-layer inkjet printing of flexible iridium-oxide based pH sensors, J. Electroanal. Chem. 819 (2018) 384–390. https://doi.org/10.1016/j.jelechem.2017.11.032.

[16] G.R. Cagnani, G. Ibáñez-Redín, B. Tirich, D. Gonçalves, D.T. Balogh, O.N. Oliveira, Fully-printed electrochemical sensors made with flexible screen-printed electrodes modified by roll-to-roll slot-die coating, Biosens. Bioelectron. 165 (2020) 112428. https://doi.org/10.1016/j.bios.2020.112428.

[17] S.J. Rowley-Neale, D.A.C. Brownson, G. Smith, C.E. Banks, Graphene oxide bulk-modified screen-printed electrodes provide beneficial electroanalytical sensing capabilities, Biosensors. 10 (2020) 27. https://doi.org/10.3390/bios10030027.

[18] I.A. de Araujo Andreotti, L.O. Orzari, J.R. Camargo, R.C. Faria, L.H. Marcolino-

Junior, M.F. Bergamini, A. Gatti, B.C. Janegitz, Disposable and flexible electrochemical sensor made by recyclable material and low cost conductive ink, J. Electroanal. Chem. 840 (2019) 109–116. https://doi.org/10.1016/j.jelechem.2019.03.059.

[19] C. Downs, A. Nejely, E. Fu, Disposable fabric-based electrochemical sensors fabricated from wax-transfer-printed fluidic cells and stencil-printed electrodes, Anal. Methods. 10 (2018) 3696–3703. https://doi.org/10.1039/C8AY01028E.

[20] A.A. Kava, C.S. Henry, Exploring carbon particle type and plasma treatment to improve electrochemical properties of stencil-printed carbon electrodes, Talanta. 221 (2021) 121553. https://doi.org/10.1016/j.talanta.2020.121553.

[21] R.R. Suresh, M. Lakshmanakumar, J.B.B. Arockia Jayalatha, K.S. Rajan, S. Sethuraman, U.M. Krishnan, J.B.B. Rayappan, Fabrication of screen-printed electrodes: opportunities and challenges, J. Mater. Sci. 56 (2021) 8951–9006. https://doi.org/10.1007/s10853-020-05499-1.

[22] J.R. Camargo, L.O. Orzari, D.A.G. Araújo, P.R. de Oliveira, C. Kalinke, D.P. Rocha, A. Luiz dos Santos, R.M. Takeuchi, R.A.A. Munoz, J.A. Bonacin, B.C. Janegitz, Development of conductive inks for electrochemical sensors and biosensors, Microchem. J. 164 (2021) 105998. https://doi.org/10.1016/j.microc.2021.105998.

[23] L.O. Orzari, R. Cristina de Freitas, I. Aparecida de Araujo Andreotti, A. Gatti, B.C. Janegitz, A novel disposable self-adhesive inked paper device for electrochemical sensing of dopamine and serotonin neurotransmitters and biosensing of glucose, Biosens. Bioelectron. 138 (2019) 111310. https://doi.org/10.1016/j.bios.2019.05.015.

[24] N. Kurra, G.U. Kulkarni, Pencil-on-paper: Electronic devices, Lab Chip. 13

(2013) 2866–2873. https://doi.org/10.1039/c3lc50406a.

[25] C.L.S.S. Chagas, L. Costa Duarte, E.O. Lobo-Júnior, E. Piccin, N. Dossi, W.K.T.T. Coltro, Hand drawing of pencil electrodes on paper platforms for contactless conductivity detection of inorganic cations in human tear samples using electrophoresis chips, 36 (2015) 1837–1844. https://doi.org/10.1002/elps.201500110.

[26] C.W. Foster, D.A.C. Brownson, A.P. Ruas De Souza, E. Bernalte, J. Iniesta, M. Bertotti, C.E. Banks, Pencil it in: Pencil drawn electrochemical sensing platforms, Analyst. 141 (2016) 4055–4064. https://doi.org/10.1039/c6an00402d.

[27] R. Kawahara, P. Sahatiya, S. Badhulika, S. Uno, Paper-based potentiometric pH sensor using carbon electrode drawn by pencil, Jpn. J. Appl. Phys. 57 (2018) 04FM08. https://doi.org/10.7567/JJAP.57.04FM08.

[28] V.N. Ataide, W.A. Ameku, R.P. Bacil, L. Angnes, W.R. De Araujo, T.R.L.C. Paixão, Enhanced performance of pencil-drawn paper-based electrodes by laser-scribing treatment, RSC Adv. 11 (2021) 1644–1653. https://doi.org/10.1039/d0ra08874a.

[29] D.S. Rocha, L.C. Duarte, H.A. Silva-Neto, C.L.S. Chagas, P. Santana, N.R.A. Filho, W.K.T. Coltro, Sandpaper-based electrochemical devices assembled on a reusable 3D-printed holder to detect date rape drug in beverages, Talanta. 232 (2021). https://doi.org/10.1016/j.talanta.2021.122408.

[30] Y. Xu, G. Zhao, L. Zhu, Q. Fei, Z. Zhang, Z. Chen, F. An, Y. Chen, Y. Ling, P. Guo, S. Ding, G. Huang, P.-Y. Chen, Q. Cao, Z. Yan, Pencil–paper on-skin electronics, Proc. Natl. Acad. Sci. 117 (2020) 18292–18301. https://doi.org/10.1073/pnas.2008422117.

[31] N. Dossi, R. Toniolo, E. Piccin, S. Susmel, A. Pizzariello, G. Bontempelli,

Pencil-drawn dual electrode detectors to discriminate between analytes comigrating on paper-based fluidic devices but undergoing electrochemical processes with different reversibility, Electroanalysis. 25 (2013) 2515–2522. https://doi.org/10.1002/elan.201300374.

[32] M. Santhiago, M. Strauss, M.P. Pereira, A.S. Chagas, C.C.B. Bufon, Direct drawing method of graphite onto paper for high-performance flexible electrochemical sensors, ACS Appl. Mater. Interfaces. 9 (2017) 11959–11966. https://doi.org/10.1021/acsami.6b15646.

[33] S. Kanaparthi, Pencil-drawn paper-based non-invasive and wearable capacitive respiration sensor, Electroanalysis. 29 (2017) 2680–2684. https://doi.org/10.1002/elan.201700438.

**[34]N. Kurra, Q. Jiang, P. Nayak, H.N. Alshareef, Laser-derived graphene: A three-dimensional printed graphene electrode and its emerging applications, Nano Today. 24 (2019) 81–102. https://doi.org/10.1016/j.nantod.2018.12.003.

An interesting review article that discussed the recent progress in the use of laser-based manufacturing of 3D printed graphene electrodes and their broad spectrum of applications.

[35] R. Ye, D.K. James, J.M. Tour, Laser-Induced Graphene, Acc. Chem. Res. 51 (2018) 1609–1620. https://doi.org/10.1021/acs.accounts.8b00084.

[36] J. Lin, Z. Peng, Y. Liu, F. Ruiz-Zepeda, R. Ye, E.L.G. Samuel, M.J. Yacaman, B.I. Yakobson, J.M. Tour, Laser-induced porous graphene films from commercial polymers, Nat. Commun. 5 (2014) 5714. https://doi.org/10.1038/ncomms6714.

[37] H. Yoon, J. Nah, H. Kim, S. Ko, M. Sharifuzzaman, S.C. Barman, X. Xuan, J. Kim, J.Y. Park, A chemically modified laser-induced porous graphene based

flexible and ultrasensitive electrochemical biosensor for sweat glucose detection, Sensors Actuators, B Chem. 311 (2020) 127866. https://doi.org/10.1016/j.snb.2020.127866.

[38] P. Nayak, N. Kurra, C. Xia, H.N. Alshareef, Highly efficient laser scribed graphene electrodes for on-chip electrochemical sensing applications, Adv. Electron. Mater. 2 (2016) 1600185. https://doi.org/10.1002/aelm.201600185.

[39] Z. You, Q. Qiu, H. Chen, Y. Feng, X. Wang, Y. Wang, Y. Ying, Laser-induced noble metal nanoparticle-graphene composites enabled flexible biosensor for pathogen detection, Biosens. Bioelectron. 150 (2020) 111896. https://doi.org/10.1016/j.bios.2019.111896.

[40] Y. Zhang, N. Li, Y. Xiang, D. Wang, P. Zhang, Y. Wang, S. Lu, R. Xu, J. Zhao, A flexible non-enzymatic glucose sensor based on copper nanoparticles anchored on laser-induced graphene, Carbon N. Y. 156 (2020) 506–513. https://doi.org/10.1016/j.carbon.2019.10.006.

[41] Z. Zhang, M. Song, J. Hao, K. Wu, C. Li, C. Hu, Visible light laser-induced graphene from phenolic resin: A new approach for directly writing graphene-based electrochemical devices on various substrates, Carbon N. Y. 127 (2018) 287–296. https://doi.org/10.1016/j.carbon.2017.11.014.