



Novel paper-based electroanalytical tools for food surveillance

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Abstract

Analytical strategies to ensure the quality and safety of food products without the need for skilled personnel are highly required. The simplicity of glucose strips for diabetes monitoring should be translated to the agri-food sector, ensuring easy evaluation of certain molecules and/or the freshness of a beverage/foodstuff. In addition to the well-known advantages that are characteristic of electroanalytical methods over other methods, paper-based materials are being used to further reduce the gap between complex laboratory testing and simple point-of-need testing. This article highlights some of the most recent advances in the development of paper-based electrochemical approaches for food surveillance, specifically focusing on the use of novel paper-based materials. Two examples are discussed: the development of a miniaturized biosensor realized on copy paper for the quantification of ethanol in commercial beers, and the measurement of ascorbic acid in food supplements within printed electronics supports. Paper-based materials have the potential to lower economic costs, simplify the operative tasks, and most importantly reduce waste generation. The continued combination of manufacturing methods and functional (smart) materials will facilitate the implementation of food analysis at the point of need.

Keywords Paper based · Food electrochemistry · Screen printing · Inkjet printing · Sustainable

Introduction

Ensuring food quality and safety is currently a hot topic globally. However, the interest arising around the word “food” relates not only to the quality of a certain product but also to the search for suitable approaches for evaluating authenticity, adulteration, toxicity, and sustainability [1]. The number of programs, including research, analysis, training, and outreach programs, the US Food and Drug Administration is developing has accelerated so as to improve the food supply chain and also to prevent acts of intentional adulteration of the food supply [2]. Institutions such as the Food and Drug Administration focus on the surveillance of food at the end stage of the supply chain. There are also huge efforts focused on the monitoring of food

at the start of the supply chain, such as the monitoring of plantations, particularly in developing countries. For example, in 2018, the Bill & Melinda Gates Foundation launched a grand challenge to solicit innovative tools and technologies for crop pest and disease surveillance in regions in low-income countries [3]. These efforts are critical to tackle malnutrition, which contributes to approximately 50% of deaths in children younger than 5 years. Thus, there is an urgent need for technologies that are able to accurately evaluate the presence of contaminants and/or the freshness/authenticity of food products at every stage of the food chain. This technology would serve to reduce waste and thus increase product yield. For example, farmers who want to measure the ethanol content of wine or look for contaminants that might spoil their corn plantation usually require the assistance of specialized personnel and, often, suitably equipped laboratories to conduct the analysis. An on-site, simple, user-friendly electrochemical sensor would allow to them to identify and solve problems earlier and therefore increase product yield [4]. These laboratory and specialized tests are not limited to the wine sector; they are also required in the meat, dairy, fish, and vegetable industries. These practices encounter several limitations, both in terms of costs and in terms of time and transportation (more than 1 day considering the sampling, transportation, analysis, and result delivery). Additional challenges

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include the recruitment of skilled personnel and sample transportation to specialized centers in remote regions.

The current state-of-the-art technologies in food analysis are primarily limited by the sample treatment step (i.e., filtration, dilution, pH adjustment, extraction, heating, etc.). They are also limited by the need for laboratory-based methods, such as high-performance liquid chromatography and chromatographic techniques coupled with mass spectrometry [5–7]. There is an increasing necessity for rapid and affordable analysis at the point of need, by providing analytical tools that can be used without expertise and bulky/expensive machinery [8]. The most commercially successful example is the diabetes strip. This tiny strip allows patients worldwide to monitor blood glucose levels [9]. This electrochemical-based device is used approximately 70 million times per day worldwide. A similar approach is desirable for the surveillance of food quality and/or the processes that are associated with food production, from farmers to industry. The use of paper as a substrate is leading the revitalization of the sensor and biosensor field [10–12]. Although work on paper does not have the same fame as trending topics such as “nano” science, its importance and novelty are uncontested within the analytical field. Paper, as a sensing solution in the field of analytical chemistry, has gained a lot of attention because of its versatility, abundance, and sustainability. However, in addition to this, it is an attractive material capable of facilitating method development, the treatment of complex matrices, and the management of waste by-products. The use of paper is not exclusive to the agri-food field: different paper-based structures have been successfully used in the development of diverse platforms, such as the detection of environmental pollutants, clinical biomarkers, and food additives [13–20]. Depending on the recognition architectures (use of enzymes and/or enzyme cocktails, agglutination, antibody-based binding, etc.), the transduction principles (colorimetric, electrochemical, fluorescence), and the influence of the matrices (amount of sample, color, turbidity, presence of gross impurities, etc.), the most suitable paper-based substrates are selected (e.g., filter paper, office paper, waxed paper, paper towel, paper-based printed electronics). Typically paper-based devices for quick monitoring rely on the colorimetric and electrochemical transduction mechanisms; however, both approaches have limitations. The main limitations of colorimetric tests are related to the sample one wishes to analyze. For instance, a colorimetric method is ineffective when one is analyzing colored matrices, even by naked-eye detection. Also, the colorimetric method is a qualitative or semiquantitative method, and is unable to quantify with accuracy the level of a specific analyte. In contrast, electrochemical devices are capable of quantitative detection, especially down to low analyte levels, because of the “blindness” to the color and turbidity of matrices (e.g., blood, tomato sauce, soil). Like the colorimetric glucometers, they can be interfaced with a battery-operated measuring device.

However, the possibility of nonspecialists to conduct autonomous and reagent-free analysis depends on the properties of paper itself. Paper-based devices can be impregnated with all the reagents used for performing a specific detection (salts, buffers, bioelements, nanomaterials) and they can filter gross matrices (blood red cells are blocked if the paper’s pores are smaller than them), and after use, paper can be incinerated (reducing the accumulation of waste).

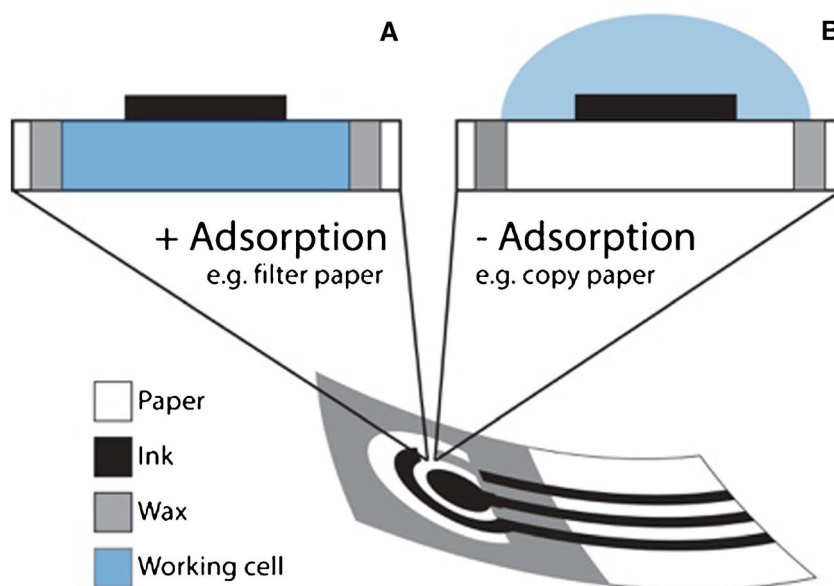
To highlight the use of electroanalytical paper-based substrates to develop user-friendly devices for food analysis, two successful illustrative examples are described in this article. To date, the realization of paper-based devices, not only electroanalytical ones, is mainly addressed by use of porous substrates such as chromatography paper. Therefore, the focus of this article is on two alternative substrates that have been successfully used, representing a further step toward the development of sustainable methods for monitoring food quality. The first example discussed here concerns the development of a miniaturized biosensor realized on copy paper for the quantification of ethanol in commercial beers [21]. As demonstrated, the use of copy paper as a substrate material for the printing of electrodes, instead of chromatography paper, leads to a 30% saving with a laboratory setup (this saving would obviously increase at the industrial-scale level), representing added value toward the realization of cheap devices. The performance of different copy papers is evaluated, also in comparison with traditional electrodes that are printed on polyester. Results and commentary are provided. The second example explores the successful integration of a single device capable of the measurement of ascorbic acid in food supplements within printed electronics supports, with the objective of printing a multifunctional single device integrated with a display and battery [22]. The creation of a single device could help to reduce generation of so-called electronic waste (e-waste) [23]. A complete paper-based device could represent a disposable solution toward the realization of fully enclosed tools for food surveillance.

Types of paper-based adsorption substrates

Paper-based electrochemical devices use mainly two types of substrates: high-adsorption and low-adsorption substrates (Fig. 1).

The former are represented by chromatography and filter paper (Whatman no. 1 filter paper, Cordenons paper (67 g/m²), cation exchanger, etc.). The advantages of using these substrates are due to the possibility of loading all of the reagents within the cellulosic structure of the paper and using a very low amount of sample (5–10 μ L). Nevertheless, the electrochemical cell is represented by the paper’s porosity itself and it is consistent with a hindered diffusion of species at the electrode surface, thus reducing sensitivity (Fig. 1, part A).

Fig. 1 Experimental setup related to the diverse types of electrochemical paper-based devices obtained by use of high-adsorption substrates (A) and low-adsorption substrates (B)



The main advantage of the latter, low adsorption, is having the printed electrodes exposed directly to the solution (higher sensitivity). With this type of paper, reagents can be loaded not within the paper but only onto the conductive ink (Fig. 1, part B). However, depending on the analytical needs, the “right” paper can be selected. Of course, both types of paper represent a sustainable alternative to the widely used plastic strips.

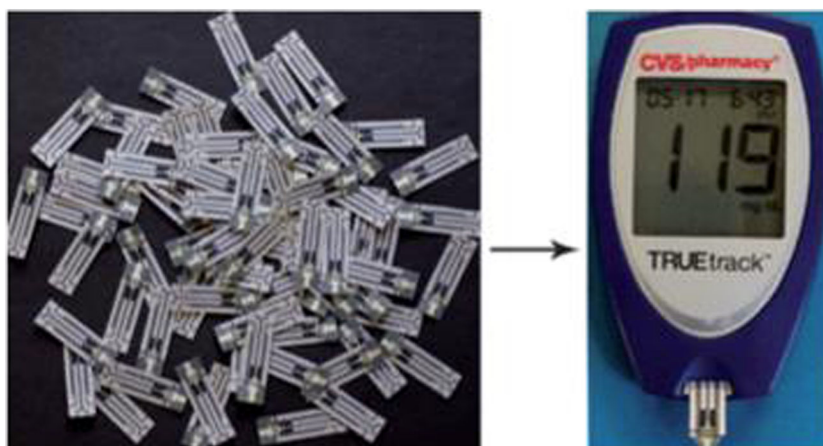
Copy paper for ethanol biosensing

The presence of ethanol in alcoholic beverages is very important because it influences the flavor. Moreover, the taxation of such alcoholic beverages strictly depends on the ethanol content [24]. The quantification of ethanol is important during the processing of an alcoholic beverage, from fermentation and monitoring the manufacture to quality control testing conducted to evaluate the expected amount. Although methods to monitor the ethanol content in alcoholic beverages are not lacking (e.g., physical methods based on refractometry, chemical methods based on gas chromatography, etc.), simple methods with low economic impact are required [25]. As described earlier, electrochemical methods have been widely reported in the literature. The absence of color interferences is the leading driver for the implementation of such approaches. Numerous sensors and biosensors have been developed that take advantage of nanomaterials, conductive polymers, and enzymes (mainly alcohol oxidase) in the search for increased sensitivity and specificity [26–28]. The use of paper-based platforms might help accelerate the entire process by reducing the tasks and removing the issues related to waste management. The use of paper helps lower the price of the final device, especially when compared with the most

traditionally used polyester for realization of the strips. The Whitesides group demonstrated a tremendous reduction of cost by switching from polyester to chromatography paper (Fig. 2) for strip manufacturing [29].

An electrochemical microfluidic paper-based analytical device (E μ PAD) was integrated into the port of a commercial glucometer (a miniaturized potentiostat). The use of Whatman no.1 filter paper allowed the inclusion on the same piece of paper of the microfluidic channels, the electrodes, the electrical interconnections, the wax insulation, and the reactants. Typically, commercially available glucose test strips are made with plastic and their price can reach \$1 per strip in the USA and €1 per strip in Europe (e.g., OneTouch Ultra, LifeScan). Currently, the Whitesides group claim a cost of \$0.014 to manufacture a paper-based strip for a glucose test. The compatibility between plastic- and paper-based strips was evaluated by measurement of glucose in human plasma, and a satisfactory correlation coefficient of 0.995 was obtained, which was further confirmed by analysis of glucose in whole blood (95 ± 9 mg/dL vs. 99 ± 3 mg/dL measured with E μ PADs and commercial strips, respectively) [29]. Moreover, the same technology has been applied to the detection of cholesterol and lactate in body fluids, and ethanol in water. Of course, specific enzymes need to be used to detect diverse species. In particular, to detect ethanol in water samples, the E μ PAD was preloaded with a mixture containing an enzyme and a cofactor (alcohol dehydrogenase and β -NAD⁺) and the electrochemical mediator ferricyanide. This architecture, including the use of an external potentiostat (glucometer), was capable of linearly detecting ethanol in the 0.1–3 mM range with a coefficient of variation ranging from 3.2% to 10.1%. Although these developments are still at an early stage, there is rough equivalency between the laboratory prototype and the commercial

Fig. 2 Integration of chromatography paper into commercially available devices for blood glucose monitoring. (Reprinted with permission from [29])



strip (in biomedical sensing). The analysis of alcohol is favorable with some cost-saving improvements such as lowering of the amount of chemicals (cofactor β -NAD⁺ is required for alcohol dehydrogenase activity), thus reducing the cost of the prototype. As reported already, the use of chromatography paper would allow test strips to be produced at lower cost with respect to the cost of plastic-based tests (impractically high for applications in the developing world). However, the use of copy paper would lead to an ultimate 30% saving [30], based on a laboratory scale and the cost of conductive inks (i.e. graphite, silver) being equal. The savings associated with the use of paper-based substrates are shown in Table 1.

The quantification of ethanol in commercial beers is an example of a technology that would hugely benefit from use of all-in-one strips made with copy paper and a customized biohybrid nanocomposite [21], and by the merging of easy procedures such as wax printing, screen printing, and drop casting. As reported in Fig. 3, a wax pattern was designed (15 mm \times 13 mm rectangle, with a 10-mm-diameter semicircle) with drawing software (Adobe Illustrator) to define the testing area, and the pattern was printed with a solid-ink printer (ColorQube 8580, Xerox, USA) onto rectangular 35 mm \times 20 mm copy paper (Copy 2, 80 g/m², Fabriano, Italy). Then the wax-printed copy paper was thermally cured at 100 °C for 4 min to allow the wax to diffuse through the cellulose and

form a hydrophilic testing area surrounded by a hydrophobic barrier. Subsequently, the three-electrode system was manually screen-printed with a squeegee and two stencils. Ag/AgCl ink was used to realize the connections and the pseudo-reference electrode, whereas graphite ink was used to produce both the working electrode and the counter electrode.

Another example of the use of paper for a multitask platform is the modification of paper with carbon black (CB), Prussian blue nanoparticles (PBNPs), and alcohol oxidase [21]. In this work, 2 μ L CB/PBNP nanocomposite was drop-cast onto the surface of the working electrode. Following the evaporation of solvent, the enzymatic mixture was immobilized. Briefly, 2 μ L of a mixture of alcohol oxidase, bovine serum albumin, Nafion[®], and glutaraldehyde was drop-cast onto the CB/PBNP-modified working electrode and allowed to dry for 1 h. Besides its economic convenience, copy paper is a suitable substrate. This was demonstrated by its resistance to mechanical stress: after 50 bending cycles, the paper-based strip did not exhibit a decrease of the peak intensities (anodic and cathodic) during cyclic voltammetry in the presence of the ferrocyanide/ferricyanide redox couple (Fig. 3b). The electrochemical response appeared unchanged. Thus, common 80 g/cm² copy paper allows the reliable manufacture of robust strips, not only for food applications but also for the detection of species in both clinical and environmental

Table 1 Costs of the components for producing one device (all costs are in euros)

Substrate	Ag/AgCl ink	Carbon ink	Insulator	Substrate	Total cost	Saving (%) ^c
Polyester	0.010	0.007	0.003 ^a	0.013	0.033	45
Whatman no. 1 filter paper			0.001 ^b	0.007	0.025	28
Office paper			0.001 ^b	0.0001	0.018	–

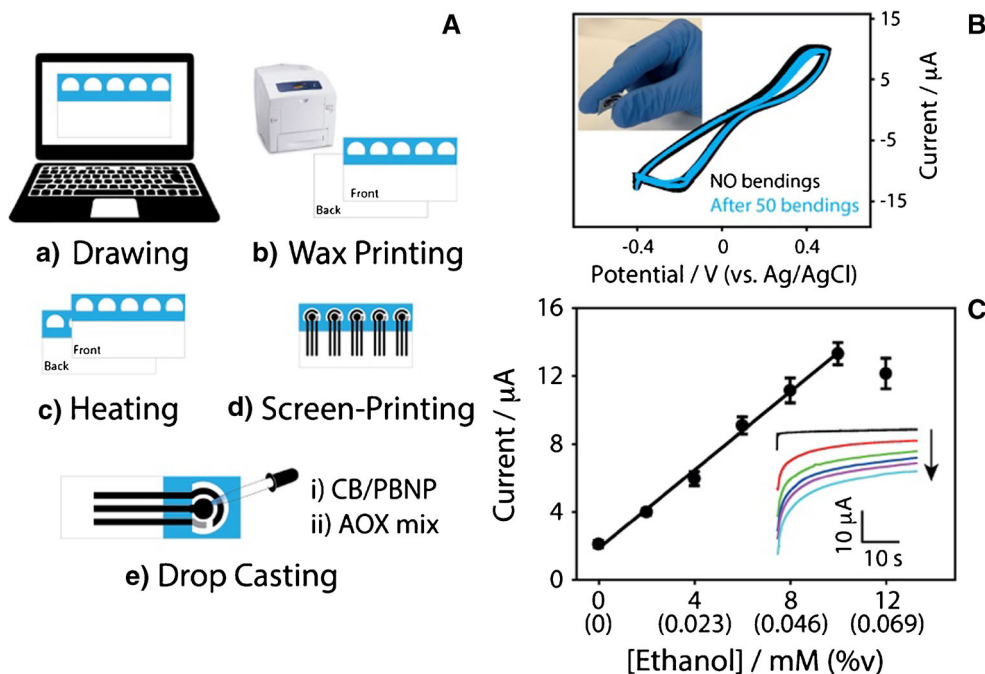
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^a Insulator ink

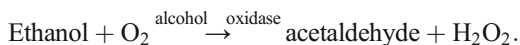
^b Wax

^c Calculated as $1 - (\text{cost for office paper}/\text{cost for other substrate}) \times 100$

Fig. 3 Use of copy paper to make printed strips for ethanol detection in beers. **a** Experimental procedure for obtaining paper-based strips on copy paper. **b** Evaluation of the mechanical robustness of the paper-based platform after 50 bending cycles by cyclic voltammetry in the presence of ferrocyanide/ferricyanide. **c** Calibration plot for ethanol in the 0–12 mM range obtained by chronoamperometry detection with an applied potential of 0.1 V (vs. Ag/AgCl). AOX alcohol oxidase, CB carbon black, PBNP Prussian blue nanoparticle. (Reprinted with permission from [21])



samples (e.g., sweat, serum, river water) [15, 30–32]. The detection of ethanol is based on the following reaction:



Ethanol is oxidized by the enzyme alcohol oxidase in the presence of molecular oxygen (O_2), and the by-products are acetaldehyde and hydrogen peroxide. Even though the latter is an electroactive compound, it requires a high anodic potential to be detected at bare electrodes, and there is the possibility of interference by other oxidizable species in real matrices. Instead, one can use PB (an electrocatalyst), which is capable of reducing the hydrogen peroxide formed at low interfering potentials. In particular, the reduced form of PB is capable of electrocatalytically reducing hydrogen peroxide ($\text{PB}_{\text{red}} + \text{H}_2\text{O}_2 \rightarrow \text{PB}_{\text{ox}} + 2\text{OH}^-$) [33], whereas the use of CB increases the conductivity at the electrode surface, ensuring high sensitivity [34]. As shown in Fig. 3c, an increase of the ethanol level is reflected in a higher cathodic (reducing) current at the electrode surface. The presence of this electrochemical mediator (already present on the working electrode) facilitated the linear detection of hydrogen peroxide (by application of a potential of -0.1 V vs. Ag/AgCl) up to 10 mM with a detection limit of 0.4 mM. Without it, at the same working potential, hydrogen peroxide was not detected at the bare electrode. However, for the subsequent ethanol detection, with the presence of all modifiers on the working electrode (electrochemical mediator and enzymatic mixture), the tasks for the consumer are reduced to zero. The reduced size of the copy-paper-based strips allowed determination of very low amounts

of samples, with just 100 μL being enough to obtain satisfactory quantification of the chosen analyte. Ethanol was determined in standard solution up to 10 mM with a sensitivity of 9.13 $\text{mA}/\text{mM cm}^2$ and a detection limit of 0.52 mM (calculated as $3\sigma b/\text{sensitivity}$). Moreover, even though the entire platform was manually produced, the repeatability was evaluated to be approximately 7%. The final paper-based strip was applied for the detection of ethanol in commercial beers: Best Bräu lager, Franziskaner wheat bear, Ceres pilsner, and alcohol-free Tourtel. The determinations were performed without chemical operations such as pH adjustment and/or removal of the interfering species being performed. Even though the method was not validated, the ethanol quantification obtained with these small paper-based strips was in high agreement with the content on the labels of such commercial beers, as reported in Table 2.

However, this sustainable and low-cost technology is capable of providing sensitive, portable, and reliable detection tools for ethanol not only in the traditional food and clinical industries but also for monitoring ethanol content step-by-step in the production chain (fermentation, processing, and quality control).

Printed electronics for ascorbic acid quantification

The field of paper-based devices can be further enhanced beyond the realization of sustainable and reagentless platforms (with all the reagents included) to serve as the substrate, where

Table 2 Agreement between the label value of commercial beers and amount of ethanol detected with the paper-based strips

Beer type	Ethanol (%)		Agreement (%)
	Label	Found	
Lager, Best Bräu, Poland	4.7	4.7 ± 0.4	100 ± 9
Wheat beer, Franziskaner, Germany	5	5 ± 0.4	100 ± 8
Pilsner, Ceres, Denmark	4.6	4.4 ± 0.2	96 ± 5
Alcohol-free, Tourtel, Italy	<0.5	0.34 ± 0.03	— ^a

^a With the paper-based method, (0.34 ± 0.03)% ethanol was found; however, an agreement cannot be provided because the label indicates “<0.5%”

also a potentiostat has been built. Further improvements of user-friendly tools for rapid monitoring should focus on fully integrated devices, in which each component is printed onto one platform. With the rise of printed electronics and with affordable fabrication methods that are associated with them, the opportunity to manufacture all-printed paper-based devices is ever closer to reality. Although some fabrication methods are still expensive and complex (photolithography), easier and cheaper methods exist (e.g., inkjet printing). Inkjet printing is based on a drop-on-demand technology that depends on the fluid jetting mechanism (piezoelectric or thermal). The water-based ink contained in the cartridge can be jetted for a short time, approximately 10 μs: this allows the deposition of picoliter volumes of low-viscosity inks with high accuracy and repeatability [35]. The concept for realizing fully integrated tools focuses on the use of both a common substrate (paper) and a common technology (inkjet/screen printing) to obtain devices that are equipped with all the parts the end user needs to quantify compounds (i.e., sensor, display, battery, etc.). A similar approach on plastic was recently reported by the group of Killard [36]. They tried to combine all the components necessary for building a point-of-care device for the measurement of total cholesterol in serum samples, as shown in Fig. 4.

They tried to merge within the same monolithic device all the organic substrates, all of the printed electronics, and the electrodes for quantification. In detail, the electrochromic display was printed onto seven stacked layers; the battery, based on zinc/manganese dioxide, was printed onto eight stacked layers as the connections of three single cells (4.5 V). The electrochemical sensor was screen-printed, including four-configured electrodes (i.e. dual working, counter, and reference electrodes). The entire device was integrated into a 22-layer stack, and approximately 8 μL of sample was analyzed in the microfluidic sample chamber. Cholesterol was measured in the presence of the inkjet-printed cholesterol esterase, cholesterol oxidase, and stabilizers. The combination of screen printing, inkjet printing, and lamination processes allowed the fabrication of the concept device. This novel integrated device could represent the basis of a new method for the fabrication of analytical tools, not only in the clinical field.

This approach also leads to an obvious reduction of e-waste production, which is the result of the accumulation of used electronic appliances, which are abundant because of their short lifespan. Starting from these achievements on plastic, the development of a single entity by exploiting the advantages of paper-based substrates could represent a further step forward. The positive qualities of paper such as its low cost, biodegradability, and easy incineration after use might promote the development of sustainable devices. The interaction of all these components is tied with the technological evolution of paper processing. There are a range of materials that might be amenable for integration into a fabrication process. The appropriate functionality and processability of the selected components to be integrated in the monolithic device could be significant challenges. Inkjet printing could be used as the technology for realizing these innovative platforms, from which all components are fabricated. The multilayer architecture proposed for cholesterol detection in biofluids described in Fig. 4 represents a prelude for future full integration. The new paradigm might be represented by switching from a multilayer device to a single-layer device, avoiding the need for different manufacturing methods and thus reducing the time for fabrication and also the amount of materials (organic substrates, adhesives, etc.). Even though inkjet printing is widely used to realize circuits, its adoption in chemical sensing is not widespread.

An example of the successful use of inkjet printing in food analysis was reported by Felix Schoeller Group, Germany. Electrodes were inkjet-printed onto a paper-based substrate (p_e:smart type 2), usually used for printed electronics, and were applied for reagent-free determination of ascorbic acid in food supplements [22]. Printable conductive inks can be easily formulated and customized depending on the analytical focus, and the possibility to modify the platform with nanomaterials for enhancement of the sensitivity and specificity is even more appealing. A Dimatix inkjet printer (Fujifilm Dimatix, USA) equipped with a piezoelectric printhead (16 nozzles) was used to produce the bare electrodes. The accuracy of the process was assured by the tiny space between each droplet; a drop spacing of 20 μm was used to obtain the electrical connections by the jetting of nominal 10 pL of liquid. Differently from

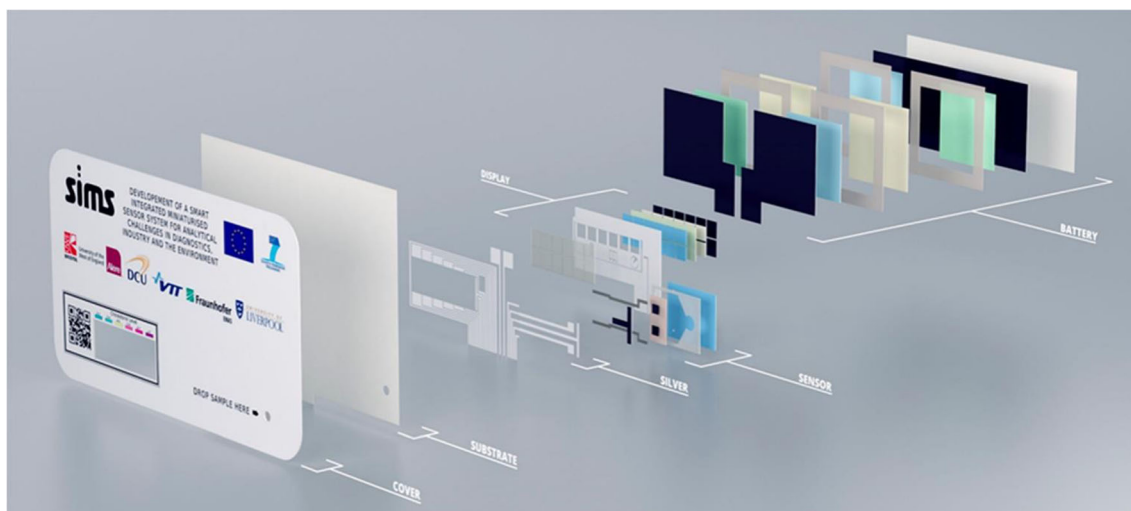


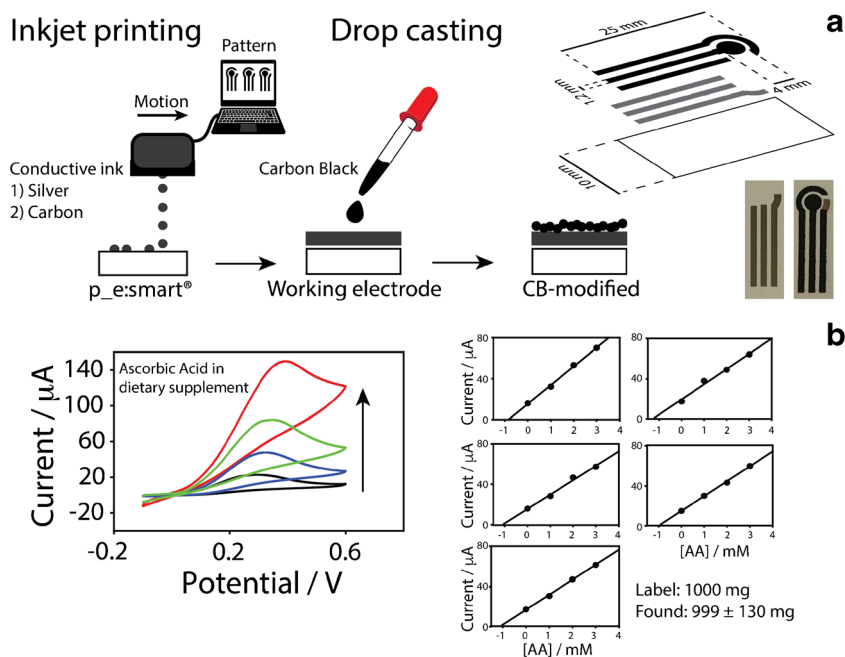
Fig. 4 Serum cholesterol detection with a single-use device with a printed electrochemical sensor for hydrogen peroxide electroreduction integrated with a printed electrochromic display and battery. (Reprinted with permission from [36])

serigraphy, the conductive inks used with an inkjet printer are water based. In the example described here, first two layers of silver nanoparticle ink were printed, and after drying and sintering at 140 °C, two layers of a nanocarbon ink were printed. The electrochemical properties of the platform were enhanced with the use of CB nanoparticles (Fig. 5a).

The amorphous nanomaterial CB, as widely reported in the literature [37–39], is capable of increasing the surface area and even enhancing electrocatalysis at the electrode surface. This was reflected by an increase of the voltammetric peak intensity and increased electron transfer in the presence of ferricyanide as the redox probe. The use of CB as a nanomodifier also led to a decrease of peak-to-peak separation, ΔE , from

0.65 to 0.30 V for a bare electrode and a CB-modified electrode, respectively. Although the conductivity of the electrochemical platform was positively influenced by the presence of CB, the resistance to charge transfer calculated with use of electrochemical impedance spectroscopy decreased from approximately 3800 Ω to 270 Ω . Moreover, the presence of CB allowed the electrochemical inkjet-printed platform to decrease the overpotential regarding the oxidation of ascorbic acid, from 0.47 to 0.28 V for bare and CB-electrodes, respectively; the use of a few microliters of CB also increased the sensitivity (more than three times) with respect to the unmodified platform. Because of the finely controlled manufacturing technology, a relative standard deviation of 8% was calculated

Fig. 5 Use of an entirely inkjet-printed sensor on printed electronics paper, modified with carbon black (CB) for the detection of ascorbic acid (AA) in food supplements. **a** Fabrication of paper-based strips. **b** Quantification of ascorbic acid in food supplements with the standard addition method. (Reprinted with permission from [22])



($n = 5$). However, to determine ascorbic acid in real foodstuff, some considerations should be kept in mind. For example, the presence of interfering species (i.e., excipients) might cause experimental problems regarding the quantification. For instance, many colorimetric methods and reference methods are based on a titration with dichlorophenolindophenol [40], which might be affected by the presence of bubbling (presence of bicarbonate) and/or the color of the matrices. These issues did not affect the inkjet-printed platform; in fact, most excipients in dietary supplements (i.e., sodium bicarbonate, sucrose, citric acid, sorbitol) did not show appreciable interference during measurement of ascorbic acid. The label of the analyzed dietary supplement (Supporvit, Èqui, Italy) reported an amount of ascorbic acid equal to 1000 mg. After the dissolution of the tablet in water, ascorbic acid was determined by use of the standard addition method as shown in Fig. 5b. Five measurements were performed with five different devices, and three measurements were performed following addition of 1 mM ascorbic acid to the sample, resulting in a mean value of 999 ± 130 mg with respect to the nominal 1000 mg declared in a single tablet. The accordance between the value found with the paper-based inkjet-printed strip and the label value was calculated to be $(100 \pm 13)\%$.

Outlook

A huge amount of effort is required to transform miniaturized analytical devices from a laboratory concept to a widely used analytical tool that may be used at the site of care. The benchmark solution provided by the strips for diabetes patients still represents a unique example. However, the increasing need to access information on-site at low cost will continue to drive the development of novel and disposable tools. The importance of having such user-friendly devices for food surveillance cannot exclude the electroanalysis. Colorimetric solutions are unable to analyze colored and turbid matrices, such as wine. This is where electrochemical sensors have major benefits over colorimetric methods. Also, colorimetric methods often require reagent and sample handling without quantitative results. Electrochemical paper-based tests could provide a platform for quantitative measurements where sample preparation (such as filtering) and sample treatment (reagents dried onto the paper) can be performed on a single paper platform. Together these advantages over other analytical methods and the well-known benefits of paper, such as inexpensiveness, abundance, and sustainability, will hopefully culminate in the development of a truly integrated analytical device. Also, paper-based strips are amenable to combination with smart functional nanomaterials and biomaterials in combination with conveniently configured manufacturing processes. The two examples discussed in this article highlight the application of different substrates and fabrication routes toward the development of user-friendly and reagent-free analytical tools to

be used in food analysis. The paper-based field was viewed from two different angles. The first looked at the use of copy paper to realize a reagent-free ethanol biosensor for beer, highlighting how even common materials (so-called everyday materials) can be easily and successfully processed to provide rapid monitoring. The second looked at the combination of printed electronics with inkjet printing to produce a paper-based sensor to detect ascorbic acid in dietary supplements. This example represents a convincing starting point toward the realization of fully disposable paper-based tools.

The search for new and more sustainable solutions in the analytical field has resulted in major research and commercial efforts. The choice of paper as an alternative to plastic-based substrates has obvious social, economic, and environmental benefits: (1) socially, reagent-free tools allow measurements to be performed by unskilled personnel without requiring a laboratory setup, delivering useful devices especially in resource-limited areas; (2) economically, switching from plastic to paper allows a high cost saving, i.e. utilizing office paper allows a 45% cost saving; and (3) environmentally, nontoxic materials are used for fabrication, a low sample volume is needed (microliters), and no waste is produced. The implementation of user-friendly and disposable paper-based electro-analytical tools in the agri-food field might help to offer an innovative and transformative solution for surveillance and early detection of food quality both during industrial processes and at the consumer stage. Moreover, the use of paper offers potential for dramatic cost reductions compared with current strategies based on plastic-based supports. Even though cost will continue to be a primary driver, the greener label of paper is growing in importance. Merging smart nanomaterials, biological elements, processing methods, and functional integration will facilitate widespread use of this technology.

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Compliance with ethical standards

Conflict of interest The author declares that he has no competing interests.

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