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**A novel disposable self-adhesive inked paper device for
electrochemical sensing of dopamine and serotonin neurotransmitters
and biosensing of glucose**

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Abstract

In this work, we detail the progress of a novel electrochemical disposable device, which has a relative low cost and easy production, with a novel conductive ink, that consists of graphite and automotive varnish mixture, deposited over a self-adhesive paper, granting an easy production with relative low cost. The electrode surface was characterized by scanning electron microscopy, X-ray powder diffraction and Fourier transforms infrared and Raman, cyclic voltammetry and electrochemical impedance spectroscopies. In addition, the proposed electrode was applied for individual electrochemical determination of dopamine and serotonin. The device achieved a linear response between 30 to 800 $\mu\text{mol L}^{-1}$ and a limit of detection (LOD) of 0.13 $\mu\text{mol L}^{-1}$, by square wave voltammetry for dopamine and a linear range from 6.0 to 100 $\mu\text{mol L}^{-1}$, with a LOD of 0.39 $\mu\text{mol L}^{-1}$, by differential pulse voltammetry. Later, the working electrode was modified with glucose oxidase and dihexadecyl phosphate film in order to obtain a biosensor. At this stage, CV was applied to detect glucose in the range of 1.0 to 10 $\mu\text{mol L}^{-1}$ and LOD of 0.21 $\mu\text{mol L}^{-1}$. By three different techniques and analytes, the sensing and biosensing processes presented high reproducibility. The proposed adhesive electrode is easy to prepare, disposable, within non-restrictive nature, which allows an approach of a new device for electrochemical sensing and biosensing.

Keywords: disposable electrode, point-of-care, self-adhesive paper, neurotransmitter, glucose.

1. Introduction

The recent interest in ridding patients of the dependence on a sophisticated laboratory and more advanced and expensive technologies for a diagnosis has been moving global science, which, allied to innovative advances, such as the use of miniaturized devices, wireless communication and nanomaterials, prove the point-of-care (POC) potentiality. (Goldstein, 2003; Schultz, 2001; Syedmoradi et al., 2017; Zarei, 2017). In this context, they present attractive qualities, such as practicality, ease of use, relative low cost and short time response, while also require minimal equipment operation and specific knowledge. As one of the major uses of such devices is the monitoring of diseases that requires routine tests, such as diabetes, POC devices can be performed with focus on the patient, increasing the healthcare system efficiency (Goldstein, 2003; Schultz, 2001; Syedmoradi et al., 2017; Zarei, 2017). Another advantage of POC devices is the acceptance of common and disposable materials, favoring a more sustainable approach (Janegitz et al., 2014; Johari-Ahar et al., 2018; Lamas-Ardisana et al., 2018; Xu et al., 2018). Some recent works have been proposed by using disposable inert substrates for electrochemical devices (Dunn, 2003; Mittal, 2003; Orzari et al., 2018; Tortorich et al., 2018), with a relative lower cost than conventional electrodes with similar performance. Nowadays, adhesives are commonly found in our environment, being frequently attached to cellulose substrates, as stamps, stickers, many everyday tapes and even medical plasters (Negash et al., 2015; Purushothama et al., 2018; Sharp and Burkitt, 2015). In this regard, adhesive paper is an interesting alternative for different applications (Dunn, 2003; Mittal, 2003). Paper is a substrate for novel electrochemical systems that has been gaining attention in the last years (Ge et al., 2012; Lee et al., 2018; Tortorich et al., 2018; Wang et al., 2017), which presents biocompatibility and biodegradability. Therefore, adhesive papers can increase

the field application, as no set place is needed for the measurement, also bringing all advantages of regular paper.

Being highly known for its conductive properties, graphite is a superposition of sp^2 carbon sheets, which produces interesting anisotropic effects. In graphite, the carbon interactions may be limited between planes, conferring some resistivity, being an excellent thermal and electrical insulator (Fukushima et al., 2006; Pierson, 2012; Tian et al., 2013). Also, it is easily sheared, as the interlaminar strength is considerable low. However, due to its σ bonds, graphite has admirable electrical conductivity at laminar planes, being even higher than some metals (Fukushima et al., 2006; Pierson, 2012; Tian et al., 2013). Also, its whole structure can present vacancies, dislocations and other structural imperfections, which could lead to a lower density, allowing it to be considered one of the lighter refractories (Khomenko et al., 2008; Li et al., 2005; Pierson, 2012). These properties, along with its chemical inertia (Pierson, 2012), favor the development of electrodes (Negash et al., 2015; Purushothama et al., 2018; Sharp and Burkitt, 2015) and conductive polymeric inks (Grisales et al., 2016; Phillips et al., 2017), in a raw proportion of up to 50% (1:1 graphite:polymer) of final composition, without any electrical conduction loss (Pekarovicova and Fleming, 2005). The literature has been presenting a range of works in which carbon-based inks are used in the development of different electrochemical systems (Jaiswal and Tiwari, 2017; Sharp and Burkitt, 2015).

As proof of concept, we applied the device towards the individual detection of dopamine and serotonin, two of the most electrochemically well acknowledged and important catecholamines in mammals, mainly due to their connection with several behavioral dependence and Parkinson disease (Baccarin et al., 2017; Berridge and Kringelbach, 2013; Fox et al., 2009; Goldstein, 2003; Iversen et al., 1977; Mayeux et

al., 1984; Schultz, 2001; Sharma et al., 2018; Vyazovskiy et al., 2007). After, we modified its surface with glucose oxidase (GOx), demonstrating the sensitive detection of glucose, an essential monosaccharide, that has substantial impact in the brain behavior, as well as its discrepant rates are the cause of diabetes (Arslan et al., 2011; Blair, 2016; Cryer, 2016; Dwier, 2002; Janegitz et al., 2011; Pisoschi, 2012; Wang and Lee, 2015).

2. Experimental

2.1. Chemicals and reagents

All reagents used in the present work were purchased from Sigma-Aldrich (Germany) and/or Fluka (Brazil), in analytical grade. Ultrapure water (Megapurity, USA), with resistivity $> 18 \text{ M}\Omega \text{ cm}$, was used for the preparation of all electrolytic solutions. Dopamine and serotonin were purchased from Sigma-Aldrich (Germany). The graphite powder (Gr) used was obtained from Fischer Chemical (USA), and the ink polymeric vehicle was the automotive varnish from Sherwin-Williams (Brazil), Lazzudur 2K Auto Brilho (AV). For electrochemical characterization of the device, as well as dopamine detection, 0.1 mol L^{-1} phosphate buffer (Sigma-Aldrich, Germany) (PB) solution (pH 6.1) was used as supporting electrolyte. Glucose biosensing was performed in 0.05 mol L^{-1} PB solution with pH 7.0 for optimal glucose oxidase (Sigma-Aldrich, Germany) (78 units per electrode) activity.

2.2. Apparatus

A Tescan VEGA-3 LMU microscope was used to perform the scanning electron microscopy (SEM), with an accelerating voltage of 10 kV, in low vacuum mode with 50

Pa. Fourier transform infrared spectroscopy (FTIR) was carried out with a Tensor II spectrophotometer (Bruker), in 4.0 cm^{-1} resolution and transmittance mode from 400 to 4000 cm^{-1} . Raman spectra were obtained using a B&W Tek *i*-Raman BWS 415-785H spectrometer, with an excitation laser of 785 nm. A B&W Tek microscope BAC151A, coupled to the spectrometer was used, with 3.5 cm^{-1} spectral resolution, in the spectral range of 0.5 to 2500 cm^{-1} , acquisition time of 120 s, and laser power of 70 mW. The x-ray diffraction (XRD) was performed by a Miniflex II x-ray diffractometer (Rigaku), with a $\text{CuK}\alpha$ radiation source ($\lambda = 0.15406\text{ nm}$), from 3° to 90° angular recording range, with a step of 0.02° and 10° min^{-1} analysis time. All electrochemical measurements were performed with an Autolab PGSTAT204 (Eco Chemie) potentiostat/galvanostat (with FRA32M module) managed by NOVA 2.1.3 software. An 827 pH-meter (Metrohm) was used for pH measurements. The conductive ink was mixed and milled with a dual asymmetric centrifuge (DAC) SpeedMixer™ Dac 150.1 FVZ-K (FlackTec Inc) and the electrodes were cut with a cutting printer (Silhouette, Cameo 3).

2.3. Preparation of the conductive ink

In the ink technology field, it is common to prepare inks by suspending pigments in a resin/polymer vehicle. For the electrically conductive materials, specific components are added to the formulation, granting the desired aspects. Here, as well as the colorant, Gr was used as the conductive material. Therefore, 7.2 g of Gr was added to an aliquot of 18 g of AV, corresponding to 30% of desired final ink mass. Afterwards, the suspension was mixed for 3 cycles of 3500 rpm for 90 seconds in SpeedMixer. It is important to highlight that this mixer has dual asymmetric centrifuge that milling, mixing and consequent heating of particles. Therefore, there was no need a polymeric curing agent, and the ink could easily solidify at room temperature.

2.4. Preparation of the disposable electrode, modification and electrochemical measurements

Figure 1 illustrates the disposable device (Gr-AV) preparation. Firstly, the graphite ink is applied on an adhesive paper sheet, respecting the edges of the cutting printer (Silhouette, Cameo 3), which is used to cut the format of the devices. Then, it was waited 40 minutes for the ink to dry. Succeeding, the excess is removed and properly discarded, resulting in a sheet full of disposable devices with working ($\varnothing = 4.5$ mm diameter), counter and reference electrodes, ready to use. A video of this process is also available in the supplementary data (Video 1). For the biosensor (Figure 1B), 10 μL of GOx and dihexadecyl phosphate (DHP, 1.0 g L⁻¹) solution was previously prepared and casted on the working electrode surface and let dry overnight at 10 °C.

FIGURE 1

To execute the electrochemical measurements, the device was adhered to a polyethylene terephthalate substrate and later to a connector previously built by the group (Orzari et al., 2018), as observed in Figure S1A (supplementary material). 70 μL of supporting electrolyte and analyte solution was cast on the disposable system and 10 s of waiting time was standardized in dopamine analysis, due to the absorptive nature of paper. The waiting time for glucose biosensing was 60 s, aiming the enzyme reaction and to standardize the water absorption problem.

2.5. Detection and samples preparation

As previously mentioned, both analytes have interesting impacts in modern human society: dopamine has a well-known relation to human desire, behavior and many

neurological disorders; and glucose is a biomarker for diabetes in its metabolism process. In this regard, all detections were performed by addition and recovery method, in synthetics urine, saliva and saline samples. The synthetic samples were prepared as presented in the literature (Baccarin et al., 2017; Campos et al., 2018; Raymundo-Pereira et al., 2016). The saline solution is a notable biological media, necessary for many optimal biological processes (Kent et al., 1985; Ozawa et al., 2006; Steinbusch et al., 1978) and was also chosen to be a detection media. In addition, all samples were prepared in PB solution, with optimal pH and ionic strength (μ) (saline samples: 6.1 (Gr-AV) and 7.0 (GOx-DHP/Gr-AV); synthetic urine: 6.1; synthetic saliva: 7.0; all with $\mu = 0.2 \text{ mol L}^{-1}$) and Table S1 shows all reactants and its concentrations used for each sample, as found in literature (Baccarin et al., 2017; Campos et al., 2018; Raymundo-Pereira et al., 2016).

3. Results and discussion

3.1. Morphological and electrochemical characterization of the disposable device

Nowadays, there is a great focus on the electroanalytical field to develop novel electrodes structures. This interest is mainly motivated by the possibility of further miniaturization of the system, allowing more practicality and the use of relative cheaper materials. The AV was selected as ink vehicle due to its polymeric composition, great accessibility and relative low cost. Even though its precise composition is not given by the manufacturers, polyurethane plays a major role in this varnish, which is an interesting compound for application in sensors development. Also, as described, here we present the use of self-adhesive paper as a support for electrodes, and, as a new electrochemical tool, its properties must be known for a better application. Therefore, the self-adhesive device surface was characterized by microscopy and spectroscopy

techniques, as shown in Figures 2-4. Figure 2 shows the SEM images obtained for the self-adhesive paper sheet (Figure 2A1-3), the self-adhesive device (Figure 2B1-3) and the same device with GOx-DHP addition (Figure 2C1-3). It can be observed the difference between these stages, with the adhesive paper presenting an expected homogeneous fibrous surface being highly rough and porous. This surface is completely changed by the ink, as only graphite associated morphology can be seen in the correspondent image. Then, the casting of GOx-DHP on the working electrode also alters the surface, making it smoother with distributed small enzyme clusters.

FIGURE 2

Figure 3A shows the FTIR spectra obtained for the AV (blue line), Gr-AV conductive ink (red line) and Gr (black line). The varnish spectrum presents expected bands, such as the carbonyl stretching at 1730 cm^{-1} , the asymmetric stretching of the ester at 1145 cm^{-1} , the broad band of O–H stretching and N–H vibration at 3450 cm^{-1} , as well as many bands associated with aromaticity, related to a major presence of polyurethane in the varnish composition (Smith, 1982). The bands at 1630 and 1380 cm^{-1} , respectively associated to the C=C bonds of the sp^2 hybridization and skeletal C–C carbon rings vibrations are found in the graphite spectrum. It is interesting to note that the ink spectrum portrays the same profile as the graphite one. This indicates that not only the paper surface is completely covered, but also that the polymeric part, as well as other organic components, does not interfere in the bulk electrode surface.

Figure 3B shows the Raman spectra for AV (blue), Gr-AV conductive ink (red) and Gr only (black). Even though the AV spectrum presents a similar profile when compared to polyurethane spectra in literature (Naumenko et al., 2012; Parnell et al., 2003; Xu et al., 2000), significant shifts can be found. These results are attributed to the

several other components in the varnish, which can promote such a behavior. Due to limitations of the available equipment, not so normally useful spectra were obtained for Gr and Gr-AV. However, these data also suggest that the dried ink surface is mainly composed by Gr that promotes high conductive characteristics.

XRD measurements were carried out for AV, Gr-AV and Gr, as shown in Figure 3C, in order to analyze the crystallinity of these materials, since conductive properties are directly correlated. As observed in the literature (Armelin et al., 2017; Blackwell et al., 1981; Pradhan and Nayak, 2012), polymeric materials have presented amorphous structures with few crystalline phases, because the non-uniformity of interactions. Therefore, the obtained AV profile corroborated with this, showing a diffused broad diffraction, with a maximum at $2\theta = 17.7^\circ$, as shown in the figure inset. The Gr profile demonstrated a characteristic (Johra et al., 2014; Popova, 2017) (002) plane peak at $2\theta = 26.3$, with crystal size of 26.96 nm and layer spacing of 1.98 Å. The ink profile showed similarities with the Gr one, but with considerable changes. The plane peak slightly shifts to $2\theta = 26.2$, which presented a higher crystal size, 31.54 nm, and closer layers, with a 1.74 Å spacing. This is attributed to the agglomeration effect and interaction between Gr and AV, by reason of ink formation process.

FIGURE 3

Generally, electroanalytical sensors are applied in aqueous media. Consequently, it became necessary, and of great importance, to define the useful working potential window, where water does not suffer electro-induced reactions (Orzari et al., 2018). Figure 4A shows the cyclic voltammograms obtained by the adhesive sensor in different media: HCl, NaOH and KCl; maintaining the μ (all in 0.1 mol L⁻¹), at 50 mV s⁻¹. A variation in the electrochemical behavior is expected, because the different activity of

hydronium and hydroxyl ions (Calegari et al., 2017; Kang et al., 2017). For example, Kang et al. (Kang et al., 2017) have studied the effect of water electrolyzation at different pHs by cyclic voltammetry with a copper based electrode, in which the production of H_2 is electrocatalyzed by lowering the pH. In the acidic media, the hydrogen formation can be observed in the proposed electrode, as expected, towards more positive potentials, with the useful region being between -0.45 and 1.3 V. In alkaline media, this potential region was limited between -0.15 and 0.75 V, since the oxygen production is early favored. In neutral media (KCl), by reason of equivalent presence of water ions, a broader range is expected. However, the non-interfering region corresponded to -0.10 to 1.3 V. Thus, it was possible to observe when lower the pH, the proposed device presented less interference.

To study the electrochemical behavior, Gr-AV electrode was submitted to cyclic voltammetry, at 50 mV s^{-1} , in the presence of $500\text{ }\mu\text{mol L}^{-1}$ dopamine. The Figure 4B shows a notorious difference between the magnitude of the anodic (334 mV) and cathodic (44.8 mV) peaks, revealing a greater sensitivity for the oxidation reaction. To estimate the electroactive area, cyclic voltammetry studies were carried out in this configuration, at varying scan rates, ranging from 10 to 200 mV s^{-1} , as shown in Figure S2. Data were applied in Randles-Sevcik equation, resulting in an electroactive area of 0.22 cm^2 . Figure 4C shows the Nyquist diagrams of EIS experiments, performed in presence of $500\text{ }\mu\text{mol L}^{-1}$ dopamine, in 0.1 mol L^{-1} PB solution (pH 6.1), for Gr-AV, as well as the obtained equivalent circuit (Cesiulis et al., 2016), with a χ^2 of 2.84×10^{-4} . The relevant data can be found in the circuit. This experiment implies that the obtained plot contains superposition of different phases, Gr-AV, Gr-Gr and Gr-dopamine. The proposed electrode based on new conductive ink presented greater resistivity when

compared to other published works (Amin et al., 2014; Cinti et al., 2017). Nonetheless, it is valid to observe its adhesion-based nature (Figure S1B-D). All these characterization data, together, suggests a novel electrochemical device category.

FIGURE 4

3.2. Electrochemical determination of dopamine with Gr-AV disposable electrode

The square wave voltammetry technique was chosen for dopamine detection, because the reversibility of the reaction and its known sensitivity. So, its parameters were studied and optimized, where the selected values were: 8.0 mV for step (s), 100 mV for amplitude (a) and 10 Hz for frequency (f). In addition, tests to evaluate the Gr-AV response in different pHs were carried out, as shows Figure S3. It was expected that the optimal response would be around pH 7.0 for biological processes; however, a shoulder peak can be observed around 145 mV, which increased in magnitude in more alkaline media, and could be related to the dopamine deprotonation (Breczko et al., 2012; Martínez-Huitle et al., 2009). Therefore, pH 6.1 was chosen as optimal, as it not only presented a better peak resolution, as it also showed less current deviation and no significant peak magnitude was lost.

After the optimization, the analytical curve was constructed for dopamine (oxidation reaction in Figure S4A), by varying its concentration from 30 to 800 $\mu\text{mol L}^{-1}$, in which was observed an increase in the peak current proportional to the concentration, being presented in Figure 5A. Therefore, it was obtained the linear equation $I_p \text{ (mA)} = 1.05 \times 10^{-6} \pm 5.30 \times 10^{-7} + 0.0530 \pm 1.21 \times 10^{-3} C_{\text{Dopamine}} \text{ (mmol L}^{-1}\text{)}$ with $R^2 = 0.997$ and the detection limit (LOD) was calculated by three times the

standard deviation of the blank divided by the slope of the analytical curve, resulting in a value of $0.13 \mu\text{mol L}^{-1}$. The Gr-AV electrode was applied for dopamine detection in spiked synthetic urine and saline samples where, respectively, a recuperation ranging from 83.2 to 109% and 90.3 to 102% was obtained, as presented in Table 1.

TABLE 1

The reproducibility study (four electrodes, one measurement per day, $n=4$) was carried out in presence of $500 \mu\text{mol L}^{-1}$ dopamine, in 0.05 mol L^{-1} PB solution (pH 6.1). The relative standard deviation (RSD) obtained was 4.46%. For the interference study, possible interfering species were used to evaluate the discrepant effects, being carried out with $500 \mu\text{mol L}^{-1}$ dopamine. In this context, ascorbic (Figure S5A) and uric (Figure S5B) acid presented no significant interference (2.0 and 1.0%, respectively) at concentration of 0.05 mol L^{-1} . For creatinine (Figure S5C), tests with $5.0 \times 10^{-3} \text{ mol L}^{-1}$ demonstrated an 8.0% peak deviation. However, we highlight that human urinary creatinine concentration ranges from 5.3×10^{-5} to $1.1 \times 10^{-4} \text{ mol L}^{-1}$ (Alessio et al., 1985; Barr et al., 2004; Boeniger et al., 1993). As the concentration is higher than the one found in human urine, it was obtained an acceptable deviation.

3.3. Electrochemical determination of serotonin with Gr-AV disposable electrode

To demonstrate the device electroanalytical versatility, Gr-AV was also applied towards the serotonin determination. Tests with cyclic voltammetry, at 50 mV s^{-1} , in presence of $100 \mu\text{mol L}^{-1}$ serotonin, in 0.1 mol L^{-1} PB solution were performed, as shown in Figure S6. An irreversible anodic peak was observed at 358 mV, with a current magnitude of 0.57 mA, corresponding to the species oxidation (Figure S4B).

Sequentially, a calibration curve was built by differential pulse voltammetry, in order to also demonstrate the device application. A linear behavior was found in the range of 6.0 to 100 $\mu\text{mol L}^{-1}$, in 0.1 mol L^{-1} PB solution, corresponding to the $I_p(A) = 4.37 \times 10^{-8} \pm 3.90 \times 10^{-9} + 1.75 \times 10^{-3} \pm 6.50 \times 10^{-5} C_{\text{Serotonin}} (\mu\text{mol L}^{-1})$ relation, with $R^2 = 0.993$. As aforementioned, the LOD of the sensor was calculated to be 0.39 $\mu\text{mol L}^{-1}$. The corresponding data is present in Figure 5B. Then, saline solution and synthetic urine samples were spiked with known concentrations of serotonin, in order to evaluate the matrix effect in this case as well. Table 1 presents the recovery data, ranging from 92.7 to 105% for the saline solution and from 96.6 to 108% for the synthetic urine samples.

FIGURE 5

3.4. Electrochemical determination of glucose with the GOx-DHP/Gr-AV disposable electrode

Figure S7 shows an attempt of detecting 10 $\mu\text{mol L}^{-1}$ glucose with the bare Gr-AV device (red), in 0.05 mol L^{-1} PB solution. As observed, no present component of the sensor can achieve the catalytic effect desired for the glucose reactions (Figure S4C and D). Therefore, a surface modification was evaluated, and the adhesive device was modified with a GOx-DHP dispersion, to obtain an electrocatalytic glucose biosensor. As a surfactant, it is expected (Janegitz et al., 2015) that DHP aqueous dispersions have lipophilic core micelles and hydrophilic heads in contact with the sample solution. In this way, we assure a better wettability, which can improve the electrochemical response (Ibanez-Redin et al., 2018; Ricci et al., 2009; Robinson et al., 2006). Figure 5C shows cyclic voltammograms obtained with GOx-DHP/Gr-AV in 0.05 mol L^{-1} PB solution (pH 7.0), at $v = 100 \text{ mV s}^{-1}$, in increasing concentration of glucose. With the catalyzed consumption of O_2 , the cathodic baseline decreases linearly to the available

glucose, until enzyme site saturation (reactions in Figure S4C and D). Therefore, all data were collected at 0.6 V and the Figure 5C inset shows the obtained behavior. The system presented linear response in the range of $1.0 \mu\text{mol L}^{-1}$ to $10 \mu\text{mol L}^{-1}$, with the equation $I_p(\text{A}) = 3.74 \times 10^{-6} \pm 1.82 \times 10^{-7} + 1.52 \pm 0.0536 C_{\text{Glucose}} (\mu\text{mol L}^{-1})$ and $R^2 = 0.991$. As aforementioned, the LOD of the sensor was calculated to be $0.21 \mu\text{mol L}^{-1}$. Also, a second linear response can be observed in the range of 10 to $40 \mu\text{mol L}^{-1}$ glucose. This considered low sensitivity is due to the effective saturation of enzyme catalytic sites, leading to a detection plateau. Therefore, further additions of glucose would cause an insignificant current variation. The device was applied in the detection of known concentrations of glucose in synthetic saliva and saline samples. For the synthetic saliva, the recovery ranged from 92.1 to 113%, and for the saline samples, from 95.8 to 103%, as shown in Table 1. In order to further evaluate the biosensor, *intradays* (four electrodes, one measurement per electrode, $n = 4$) and *interdays* (four electrodes, one measurement and electrode per day, $n = 4$) reproducibility tests were carried out, under optimal conditions, in presence of $5.0 \mu\text{mol L}^{-1}$ glucose. The calculated RSD values of 2.5 and 7.4% were obtained for *intra* and *interdays*, respectively.

Evaluations of the electrochemical behavior of dopamine in presence of glucose were also performed by using the proposed electrode. Figure S5D shows the data of Gr-AV in presence of $500 \mu\text{mol L}^{-1}$ dopamine and $8.0 \mu\text{mol L}^{-1}$ glucose, in 0.1 mol L^{-1} PB solution (pH 7.0), at same configurations as the calibration curve for dopamine. Even though the peak presented a large profile, suggesting a slower kinetic effect, the current magnitude does not have significant changes, with an RSD of 1.5% with $n = 4$. The opposite analysis was carried out with same concentrations, in 0.05 mol L^{-1} PB solution (pH 7.0), by cyclic voltammetry with $v = 100 \text{ mV s}^{-1}$ (Figure S5E) by using the

proposed biosensor. A significant alteration can be observed, with RSD of 10%, with $n = 4$. However, as literature introduces (Bergman et al., 2018; Zen et al., 1997) GOx can interact with dopamine, as many neuronal systems correlate the activities of both analytes. No tests were made in simultaneous presence of dopamine and serotonin, since both catecholamines have very similar molecular structure and oxidation pattern, and, by this reason, a significant influence is expected.

In Table S2, both Gr-AV and GOx-DHP/Gr-AV are compared to others already reported disposable electrochemical devices, towards the detection of dopamine and glucose. The focus was to compare with other disposable sensors, as this is the most commonly equal aspect of this work and the literature. The data demonstrate an equal efficiency between this and other works, validating this approach.

4. Conclusions

This work reports the development and application of a novel self-adhesive based electrochemical device, made by conductive ink of Gr and AV, towards the detection of dopamine (from 30 to 800 $\mu\text{mol L}^{-1}$, with a LOD of 0.13 $\mu\text{mol L}^{-1}$), serotonin (from 6.0 to 100 $\mu\text{mol L}^{-1}$, LOD corresponding to 0.39 $\mu\text{mol L}^{-1}$) and glucose (from 1.0 to 10 $\mu\text{mol L}^{-1}$, with a LOD of 0.21 $\mu\text{mol L}^{-1}$) by using three different voltammetric techniques (cyclic; square wave and differential pulse). As proof of concept, these experiments demonstrated acceptable analytical efficiency, with further tests in regards of its reproducibility, possible interfering species and operation in environmental and biological samples, in which both showed promising outcomes in all aspects. This work widens the scope of further surface modification, while simultaneously building interest in the field of electrochemical sensing and biosensing.

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5. References

- Alessio, L., Berlin, A., Dell'Orto, A., Toffoletto, F., Ghezzi, I., 1985. Reliability of urinary creatinine as a parameter used to adjust values of urinary biological indicators. *International archives of occupational and environmental health* 55(2), 99-106.
- Amin, S., Soomro, M.T., Memon, N., Solangi, A.R., Qureshi, T., Behzad, A.R., 2014. Disposable screen printed graphite electrode for the direct electrochemical determination of ibuprofen in surface water. *Environmental Nanotechnology, Monitoring & Management* 1, 8-13.
- Armelin, E., Whelan, R., Martínez-Triana, Y.M., Alemán, C., Finn, M., Díaz, D.D., 2017. Protective coatings for aluminum alloy based on hyperbranched 1, 4-polytriazoles. *ACS Appl. Mater. Interfaces* 9(4), 4231-4243.
- Arslan, F., Ustabaş, S., Arslan, H., 2011. An Amperometric Biosensor for Glucose Determination Prepared from Glucose Oxidase Immobilized in Polyaniline-Polyvinylsulfonate Film. *Sensors (Basel, Switzerland)* 11(8), 8152-8163.
- Baccarin, M., Santos, F.A., Vicentini, F.C., Zucolotto, V., Janegitz, B.C., Fatibello-Filho, O., 2017. Electrochemical sensor based on reduced graphene oxide/carbon black/chitosan composite for the simultaneous determination of dopamine and paracetamol concentrations in urine samples. *J. Electroanal. Chem.* 799, 436-443.
- Barr, D.B., Wilder, L.C., Caudill, S.P., Gonzalez, A.J., Needham, L.L., Pirkle, J.L., 2004. Urinary creatinine concentrations in the US population: implications for urinary biologic monitoring measurements. *Environmental health perspectives* 113(2), 192-200.
- Bergman, J., Mellander, L., Wang, Y., Cans, A.-S., 2018. Co-Detection of Dopamine and Glucose with High Temporal Resolution. *Catalysts* 8(1), 34.
- Berridge, K.C., Kringelbach, M.L., 2013. Neuroscience of affect: brain mechanisms of pleasure and displeasure. *Current opinion in neurobiology* 23(3), 294-303.
- Blackwell, J., Nagarajan, M., Hoitink, T., 1981. Structure of polyurethane elastomers. X-ray diffraction and conformational analysis of MDI-propandiol and MDI-ethylene glycol hard segments. *Polymer* 22(11), 1534-1539.
- Blair, M., 2016. Diabetes Mellitus Review. *Urologic nursing* 36(1), 27-36.
- Boeniger, M.F., Lowry, L.K., Rosenberg, J., 1993. Interpretation of urine results used to assess chemical exposure with emphasis on creatinine adjustments: a review. *American Industrial Hygiene Association Journal* 54(10), 615-627.

- 412 Breczko, J., Plonska-Brzezinska, M.E., Echegoyen, L., 2012. Electrochemical oxidation
413 and determination of dopamine in the presence of uric and ascorbic acids using a carbon
414 nano-onion and poly (diallyldimethylammonium chloride) composite. *Electrochim.*
415 *Acta* 72, 61-67.
- 416 Calegari, F., de Souza, L.P., Barsan, M.M., Brett, C.M., Marcolino-Junior, L.H.,
417 Bergamini, M.F., 2017. Construction and evaluation of carbon black and poly (ethylene
418 co-vinyl) acetate (EVA) composite electrodes for development of electrochemical (bio)
419 sensors. *Sensors and Actuators B: Chemical* 253, 10-18.
- 420 Campos, A.M., Raymundo-Pereira, P.A., Mendonça, C.D., Calegari, M.L., Machado,
421 S.A., Oliveira Jr, O.N., 2018. Size Control of Carbon Spherical Shells for Sensitive
422 Detection of Paracetamol in Sweat, Saliva, and Urine. *ACS Applied Nano Materials*
423 1(2), 654-661.
- 424 Cesiulis, H., Tsyntsar, N., Ramanavicius, A., Ragoisha, G., 2016. The study of thin
425 films by electrochemical impedance spectroscopy, *Nanostructures and Thin Films for*
426 *Multifunctional Applications*. Springer, pp. 3-42.
- 427 Cinti, S., Mazzaracchio, V., Cacciotti, I., Moscone, D., Arduini, F., 2017. Carbon black-
428 modified electrodes screen-printed onto paper towel, waxed paper and parafilm m®.
429 *Sensors* 17(10), 2267.
- 430 Cryer, P.E., 2016. Chapter 34 - Hypoglycemia A2 - Melmed, Shlomo, in: Polonsky,
431 K.S., Larsen, P.R., Kronenberg, H.M. (Eds.), *Williams Textbook of Endocrinology*
432 (Thirteenth Edition). Content Repository Only!, Philadelphia, pp. 1582-1607.
- 433 Dunn, D.J., 2003. *Adhesives and Sealants Technology, Applications and Markets*.
434 Rapra Technology Limited.
- 435 Dwier, D.S., 2002. Glucose metabolism in the brain.
- 436 Fox, S.H., Chuang, R., Brotchie, J.M., 2009. Serotonin and Parkinson's disease: On
437 movement, mood, and madness. *Movement disorders: official journal of the Movement*
438 *Disorder Society* 24(9), 1255-1266.
- 439 Fukushima, H., Drzal, L., Rook, B., Rich, M., 2006. Thermal conductivity of exfoliated
440 graphite nanocomposites. *Journal of thermal analysis and calorimetry* 85(1), 235-238.
- 441 Ge, S., Ge, L., Yan, M., Song, X., Yu, J., Huang, J., 2012. A disposable paper-based
442 electrochemical sensor with an addressable electrode array for cancer screening.
443 *Chemical Communications* 48(75), 9397-9399.
- 444 Goldstein, D.S., 2003. Catecholamines and stress. *Endocrine regulations* 37(2), 69-80.
- 445 Grisales, C., Herrera, N., Fajardo, F., 2016. Preparation of graphite conductive paint and
446 its application to the construction of RC circuits on paper. *Physics Education* 51(5),
447 055011.
- 448 Ibanez-Redin, G., Silva, T.A., Vicentini, F.C., Fatibello-Filho, O., 2018. Effect of
449 carbon black functionalization on the analytical performance of a tyrosinase biosensor
450 based on glassy carbon electrode modified with dihexadecylphosphate film. *Enzyme*
451 *and microbial technology* 116, 41-47.

- Iversen, L., L., Iversen, S.D., Snyder, S.H., 1977. Handbook of psychopharmacology - Volume 8: Drugs, Neurotransmitters, and Behavior.
- Jaiswal, N., Tiwari, I., 2017. Recent build outs in electroanalytical biosensors based on carbon-nanomaterial modified screen printed electrode platforms. *Anal. Methods* 9(26), 3895-3907.
- Janegitz, B.C., Baccarin, M., Raymundo-Pereira, P.A., dos Santos, F.A., Oliveira, G.G., Machado, S.A.S., Lanza, M.R.V., Fatibello-Filho, O., Zucolotto, V., 2015. The use of dihexadecylphosphate in sensing and biosensing. *Sensors and Actuators B: Chemical* 220, 805-813.
- Janegitz, B.C., Cancino, J., Zucolotto, V., 2014. Disposable biosensors for clinical diagnosis. *J. Nanosci. Nanotechnol.* 14(1), 378-389.
- Janegitz, B.C., Pauliukaite, R., Ghica, M.E., Brett, C.M.A., Fatibello, O., 2011. Direct electron transfer of glucose oxidase at glassy carbon electrode modified with functionalized carbon nanotubes within a dihexadecylphosphate film. *Sens. Actuator B-Chem.* 158(1), 411-417.
- Johari-Ahar, M., Karami, P., Ghanei, M., Afkhami, A., Bagheri, H., 2018. Development of a molecularly imprinted polymer tailored on disposable screen-printed electrodes for dual detection of EGFR and VEGF using nano-liposomal amplification strategy. *Biosensors and Bioelectronics* 107, 26-33.
- Johra, F.T., Lee, J.-W., Jung, W.-G., 2014. Facile and safe graphene preparation on solution based platform. *Journal of Industrial and Engineering Chemistry* 20(5), 2883-2887.
- Kang, W., Pei, X., Rusinek, C.A., Bange, A., Haynes, E.N., Heineman, W.R., Papautsky, I., 2017. Determination of lead with a copper-based electrochemical sensor. *Anal. Chem.* 89(6), 3345-3352.
- Kent, M.C., Light, N.D., Bailey, A.J., 1985. Evidence for glucose-mediated covalent cross-linking of collagen after glycosylation in vitro. *Biochemical Journal* 225(3), 745-752.
- Khomenko, V., Raymundo-Piñero, E., Béguin, F., 2008. High-energy density graphite/AC capacitor in organic electrolyte. *Journal of Power Sources* 177(2), 643-651.
- Lamas-Ardisana, P., Martínez-Paredes, G., Añorga, L., Grande, H., 2018. Glucose biosensor based on disposable electrochemical paper-based transducers fully fabricated by screen-printing. *Biosensors and Bioelectronics* 109, 8-12.
- Lee, V.B.C., MOHD-NAIM, N.F., Tamiya, E., Ahmed, M.U., 2018. Trends in paper-based electrochemical biosensors: From design to application. *Analytical Sciences* 34(1), 7-18.
- Li, L., Reich, S., Robertson, J., 2005. Defect energies of graphite: Density-functional calculations. *Physical Review B* 72(18), 184109.
- Martínez-Huitle, C.A., Cerro-Lopez, M., Quiroz, M.A., 2009. Electrochemical behaviour of dopamine at covalent modified glassy carbon electrode with l-cysteine: preliminary results. *Materials Research* 12(4), 375-384.

- 493 Mayeux, R., Stern, Y., Cote, L., Williams, J.B., 1984. Altered serotonin metabolism in
494 depressed patients with Parkinson's disease. *Neurology* 34(5), 642-642.
- 495 Mittal, K.L., 2003. Adhesion aspects of polymeric coatings. VSP BV.
- 496 Naumenko, A., Yashchuk, V., Bliznyuk, V., Singamaneni, S., 2012. Peculiarities of
497 Raman spectra of polyurethane/carbon nanotube composite. *The European Physical*
498 *Journal B* 85(4), 120.
- 499 Negash, N., Alemu, H., Tessema, M., 2015. Electrochemical Characterization and
500 Determination of Phenol and Chlorophenols by Voltammetry at Single Wall Carbon
501 Nanotube/Poly(3,4-ethylenedioxythiophene) Modified Screen Printed Carbon
502 Electrode. *International Scholarly Research Notices* 2015, 459246.
- 503 Orzari, L.O., de Araujo Andreotti, I.A., Bergamini, M.F., Junior, L.H.M., Janegitz,
504 B.C., 2018. Disposable electrode obtained by pencil drawing on corrugated fiberboard
505 substrate. *Sensors and Actuators B: Chemical* 264, 20-26.
- 506 Ozawa, K., Hashimoto, K., Kishimoto, T., Shimizu, E., Ishikura, H., Iyo, M., 2006.
507 Immune activation during pregnancy in mice leads to dopaminergic hyperfunction and
508 cognitive impairment in the offspring: a neurodevelopmental animal model of
509 schizophrenia. *Biological psychiatry* 59(6), 546-554.
- 510 Parnell, S., Min, K., Cakmak, M., 2003. Kinetic studies of polyurethane polymerization
511 with Raman spectroscopy. *Polymer* 44(18), 5137-5144.
- 512 Pekarovicova, A., Fleming, P.D., 2005. Innovations in Ink and paper technology to
513 improve printability. Pira international Ltd.
- 514 Phillips, C., Al-Ahmadi, A., Potts, S.-J., Claypole, T., Deganello, D., 2017. The effect
515 of graphite and carbon black ratios on conductive ink performance. *Journal of Materials*
516 *Science* 52(16), 9520-9530.
- 517 Pierson, H.O., 2012. Handbook of carbon, graphite, diamonds and fullerenes:
518 processing, properties and applications. William Andrew.
- 519 Pisoschi, A.M., 2012. Glucose Determination by Biosensors. *Biochem. Anal. Biochem.*
520 1, 1-2.
- 521 Popova, A.N., 2017. Crystallographic analysis of graphite by X-Ray diffraction. *Coke*
522 *and Chemistry* 60(9), 361-365.
- 523 Pradhan, K.C., Nayak, P., 2012. Synthesis and characterization of polyurethane
524 nanocomposite from castor oil-hexamethylene diisocyanate (HMDI). *Advances in*
525 *Applied Science Research* 3(5), 3045-3052.
- 526 Purushothama, H.T., Nayaka, Y.A., Vinay, M.M., Manjunatha, P., Yathisha, R.O.,
527 Basavarajappa, K.V., 2018. Pencil graphite electrode as electrochemical sensor for the
528 voltammetric determination of chlorpromazine. *Journal of Science: Advanced Materials*
529 *and Devices*.
- 530 Raymundo-Pereira, P.A., Shimizu, F.M., Coelho, D., Piazzeta, M.H., Gobbi, A.L.,
531 Machado, S.A., Oliveira Jr, O.N., 2016. A nanostructured bifunctional platform for

- 532 sensing of glucose biomarker in artificial saliva: Synergy in hybrid Pt/Au surfaces.
533 Biosensors and Bioelectronics 86, 369-376.
- 534 Ricci, F., Zari, N., Caprio, F., Recine, S., Amine, A., Moscone, D., Palleschi, G.,
535 Plaxco, K.W., 2009. Surface chemistry effects on the performance of an electrochemical
536 DNA sensor. Bioelectrochemistry (Amsterdam, Netherlands) 76(1-2), 208-213.
- 537 Robinson, L., Isaksson, J., Robinson, N.D., Berggren, M., 2006. Electrochemical
538 control of surface wettability of poly(3-alkylthiophenes). Surface Science 600(11),
539 L148-L152.
- 540 Schultz, W., 2001. Reward signaling by dopamine neurons. The Neuroscientist : a
541 review journal bringing neurobiology, neurology and psychiatry 7(4), 293-302.
- 542 Sharma, S., Singh, N., Tomar, V., Chandra, R., 2018. A review on electrochemical
543 detection of serotonin based on surface modified electrodes. Biosensors and
544 Bioelectronics 107, 76-93.
- 545 Sharp, D., Burkitt, R., 2015. Carbon materials for analytical electrochemistry: printed
546 carbon materials and composites. Materials Technology 30(sup7), B155-B162.
- 547 Smith, A., 1982. The coblenz society desk book of infrared spectra. The Coblentz
548 society desk book of infrared spectra, 1-24.
- 549 Steinbusch, H., Verhofstad, A., Joosten, H., 1978. Localization of serotonin in the
550 central nervous system by immunohistochemistry: description of a specific and
551 sensitive technique and some applications. Neuroscience 3(9), 811-819.
- 552 Syedmoradi, L., Daneshpour, M., Alvandipour, M., Gomez, F.A., Hajghassem, H.,
553 Omidfar, K., 2017. Point of care testing: The impact of nanotechnology. Biosensors and
554 Bioelectronics 87, 373-387.
- 555 Tian, X., Itkis, M.E., Bekyarova, E.B., Haddon, R.C., 2013. Anisotropic thermal and
556 electrical properties of thin thermal interface layers of graphite nanoplatelet-based
557 composites. Sci Rep 3, 1710.
- 558 Tortorich, R., Shamkhalichenar, H., Choi, J.-W., 2018. Inkjet-Printed and Paper-Based
559 Electrochemical Sensors. Applied Sciences 8(2), 288.
- 560 Vyazovskiy, V.V., Riedner, B.A., Cirelli, C., Tononi, G., 2007. Sleep homeostasis and
561 cortical synchronization: II. A local field potential study of sleep slow waves in the rat.
562 Sleep 30(12), 1631-1642.
- 563 Wang, H.-C., Lee, A.-R., 2015. Recent developments in blood glucose sensors. Journal
564 of Food and Drug Analysis 23(2), 191-200.
- 565 Wang, P., Wang, M., Zhou, F., Yang, G., Qu, L., Miao, X., 2017. Development of a
566 paper-based, inexpensive, and disposable electrochemical sensing platform for nitrite
567 detection. Electrochem. Commun. 81, 74-78.
- 568 Xu, G., Jarjes, Z.A., Desprez, V., Kilmartin, P.A., Travas-Sejdic, J., 2018. Sensitive,
569 selective, disposable electrochemical dopamine sensor based on PEDOT-modified laser
570 scribed graphene. Biosensors and Bioelectronics 107, 184-191.

Xu, L., Li, C., Ng, K.S., 2000. In-situ monitoring of urethane formation by FTIR and Raman spectroscopy. *The Journal of Physical Chemistry A* 104(17), 3952-3957.

Zarei, M., 2017. Portable biosensing devices for point-of-care diagnostics: Recent developments and applications. *TrAC Trends in Analytical Chemistry* 91, 26-41.

Zen, J.-M., Lo, C.-W., Chen, P.-J., 1997. An Enzymatic Clay-Modified Electrode for Aerobic Glucose Monitoring with Dopamine as Mediator. *Anal. Chem.* 69(8), 1669-1673.

Legend of figures

Figure 1. (A) Gr-AV electrode preparation: (I) Conductive ink preparation and application over a self-adhesive paper sheet. (II) After drying for 40 min, the electrodes are printed. (IV) ink excess is removed and the device is ready for (V) electrochemical analyses. (B) Casting of GOx-DHP over the working electrode surface. (C) The device ready to use

Figure 2. SEM images of (A) self-adhesive paper, (B) Gr-AV and (C) GOx-DHP/Gr-AV at (1) 200, (2) 1000 and (3) 5000 times increase

Figure 3. (A) FTIR spectra of automotive varnish (blue line), conductive ink (red line) and graphite powder (black line). (B) Raman spectra of AV (blue), Gr-AV ink (red) and Gr powder (black). (C) X-ray diffraction patterns of AV (blue), Gr-AV ink (red) and Gr (black). Inset shows a closer view of AV pattern

Figure 4. (A) Cyclic voltammograms obtained by Gr-AV in equimolar concentration of 0.1 mol L⁻¹ acidic (HCl pH 1.0, red line), alkaline (NaOH pH 13, black line) and neutral media (KCl pH 6.1, blue line); $\nu = 50 \text{ mVs}^{-1}$. (B) Cyclic voltammogram obtained by the disposable adhesive device Gr-AV in presence of 500 $\mu\text{mol L}^{-1}$ dopamine (green) and in absence of it (black), in 0.1 mol L⁻¹ PB solution (pH 6.1); $\nu = 50 \text{ mV s}^{-1}$. (C) Nyquist plot of the adhesive device (green) in presence of 500 $\mu\text{mol L}^{-1}$ dopamine, in 0.1 mol L⁻¹ PB solution (pH 6.1). Inset shows the equivalent fitting circuit

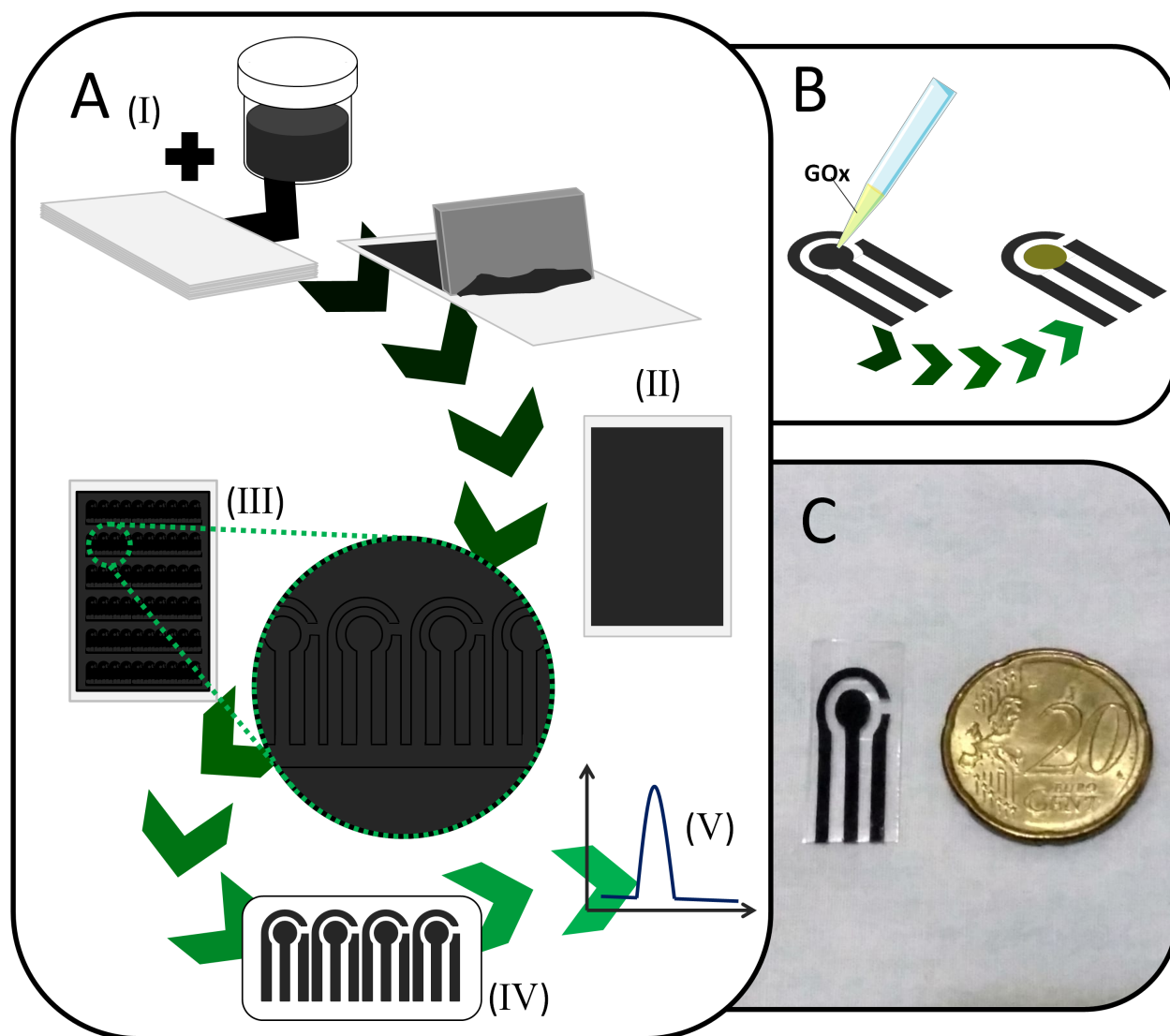
Figure 5. (A) Square wave voltammograms obtained with sticker electrode in presence of different concentrations of dopamine, varying from 30 to 800 $\mu\text{mol L}^{-1}$, in 0.1 mol L⁻¹ PB solution (pH 6.1); $s = 8 \text{ mV}$; $a = 100 \text{ mV}$; $f = 10 \text{ Hz}$. Inset shows the analytical curve. (B) Differential pulse voltammograms obtained by Gr-AV in presence of varying concentrations of serotonin, ranging from 0.6 (black) to 100 (lighter red) $\mu\text{mol L}^{-1}$ and

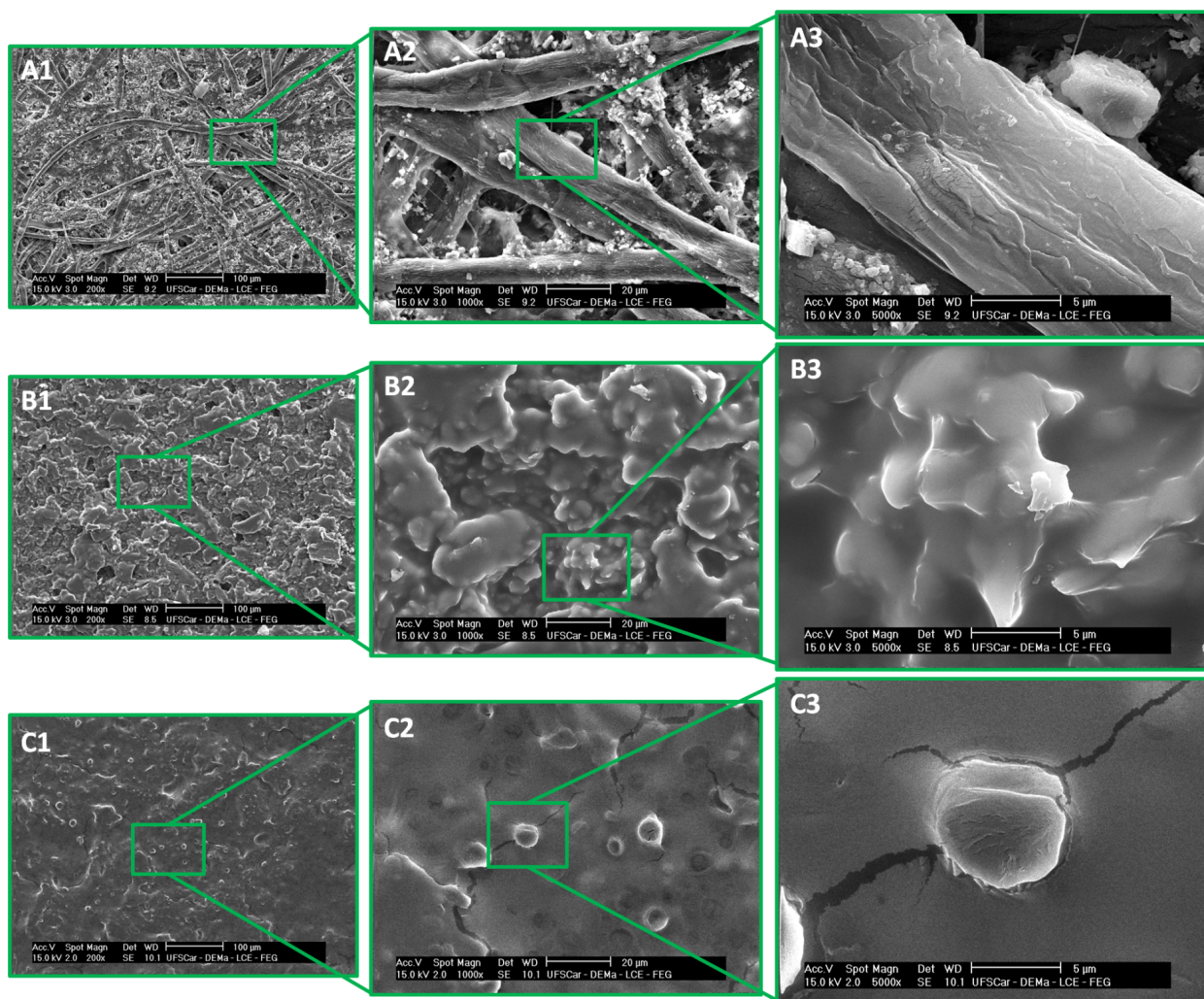
in absence of it (orange), in 0.1 mol L⁻¹ PB solution (pH 6.1); $\nu = 10 \text{ mV s}^{-1}$, $a = 25$
mV, modulation time = 5 ms. Inset shows the analytical curve. (C) Cyclic
voltammograms obtained by GOx-DHP/Gr-AV disposable device in presence of
varying glucose concentrations, ranging from 1.0 to 40 $\mu\text{mol L}^{-1}$, in 0.05 mol L⁻¹ PB
solution (pH 7.0); $\nu = 100 \text{ mV s}^{-1}$, $t = 60 \text{ s}$. Inset shows the analytical curve

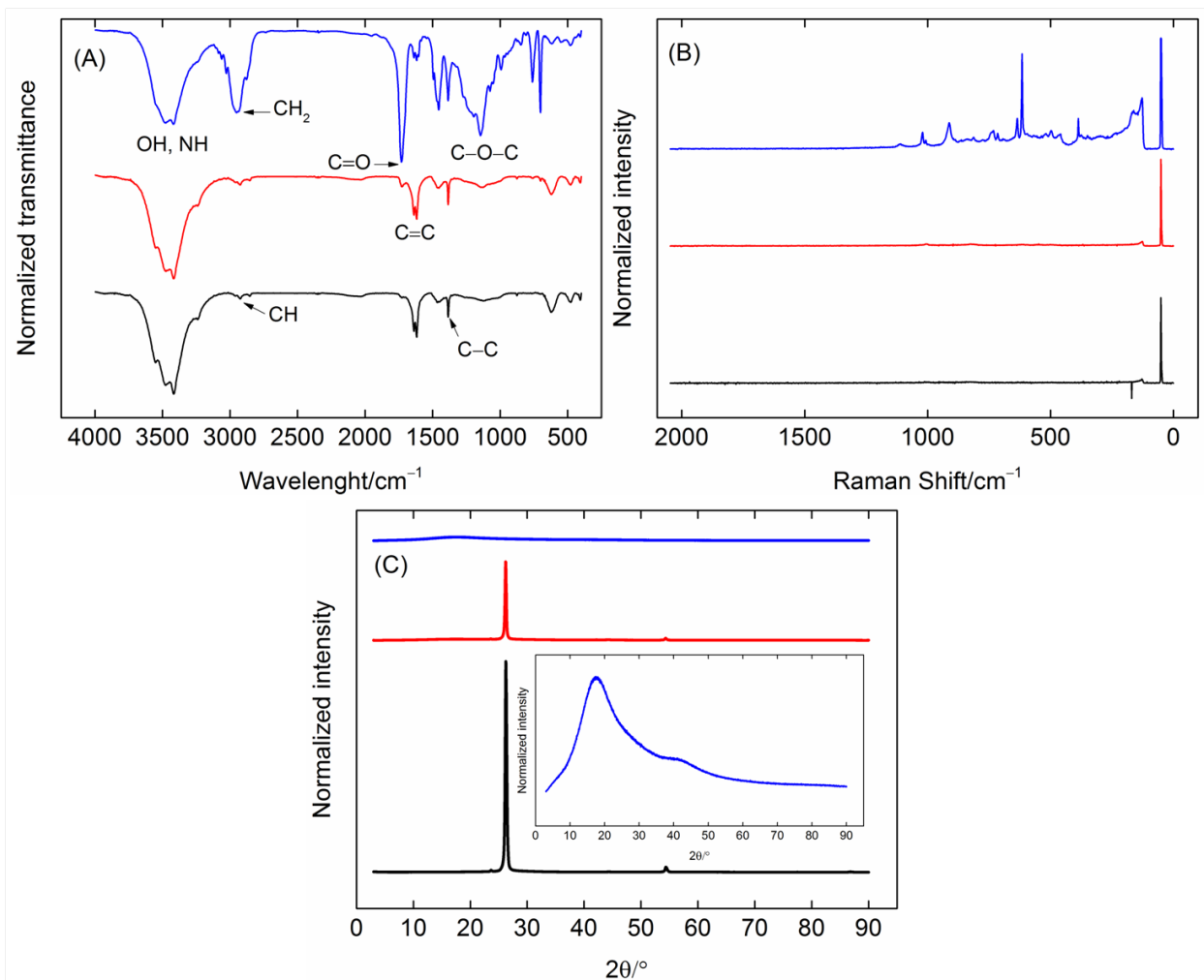
Tables

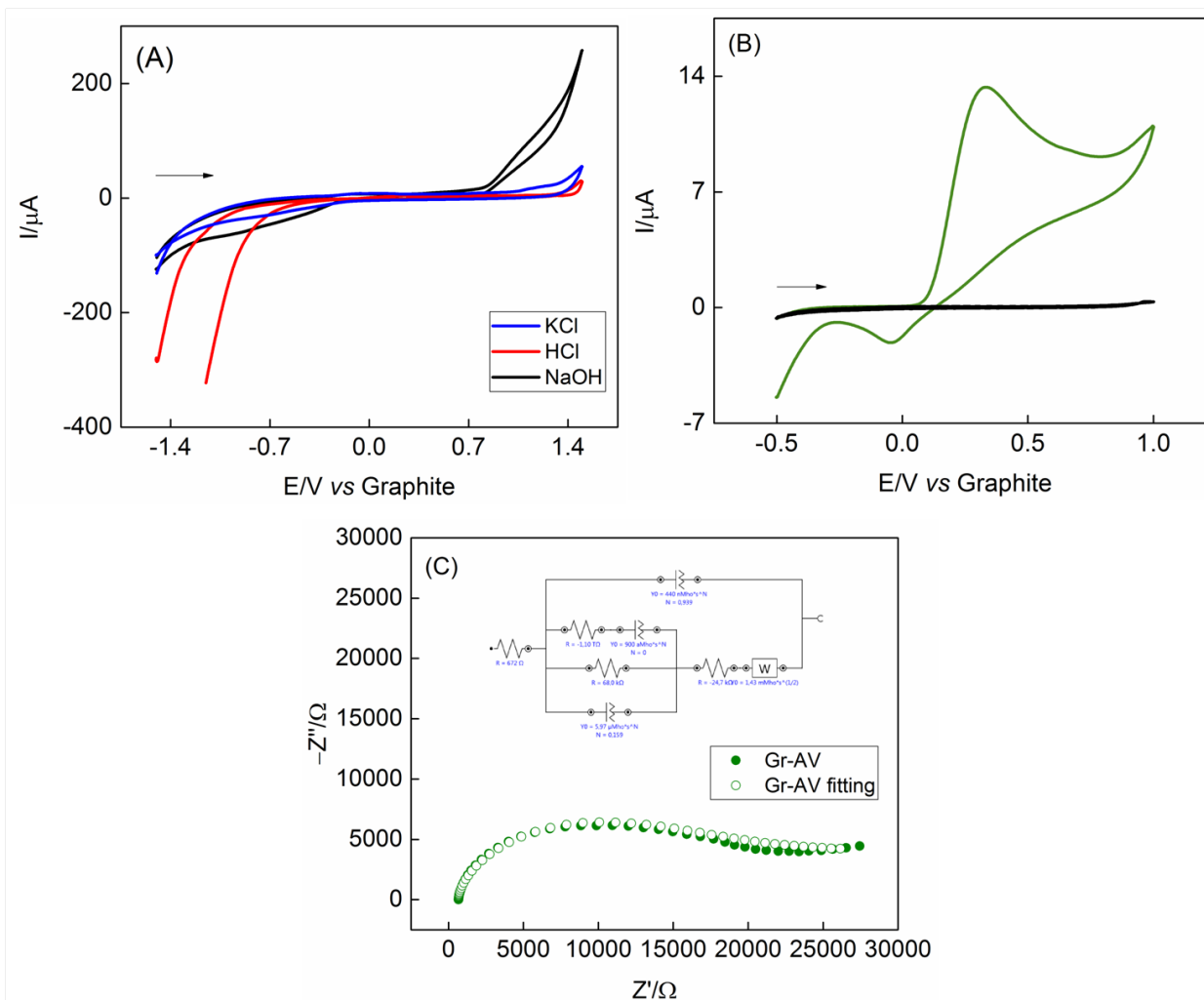
Table 1. Dopamine and serotonin determinations in spiked saline and synthetic urine samples by Gr-AV; and glucose determination in spiked saline and synthetic saliva samples by GOx-DHP/Gr-AV

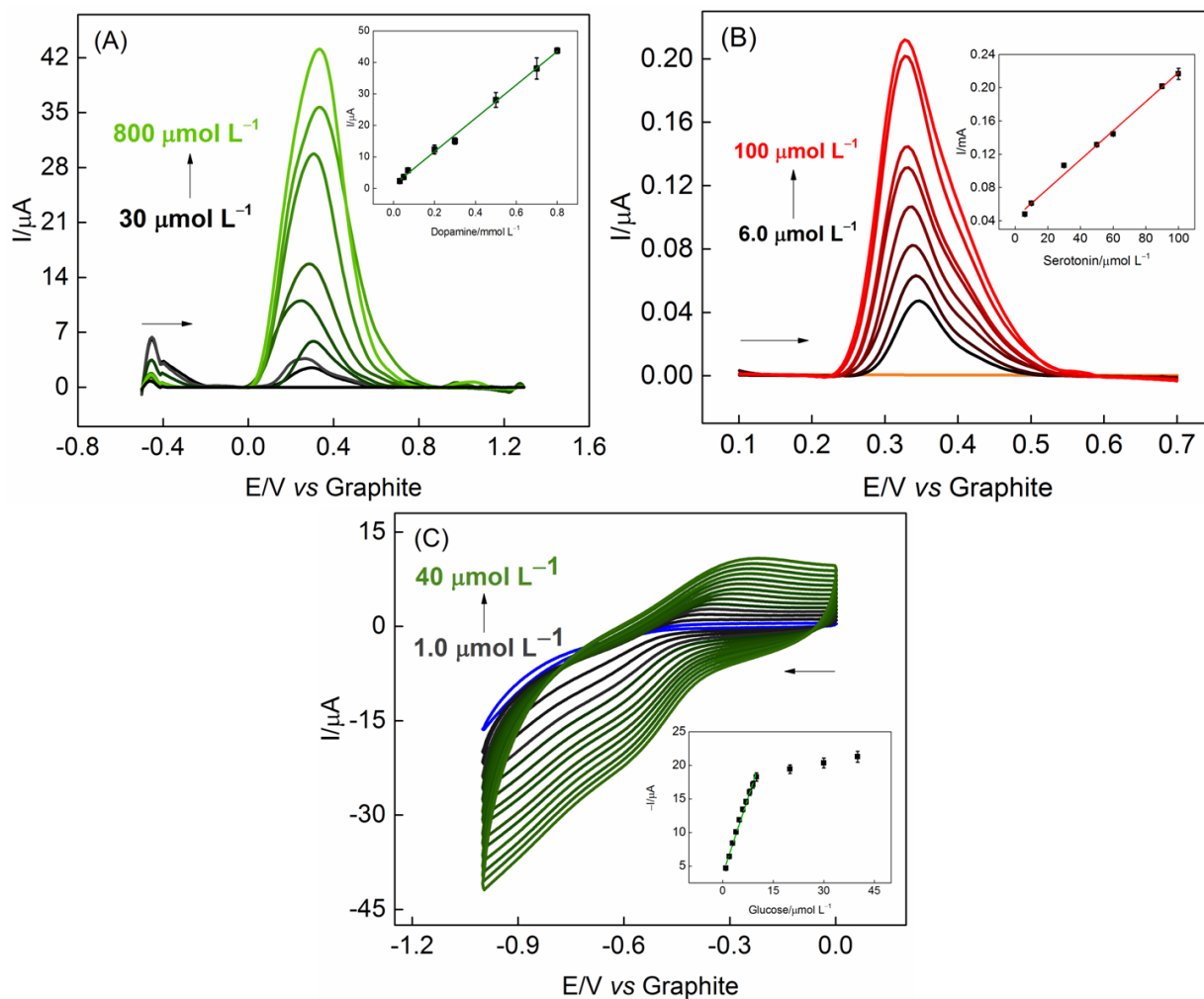
Gr-AV							
DOPAMINE				SEROTONIN			
Saline							
Sample	Added ($\mu\text{mol L}^{-1}$)	Detected ($\mu\text{mol L}^{-1}$)	Recovery%	Sample	Added ($\mu\text{mol L}^{-1}$)	Detected ($\mu\text{mol L}^{-1}$)	Recovery %
A	300	305	102	A	60	56	93.1
B	300	271	90.3	B	50	52	105
C	800	731	91.4	C	10	9.3	92.7
Synthetic Urine							
A	30.0	26.6	83.2	A	20	22	108
B	50.0	51.8	104	B	30	29	97.0
C	70.0	76.4	109	C	90	87	96.6
GOx-DHP/Gr-AV							
GLUCOSE							
Saline				Synthetic saliva			
A	9.00	8.83	95.8	A	9.00	8.83	92.1
B	8.00	8.12	99.9	B	8.00	8.12	102
C	7.00	7.92	103	C	7.00	7.92	113











Highlights

- New conductive ink using graphite and automotive varnish is proposed
- We have proposed an innovative self-adhesive electrochemical sensor
- Novel disposable electrochemical device was applied for dopamine, serotonin and glucose detection



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Credit Author Statement

I, Bruno Campos Janegitz , corresponding author, behalf of other co-authors, declare that all authors participated of the conceptualization; data curation; formal analysis; funding acquisition; investigation; methodology; project administration; resources; validation; writing - original draft; writing - review & editing of the manuscript entitled “A novel disposable self-adhesive inked paper device for electrochemical sensing of dopamine and serotonin neurotransmitters and biosensing of glucose” by Luiz Otávio Orzari, Rafaela Cristina de Freitas, Isabela Aparecida de Araujo Andreotti, Alexandre Gatti and Bruno Campos Janegitz.

Sincerely yours,

April 17th, 2019.

Dr. Bruno C Janegitz