Journal of Electroanalytical Chemistry 717-718 (2014) 61–68

Contents lists available at ScienceDirect

Journal of Electroanalytical Chemistry

journal homepage: www.elsevier.com/locate/jelechem

Large scale inkjet-printing of carbon nanotubes electrodes for antioxidant assays in blood bags

Andreas Lesch [a], Fernando Cortés-Salazar [a], Michel Prudent [b], Julien Delobel [b], Shokoufeh Rastgar [c], Niels Lion [b], Jean-Daniel Tissot [b], Philippe Tacchini [d], Hubert H. Girault [a,*]

[a] Ecole Polytechnique Fédérale de Lausanne, Laboratoire d'Electrochimie Physique et Analytique, Station 6, CH-1015 Lausanne, Switzerland
[b] Service Régional Vaudois de Transfusion Sanguine, Unité de Recherche et Développement, CH-1066 Epalinges, Switzerland
[c] Department of Chemistry, Sharif University of Technology, Tehran 11155-9516, Iran
[d] Edel for Life, PSE-B/EPFL, CH-1015 Lausanne, Switzerland

ARTICLE INFO

Article history:
Received 7 December 2013
Received in revised form 19 December 2013
Accepted 20 December 2013
Available online 8 January 2014

Keywords:
Carbon nanotubes
Amperometric sensors
Inkjet printing
Antioxidants
Erythrocyte concentrates

ABSTRACT

Herein, we present the large scale fabrication of carbon nanotubes (CNT) electrodes supported on flexible polymeric sheets by subsequent multilayer inkjet printing of a silver layer for electrical connection, CNT layers as active electrode material and an insulation layer to define a stand-alone CNT active electrode area with high accuracy. Optical and electrochemical characterization using several redox mediators demonstrates the reproducibility of the electrode surfaces and their functionality even with a single inkjet printed CNT layer. These electrodes are targeted to the clinical sector for the determination of the antioxidant power (AOP) of biologically relevant fluids by pseudo-titration voltammetry. As a proof-of-concept, the AOP of ascorbic acid solutions and biological samples such as erythrocyte concentrates (ECs) from different blood donors were determined demonstrating the potential use of the presented CNT sensors on ECs for blood transfusion purposes and the clinical sector.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

There is a great interest in measuring the antioxidant (AO) defense system directly or indirectly in a fast, non-expensive and reliable way, not only in the human health sector (*i.e.* in blood and saliva), but also in the pharmaceutical and food industries. The reason for such interest is the fact that the antioxidant defense system, which is composed of enzymes, metal chelators, drugs and dietary AOs, normally counterbalances oxidative stress conditions by transforming excess of reactive oxygen species (ROS) into harmless compounds and complexes. Otherwise, an enhanced ROS level or a reduced activity of the AO defense system can lead to the damage of intracellular proteins, lipids, DNA, cellular membranes, cells and the development of several pathological conditions such as Parkinson, Alzheimer and cardiovascular diseases, neurological disorders, diabetes and cancer [1–3]. Therefore, monitoring the AO content in blood and saliva can provide relevant information about the status of the AO system and the overall health condition of a specific person. The latter implicates the measurement of a global AO content, denoted as total antioxidant capacity (TAC), rather than of single compounds. It is usually determined by several assays including the trolox equivalent antioxidant capacity (TEAC) [4,5], the ferric reducing antioxidant power (FRAP) [6], the 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical scavenging [7] and the oxygen radical absorbance capacity (ORAC) [8]. These approaches are based on the monitoring of colored or fluorescent compounds that are generated or degraded in the presence of AOs by means of electron transfer (ET) or hydrogen atom transfer (HAT) reactions. Comparing the obtained values with respect to a sample with a known concentration of a model AO, *e.g.* Trolox, the TAC can be expressed in several matrixes in terms of a common unit [4–8].

Electrochemical assays that record signals of redox active species including AOs represent an alternative approach, which has been summarized in some recent reviews [9–11]. Cyclic voltammetry (CV) or linear sweep voltammetry (LSV) [12–14], differential pulse voltammetry [15,16], square wave voltammetry [17,18], the combination with HPLC detection [19,20] and flow systems such as flow injection analysis (FIA) [9] have been used over a broad range of biological fluids. More recently, other assays such as the reducing antioxidant capacity evaluated by electrolysis (RACE) [21], the rapid electrochemical screening of antioxidant capacity (RESAC) [22] or microfluidic devices [23] were proposed. In addition, a new pseudo-titration voltammetry (PTV) concept where AOs that are rapidly oxidized at low oxidation potentials are mathematically separated from slow reacting and less relevant compounds has also been described [24].

* Corresponding author. Tel.: +41 (0)21 693 31 51; fax: +41 (0)21 693 36 67.
E-mail address: hubert.girault@epfl.ch (H.H. Girault).

1572-6657/$ - see front matter © 2014 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.jelechem.2013.12.027

In terms of electrode materials, carbon-based electrodes like carbon paste, glassy carbon or modified carbon composites are mainly used for electrochemical detection of AOs due to their well-known affinity for organic molecules, biocompatibility and low cost. Recently, carbon nanotubes (CNT) materials have attracted a lot of attention due to their high surface area, mechanical stability, electrical conductivity and electrochemical reactivity [25–27]. Indeed, it is expected that such type of materials will overcome drawbacks encountered in graphite electrodes such as inhomogeneous electrode surfaces, electrode fouling and high overpotentials for some electrode processes [27]. However, the properties and electrochemical characteristics of CNTs are still somewhat controversial [28–30]. A CNT is composed of sp^2-hybridised, tubular carbon networks and can be of single wall (SWNT), double wall (DWNT) or multi wall (MWNT) type. Differences in their electrochemical behavior normally occur due to the nature of the CNTs, present functional groups, employed synthesis strategy, cleaning procedures and remaining metallic and carbonaceous impurities, storage conditions and aging [26,31–35].

Both graphite and CNT electrodes have been prepared by using screen-printed electrodes, thus allowing for batch fabrication of a multitude of disposable sensors [36]. A high resolution in the micrometer range is not easy to achieve by this technology and therefore a more precise technique is required to obtain highly reproducible sensors. Nowadays, inkjet printing of functional materials has become one very promising mask-free microfabrication technique that allows for a precise deposition of different materials such as CNTs [37–41]. Most of the functional CNT sensors have been deposited so far over a conductive metallic or carbon-based layer to ensure sufficient electrical conductivity among all the deposited CNTs. In addition, multiple printed layers (>10–30 printed layers) are employed with the same aim. For instance, very recently, a Nafion/MWNT inkjet printed electrode (*i.e.* 15 inkjet printed layers) deposited on an indium-tin-oxide (ITO) coated polyethylene terephthalate (PET) substrate was employed for the detection of dopamine in blood with a high sensitivity (0.1 μM) and a mean error less than 10% for 5 tested electrodes [39]. To date, stand-alone inkjet printed CNT electrodes with proper electrochemical behavior and fully functional as amperometric sensors after a single printed layer are still rare.

In this study, we use a commercially available DWNTs dispersion suitable for inkjet printing (IJP) for the fabrication of stand-alone CNT electrodes with high electrical conductivity and electrochemical activity thanks to the percolation of the randomly spread and flat CNT networks as corroborated by scanning electron microscopy (SEM) analysis. A silver layer for electrical connections and an insulating layer to precisely define the active electrode area complete the CNT sensor. Optical microscopic, electrical and electrochemical analysis demonstrated a high reproducibility of the fabrication process of more than 350 units per batch. The developed amperometric sensors were applied in combination with the PTV method for measuring the AOP of ascorbic acid solutions (*i.e.* a model AO) and in real samples such as erythrocyte concentrates (ECs), a labile blood product widely used in transfusion medicine, donated by two healthy persons and targeted for blood donation. The latter is of high relevance in blood product research and management since erythrocytes suffer from storage lesions, during storage at 4 °C, which alter lipids, proteins, metabolism, and biomechanical properties of the cells [42–48], resulting in potential adverse effects upon transfusion [47–52]. Erythrocytes are enucleated cells not able to synthesize proteins, which requires a solid system (like the repair or destroy box) [53,54] to protect the cells against oxidative stress. Thus in order to take the proper actions to diminish such effects, the measurement of AOs in ECs has to be performed in a reliable, cost-effective and precise way.

2. Experimental section

2.1. Chemicals

Ferrocenemethanol (FcMeOH; Sigma–Aldrich, Buchs, Switzerland), hexaammine ruthenium chloride ($[Ru(NH_3)_6]Cl_3$; Acros, Renningen, Germany), potassium hexachloroiridate(III) (K_3IrCl_6; Alfa Aesar, Karlsruhe, Germany), potassium chloride (KCl; Sigma–Aldrich, Buchs, Switzerland) and L(+)-Ascorbic acid (AA; Riedel-de Haën) were used as received and were of analytical grade. Deionized water was produced by a Milli-Q plus 185 model (Millipore, Zug, Switzerland). Jettable nano silver EMD5603 (w/w 20%), jettable insulator EMD6201 (both Sun Chemical, Carlstadt, USA) and the CNT dispersion BSI.B12212 (Brewer Science, Rolla, USA) were used as inks for IJP. As specified by the manufacturer, the CNTs are mostly DWNTs with an unknown amount of SWNTs with lengths $L = 763.08 \pm 595.16$ nm, diameters $d = 1.56 \pm 0.56$ nm and an aspect ratio $L/d = 322.6 \pm 454.5$. The CNTs were functionalized by the manufacturer with 1-pyrenemethylamine-3,6,8-trisulfonic acid groups to ensure a stable dispersion free of agglomerates.

2.2. Preparation of CNT sensors

CNT sensors were prepared on Kapton HN® foils (polyimide (PI); 125 μm-thick; Goodfellow, Huntingdon, England) by a multilayer inkjet printing process (*vide infra*) and by using the drop-on-demand DMP-2831 materials printer (Dimatix Fujifilm, Santa Clara, CA, USA). This printer allows simultaneous printing with up to 16 nozzles and providing droplet volumes of 1 and 10 pL. During IJP of both conductive inks, *i.e.* Ag and CNT, the substrate temperature was increased for a faster evaporation of the ink carrier solvents and therefore reaching an increase in the pattern resolution. The Ag and CNT patterns were cured for 30 min at 200 °C and 120 °C, respectively, with a heating and cooling rate of 1–2 °C min^{-1}. The insulating UV curable ink was printed under simultaneous UV exposition. The latter was achieved by a custom-made modification of the DMP-2831 system by mounting a light guide from an Omnicure® S2000 mercury UV lamp (Lumen Dynamics, Mississauga, Ontario, Canada) into the DMP-2831 print head. The Dimatix software allows opening of the UV light guide shutter during printing in order to polymerize the UV curable ink just after it has been deposited on the substrate. Printing parameters such as jetting frequency, waveform, voltage, cartridge temperature, and cleaning cycles were adjusted for optimum printing performance of each ink. The dimensions of the patterns were investigated by laser scanning microscopy (LSM) in reflection mode using a Keyence VK 8700 (Keyence, Osaka, Japan) and by SEM using a LEO 1550 (Carl Zeiss, Jena, Germany).

2.3. Preparation of erythrocyte concentrate

ECs were prepared from whole blood donations. Briefly, 450 ± 50 mL of blood from two healthy volunteer donors were mixed with 63 mL of citrate phosphate dextrose anticoagulant solution and left at 22 °C overnight. All blood components (*i.e.* erythrocytes, plasma and white blood cell- and platelet-containing buffy coat) were separated upon centrifugation at 3500 *g* for 14 min. The separated components were then distributed among the sterile inter-connected blood bags by applying a semi-automated pressure on the centrifuged original blood donation bag. Erythrocytes were then transferred into a SAG-M™-containing bag to a total volume of 275 ± 75 mL and a hematocrit level of 0.6 ± 0.1 *v*/*v*. A leukodepletion step was performed by filtration. ECs that did not meet the quality criteria for blood transfusion, *i.e.* a low hemoglobin content or a too small volume, were used un-

Download English Version:

https://daneshyari.com/en/article/6662590

Download Persian Version:

https://daneshyari.com/article/6662590

Daneshyari.com