



Non-enzymatic electrochemical detection of glucose with a disposable paper-based sensor using a cobalt phthalocyanine–ionic liquid–graphene composite

Sudkate Chaiyo^a, Eda Mehmeti^{b,c}, Weena Siangproh^d, Thai Long Hoang^e, Hai Phong Nguyen^e, Orawon Chailapakul^{a,f,*}, Kurt Kalcher^{b,**}

^a Electrochemistry and Optical Spectroscopy Center of Excellence (EOSCE), Department of Chemistry, Chulalongkorn University, 254 Phayathai Road, Patumwan, Bangkok, Thailand

^b Institute of Chemistry, Analytical Chemistry, Karl-Franzens University, Universitätsplatz 1, Graz A-8010, Austria

^c UBT-Higher Education Institution, Lagjia Kalabria p.n., 10000 Prishtina, Kosovo

^d Department of Chemistry, Srinakharinwirot University, Sukhumvit 23, Wattana, Bangkok, Thailand

^e College of Sciences, Hue University, 77 Nguyen Hue Str., Hue, Vietnam

^f Nanotec-CU Center of Excellence on Food and Agriculture, Chulalongkorn University, Bangkok 10330, Thailand

ARTICLE INFO

Keywords:

Paper-based analytical devices
Non-enzymatic glucose sensor
Cobalt phthalocyanine
Graphene
Ionic liquids

ABSTRACT

We introduce for the first time a paper-based analytical device (PAD) for the non-enzymatic detection of glucose by modifying a screen-printed carbon electrode with cobalt phthalocyanine, graphene and an ionic liquid (CoPc/G/IL/SPCE). The modifying composite was characterized by UV–visible spectroscopy, energy dispersive X-ray spectroscopy (EDX), transmission electron microscopy (TEM) and scanning electron microscopy (SEM). The disposable devices show excellent conductivity and fast electron transfer kinetics. The results demonstrated that the modified electrode on PADs had excellent electrocatalytic activity towards the oxidation of glucose with NaOH as supporting electrolyte (0.1 M). The oxidation potential of glucose was negatively shifted to 0.64 V vs. the screen-printed carbon pseudo-reference electrode. The paper-based sensor comprised a wide linear concentration range for glucose, from 0.01 to 1.3 mM and 1.3–5.0 mM for low and high concentration of glucose assay, respectively, with a detection limit of 0.67 μ M (S/N = 3). Additionally, the PADs were applied to quantify glucose in honey, white wine and human serum. The disposable, efficient, sensitive and low-cost non-enzymatic PAD has great potential for the development of point-of-care testing (POCT) devices that can be applied in healthcare monitoring.

1. Introduction

Glucose detection is an important issue because diabetes mellitus is one of the leading causes of death and disability in the world (Matz et al., 2006; Shepherd and Kahn, 1999). Abnormality of the glucose level in human blood causes several disorders such as blindness, nerve degeneration and kidney failure (Garg et al., 2004; Zhu et al., 2012). As an example, about 300 million people suffer from diabetes worldwide and this number is estimated to be almost double in 2030 (King et al., 1998; Zhang et al., 2010). The diagnosis and management of diabetic patients require exact monitoring and control of the glucose level in the body. Therefore, an enormous amount of glucose assays have been proposed, such as spectrophotometric (Ali et al., 2016; Pham et al.,

2016), chromatographic (Agblevor et al., 2004; Monti et al., 2017) and electrochemical methods (Liu et al., 2008; Pla-Tolós et al., 2016). Nevertheless, many approaches suffer from limitations of complicated pretreatment steps, equipment costs and expensive time-consuming labor resources. Apart from medicinal aspects glucose is an important analyte in foodstuff with respect to taste and nutritional value. Obviously, effective analytical methods for rapid determination of glucose are still directly needed.

Recently, microfluidic paper-based analytical devices (μ PADs) were reported by Whiteside's group as alternative devices for point-of-care testing for developing countries (Martinez et al., 2007). μ PADs provide simultaneous detection of multiple analytes on the same device, in which the platform is inexpensive, easy-to-use, and portable (Agblevor

* Corresponding author at: Electrochemistry and Optical Spectroscopy Center of Excellence (EOSCE), Department of Chemistry, Chulalongkorn University, 254 Phayathai Road, Patumwan, Bangkok, Thailand.

** Corresponding author.

E-mail addresses: corawon@chula.ac.th (O. Chailapakul), kurt.kalcher@uni-graz.at (K. Kalcher).

et al., 2004; Chaiyo et al., 2015; Dungchai et al., 2009; Martinez et al., 2007, 2008). Henry's group has already demonstrated the integration of electrochemical detections in paper-based analytical devices for the determination of glucose, lactate, and uric acid in biological samples using oxidase-based enzymatic reactions (Dungchai et al., 2009). Moreover, most previous studies on the electrochemical paper-based analytical devices of glucose are related to the biocatalysis of natural enzymes (e.g., glucose oxidase, and glucose dehydrogenase) toward the zymolyte with fast, accurate, and specific responses (Amor-Gutiérrez et al., 2017; Noiphung et al., 2013). However, there is some disadvantage for the use of enzymes originating from their sensitivity to temperature which may result in a reduced shelf lifetime by improper storage conditions (Park et al., 2006; Wang et al., 2008). Thus, fabrication of non-enzymatic glucose sensors has continuously been motivating research interests.

Metallophthalocyanines (MPcs) (e.g., cobalt(II) phthalocyanine, CoPc, copper(II) phthalocyanine, CuPc, and iron(II) phthalocyanine, FePc) are known to exhibit good electrocatalytic activities towards the oxidation of common chemical substances such as hydrogen peroxide (Foster et al., 2014), cysteine (Kuhnline et al., 2006), nitrite (Santos et al., 2006), nitric oxide (Vilakazi and Nyokong, 2001) and ascorbic acid (Agboola et al., 2009). Furthermore, metallophthalocyanine-based electrodes have been used as redox mediators for enzyme based glucose sensors (Barrera et al., 2006; Devasenathipathy et al., 2015; Özcan et al., 2008). However, MPc is not very appropriate on the electrode surface due to its low conductivity along with poor electrochemical activity (Cui et al., 2013). Recently, Agboola et al. have reported that carbon nanomaterials can combine with MPc to form a more stable composite due to the strong π interaction between MPc and the carbon nanomaterial (Agboola et al., 2009). Graphene (G) is an important member of the family of carbon nanomaterials with hexagonal aromatic structures. With outstanding properties such as good mechanical strength, high carrier mobility and large surface area (Guo and Dong, 2011; Li and Kaner, 2008), G is a promising candidate for forming a composite with MPc for non-enzymatic based glucose sensors. Furthermore, ionic liquids (IL) are either organic salts or mixtures of salts. Due to the excellent physicochemical properties such as high ionic conductivity, wide electrochemical windows, negligible vapor pressure, chemical and thermal stability, good antifouling ability, good biocompatibility, and inherent catalytic ability (Xu et al., 2009; Zhu et al., 2012).

In the present study, for the first time, a non-enzymatic electrochemical paper-based sensor using a cobalt(II) phthalocyanine/ionic liquid/graphene composite (CoPc/IL/G) was developed and applied for the determination of glucose.

2. Experimental

2.1. Materials and chemicals

Carbon ink was purchased from Acheson™ (California, USA). Glucose, sucrose, galactose, ascorbic acid, fructose, dopamine, lactose,

sodium hydroxide (NaOH) and potassium chloride (KCl) were purchased from Merck (Darmstadt, Germany). Industrial-quality graphene was obtained from ACS Material, LLC (Medford, USA). 1-Butyl-2,3-dimethylimidazolium tetrafluoroborate (IL), cobalt(II) phthalocyanine (CoPc), Whatman chromatography paper #1 (58 cm × 60 cm), N,N-dimethylformamide (DMF), paracetamol, uric acid, dopamine and potassium hexacyanoferrate(III) ($K_3[Fe(CN)_6]$) were purchased from Sigma–Aldrich (Buchs, Switzerland). All reagents were of analytical grade, and were used without further purification. All solutions were prepared using ultra-purified water ($> 18\text{ M}\Omega\text{ cm}$) refined by a cartridge purification system (Millipore, UK).

2.2. Instrumentation

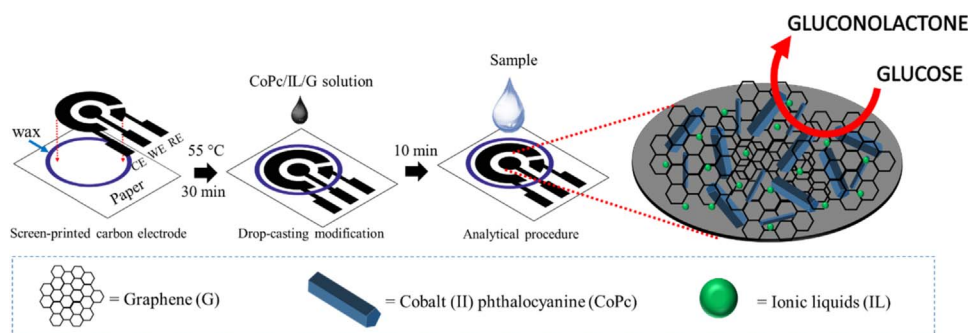
The electrochemical experiments were performed using an Autolab electrochemical system with a potentiostat PGSTAT 128 N (EcoChemie, Utrecht, Netherlands) controlled by corresponding software (NOVA 10.1). A scanning electronic microscope (SEM) (JEOL, Ltd., Japan) was used for microscopic analysis of paper devices. Transmission electron microscopic images (TEM) were recorded with an H-7650 device (Hitachi, Japan). The screen-printing block was fabricated by Chaiyaboon Co. Ltd. (Bangkok, Thailand). Absorbance measurements were conducted by a UV–visible spectrophotometer (HP HEWLETT PACKARD 8453, UK) using a 1.0 cm path length quartz cell.

2.3. Preparation of the CoPc/IL/G casting solution

For the preparation of the CoPc/IL/G composite, 1.0 mg of the graphene was dispersed in a closed polyethylene vial in DMF (1 mL) by ultrasonication (Transsonic T700/H, 560 W, 35 kHz, water volume 6 L) for about 3 h. The temperature was not controlled and was around 37 °C after the sonication time. Then, 5.0 mg of CoPc and 20 μL of the ionic liquid solution (0.5 mg mL^{-1} in DMF) were added to the graphene dispersion and sonicated for further 30 min.

2.4. Preparation of PADs and modified electrode

In this work, PADs were fabricated by wax-printing. First, the patterned paper-based sensor was designed by a graphic program (Adobe Illustrator) and then printed onto filter paper (Whatman no. 1) using a wax printer (Xerox ColorQube 8570, Japan). Next, the printed paper-based sensor was placed on a hot plate at 175 °C for 40 s to melt the wax. The area covered with wax was hydrophobic, and the area without wax was hydrophilic. For the three electrode system on the PADs, a working electrode (WE, area = 0.196 cm^2) a counter electrode (CE) and a pseudo-reference electrode (RE) were screen-printed in-house using carbon ink. The sensor was allowed to dry at 55 °C for 1 h. Then, about 2.0 μL of CoPc/IL/G composite solution was drop cast on the working electrode surface and dried at room temperature and atmospheric pressure for approximately 10 min as shown in Scheme 1. The CoPc- and graphene-modified screen-printed carbon electrodes were prepared analogously with suspensions containing the ionic liquid plus



Scheme 1. Schematic representation of the fabrication, modification and analytical procedure for the glucose sensor based on the coupling of the CoPc/IL/G SPCE with PADs.

Download English Version:

<https://daneshyari.com/en/article/7229872>

Download Persian Version:

<https://daneshyari.com/article/7229872>

[Daneshyari.com](https://daneshyari.com)