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A Brief Review on Fabrication of Screen-Printed Carbon Electrode: Materials and Techniques

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Abstract

Screen-printed carbon electrode (SPCE) is one of the most interesting designs to combine a working (from carbon based material), reference, and counter electrode in a single-printed substrate. SPCE has been used in many electrochemical measurements due to its advantages for analysis in microscale. This paper summarises the main information about SPCE fabrication from the material and fabrication technique aspect on the flat substrate based on the work that has been published in the last 30 years. The success of SPCE fabrication is highly dependent on the composition of conductive ink which consists of conductive materials, binder, and solvents; substrate; and fabrication techniques. Among the carbon-based materials, the most widely used for SPCE fabrications are graphite, graphene, and carbon nanotubes. The frequent binder used are polymer-based materials such as polystyrene, polyaniline, poly 3,4-ethylenedioxythiophene:polystyrene sulfonate (PEDOT:PSS), and polyvinyl chloride. The solvents used for SPCE fabrication are varied including water and various organic solvents. The main characteristics of the SPCE substrate should be inert in order to avoid any interferences during electrochemical measurements. The screen printing and inkjet printing technique are preferred for SPCE fabrication due to easy fabrication and the possibility for mass production of SPCE.

Keywords: Electrode, fabrication, inkjet, polymer, screen printing

INTRODUCTION

The development of electrochemical sensors is increasing rapidly every year. Those electrochemical sensors are applied for in-situ monitoring and point of care testing in pharmaceutical, biomedical, industrial, and environmental analysis. Since the in-situ monitoring and point of care analysis need a rapid and simple procedure, a disposable electrode is required. Screen printing-based electrodes is a disposable electrode for electrochemical sensors that suits the requirement. The screen-printed electrode combines working, reference, and a counter electrode in a substrate that supports the miniaturization and portability aspect of the sensor. Furthermore, due to the ease in its fabrication mass production of a screen-printed electrode is possible to conduct.

Several types of screen-printed electrodes have been already developed, among other screen-printed electrodes fabricated from carbon-based material gain a lot of attention. Screen-printed carbon electrode (SPCE) combines a carbon working electrode, a reference electrode, and a counter electrode in a substrate. The properties of carbon material including a wide potential window, low background current, inertness (Matters et al., 2011), and low cost (Liu et al., 2012) drives the massive usage of SPCE for the electrochemical sensor.

Various methods for SPCE fabrication techniques have been investigated and studied in numerous scientific papers. In general, this fabrication process includes the preparation of conductive inks for working, reference, and counter electrodes as well as the printing process of prepared conductive ink onto the substrate. The fabrication method is adjusted with the character of the conductive ink, the character of the substrate, and the intended application of the fabricated electrodes. In addition, it is important to consider the control over the precision of the process and the device in the fabrication process.

A comprehensive study of SPCE fabrication covering materials and techniques is required to provide information and references for the success of the SPCE fabrication process. Several studies on screen-printed carbon electrode fabrication have been reported (Figure 1), otherwise to the best of authors knowledge, the review focus on SPCE fabrication covering materials and fabrication techniques is still limited. This review focuses on the aspect of material for SPCE fabrication including a conductive material, substrate, solvent, binder, and the techniques of SPCE fabrication including printing and screen printing. This review is expected to be a reference for SPCE fabrication.

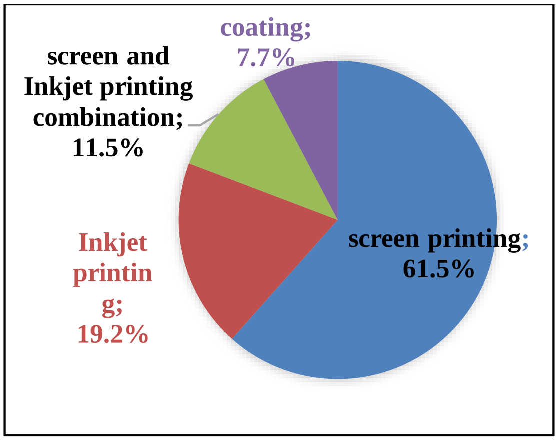

Figure 1. Variety of fabrication technique of SPCE.

Components for the fabrication of SPCE

Conductive materials for SPCE fabrication

The materials for SPCE fabrication have been selected by considering the properties to conduct electrons in the electrochemical systems. The material conductivity will impact the performance of the electrochemical system. Beside the conductivity properties, the other influencing factors of material can be used as an electrode such as potential window, background current, and corrosion stability. In general, a screen-printed electrode employs carbon material as the main component for its working electrode. The initial step of SPCE fabrication is the production of conductive ink for the working and reference electrode. The main component to fabricate a working electrode can be carbon derivatives materials such as graphite (Petroni et al., 2017) which can be obtained from nature (Latupeirissa et al., 2017; Tanasale et al., 2014; Sekewael et al., 2015), graphene (Karuwan et al., 2012), single-walled carbon nanotube (Tortorich et al., 2014), or multi-walled carbon nanotube (Da Costa et al., 2015). While the major component of conductive ink to produce a reference electrode is silver ink.

Graphite has potency as an electrode material to be used for electrochemical sensors fabrication regarding its better conductivity than carbon material. Graphite has a high surface areas, low molecular weight, and behave either as conductor or semiconductor (Tortorich and Choi, 2013), a wide range of potential window which can be used as a good mediator for electron transfer process in the working electrode (Kochmann et al., 2012). In addition to the good conductivity of graphite combined with its corrosion stability can be used as the main component for the working electrode as well as composite working electrode (Borenstein et al., 2017).

Graphene can be used as a material alternative for the working electrode due to its high conductivity, high surface area, and good mediator for the electron transfer process (Karuwan et al., 2012; Karuwan et al., 2013). In addition of graphene has a hexagonal structure with sp^2 hybridization makes it a good material for electron transfer mediators. Graphene has been used in many electrochemical sensors such as for hydroquinone detection in the cosmetic sample (Duekhuntod et al., 2019) detection for carcinoembryonic antigen (Chan et al., 2015), and simultaneous detection of quercetin and rutin (Elinda et al., 2019). The use of graphene to quercetin and rutin simultaneously using modified SPCE showed better linearity, precision, the detection dan quantitation limit, and stability compared to commercial SPCE. In the development of a screen-printed electrode, graphene (Karuwan et al., 2012; Karuwan et al., 2013), graphene oxide (Kudr et al., 2020), single-walled carbon nanotube (Tortorich et al., 2014), or multi-walled carbon nanotube (Da Costa et al., 2015) also can be used as a carbon substitute in the working electrode.

Substrate for SPCE fabrication

The screen-printed electrode can be modified using different materials in order to adjust with various substrates. The good substrate for the fabrication of the screen-printed electrode should not give any current response when it is contacted with a sample solution due to avoiding any electrochemical reaction on the electrode surface. The surface tension effect, substrate hydrophilicity, and hydrophobicity also giving an influence for the electrochemical performance of SPCE (Du et al., 2016). In general, the substrate should have water-resistant properties to avoid any damage issues during the measurement. The rough surface of the substrate is preferred due to ease in the ink printing process and produced a good quality of screen-printed electrode. The substrates have been used to fabricate SPCE are ceramic (Taleat et al., 2014), polyethylene terephthalate (PET) film (Du et al., 2016), Whatman filter paper (Nontawong et al., 2018), ink press transparent film [5], poly(ethylene 2,6-naphthalate) (PEN) (Kudr et al., 2020), and polyimide (Lesch et al., 2014) and polyvinyl chloride

(Karuwan et al., 2013; Duekhuntod et al., 2019; Khaled et al., 2008; Khaled et al., 2010; Promphet et al., 2015; Tirawattanakoson et al., 2016; Wahyuni et al., 2021).

Combination of binder materials with solvent in SPCE fabrication

The fluid properties such as density, viscosity, and surface tension are the prime parameters for ink chloride (Khaled et al., 2008; Khaled et al., 2010), cellulose acetate (Wring and Hart, 1992; Miserere et al., 2006), PEDOT:PSS (Sriprachuabwong et al., 2012), and polyaniline (Maity et al., 2019). The ohmic resistance and thickness of the printed electrode will increase with an increasing of binder concentration in the ink formula. In a certain case, it might result to produce a highly viscous ink. This will lead to the increasing of electrical resistance and resulted a

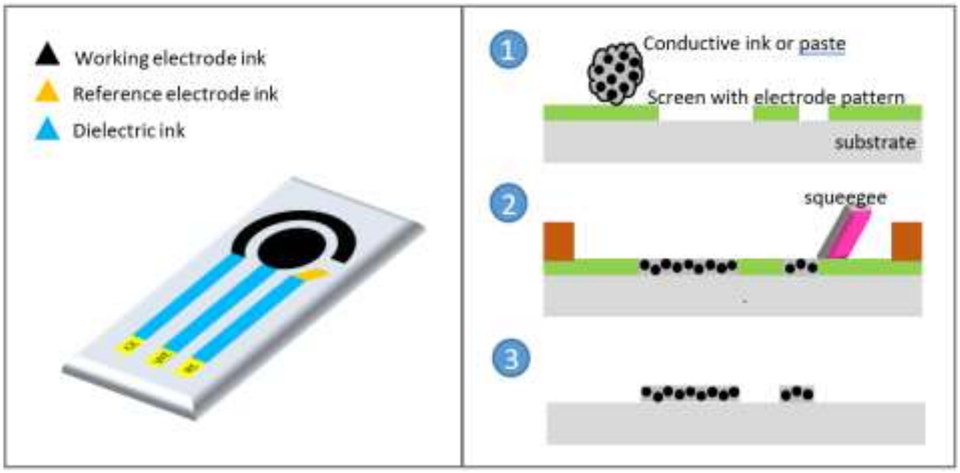

Figure 2. SPCE pattern and illustration of SPCE fabrication by a screen printing technique.

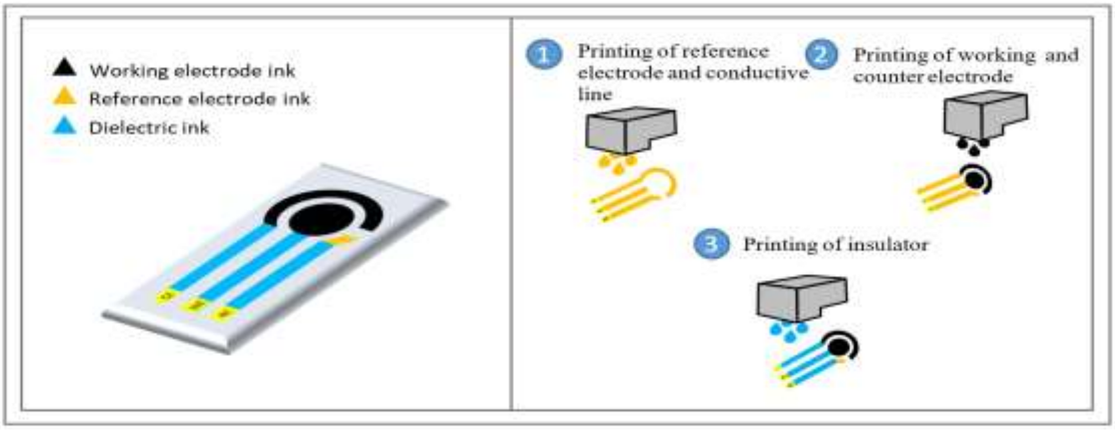

Figure 3. Pattern of SPCE and illustration of SPCE fabrication by inkjet printing technique

selection (Cummins and Desmulliez, 2019). The quality of conductive ink for SPCE fabrication determines its performance in the electrochemical system. In order to obtain a good quality of electrode ink, binder substance to maintain conductive materials on the surface of the electrode. Therefore, it can improve the quality of the screen-printed electrode and thus obtaining a good response from the electrochemical measurement. Binder material for SPCE fabrication can be used from polystyrene (Petroni et al., 2017; Wahyuni et al., 2019), polyvinyl diminished in the current signal.

Besides binder materials, solvent also helps to preserve the ink consistency in the liquid form since its mixing until its application to the substrate surface. The process for solvent selection affected by conductive and binder materials, type of substrates and the screen-printed electrode fabrication techniques. The properties of solvents that suitable for screen printing technique is a solvent with moderate points of boiling temperature and optimum viscosity. These solvent properties will impact to a better ink adhesion to the substrate and an efficient of ink drying process. Several organic solvents have been applied for SPCE fabrication such as dichloromethane (Petroni et al., 2017), cyclohexanone (Miserere et al., 2006), cyclohexanone-acetone (Khaled et al., 2008; Khaled et al., 2010), chloroform (Wahyuni et al., 2019), and diacetone hexanol (Tirawattanakoson et al., 2016).

Fabrication Techniques of Screen-Printed Carbon Electrode

Screen-printed carbon electrode combines working, reference, and counter electrodes in one design. This type of electrode provides a simple, portable, and disposable electrochemical measurement for sensing purposes. Since this electrode is disposable, the fabrication techniques should be able to facilitate fast mass production. Several fabrication techniques for SPCE fabrication were reported in scientific papers. Most of them are using printing techniques (inkjet printing and screen printing) on flat substrate and the other was a coating technique (Table 1).

The screen-printing technique is a simple approach for SPCE fabrication. This technique is easy to follow by anyone without special expertise. In the screen-printing technique, three main components in SPCE fabrication including screen with electrode pattern, conductive ink, and squeegee like part for spread the ink. The screen is available in various pore sizes. The screen is selected based on the particle size of conductive material for ink composition. The pore size of screen material should bigger than the particle size of conductive material. Several papers reported the use of a screen with a pore size of 36 mesh (Khaled et al., 2008), 100 mesh (Wring and Hart, 1992), dan 200 mesh (Bishop et al., 2016).

Figure 2 shows the scheme of SPCE fabrication by a screen-printing technique. The steps are 1) Screen contains SPCE pattern is placed on top of the substrate, 2) The conductive ink is placed on top of the screen, 3) The ink is spread to all part of screen with constant pressure. Step 3 is crucial to produce a homogeneous surface of SPCE from the screen-printing fabrication process. Screen printing technique could be applied for SPCE fabrication both on flexible and rigid substrates. For instance, Kit-Anan and colleagues applied screen printing technique for SPCE fabrication on Whatman paper filter (Kit-Anan et al., 2012) while develop fabrication of SPCE on PET substrate (Du et al., 216). As a comparison, fabrication of SPCE by inkjet printing technique offers more precise result. The technology of inkjet printing machine provides precise control on ink dispensing.

Furthermore, the Inkjet printing technique consumes less ink compared to the screen printing technique. Several printer models are used in SPCE fabrication by inkjet printing technique, for instance Fujifilm Dimatix Materials Printer (Karuwan et al., 2012; Kudr et al., 2020; Lesch et al., 2014; Sriprachuabwong et al., 2012; Cinti et al., 2018; Kit-Anan et al., 2012), printer inkjet HP Deskjet 5650 (Tortorich and Choi, 2012), HP Deskjet D4260 (Da Costa et al., 2015), Epson EcoTank ET-2650, Epson Stylus Photo 1500W (Rosati et al., 2019), and EPSON R230 (Cai et al., 2019).

The pattern and scheme of SPCE fabrication by inkjet printing technique are illustrated in Figure 3. The electrode pattern was prepared by computer-aided design (CAD) and it was sent to the printer which able to print the object rapidly. Further, the ink was dispensed from the cartridge to the substrate through the nozzle (Moya et al., 2017). Several techniques such as piezoelectric (Wijshoff, 2010), thermal (Setti et al., 2005) dan electrodynamic (Ali et al., 2016) are used for ink dispense. The piezoelectric is preferred over other techniques due to its ability to control size and printing speed (Derby, 2010).

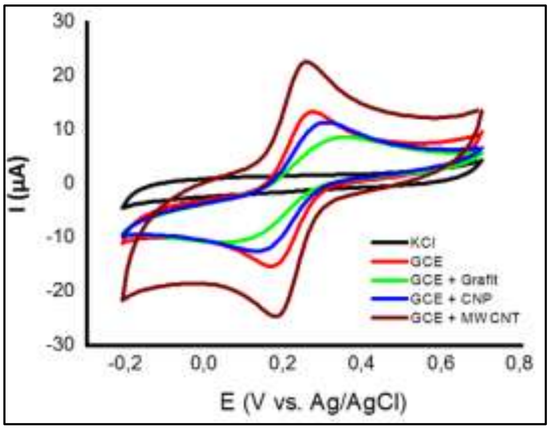

Figure 4. Cyclic voltammogram of 1 mM K_3FeCN_6 in 0.1 M KCl at glassy carbon and glassy carbon modified with conductive materials by drop coating method.

Furthermore, the inkjet printing technique provides the automatic process that able to produce SPCE on a large scale in a short time. Both screen printing and inkjet printing techniques possess the advantages to fabricate a screen-printed electrode. Screen printing technique advantages are simple and able to produce planar electrode with high precision in huge number for short duration (Metter et al., 2011).

Table 1. List of literatures reported the SPCE fabrication with inkjet printing and screen-printing technique

Technique	Analyte	Conductive CMaterial	Solvent	Binder Material	Substrate	Ref.
Screen-printing	Reduced glutathione (GSH)	Graphite	Cyclohexanone-acetone	Cellulose acetate	PVC	Wring and Hart, 1992
	NO	Graphite	Cyclohexanone	Cellulose acetate	Alumina ceramic, plastic	Miserere et al., 2006
	$[Fe(CN)_6]^{3-}$	Graphite commercial ink	-	-	Polyester film	Kadara et al., 2009
	Pb^{2+}, Cd, Cu	Carbon	Cyclohexanone-acetone	PVC	PVC	Khaled, 2010
	H_2O_2, NADH, $[Fe(CN)_6]^{3-}$	Graphene-carbon paste	-	-	PVC	Karuwan et al., 2013
	Simultaneous isoproturon &carbendazim	Graphene, graphite, and commercial carbon ink	-	Diethytlene glycol, monobutyl ether: 2-butoxyethyl acetate	PVC	Noyrod, et al., 2014
	Pb^{2+}, Cd^{2+}	commercial carbon ink	-	-	PVC	Promphet et al., 2015
	$[Fe(CN)_6]^{3-}$, $[Fe(CN)_6]^{4-}$	commercial carbon ink	-	-	PET	Du et al., 2016
	$[Fe(CN)_6]^{3-}$	commercial graphite ink	-	-	Cellulose acetate Film	Bishop et al., 2015
	Antioxidant	Commercial carbon ink, modified graphene	-	-	PVC	Tirawattanakoson et al., 2016
	Ascorbic acid, uric acid, dopamine	Graphene	Diacetone and hexanol	-	Polyester	Randviir et al., 2015
	Hydroquinone	Graphene and carbon paste	-	-	PVC	Duekhuntod et al., 2019
	Capsaicin	commercial graphite ink modified SWCNT and MWCNT	-	-	Polyester film	Metters et al., 2013
	Nitrite	Graphite	DCM	Polystyrene	Acrylic	Petroni et al., 2017
	Simultaneous of ascorbic acid, dopamine, and uric acid	Modified graphite paste	-	-	Whatman filter paper	Nontawong et al., 2018
	Phosphate	Graphite	Cyclohexanone-acetone	PVC	PVC	Khaled et al., 2008

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Table 1. List of literatures reported the SPCE fabrication with inkjet printing and screen-printing technique (*Continued*)

Technique	Analyte	Conductive Material	Solvent	Binder Material	Substrate	Ref.
Inkjet printing	Simultaneous of ascorbic acid, dopamine, and uric acid	Modified graphite paste	-	-	Whatman filter paper	Nontawong et al., 2018
	Phosphate	Graphite	Cyclohexanone-acetone	PVC	PVC	Khaled et al., 2008
	Salbutamol	Graphene	-	PEDOT: PSS	SPCEs	Karuwan et al., 2012
	Ascorbic acid	Commercial carbon ink	-	-	Smart paper	Cinti et al., 2018
	Blood antioxidant	Carbon nanotube commercial ink	-	-	Polyimide	Lesch et al., 2014
	Fe^{2+}, dopamine	Multiwalled carbon nanotube	Aquabidest	-	Paper	Da Costa et al., 2015
Screen and inkjet printing	Ascorbic acid	Graphite carbon paste modified with polyaniline	-	-	Whatman filter paper	Kit-Anan et al., 2012
	Fe_2SO_4	Single walled carbon nanotube	Aquabidest	-	Transparent film InkPress	Tortorich et al., 2014

The disadvantage of this technique is unable to reach small corner of the mold, as the consequence the printing result looks smear. In contrast, fin inkjet printing fabrication conductive ink are dispense accurately with high precision. However, homogeneity of fabricated electrode surface is low due to the very thin coating. To handle this problem, replication in printing process are needed. Both screen printing and inkjet printing techniques in SPCE fabrication are applied for mass production of SPCE. Furthermore, some papers reported the combination of both techniques to obtain the desire properties of fabricated SPCE (Tortorich et al., 2014; Kudr et al., 2020; Kit-Anan et al., 2012).

Fabrication technique with coating techniques are performed by drop-coating and drop-casting technique. This technique is widely used to modify electrodes and enhance its electrochemical properties (Wahyuni et al., 2019; Maity et al., 2019; Wahyuni et al., 2020). Conductive ink deposited on top of electrode surface by dropping. In some cases, masking technique is applied to obtain the specific pattern of coating. In addition, drop coating method could be applied to check electrochemical properties of conductive materials. For instance, comparison of electrochemical performance of carbon nanoparticles, multi-wall carbon nanotube (MWCNT), and graphite. Conductive ink from each material is drop coated on top of glassy carbon electrode (GCE). Figure 4 shows the cyclic voltammogram of potassium hexacyanoferrate at GCE modified with each material. MWCNT modified GCE provides higher current compare to other materials, indicated that MWCNT having higher conductivity compared to other tested materials.

CONCLUSION

The success of SPCE fabrication is determined by several aspects, among others are the composition of conductive ink, substrate, and fabrication technique. Conductive material for ink composition should possess high conductivity property. Several carbon-based materials are promising for SPCE fabrication, including graphite, graphene, and carbon nanotubes. Other materials that significant for conductive ink properties are binder and solvent. Binder enhances the ink properties including viscosity, homogeneity, and ink adhesivity to the substrate while the solvent should be able to disperse the conductive material and binder in a homogeneous mixture. In addition, conductive ink properties should be suitable with the substrate and fabrication technique. Based on the literature review, the most widely used SPCE fabrication techniques are screen printing and inkjet printing. Both printing technologies are capable to produce electrodes that match the specifications for electrochemical measurement and sensing.

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