CHAPTER I
INTRODUCTION

In recent years, a vast number of reports on screen-printing carbon electrode (SPCE) technology has been used to develop biosensors for determination of biological molecules in applications such as environmental (Soo *et al.*, 2010), biomedical (Rawson *et al.*, 2009), microbiological (Thanyani *et al.*, 2008), healthcare (Zheng *et al.*, 2013), and chemical/biochemical analysis (Yang *et al.*, 2010). Since SPCE is inexpensive and can be used as disposal electrodes with large scale production capability, a number of methods have been devised for increasing the surface area of SPCE to enhance the sensitivity of electrochemical detection. For example, coating with conductive polymer as polypyrrole(PPy) (Oliveira *et al.*, 2012, Wu *et al.*, 2011), polyaniline (PANI) (Chang *et al.*, 2007) and poly (3,4-ethylenedioxythiophene) : poly(styrene-sulfonate) acid (PEDOT:PSS) (Donavan *et al.*, 2011) by electropolymerization processes using potentiodynamic, potentiostatic or galvanostatic modes which can be used in combination with chemical/biological components embedded in their structures. In amperometric biosensors, the enzyme electrodes containing oxidase enzymes can catalyze the substrates such as glucose (Ahmad *et al.*, 2010) , uric acid (Kiran *et al.*, 2012) or lactate (Rawson, 2009), as reducing oxygen to form hydrogen peroxide (H_2O_2) and the oxidation current can be evaluated (Dungchai *et al.*, 2009).

Among the different techniques used to obtain nanostructured surface, electrospinning techniques are alternative methods to produce nanofiber with a large surface area; one to two orders of magnitude higher than those found in continuous films, high porosity and high features to incorporate with active composites such as metal nanoparticle(Liu *et al.*, 2008), graphene (Karuwan *et al.*, 2012) or carbon nanotube (CNT) (Ju *et al.*, 2008, Rujitanaroj *et al.*, 2008) to improve electron transfer activities. CNT is extensively applied in the areas of biosensor due to their unique tubular structure with nano-scale diameter, high conductivity and outstanding mechanical properties (Choi *et al.*, 2010).

Graphene (G) nanoparticles are a flat monolayer of carbon atoms tightly packed into a two-dimensional (2D) honeycomb lattice. They have been widely used in the fabrication of electrochemical biosensors with excellent electrical conductivity (Alwarappan *et al.*, 2009, Li *et al.*, 2012) and mechanical strength (Nataraj *et al.*, 2012), a large surface-to-volume ratio, and appropriate chemical stability (Zhu *et al.*, 2013). G extraordinary properties establish the potential for applications to determine a variety of electro-active species, including dopamine (Kim *et al.*, 2010, Liu *et al.*, 2012), ractopamine (Bai *et al.*, 2014), hydrogen peroxide (Gao *et al.*, 2013, Ruan *et al.*, 2013), and metal ions (Zhang *et al.*, 2013). In addition, G can be integrated into more complex assemblies to form other advanced composites with materials such as gold (Au) (Zhang *et al.*, 2013, Zhong *et al.*, 2013), silver (Ag) (Huang *et al.*, 2014, Zhong, 2013), and platinum (Pt) (Araque *et al.*, 2014, Eremia *et al.*, 2013), increasing its versatility as an advanced electrode material for high performance biosensors (Huang *et al.*, 2014). Gold nanoparticles have several advantages, including faster electron transfer ability, higher surface area, and the ability to link various biofunctional groups using diverse chemical reactions (Luo *et al.*, 2013, Wang *et al.*, 2013).

Carbonized composite nanofibers are also interesting for the fabrication of electrodes because of their wide variety of applications and straightforward production through electrospinning. After stabilization and carbonization, carbonized nanofibers form a ladder structure *via* nitrile polymerization (Nataraj, 2012), and this structure exhibits excellent conductivity and electrochemical activity (Patil *et al.*, 2013). Therefore, Au/G hybrid nanowire inspired this study, and the composite could successfully enable the development of more complicated electrochemical activity for biosensing materials.

Vapor-phase polymerization (VPP) has been used to synthesize and optimize intrinsically conducting polymers (ICPs). The substrates are coated with oxidizing agents such as $FeCl_3$ (Jang *et al.*, 2009), Fe(III) *p*-toluenesulfonate (FeTos) (Laforgue *et al.*, 2010), benzenesulfonic acid, *p*-dodecylbenzenesulfonic acid (DBSA), *p*-ethylbenzenesulfonic acid (Subramanian *et al.*, 2008) by simple coating methods. Then, oxidant-coated substrates were exposed to monomer of ICPs. When the monomer vapors evaporated at the place where oxidant was coated, they were rapidly polymerized to form a conductive thin film formation on substrate surface. VPP method has been reported by a number of scientists revealing the excellent electronic conductivity properties on various material surfaces. Herein as present work, VPP was chosen for coating PPy layer on electrospun nanofiber.

In this work, we have used the new technique and a novel materials to enhance the sensitivity and selectivity of disposable electrode in order to determine the bio-molecule substances such as glucose and dopamine in biological samples. All results show potential promise for the fabrication of a practical biosensor.

Scope of Research Work

Research Work 1: Modification of Disposable Screen-Printed Carbon Electrode Surfaces with Conductive Electrospun Nanofiber for Biosensor Applications

The aim of this study is to modify an SCPE surface through the preparation of a PPy/PAN-MWCNT/SPCE electrode with a PPy coating on CNT-embedded PAN electrospun nanofibers, which is key in the enhancement of the electrode electrochemical performance. First, the redox behaviors of the unmodified/modified electrode were studied in a ferri/ferrocyanide solution to optimize the electrode surface modification. Second, the H_2O_2 detection of the modified electrode and any interference were studied. Glucose detection was also studied; glucose oxidase was used as a representative enzyme without and with the addition of a mediator. The performance of PPy/PAN-MWCNT/SPCE electrodes with respect to their sensitivity, detection limits, and calibration curves are described and discussed.

Research Work 2: Development of a Disposable Electrode Modified with Carbonized, Electrospun, Polyacrylonitrile-loaded Graphene Nanoparticles for the Detection of Dopamine in Human Serum

This research aimed to develop a simple method to modify a SPCE using a combination of electrospinning and carbonization of composite nanofibers to enhance electrochemical performance. Sodium dodecyl sulfate (SDS) was also used to further improve DA detection. These modifications provide several advantages, including a small sample volume; low cost; high selectivity for DA due to electrostatic interactions with the negatively charged SDS; and promise for large-scale production. The redox responses of unmodified and modified electrodes were evaluated in ferri/ferrocyanide solution using cyclic voltammetry (CV). Square-wave voltammetry (SWV) was used to detect DA with various modified electrodes in phosphate-buffered saline (PBS). Finally, DA content was quantified in the presence of typical interfering molecules (AA and UA), as well as in real human serum.

Research Work 3: Novel Carbonized, Gold/Graphene Hybrid Nanowire-Modified Disposable Electrode for the Ultrasensitive and Selective Detection of Dopamine in Biological Samples

In this study, we described a dopamine biosensor based on carbonized hybrid Au/G nanowire (CPAN5G-Au), which was fabricated by a combination of electrospinning and carbonization processes. Characterization of the basic materials and electrochemical behavior of the carbonized products were systematically studied. The CPAN5G-Au modifications were constructed on a SPCE surface. CPAN5G-Au showed an excellent differential pulse voltammetry (DPV) response to DA in the presence of typical interfering molecules (AA and UA) in 0.1 M phosphate-buffered saline (PBS) at pH 7.4 and in human serum without a discriminating agent. The results show great promise for the fabrication of a practical DA biosensor.

Research Work 4: Development of A Novel Anti-tuberculosis Melt-Blown Polypropylene Filters Coated with Mangosteen Extracts for use as Medical Face Mask Applications

The objectives of the present study were to modify the surface of low cost commercial polypropylene melt-blown filter with various concentrations of mangosteen extract by spray coating. Physical properties of filters were studied. Bacterial filtration efficacy (BFE) and pressure drop were performed by NELSON Laboratory. Further, the antibacterial activities of coated filter against Gram-positive bacteria *Staphylococcus aureus*, Gram-negative bacteria *Escherichia coli* and MDR-TB were evaluated quantitatively as a function of exposure times.