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DEVELOPMENT OF ELECTROCHEMICAL IMMUNOSENSOR-BASED POLY(3,4-ETHYLENEDIOXYTHIOPHENE) COMPOSITES FOR CLENBUTEROL DETECTION

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By

NURUL AIN A. TALIB

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

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Chairman : Associate Professor Yusran Bin Sulaiman, PhD
Faculty : Institute of Advance Technology

Illegal usage of banned antibiotic such as clenbuterol (CLB) in food products is a big concern since this will directly affect the consumer health. World Health Organization (WHO) is forbidding any usage of CLB in the livestock animals due to health effects such as muscular tremor, increase rate of heart throb, glaucoma, fever and respiratory problems to human that influenced by CLB residue in food products. Currently, the methods used for CLB detection is expensive, time-consuming and involving complicated analysis. In this study, immunosensor modified with poly(3,4-ethylenedioxythiophene)/multi-walled carbon nanotube (PEDOT/MWCNT), poly(3,4-ethylenedioxythiophene)/graphene oxide (PEDOT/GO) and anti-clenbuterol antibody (Ab) were developed on screen-printed carbon electrode (SPCE) for detection of CLB. Sensor platforms from modification of electrode with PEDOT/MWCNT and PEDOT/GO composites were prepared by electropolymerization using chronoamperometry (CA) technique. The operating conditions (concentration of MWCNT, concentration of GO, electropolymerization potential and deposition time) were optimized by using response surface methodology (RSM) combined with central composite design (CCD) and Box-Behnken design (BBD) for PEDOT/MWCNT and PEDOT/GO composites, respectively to obtain the optimum peak current. The statistical analysis showed that the concentration of MWCNT, concentration of GO, electropolymerization potential and deposition time have significantly affected the peak current response. The coefficient of determination (R^2) for model equations of PEDOT/MWCNT and PEDOT/GO composites resulting value of 0.9973 and 0.9965, respectively. The optimized condition predicted by the software was compared with the experiments and resulting in less than 2% error, indicating that this model was reliable and able to predict the peak current response accurately. The cyclic voltammetry (CV) measurements indicated that PEDOT/MWCNT and PEDOT/GO modified electrodes had successfully enhanced the peak currents compared to PEDOT, MWCNT and GO. Incorporation of MWCNT and GO into PEDOT were proven by field emission scanning electron microscopy (FESEM) images, Fourier transform infrared

(FTIR) and Raman spectra. The optimized PEDOT/MWCNT and PEDOT/GO composites were further modified with Ab on SPCE to develop CLB immunosensors. Detection of CLB was performed through direct competitive format, whereby the CLB in sample solutions were competing with CLB conjugated with horseradish peroxide (CLB-HRP) to bind with Ab. The change in current value was analyzed through CA for quantification of CLB amount in the sample. The immunoassay conditions for these immunosensors were optimized by using RSM/CCD, whereby the incubation temperature, Ag incubation time and %blocking were determined as significant parameters. The resulting immunosensors exhibited excellent reproducibility with low standard deviation (SD) value. These immunosensors also very selective towards CLB in comparison with other antibiotics from same family group (β-agonist) and another group of antibiotics. Based on storage stability study, these immunosensors can retain its performance up to 95% after a month storage at 4 °C. Thus, highly reproducible, sensitive and stable immunosensors for detection of CLB in the real samples were developed and satisfactorily meet the requirement for actual application.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Ijazah Doktor Falsafah

PEMBANGUNAN PENDERIA IMUNO ELEKTROKIMIA BERASASKAN KOMPOSIT POLI(3,4-ETILENADIOKSITIOFENA) UNTUK PENGESANAN "*CLENBUTEROL*"

Oleh

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Julai 2018

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Penyalahgunaan antibiotik terlarang seperti *"clenbuterol"* (CLB) di dalam produk makanan amat membimbangkan memandangkan ini akan memberikan kesan terus terhadap kesihatan pengguna. Pertubuhan Kesihatan Sedunia (WHO) melarang apa-apa pengunaan CLB dalam ternakan haiwan berikutan kesannya terhadap kesihatan seperti getaran pada otot, peningkatan kadar degupan jantung, glaukoma, demam dan masalah pernafasan kepada manusia yang terkesan dengan baki CLB dalam produk makanan. Pada masa ini, kaedah yang digunakan untuk pengesanan CLB adalah mahal, memakan masa and melibatkan analisis yang rumit. Dalam pengajian ini, penderia imuno terubahsuai dengan poli(3,4-etilenadioksitiofena)/nanotiub karbon berbilang dinding (PEDOT/MWCNT), poli(3,4-etilenadioksitiofena)/grafin oksida (PEDOT/GO) dan antibodi anti-*"clenbuterol"* (Ab) telah dibangunkan di atas elektrod skrin tercetak karbon (SPCE) untuk pengesanan CLB. Platfom penderia daripada pengubahsuaian elektrod dengan komposit PEDOT/MWCNT dan PEDOT/GO telah disediakan menggunakan teknik kronoamperometri (CA) melalui pengelektropolimeran. Kondisi operasi (kepekatan MWCNT, kepekatan GO, keupayaan pengelektropolimeran dan tempoh pemendapan) dioptimumkan dengan menggunakan kaedah respon permukaan (RSM) kombinasi bersama reka bentuk komposit berpusat (CCD) dan reka bentuk Box-Behnken (BBD) masing-masing untuk PEDOT/MWCNT dan PEDOT/GO komposit bagi memperolehi puncak arus yang optimum. Analisis statistik menunjukkan kepekatan MWCNT, kepekatan GO, keupayaan pengelektropolimeran dan tempoh pemendapan mempunyai kesan yang ketara terhadap puncak arus. Pekali penentuan (R^2) persamaan model bagi PEDOT/MWCNT dan PEDOT/GO adalah masing-masing 0.9973 dan 0.9965. Kondisi optimum yang diramalkan oleh perisian telah dibandingkan dengan keputusan eksperimen dan menghasilkan keputusan ralat kurang daripada 2%, menunjukkan model ini boleh dipercayai dan mampu untuk meramal respon puncak arus dengan tepat. Pengukuran menggunakan voltammetri berkitar menunjukkan bahawa elektrod termodifikasi PEDOT/MWCNT dan PEDOT/GO berjaya meningkatkan puncak arus berbanding PEDOT, MWCNT and GO.

Penggabungan MWCNT dan GO ke dalam PEDOT telah dibuktikan oleh imej mikroskopi elektron pengimbasan pancaran medan (FESEM), infra merah transformasi Fourier (FTIR) dan Raman spektrum. Seterusnya, PEDOT/MWCNT dan PEDOT/GO yang telah dioptimumkan telah dimodifikasikan pula dengan Ab di atas SPCE untuk dibangunkan menjadi penderia imuno CLB. Pengesanan CLB dijayakan melalui format kompetitif lansung, di mana CLB di dalam larutan sampel bersaing dengan CLB yang telah dikonjugasikan dengan horseradish peroksida (CLB-HRP) untuk membentuk ikatan dengan Ab. Perubahan terhadap nilai arus dianalisis menggunakan CA untuk pengkuantitian jumlah CLB di dalam sampel. Kondisi imunoasai untuk penderia imuno ini telah dioptimumkan dengan menggunakan RSM/CCD, di mana suhu inkubasi, tempoh inkubasi antigen (Ag) dan peratusan penghalang telah dikenalpasti sebagai parameter yang ketara. Penderia imuno yang dihasilkan telah mempamerkan ciri-ciri kebolehulangan yang cemerlang dan nilai sisihan piawai (SD) yang rendah. Penderia imuno ini juga sangat selektif terhadap CLB jika dibandingkan dengan antibiotik lain daripada kumpulan keluarga yang sama (agonis beta) dan kumpulan yang berbeza. Berdasarkan kajian kestabilan penyimpanan, penderia imuno ini boleh mengekalkan prestasinya sehingga 95% selepas disimpan pada 4 °C selama sebulan. Oleh itu, penderia imuno yang berkebolehulangan, sensitif serta berkestabilan tinggi untuk pengesanan CLB dalam sampel sebenar telah dibangunkan dan memenuhi keperluan yang memuaskan untuk aplikasi sebenar.

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I certify that a Thesis Examination Committee has met on 23 July 2018 to conduct the final examination of Nurul Ain Binti A. Talib on her thesis entitled "Development of Electrochemical Immunosensor-Based Poly(3,4-Ethylenedioxythiophene) Composites for Clenbuterol Detection" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctoral of Philosophy.

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This is to confirm that:

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2D	two dimensional
3D	three dimensional
Ab	antibody
AC	alternating current
ACN	acetonitrile
Ag	antigen
ANOVA	analysis of variance
ATR	Attenuated total reflection
AuNP	gold nanoparticle
BBD	Box-Behnken design
BCA	bicinchoninic acid
BSA	bovine serum albumin
C.V.	coefficient of variation
CA	Chronoamperometry
CAC	Codex Alimentarius Commission
CCD	central composite design
CCRD	central composite rotatable design
CE	counter or auxiliary electrode
CLB	clenbuterol
CNT	carbon nanotube
CV	cyclic voltammetry
DD	Doehlert design
DF	degree of freedom
DI	deionized
DLLME	dispersive liquid-liquid microextraction
DNA	deoxyribonucleic acid
DOE	design of experiment
DPV	different pulse voltammetry
EDC	N-(3-dimethylaminopropyl)-N'-ethylcarbodiimide hydrochloride
EDOT	3,4-Ethylenedioxythiophene
EIS	electrochemical impedance spectroscopy
ELISA	enzymes-linked immunosorbent assay
EU	European union
FCA	Freund's complete adjuvant
FESEM	field emission scanning electron microscopy
FIA	Freund's incomplete adjuvant
FTIR	Fourier transform infrared
GO	graphene oxide
GC-MS	gas chromatography-mass spectrometry
HPLC	high-performance liquid chromatography
HRP	horse radish peroxidase
Ig	immunoglobulin
IgG-AP	alkaline phosphate conjugated goat affinity purified antibody to rabbit immunoglobulin G
IMS	ion mobility spectrometry
ITO	indium tin oxide
IUPAC	International Union of Pure and Applied Chemist
KLH	keyhole limpet hemocyanin

LC-MS	liquid chromatography-mass spectrometry
LC-TOFMS	liquid chromatography electrospray time-of-flight mass spectrometry
LOD	limit of detection
LSV	linear-sweep voltammetry
MIP	molecular imprinted polymer
MRL	maximum residue limit
MSe	error mean square
MWCNT	multi-walled carbon nanotube
NHSS	N-hydroxysulfosuccinimide
OLEDs	organic light-emitting diodes
OVA	ovalbumin
P(mPD)	poly(m-phenylenediamine)
p-NPP	4-nitrophenyl phosphate disodium salt hexahydrate
PANI	polyaniline
PBS	phosphate buffer solution
PEDOT	poly(3,4-ethylenedioxythiophene)
PPy	polypyrrole
PRESS	prediction error sum of squares
QCM	quartz crystal microbalance
R^2	regression coefficient
R_{ct}	charge transfer resistance
RE	reference electrode
rGO	reduced GO
RNA	ribonucleic acid
RSE	residual standard errors
RSM	response surface methodology
SD	standard deviation
SDS-PAGE	sodium dodecyl sulfate-polyacrylamide gel electrophoresis
SERS	surface-enhanced Raman scattering
SPCE	screen-printed carbon electrode
SPE	screen-printed electrode
SPR	surface plasmon resonance
SWCNT	single-walled carbon nanotube
SWV	square-wave voltammetry
TEMED	1,2-di-(dimethylamino)ethane
TMB	3,3′,5,5′-tetramethylbenzidine
tris HCl	tris(hydroxymethyl)aminomethane hydrochloride
UCP	up conversion phosphor
UHT	ultra heat treatment
WE	working electrode
WHO	World Health Organization

CHAPTER 1

INTRODUCTION

1.1 Background of study

There Antibiotics play a major role in the treatment of diseases for both humans and animals. However, the long withdrawal period of antibiotics may cause contamination of the food products based on the livestock animals due to the antibiotic residues. Some of the antibiotics have the properties to increase the leaner meat productions and milk collection rate. The misuse of the antibiotics has caused various health issues such as food poisoning, nausea, antibiotics resistant, fever and other health problems.

Clenbuterol (CLB), an antibiotic from β–agonist family, is one of the common antibiotics that is illegally introduced to the livestock animal to boost the meat and milk production (Elliott *et al.*, 1995;He *et al.*, 2009a;Li *et al.*, 2014a;Ma *et al.*, 2018;Xue and Zhang, 2013). Therefore, regular screening and monitoring of antibiotic residue in the livestock animal products by a regulatory agency is needed to control the food quality and improve the consumer health life. The analysis of trace substances in environmental science, pharmaceutical and food industries is a big challenge since many of these applications require a continuous monitoring mode. Various techniques have been developed for this purpose such as microbial inhibition assay (Pikkemaat, 2009), chromatographic methods (Lefeuvre *et al.*, 2017) and biosensors (Chen *et al.*, 2017).

Among the available methods, biosensor has emerged as the most appropriate new type of analytical instruments for these applications due to the simplicity and rapid detection offered by this method. Biosensors are one of the methods that are suitable for rapid detection of various biological compounds including antibiotics. Antibiotics such as CLB is a hapten, which is very small in size and is able to produce only small signal thus very challenging for the development of detection methods. Usually, immunological analytical methods can offer high sensitivity and good specificity (Chen *et al.*, 2017). Immunosensor (a type of biosensor) for detection of CLB residue in real samples is proposed to overcome these issues by utilizing various sensing strategies. Immunosensors are compact analytical devices in which the event of formation of antigen-antibody complexes is detected and converted by means of a transducer, to an electrical signal, which can be processed, recorded and displayed (Moina and Ybarra, 2012). The fundamental of immunosensor is the specificity of the molecular recognition of antigens by antibodies to form a stable complex. Selection of high-performance bio-recognition element such as highly specific antibody has a major effect on the development of high sensitivity and high selectivity immunosensor. The performance of the immunosensor also closely related to the transducer design. In electrochemical immunosensor, excellent electrochemical properties of the electrode are required to be used as the sensor platform (transducer) thus convert the signal to electrical current. The performance of the sensor platform can be enhanced through modification of the electrode with conducting materials to make them suitable for the development of immunosensors.

Conducting polymers and its composites are commonly used in the modification of electrode. Poly(3,4-ethylenedioxythiophene) (PEDOT) is one of the conducting polymers derived from polythiophene that is still being explored for the application in the sensor technology. The utilization of PEDOT in electrode modification has a high potential for the development of a sensor platform with improved electrochemical properties that will contribute to increasing the electrochemical performance of immunosensor. However, PEDOT needs to be doped with other materials in order to maximize its performance. Carbon materials such as multi-walled carbon nanotube (MWCNT) and graphene oxide (GO) are excellent dopants due to their excellent conductivity properties. Physical properties such as high surface area of CNT and GO will allow these materials to become a supporting matrix for high density protein attachment such as for antibody attachment and at the same time may confer good conductivity for electrochemical detection (Hu et al., 2011). CNT and GO can also act as good filler in the formation of conducting polymer composite to increase the electrochemical performance of the modified electrodes (Alvi *et al.*, 2011;Peng *et al.*, 2007). Therefore, in this study conducting polymer composites derived from PEDOT, MWCNT and GO have high potential to be developed into high-performance electrochemical immunosensors.

1.2 Statement of problems

Contamination of meat-based food products from antibiotics residue is a serious worldwide issue. The concern regarding the safety of food products has grown among consumers due to bad health effect reported regarding this issue. Contamination from antibiotics might occur due to the early slaughtering of the livestock animal, which is before the antibiotics withdrawal period end or misuse of the drugs as a growth promoter. Antibiotics such as CLB is an illegal steroid for livestock animal, but the use of this drug in livestock animal is still being reported. Therefore, constant monitoring and screening of CLB contamination in the animal-based product are very crucial. Detection of CLB based on chromatographic methods such as liquid chromatography-mass spectrometry (LC-MS), gas chromatography-mass spectrometry (GC-MS) and high-performance liquid chromatography (HPLC) can offer high sensitivity and high selectivity. However, these methods are expensive and it involves complicated operating procedures that requires only highly skilled person to handle the spacious instruments. Therefore, simple and low cost method such as electrochemical sensor that is possible to be developed up to point-of-care is a good alternative. It is more practical to be used in CLB screening and monitoring, before proceeding to the confirmatory test by chromatographic methods.

The performance of an electrochemical immunosensor is closely related to the electrochemical performance of the sensor platform. Currently, only a few electrochemical immunosensors have been developed to detect CLB. Due to its small molecular size, CLB has less binding sites hence it is difficult to be immobilized and it also has less effect on the electron transfer of the electrochemical mediator in solution. PEDOT/MWCNT and PEDOT/GO composites have high potential to improve the charge transfer due to the high conductivity possess by PEDOT, MWCNT and GO, thus enhancing the immunosensors performance. Coupling of antibody to the PEDOT/MWCNT and PEDOT/GO modified electrode may not only allow specific binding but it will also increase the sensitivity of the immunosensor. Modification of electrode surface with PEDOT, MWCNT and GO are believed to give major effect on electron transfer of fabricated immunosensors. Development of immunosensors based on this polymer composite may not only contribute in sensor field but also to environmental science, pharmaceutical and food industries. Therefore, this research would be the basis for the future findings and developments of sensor technology.

1.3 Objectives of study

The aim of this study is to develop the electrochemical immunosensors for detection of CLB using PEDOT/MWCNT and PEDOT/GO modified electrodes. This research also provides models for correlating the critical parameters optimization (*i.e.* the concentration of MWCNT, concentration of GO, electrodeposition potential, deposition time, pH, antibody incubation temperature, antigen incubation time and %blocking) for peak currents of the modified electrodes and current signal of the immunoassay. The objectives of this research are:

1. To produce and characterize polyclonal anti-CLB antibody as the bio-receptor for the immunoassay.
2. To prepare and characterize PEDOT/MWCNT and PEDOT/GO modified electrodes as the sensor platforms by using chronoamperometry (CA).
3. To model and analyze the electrochemical performance of the sensor platforms and immunoassay using response surface methodology (RSM) and optimize the operating parameters.
4. To develop PEDOT/MWCNT and PEDOT/GO sensor platforms for detection of CLB and evaluate the performance of the immunosensors.

1.4 General overview of the thesis

Modification and optimization of electrodes based on PEDOT/MWCNT and PEDOT/GO composites for development of CLB immunosensors are presented in this thesis. Background and objectives of this research are introduced in Chapter 1. The production of antibody, electrodes modification, immunosensor design, detection techniques and experimental design for optimization are reviewed in Chapter 2.

Production of polyclonal anti-CLB antibody is reported in Chapter 3. The immunization protocol and the characterization of antibody produced are described here. Evaluation of this antibody as bio-receptor is also included in this chapter. Modification of electrode with PEDOT/MWCNT composite is reported in Chapter 4. Details of experimental design used to optimize the operating parameters in order to predict the optimum electropolymerization condition are described. The characterizations of PEDOT/MWCNT modified electrode are detailed in this chapter. Chapter 5 describes the development of CLB immunosensor based on PEDOT/MWCNT composite. The optimum conditions for electrode modification with PEDOT/MWCNT are applied to fabricate sensor platform on screen-printed carbon electrode (SPCE). The SPCE/PEDOT/MWCNT is further modified with the antibody and the electrochemical assay performed is discussed in this chapter. The optimization of the immunoassay performed by utilizing response surface methodology/central composite design (RSM/CCD) method is detailed along with the characterization and sensing performance of the immunosensor.

Electropolymerization of PEDOT/GO composite on the electrode surface is presented in Chapter 6. The optimization of electrodeposition conditions performed by using response surface methodology/Box-Behnken design (RSM/BBD) is explained and the optimum condition predicted is reported along with characterizations of the optimized modified electrode. Another CLB immunosensor developed from modification of SPCE with PEDOT/GO composite is described in Chapter 7. The previous optimized condition for PEDOT/GO electrode modification (Chapter 6) and the immunoassay conditions (Chapter 5) is applied for this immunosensor. The sensing performance and the characterization of this immunosensor are reported in this chapter. Finally, the overall findings of this study are summarized in Chapter 8 and recommendations for future study are suggested.

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