PREPARATION, CHARACTERIZATION AND APPLICATIONS OF GRAPHENE OXIDE AND OIL-PALM ACTIVATED CARBON MODIFIED MAGNETIC-POLYPYRROLE SORBENTS FOR MICROEXTRACTION OF SELECTED ORGANIC POLLUTANTS

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ABSTRACT

Magnetite (Fe_3O_4) and polypyrrole (PPy) are widely used in the electrical industry. However, their uses as sorbents in analytical chemistry applications are scarce. Thus, this research investigates the preparation, development and applications of Fe_3O_4-PPy composites as sorbents for microextraction of different emerging and persistent occurring pollutants optimized with the aid of response surface methodology (RSM) approach. In the first part of study, Fe_3O_4-PPy sorbent was applied in magnetic solid phase extraction (MSPE) combined with high-performance liquid chromatography-ultraviolet detection (HPLC-UV) for the analysis of three selected non-steroidal anti-inflammatory drugs (NSAIDs), namely naproxen, diclofenac and mefenamic acid. Systematic investigations on the synthesis and Fe_3O_4-PPy MSPE efficiencies for the NSAIDs were successfully modelled using the Box-Behnken Design (BBD) (R^2 = 0.94-0.98, p < 0.001%). A comparison of the use of RSM and one-variable-at-a-time approach showed more positive implications of the former in terms of order of factor significance and result prediction. Optimized conditions for the procedure gave low limits of detection (LOD = 3s, 0.9-3.5 μg L^{-1}) and limits of quantitation (LOQ = 10s, 2.9-11.5 μg L^{-1}) with good reproducibility (relative standard deviation, RSD < 7.2%) and excellent recoveries (97.9 – 100.5%) for tap water, river water, and wastewater samples. In order to enhance the surface ability of the sorbent, Fe_3O_4-PPy was incorporated with graphene oxide, (GO) to produce GO-Fe_3O_4-PPy in the second part of study. The synthesized GO-Fe_3O_4-PPy composite was applied in MSPE of three selected neonicotinoids namely imidacloprid, thiacloprid and thiamethoxam, in water samples using HPLC-UV detection. Experimental results obtained with aid of RSM-BBD were in good agreement (97%) with the model predictions. The surface morphology of GO-Fe_3O_4-PPy showed that the synthesized Fe_3O_4 was embedded within the slits of GO and that PPy sealed the GO-Fe_3O_4 surface to produce an effective sorbent for the extraction of neonicotinoids in water matrices. Analysis showed excellent LOD and LOQ (LOD = 3s, 0.026- 0.045 μg L^{-1}; LOQ = 10s, 0.087- 0.149 μg L^{-1}) with good recoveries of 93-102%. In the quest for a rapid analytical extraction method, the third part of study was performed where oil-palm activated carbon (OPAC) was incorporated with Fe_3O_4-PPy to produce OPAC-Fe_3O_4-PPy for MSPE of two selected organochlorine pesticides (OCPs) namely endosulfan (ESO) and dieldrin (DIE) in aquatic samples. Analysis was performed using gas chromatography with microelectron capture detection. The effects of three preparation variables, namely ratio of Fe_3O_4:OPAC, amount of pyrrole monomer, and amount of $FeCl_3$ oxidant were optimized successfully using RSM-BBD (R^2 < 0.99, p < 0.001%). RSM-BBD was also used for the optimization of four numerical parameters in MSPE. The significance of MSPE parameters were salt addition > pH sample solution > extraction time > desorption time. The OPAC-Fe_3O_4-PPy MSPE method demonstrated good linearity (25-1000 ng L^{-1}) with good correlation (R^2 > 0.991) and low LODs and LOQs (LOD = 3S/N, 6.5 ng L^{-1} for ESO and 7.3 ng L^{-1} for DIE; LOQ = 10S/N, 25 ng L^{-1} for ESO and DIE) with high recoveries (98-105%) of OCPs from tap water, and palm oil effluent samples. A comparative study showed that the incorporation of Fe_3O_4 and PPy separately to GO and OPAC, showed different behaviours in accordance to the particle size and surface chemistry. The adsorption capacities of the sorbents, represented by q_m is in the order of GO-Fe_3O_4-PPy > OPAC-Fe_3O_4-PPy > Fe_3O_4-PPy. Thus this study proved that the adsorption strength of sorbents could be reinforced by the presence of OPAC and GO. Analytical Eco-Scale analysis showed that all the developed methods achieved acceptable green analysis scale (AES value: 77-81). The proposed methods offered good features such as sustainable, simple, and rapid extraction that meet the green chemistry concept.

ABSTRAK

Magnetit (Fe_3O_4) dan polipirol (PPy) digunakan dengan meluas dalam industri elektrik. Walau bagaimanapun, penggunaannya sebagai penjerap dalam aplikasi kimia analisis masih tidak banyak. Oleh itu, penyelidikan ini mengkaji penyediaan, pembangunan dan penggunaan komposit berasaskan Fe_3O_4-PPy sebagai penjerap untuk pengekstrakan mikro bahan pencemar baharu dan kekal berterusan yang berbeza dengan berbantukan pendekatan metodologi tindak balas permukaan (RSM). Dalam bahagian pertama kajian, penjerap Fe_3O_4-PPy telah digunakan dalam pengekstrakan fasa pepejal bermagnet (MSPE) yang digabungkan dengan kromatografi cecair berprestasi tinggi dengan pengesan ultra lembayung (HPLC-UV) untuk analisis tiga dadah anti-radang bukan steroid (NSAID) yang terpilih iaitu naproksen, diklofenak dan asid mefenamik. Penyelidikan sistematik ke atas sintesis dan kecekapan Fe_3O_4-PPy MSPE bagi NSAID telah berjaya dimodelkan menggunakan reka bentuk Box-Behnken (BBD) (R^2 = 0.94-0.98, p < 0.001%). Perbandingan penggunaan RSM dan pendekatan satu pemboleh ubah pada satu masa menunjukkan implikasi positif bagi RSM dari segi susunan faktor signifikan dan ramalan hasil. Keadaan optimum prosedur memberi had pengesanan (LOD = 3s, 0.9-3.5 μg L^{-1}) dan had kuantitatif (LOQ = 10s, 2.9-11.5 μg L^{-1}) yang rendah dengan kebolehulangan yang baik (< 7.2% sisihan piawai relatif, RSD) serta had pemulihan yang sangat baik (97.9-100.5%) bagi air paip, air sungai, dan sampel sisa air buangan. Untuk meningkatkan kemampuan permukaan penjerap, Fe_3O_4-PPy telah digabungkan dengan grafin oksida (GO) dalam bahagian kedua kajian untuk menghasilkan GO-Fe_3O_4-PPy. Komposit GO-Fe_3O_4-PPy yang disintesis digunakan untuk MSPE tiga neonikotinoid terpilih iaitu imidakloprid, tiakloprid dan tiametoksam di dalam sampel air menggunakan analisis HPLC-UV. Hasil eksperimen berbantukan RSM-BBD yang diperoleh sangat bertepatan dengan model ramalan (97%). Morfologi permukaan GO-Fe_3O_4-PPy menunjukkan bahawa Fe_3O_4 yang disintesis tertanam antara lapisan GO dan PPy menutup permukaan GO-Fe_3O_4 untuk menghasilkan penjerap yang berkesan dalam pengekstrakan neonikotinoid dari matriks air. Analisis menunjukkan LOD dan LOQ yang sangat baik (LOD = 3s, 0.026-0.045 μg L^{-1}; LOQ =10s, 0.087-0.149 μg L^{-1}) serta had pemulihan yang tinggi (93-102%). Dalam usaha mencari kaedah pengekstrakan analisis yang pantas, kajian ketiga dijalankan dengan karbon teraktif kelapa sawit (OPAC) yang digabungkan dengan Fe_3O_4-PPy bagi menghasilkan OPAC-Fe_3O_4-PPy untuk digunakan dalam MSPE dua racun perosak organoklorin terpilih (OCP) iaitu endosulfan (ESO) dan dieldrin (DIE) di dalam sampel akueus. Analisis dijalankan menggunakan kromatografi gas dengan pengesan penangkapan mikroelektron. Kesan tiga pembolehubah penyediaan penjerap, iaitu nisbah Fe_3O_4:OPAC, jumlah monomer pirol, dan jumlah oksidan $FeCl_3$ telah dioptimumkan dengan jayanya menggunakan RSM-BBD (R^2 < 0.99, p < 0.001%). RSM-BBD juga digunakan untuk pengoptimuman empat parameter numerik dalam MSPE. Tahap signifikan parameter MSPE adalah penambahan garam > pH larutan sampel > masa pengekstrakan dan masa nyahjerap. Kaedah OPAC-Fe_3O_4-PPy MSPE menunjukkan kelinearan yang baik (25-1000 ng L^{-1}) dengan korelasi yang baik (R^2 > 0.991) dan LOD dan LOQ yang rendah (LOD = 3S/N, 6.5 ng L^{-1} untuk ESO dan 7.3 ng L^{-1} untuk DIE; LOQ = 10S/N, 25 ng L^{-1} untuk ESO dan DIE) dan had pengembalian semula OCP yang tinggi (98-105%) dari sampel air paip dan efluen sawit terawat. Kajian perbandingan penambahan kedua-dua Fe_3O_4 dan PPy kepada GO dan OPAC secara berasingan menunjukkan perbezaan kelakuan selaras dengan saiz zarah dan kimia permukaan. Kapasiti penjerapan penjerap, yang diwakili oleh q_m adalah GO-Fe_3O_4-PPy > OPAC-Fe_3O_4-PPy > Fe_3O_4-PPy. Kajian ini membuktikan bahawa kekuatan penjerapan dapat diperkukuhkan dengan kehadiran OPAC dan GO. Analisis skala eko-analisis menunjukkan bahawa kesemua kaedah yang dibangunkan mencapai skala analisis hijau (nilai AES: 77-81). Kaedah yang dicadangkan ini menawarkan ciri yang baik seperti mampan, mudah dan pantas serta memenuhi konsep kimia hijau.

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LIST OF ABBREVIATIONS

ACN	-	Acetonitrile
AES	-	Analytical Eco Scale
ANOVA	-	Analysis of variance
ATR	-	Attenuated total reflection
BBD	-	Box Behnken design
BET	-	Brunauer-Emmett-Teller
C_{18}	-	Octadecylsilica
CCD	-	Central composite design
CFME	-	Composite film microextraction
CH_3COONa	-	Sodium acetate anhydrous
CNT	-	Carbon nanotubes
CTABr	-	Cetyltrimethylammonium bromide
DAD	-	Diode array detector
DIC	-	Diclofenac
DIE	-	Dieldrin
DM	-	Doehlert matrix
DoE	-	Design of experiments
DSPE	-	Dispersive solid phase extraction
DX7	-	Design Expert 7 software
ECD	-	Electron Capture Detector
EDAX	-	Energy dispersive analysis of X-rays
EFB	-	Empty fruit bunches
EME	-	Electromembrane extraction
ESO	-	Endosulfan
EtOH	-	Ethanol
FESEM	-	Field emission scanning electron microscopy
FTIR	-	Fourier transform infra-red
G	-	Graphene
GC	-	Gas chromatography
GC-MS	-	Gas chromatography mass spectrometry

GO	-	Graphene oxide
HLB	-	Hydrophilic–lipophilic balance
HPLC	-	High performance liquid chromatography
I.D	-	Internal diameter
IMI	-	Imidacloprid
IPA	-	Iso-propanol
LC	-	Liquid chromatography
LC-EI/MS	-	Liquid chromatography electrospray ionization mass spectrometry
LC-MS	-	Liquid chromatography mass spectrometry
LLE	-	Liquid-liquid extraction
LOD	-	Limit of detection
LOQ	-	Limit of quantification
MAA	-	Methyl acryl acid
MEF	-	Mefenamic acid
MeOH	-	Methanol
MISPE		Molecular imprinted solid phase extraction
MNP	-	Magnetic nanoparticle
MOF	-	Molecular orbital framework
MS	-	Mass spectrometry
MS/MS	-	Tandem mass spectrometry
MSPE	-	Magnetic solid phase extraction
MWCNT	-	Multi walled carbon nanotubes
NAP	-	Naproxen
NMR	-	Nuclear magnetic resonance
NSAIDs	-	Non-steroidal anti-inflammatory drugs
OCPs	-	Organochlorine pesticides
ODS	-	Octadecyl-silane
OPAC	-	Oil palm activated carbon
OPEFB	-	Oil palm empty fruit bunch
OVAAT	-	One variable at a time
PDF	-	Powder diffraction file
POPs	-	Persistent organic pollutants

PPy	-	Polypyrrole
RSM	-	Response surface methodology
S/N	-	Signal to noise ratio
SEM	-	Scanning electron microscopy
SPE	-	Solid phase extraction
SPME	-	Solid phase microextraction
TEM	-	Transmission electron microscopy
TGA	-	Thermogravimetric analysis
TIC	-	Thiacloprid
TIM	-	Thiamethoxam
UHPLC	-	Ultra high performance liquid chromatography
UV	-	Ultraviolet
VSM	-	Vibrating sample magnetometry
XPS	-	X-ray photoelectron spectroscopy
XRD	-	X-ray diffraction

LIST OF SYMBOLS

°C	-	Degree Celsius
Å	-	Armstrong unit
C_o		Initial concentration
C_e		Experimental concentration
C_f		Final concentration
cm^{-1}	-	Wave length
cm^3/g	-	Centimetres cubic per gram
$g\ L^{-1}$	-	Gram per liter
$K_{o/w}$	-	Coefficient ratio of solubility octanol:water
K_{P1}		Pseudo first order rate of adsorption
K_{P2}		Pseudo second order rate of adsorption
k_T		Temkin rate of adsorption
$m^2\ g^{-1}$	-	Meter square per gram
$mg\ kg^{-1}$	-	Miligram per kilogram
min	-	Minute
mL	-	Mililiter
mm	-	Milimeter
mM	-	Milimolar
$ng\ L^{-1}$	-	Nanogram per litre
nm	-	Nanometer
P/P^o	-	Relative vapor pressure initial-final
pK_a	-	Acid dissociation constant
q_e		Adsorption capacity
$q_{e(cal)}$		Calculated adsorption capacity
$q_{e(exp)}$		Experimental adsorption capacity
q_{max}		Maximum adsorption capacity
q_t		Adsorption capacity at time, t
r	-	Correlation coefficients
R^2		Coefficient of determination
R_E		Elovich distribution equilibrium parameter

R_L		Langmuir separation factor
rpm	-	Rate per minute
RSD	-	Relative standard deviation
v		Volume
w		Weight
w/v	-	Weight over volume
Δq		Normalized standard deviation
$\mu g\ g^{-1}$		Microgram per gram
$\mu g\ L^{-1}$	-	Microgram per litre
$\mu g\ mL^{-1}$	-	Microgram per mililiter
µL	-	Microliter
µm	-	Micrometer

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CHAPTER 1

INTRODUCTION

1.1 Background of study

The rising of contaminants of emerging concern (CECs) and persistent organic pollutants (POPs) in the environment that could not be treated by conventional wastewater treatment is a big issue as they are groups of ubiquitous, persistent and biologically active compounds with some unrecognized toxicities and endocrine disruptor functions. The presence of high levels of interfering compounds responsible for non-specific detection makes quantifications of the analytes difficult and imprecise, thus sample preparation is required prior to quantitative analysis.

In any trace analysis, sample preparation is a critical step before instrumental analysis. The sample preparation procedure for the determination of residues in environmental samples typically involves several steps such as extraction or dilution of sample, preconcentration of analytes, and removal of interfering substances. Liquid-liquid extraction (LLE) and solid phase extraction (SPE) have been widely used for analysis but both have significant drawbacks. While liquid phase microextraction (LPME) and solid phase microextraction (SPME) methods offer miniaturized greener alternatives, they are limited by the time frame given for a rapid analysis. Another alternative is to use dispersive solid phase extraction (DSPE) which offers a more rapid and simple extraction compared to others. As an added value to DSPE, magnetic solid phase extraction (MSPE) was introduced for its ease of separation via external magnet, thus, reducing cost, and labour of filtration (Wan Ibrahim *et al.*, 2015). The sorbent was combined with magnetite of choice for adsorption or extraction of organic pollutants. Most of the current literatures used carbon and silica based magnetic nanoparticles (MNP) (Sha *et al.*, 2008; Wan Ibrahim *et al.*, 2013; Xie *et al.*, 2014). Nevertheless, there have been search of

compatible sorbent to be combined with magnetite for extraction of pollutants from aqueous samples.

Conducting polymers have been in the spotlight as an advanced nanomaterial for its unique properties such as conductivity, resistivity and stability. Among those conducting polymers, polypyrrole (PPy) is especially promising due to good environmental stability, facile synthesis, and higher conductivity. It is typically used for coating on SPME (Saraji *et al.*, 2013) and sensors for metal detection (Bhaumik, *et al.*, 2011b; Babaei and Alizadeh, 2013; Tiwari *et al.*, 2015). These attributions have led to its usage in separation science.

Multivariate analysis cannot be separated from analytical chemistry, but the integration of multivariate statistical software with analytical chemistry has only recently been established. Multivariate analysis has been used in the optimization of chemical factors during development of analytical strategies involving pre-concentration systems using SPE, LLE, and other chromatographic methods (Aguilar *et al.*, 1999; Tadjarodi *et al.*, 2015; do Carmo *et al.*, 2017). It allows the development of mathematical models that permit assessment of the relevance as well as evaluate the interaction between optimization factors. It can also be used to predict and tailor the optimization based on the method desirability or aim.

1.2 Problem Statement

Various emerging contaminants used by humans have been infesting our waters, posing a big threat either via ecotoxicity or mutagenicity. A rapid and effective approach is needed to regulate and monitor these contaminants. Numerous techniques to cater these residues have been developed consisting of LLE and LPME, which uses substantial amounts of hazardous solvents. SPE has been applied yet uneconomical due to channellings and clogging of expensive cartridges. In addition several pre-concentration steps needed in analysis is time-consuming. Therefore, current researches focused on developing a rapid and effective method to regulate and monitor these pharmaceutical residues, thus MSPE comes into the

picture. MSPE offers an easy approach to sample preparation as small amount of sorbents are used (Wan Ibrahim *et al.*, 2015). It involves easy separation of analytes from solution by the mere use of external magnet, followed by desorption of analytes (minimal solvent usage) from sorbent prior to analysis. Most of the current literatures used carbon and silica-based MNP (Sha *et al.*, 2008; Sarafraz-Yazdi *et al.*, 2010; Wan Ibrahim *et al.*, 2013; Xie *et al.*, 2014) and tailored generally towards non-polar analytes. Conducting polymers was then introduced in order to cater to mid-polar range pollutants.

Polypyrrole (PPy) is a promising conductive polymer well known for its ruggedness and ease of synthesis, attributed by its ability to simultaneously attract non-polar and polar compounds. In recent years, conductive polymers have attracted great attention for the extraction of polar compounds due to the charge of PPy that can be readily controlled by oxidation and reduction of polymers. PPy has been used as modifier for sorbent in many studies because of its thermal stabililty (Mohammadi *et al.*, 2005; Ramoa *et al.*, 2015) and non-toxicity (Mehdinia *et al.*, 2007; Sharma *et al.*, 2013). It is recognized having adsorption ability through ion exchange or electrostatic interaction, largely owing to the existence of positively charged nitrogen atoms in PPy matrix. Moreover PPy can undergo protonation or deprotonation process using acid or basic solution, depend on its counter ions. (Junjie *et al.*, 2013), which results in the change of its surface charges. The capability of reversible transformation of PPy makes it possible that the ions could be absorbed or desorbed from PPy, hence, we predict that it would have an excellent adsorption-desorption property. In this regard, PPy can be considered as a potential reinforcer to improve the adsorption capacities of sorbent.

However, this unidimensional polymer lacks dispersity as it is lightweight, and typically used for coating on SPME (Saraji *et al.*, 2013) and sensors for metal detection (Bhaumik *et al.*, 2011b; Babaei and Alizadeh, 2013; Tiwari *et al.*, 2015). The conductiveness of PPy, that produces its ion-exchange ability, heavily relies on the optimization of the polymer synthesis. Thus a balance of preparation of the sorbent and magnetization carrier is needed to produce efficient sorbent for magnetic extraction. To the best of our knowledge, there has not been any in-depth research

regarding the extraction of pharmaceutical residues using PPy reinforced magnetic sorbent prior to analysis.

In order to improve the overall adsorption capacity and increase its dispersion of the Fe_3O_4-PPy, it was enhanced with graphene oxide (GO), which is known to have high surface area and polar and nonpolar groups (-OH and –COOH) (Stankovich *et al.,* 2006). Neonicotinoids, a polar non-volatile pesticide was selected as model compounds for extraction as they can benefit from the electrostatic interactions from PPy and Fe_3O_4. PPy has been combined with GO for the preconcentration of organic contaminants such as terpenes (Zhang *et al.*, 2014), and phenols (Zou *et al.,* 2011). A balance in the preparation of the sorbent is needed for maximum extraction efficiency of polar emerging contaminant, neonicotinoids, by anion exchange from PPy lone pair site. It is expected that this research will produce new efficient MSPE adsorbents based on Fe_3O_4-PPy reinforced GO nanocomposite for the extraction of selected neonicotinoids.

In exploring the wide-range of green sorbents, biowaste from agriculture industries was considered as a viable source of sorbents with major advantages of biodegradability and sustainability. The widely-available oil palm waste in Malaysia is an advantage to be studied as an alternative green carbon based sorbent. Oil palm empty fruit bunches waste (OPEFB) that accounted for 19.5 million tons in 2008 (Wirasnita *et al.*, 2015) contains lignin and cellulosic materials that prevents the usage for extraction purposes. Normally OPEFB is used as feed for energy and converted to oil palm activated carbon (OPAC). OPAC based on OPEFB have been studied for removal and absorption of chlorophenols (Shaarani and Hameed, 2011) and lead (Daneshfozoun *et al.*, 2016), but none on modifications of OPAC and Fe_3O_4-PPy for magnetic extraction of organochlorine pesticides in aqueous matrices. The development of this sorbent may bridge the gap for a greener and more sustainable sorbents for MSPE.

The development of method based on sorbent reinforced with Fe_3O_4-PPy is promising, less laborious and offer fast extraction time. It is expected that this research will produce new efficient MSPE sorbents based on modifications of Fe_3O_4-PPy composite for the extraction of selected organic pollutants. With the aid of

RSM-BBD, the lab work could be minimized, and the interaction between factors that affect the extraction of organic pollutants of choice could be better understand with minimum amount of lab work.

1.3 Aims and objectives

The study aims to produce simple, fast and efficient extraction techniques that use Fe_3O_4 as the base for separation purpose. Modifications were performed on Fe_3O_4 using PPy to produce Fe_3O_4-PPy. Two other sorbents were produced based on modification of Fe_3O_4-PPy with GO to produce GO-Fe_3O_4-PPy and modification with OPAC to produce OPAC-Fe_3O_4-PPy. These three sorbents were used for the development of three new MSPE methods for the analysis of selected organic pollutants, namely non-steroidal anti-inflammatory drugs (NSAIDs), neonicotinoids (NEOs) and organochlorine pesticides (OCPs) in aqueous matrices.

The objectives of this study are to;

i. prepare, characterize and apply magnetic polypyrrole sorbent (Fe_3O_4-PPy) using response surface methodology-Box Behnken design (RSM-BBD) as optimization tool, catered to the extraction of selected pharmaceutical residues (NSAIDs) in aqueous matrices.
ii. study the incorporation of carbon with high surface area (graphene oxide-GO) through synthesis, characterization, and application of GO-Fe_3O_4-PPy using RSM-BBD as optimization tool, catered to the extraction of selected neonicotinoids (NEOs) in aqueous matrices.
iii. To study the incorporation of biosorbent, oil palm waste-based activated carbon (OPAC) through synthesis, characterization and application of OPAC-Fe_3O_4-PPy sorbent using RSM-BBD as optimization tool, catered to the extraction of selected organochlorine pesticides (OCPs) in aqueous matrices.

1.4 Scope of study

This study explores the possibility of Fe_3O_4 magnetic nanoparticle (MNP) and PPy-reinforced composite in the enhancement of the extraction of selected analytes using MSPE method with the aim of high extraction efficiency and rapid microextraction. Response surface methodology – Box Behnken Design (RSM-BBD) was chosen for the optimization of the material for the extraction method.

The incorporation of PPy to Fe_3O_4 magnetic nanoparticle (MNP) was optimized with the aid of RSM-BBD to produce Fe_3O_4-PPy sorbent that was applied to the extraction of pharmaceutical residues, non-steroidal anti-inflammatory drugs (NSAIDs) in aqueous matrices using high performance liquid chromatography with ultraviolet detector (HPLC-UV). The potential applications of the optimized absorbents were studied for microextraction of selected NSAIDs namely naproxen, diclofenac sodium salt and mefenamic acid from aqueous samples.

To further enhance its extraction capacity, GO, the selected carbon-based material, well known for its high surface area, was incorporated to Fe_3O_4-PPy to form Fe_3O_4-GO-PPy, and further optimized using RSM-BBD. The aim was to achieve high selectivity towards newly-developed herbicides, neonicotinoids, combined with HPLC-UV determination.

The incorporation of Fe_3O_4 and PPy to a more sustainable biosorbent; oil palm-biomass waste activated carbon (OPAC) was studied in terms of its ability to extract organochlorine pesticides (OCPs) that is commonly used as an insecticide in oil palm industry. RSM-BBD was also used to assist the synthesis optimization as well as extraction method and analysis using gas chromatography equipped with micro electron capture detector (GC-μECD).

The physical and chemical properties of the synthesized sorbent were characterized using Fourier-transform infrared spectroscopy (FTIR), field emission scanning electron microscopy (FESEM), transmission electron microscopy (TEM), vibrating sample magnetometry (VSM) and nitrogen adsorption analysis.

For the application of the sorbent for MSPE method, several important extraction parameters were optimized comprehensively and analytical performance of the developed method were evaluated, validated and applied assisted by RSM-BBD tool to the determination of these pollutants in environmental aqueous samples, namely tap water, waste water (effluent) and river water.

This study has potential limitations. The RSM-BBD models are based on interventional and prospective observational studies. They are therefore subject to biases and confounding that may have influenced out model estimates. However, the optimization effects of development of sorbent and method were estimated from meta analysis with confirmatory validity analysis. In order for the method to be used commercially, low-end instruments were used in this study, such as HPLC-UV and GC-μECD. For more sensitive detection, high-end instrument such as gas chromatography–mass spectrometry (GC-MS) or liquid chromatography-mass spectrometry (LC-MS) is suggested. Other than that, the sample for the study is limited to the samples collected by the Department of Environment in Johor State only.

1.5 Significance and original contributions of the study

Whilst the occurrence of environmental pollution is on the rise, there is increasing demand for faster and greener solutions to minimize the impact on the human health via excessive use of organic solvents in chemical analysis. To this end, this work would be able to contribute using different approaches for the preparation of Fe_3O_4-PPy and Fe_3O_4-PPy-reinforced sorbent. The prepared Fe_3O_4-PPy and Fe_3O_4-PPy-reinforced sorbent are expected to be useful in the analysis of a wide range of emerging contaminants, and persistent organic pollutants. It is also expected to offer a simple, fast and cost-effective extraction technique as an alternative that is greener and safer for the environment.

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LIST OF PUBLICATIONS

Journal with Impact Factor

1. **Mohd Marsin, F.,** Wan Ibrahim, W. A., Abdul Keyon, A. S., and Sanagi, M. M. (2018). Box–Behnken Experimental Design for the Synthesis of Magnetite–Polypyrrole Composite for the Magnetic Solid Phase Extraction of Non-steroidal Anti-inflammatory Drug Residues. *Analytical Letters*, *51*(14), 2221-2239. **(Q3: IF: 0.34)**
2. **Mohd Marsin, F.,** Wan Ibrahim, W.A., Nodeh, H. R., and Sanagi, M.M. (2019). New Magnetic Oil Palm Fiber Activated Carbon-Reinforced Polypyrrole Solid Phase Extraction Combined With Gas Chromatography-Electron Capture Detection for Determination of organochlorine pesticides in Water Samples. *Journal Chromatography A,* 1612, 460638. **(Q1: IF: 3.858)**

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