REMOVAL OF BISPHENOL A FROM DRINKING WATER USING HYBRID SYSTEM OF PALM KERNEL SHELL BIOSORBENT AND HEMATITE NANOCOMPOSITE MEMBRANE FILTRATION

By

KATIBI KAMIL KAYODE

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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DEDICATION

This work is dedicated to ALLAH (*Subhanahu Wataala*); and to my beloved late Father Sheikh Dr. Ahmad Sa'adudeen Katibi Ibrahim. May the Almighty *ALLAH* continue to increase His abundant rahmah and blessings on him till the day of resurrection.

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

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Chairman : Associate Professor Khairul Faezah Md. Yunos, PhD
Faculty : Engineering

Water is an essential resource required for sustainable living, and its availability should be safeguarded, appropriated, and freely accessible. In recent years, the persistent detection of endocrine-disrupting chemicals (EDCs), including Bisphenol A (BPA) (µg/L or ng/L) in wastewater as well as drinkable water has drawn pressing concerns among the public, environmentalists as well as regulatory bodies owing to its related ecological risks, a hazard to global water quality in addition to public health. The most widely applied conventional biological treatment technique is inefficient and often failed to reduce the concentration of recalcitrant BPA to an acceptable discharge limit (10 µg/L) of National Water Quality Standards for Malaysia and Drinking Water Hygienic Standard (GB5749-2006). This was due to the complex aromatic ring structure of BPA resulting in high resistance to oxidation and microbial degradation, requiring the need to develop alternate efficient treatment technologies. Therefore, the main objective of this study is to develop a novel integrated system and physical technique which comprises the adsorption using magnetic biochar derived from palm kernel shell (PKS) as biosorbent and nanocomposite membrane filtration for removing Bisphenol A (BPA) and other contaminants present in drinking water. Initially, magnetic biochar was synthesized using high-energy ball milling and co-precipitation techniques using ferric chloride and ferrous chloride as the magnetic medium. The synthesized biochar was applied in the batch system to investigate the remediation of BPA from aqueous solution. Hematite (Fe_2O_3) nanoparticles (NPs) were synthesized using the facile sol–gel technique, modified by 3-Aminopropyl tri-ethoxy silane (APTES), and analyzed to identify the structural properties, functional groups, elemental constituents, and the surface modification of Fe_2O_3 NPs. Then, pristine polyvinylidene Fluoride (PVDF)-Polyethylene glycol (PEG) and nanocomposite flat sheet membrane blended with modified Fe_2O_3 NPs were fabricated via non-solvent induced phase inversion (NIPs) technique. The resultant flat sheet membranes were analyzed based on the contact angle, FESEM, EDX, surface zeta-potential, and porosity analyses, and evaluated for permeate flux, resistance to fouling, Bisphenol A, Bisphenol F and Bisphenol S (BPF) removal in aqueous solution. The result of the batch adsorption indicated that the magnetic biochar

demonstrates superior adsorption capacity of 37.64 mg/g and maximum removal efficiency of BPA with 94.2%, over the neat biochar using aqueous solution. The flat sheet nanocomposite membrane blended with 1.5 wt.% of modified Fe_2O_3 NPs exhibited a superior performance with permeation flux of 125.47 L/m^2.h, least contact angle of 58.5°, remarkable removals of BPA (94.73%), BPS (92.19%), BPF (90.69%), superior flux recovery ratio (FRR) and relative flux reduction (RFR) of 78.83% and 14.05%, following three cycles of filtration for a complete 5.6 hours compared to pristine membranes. For the hybrid system, initially, the untreated water sample was subjected to an adsorption process for 200 minutes using magnetic biochar as a biosorbent. The treated effluent was analyzed, and BPA removal of 66.98% was achieved under 200 minutes contact time, with a BPA concentration of 12.86 μg/L, which was above (10 μg/L) of the National Water Quality Standards for Malaysia and Drinking Water Hygienic Standard (GB5749-2006) discharge limit for phenolic compounds. Hence, the application of further treatment process became imperative. The magnetic biochar-treated effluent was led into the membrane section. On this basis, hematite nanoparticles were blended into the membrane dope formulations with varied Fe_2O_3 NPs concentrations (0, 1.0, 1.25, 1.50, and 2.0 wt.%), and the resulting dopes were distinctly swirled using phase inversion procedure. The resultant hollow fibre membranes were analyzed based on surface charge characteristics, structural morphology, hydrophilicity, elemental compositions, porosity, as well as thermal stability. Observably, the fouling resistance, permeation flux, and BPA rejection were enhanced with the rise in Fe_2O_3 NPs loading. PVDF-PEG hollow fibre with 1.50 wt.%-Fe_2O_3 NPs loading exhibited the most hydrophilic with contact angle of 56.3°, the most negatively charged surface with -43.7mV, and excellent performance with 191.85 L/m^2-h of water permeation flux. Furthermore, the 1.5 wt-Fe_2O_3 NPs composite membrane demonstrated a superior antifouling performance upon the completion of the third cycle of filtration (cycle 3), the modified membrane with 1.5 wt%- Fe_2O_3 NPs still accomplished a higher percent of FRR (77.35%) along with RFR of 21.29%, respectively. In general, the %FRR and %RFR diminished together with the filtration cycles. However, a decline in the permeation flux and fouling resistance were observed at an elevated loading at excess loading of 2.0 wt.%-Fe_2O_3 NPs. This was a consequence of the agglomerated nanoparticles within the dope matrix upon elevated loading. As a result of the upsurge performance and remarkable resistance to fouling of the PVDF-PEG with 1.50 wt.% Fe_2O_3 NPs nanocomposite membrane, it was considered and incorporated into the integrated system to further advance the removal of BPA from the magnetic biochar-treated tap water. The incorporated Fe_2O_3 at 1.50 wt.% loading enhanced BPA removal and the antifouling properties of the modified nanocomposite PVDF-PEG membrane. Conclusively, the physicochemical analysis of the treated final permeate by the hybrid system revealed that the BPA concentration was reduced to 5.02μg/L which was below 10 μg/L of the National Water Quality Standards for Malaysia and Drinking Water Hygienic Standard (GB5749-2006) discharge limit. It can be deduced that the successfully developed and evaluated novel hybrid system was able to reduce the concentration of BPA to comply with the discharge limits. Thus, the implication of this study demonstrated a full capacity of deterring all possible adverse effects of BPA on the environment. Therefore, the implementation of this hybrid system will assist the relevant industries and water treatment authorities to fully comply with the discharge limit.

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

PENGASINGAN BISPENOL A DARIPADA AIR MINUMAN MENGGUNAKAN SISTEM HIBRID BIOSORBEN KERNEL KELAPA SAWIT DAN PENAPISAN MEMBRAN NANOKOMPOSIT HEMATIT

Oleh

KATIBI KAMIL KAYODE

Februari 2023

Pengerusi : Profesor Madya Khairul Faezah Md Yunos, PhD
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Air adalah sumber penting yang diperlukan untuk kehidupan yang mampan, dan ketersediaannya harus dilindungi, diperuntukkan, dan boleh diakses secara bebas. Dalam beberapa tahun kebelakangan ini, pengesanan berterusan bahan kimia pengganggu endokrin (EDC), termasuk Bisphenol A (µg/L atau ng/L) dalam air sisa dan air minuman telah menimbulkan kebimbangan yang mendesak di kalangan pihak berkuasa tempatan dan kawal selia berikutan risiko ekologi yang berkaitan, ancaman kepada kualiti air global dan juga kesihatan awam.Teknik rawatan biologi konvensional yang paling meluas digunakan adalah tidak cekap dan sering gagal untuk mengurangkan kepekatan BPA yang sukar dikawal kepada had pelepasan yang boleh diterima (10 µg/L) oleh Piawai Kualiti Air Kebangsaan untuk Malaysia dan Piawai Kebersihan Air Minuman (GB5749-2006). Hal ini disebabkan oleh struktur cincin aromatik BPA yang kompleks menyebabkan rintangan yang tinggi terhadap oksidasi dan degradasi mikrob dan memerlukan keperluan untuk membangunkan teknologi rawatan alternatif yang cekap. Sehubungan itu, fokus utama kerja penyelidikan ini adalah untuk membangunkan sistem bersepadu baru dan teknik fizikal yang terdiri daripada penyepaduan penjerapan menggunakan biochar magnetik yang diperolehi daripada kulit isirong sawit sebagai penjerap dan penapisan membran untuk menyingkirkan Bisphenol A dan bahan cemar lain yang ada di dalam air. Pada permulaannya, biochar magnetik telah disintesis menggunakan teknik pengilingan bebola tenaga tinggi dan teknik kerpasan bersama menggunakan ferik klorida dan ferus klorida sebagai medium magnet. Biochar yang telah disintensis telah diaplikasikan dalam sistem kelompok untuk menyiasat pemulihan BPA daripada larutan akueus. Nanopartikel (NP) Hematit (Fe_2O_3) telah disintesis menggunakan teknik sol-gel mudah, diubah suai oleh 3-Aminopropyl tri-ethoxy silane (APTES), dan dianalisa untuk mengenalpasti sifat struktur, kumpulan berfungsi, unsur asas, dan pengubahsuaian permukaan NP Fe_2O_3. Kemudian, Polivinilidena Fluorida (PVDF)-Polyethylene glycol (PEG) tulen dan nanokomposit membran kepingan rata dicampur dengan NP Fe_2O_3 yang telah diubah suai telah direka melalui teknik penyongsangan fasa teraruh bukan pelarut . Membran kepingan rata yang terhasil telah dianalisa berdasarkan sudut sentuhan, FESEM, EDX, potensi permukaan zeta dan analisis keliangan. Ia juga telah diuji untuk fluk resapan, rintangan terhadap kekotoran, penyingkiran Bisphenol A, Bisphenol F and Bisphenol S (BPF) dalam larutan akueus. Hasil daripada pejerapan kelompok menunjukkan bahawa biochar magnetik menunjukkan kapasiti penjerapan yang besar dengan nilai 37.64 mg/g dan kecekapan penyingkiran BPA yang maksimum dengan nilai 94.2%, ke atas biochar asli menggunakan larutan akueus. Membran nanokomposit kepingan rata yang diadun dengan 1.5 wt.% NP Fe_2O_3 yang telah diubah suai mempamerkan prestasi unggul dengan fluks resapan 125.47 L/m2.h, sudut sentuhan terkecil 58.5°, penyingkiran BPA yang luar biasa (94.73%), BPS (92.19%), BPF (90.69%), kadar pemulihan fluks (FRR) unggul dan pengurangan fluks relatif (RFR) sebanyak 78.83% dan 14.05%, berikutan tiga kitaran penapisan selama 5.6 jam lengkap berbanding dengan membran tulen. Bagi sistem hibrid, pada mulanya, sampel air yang tidak dirawat tertakluk kepada proses penjerapan selama 200 minit menggunakan biochar magnetik sebagai biosorben.Sisa buangan terawat telah dianalisis, dan BPA yang disingkirkan mencapai 66.98% dengan masa sentuhan kurang dari 200 minit dengan kepekatan BPA 12.86 μg/L, dimana ia melebihi dari (10 μg/L) had pelepasan sebatian fenolik yang ditetapkan oleh Piawaian Kualiti Air Kebangsaan Malaysia dan Piawaian Kebersihan Air Minuman (GB5749-2006). Oleh itu, aplikasi proses rawatan selanjutnya adalah sangat penting. Sisa buangan terawat biochar magnetik dibawa ke bahagian membran. Atas dasar ini, nanopartikel hematit dicampur kedalam formulasi dope membran dengan kepekatan Fe_2O_3 NP yang berbeza (0,1.0,1.25,1.50, dan 2.0 wt.%) dan menghasilkan dope yang berlingkar menggunakan fasa penyonsangan. Gentian berongga yang terhasil dianalisis berdasarkan ciri cas permukaan, morfologi struktur, hidrofilik, komposisi unsur, keliangan, serta kestabilan terma. Berdasarkan pemerhatian, rintangan fouling, fluks resapan, dan penolakan BPA telah dipertingkatkan dengan peningkatan dalam pemuatan NP Fe_2O_3. PVDF-PEG dengan pemuatan 1.50 wt.%-Fe_2O_3 NPs mempamerkan kebanyakan hidrofilik dengan sudut sentuhan 56.3°, permukaan paling bercas negatif dengan -43.7mV dan prestasi cemerlang dengan 191.85 L/m^2-j fluks resapan air. Tambahan pula, membran komposit 1.5 wt-Fe_2O_3 NPs menunjukkan prestasi antifouling yang unggul walaupun pada penyiapan penapisan ketiga (kitaran 3), membran yang diubah suai dengan 1.5 wt% - NPs Fe_2O_3 masih mencapai peratusan FRR yang lebih tinggi (77.35%) sepanjang dengan RFR 21.29%, masing-masing. Secara umum, %FRR dan %RFR berkurangan bersama-sama dengan kitaran penapisan. Walau bagaimanapun, penurunan dalam fluks resapan dan rintangan fouling telah diperhatikan pada beban tinggi pada beban berlebihan 2.0 wt.%- Fe_2O_3 NPs. Ini adalah akibat daripada nanopartikel terkumpul dalam matriks dope apabila pemuatan tinggi. Hasil daripada prestasi peningkatan dan rintangan yang luar biasa terhadap fouling membran komposit 1.50 wt.% -Fe_2O_3 NPs, ia telah dipertimbangkan dan digabungkan ke dalam sistem bersepadu untuk memajukan lagi penyingkiran BPA daripada air paip yang dirawat biochar magnetik. Gabungan pemuatan 1.5 wt. % Fe_2O_3 meningkatkan penyingkiran BPA dan sifat antifouling membran PVDF-PEG komposit nano diubahsuai. Secara konklusinya, analisis fizik-kimia untuk resapan akhir yang dirawat oleh sistem hibrid mendedahkan bahawa kepekatan BPA dikurangkan kepada 5.02 μg/L iaitu di bawah has pelepasan (10 μg/L) Piawaian Kualiti Air Kebangsaan untuk Malaysia dan Piawaian Kebersihan Air Minuman (GB5749-2006). Dapat disimpulkan bahawa sistem hibrid novel berjaya dibangunkan dan dinilai mampu mengurangkan kepekatan BPA untuk mematuhi had pelepasan. Justeru, implikasi kajian ini menunjukkan kapasiti penuh untuk menghalang semua kemungkinan kesan buruk BPA terhadap alam sekitar. Oleh itu, pelaksanaan sistem hibrid ini akan membantu industri berkaitan dan pihak berkuasa rawatan air untuk mematuhi sepenuhnya had pelepasan.

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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Declaration by the Members of Supervisory Committee

This is to certify that:

- The research conducted and thee writing of the thesis was under our supervision
- Supervision of responsibilities as slated in Rule 41 in Rule 2003 (Revision 2012-2013) were adhered to.

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LIST OF ABBREVIATIONS

ANOVA	Analysis of variance
APTES	3-Aminopropyl tri-ethoxy silane
BET	Brunauere Emmentte Teller
BPA	Bisphenol A
BPF	Bisphenol F
BPS	Bisphenol S
EAD	electron acceptor–donor
EDC	Endocrine disrupting compounds
EDX	Energy Dispersive X—ray Spectroscopy (EDX)
Fe_2O_3	Hematite iron oxides
FRR	Flux Reduction Ratio
FTIR	Fourier Transform Infra-Red
Gramme	g
H_2SO_4	Sulfuric acid
HEBM	High Energy Ball Milling
hour	h
J	Flux
kg	Kilogram
L	Litre
m	Meter
mg/L	Milligram per liter
NaOH	Sodium hydroxide
NP	Nonylphenol
NPs	Nanoparticles

PCPPs	Personal care products and pharmaceuticals
PEG	Polyethylene glycol
PFO	Pseudo first order
PKS	Palm kernel shell
PSO	Pseudo second order
PVDF	Polyvinylidene Fluoride
RFR	Relative Flux Recovery
RSM	Response Surface Methodology
SEM	Scanning Electron Microscopy
TMP	Transmembrane pressure
VSM	Vibrating sample magnetometer

CHAPTER 1

INTRODUCTION

1.1 Research Background

Over the past years, the rapid expanding global population has exerted unmatched pressure on the safety of clean water supply. The World Health Organization forecast that half of the global population will live in water-stressed regions by 2025 (Levallois & Villanueva, 2019; WHO/UNICEF, 2017). Safeguarding current freshwater sources and expanding technologies to enhance existing water supplies are highly indispensable (Van Rees et al., 2021). Leaching of endocrine disrupting compounds as well as possibly toxic constituents into the surroundings from plastics causes plastic pollution enigma which is a severe environmental challenge (Wang et al., 2018b).

The global plastic production reached 7800 million tons, while over 6000 million tons were landfilled otherwise neglected in nature (Ross Brown et al., 2019). Furthermore, it is estimated that the world-wide output of thermoplastics will attain nearly 445.25 million metric tons by 2025 and the annual manufacture magnitudes are likely to keep on expanding in the succeeding years, expanding to nearly 590 million metric tons in 2050. This would constitute over 30 percent surge as compared to 2025. During the last decade, the worldwide plastics production has expanded from 270 million metric tons to nearly 370 million metric tons (Xu et al., 2021). The occurrence of endocrine disrupting compounds in drinking water supply systems, from the source to the supply, stimulates concerns relating to the sustainability and security of the system. This is in taking into consideration not only the contamination level on the other hand also human vulnerability to EDCs, through water consumption (Talib et al., 2020). EDCs are routinely recognized to influence the endocrine operation, affecting ecological sustainability, human health (provoking both severe and incessant disorders), as well as economic prosperity (Wee & Aris, 2019).

Recently EDCs have surfaced as pollutants deserving significant issue due to their widespread use and high-minded pollution in numerous environmental media (Wee et al., 2019). These environmental pollutants can be categorized on the basis of their possible endocrine disruption effects (for instance, genotoxic, carcinogenic, neurotoxic as well as cytotoxic), affecting population growth as well as expansion together with social and economic values. Despite that, there is a deficient international and national degree of concern and practice in spite of continuous scientific work to unravel the nature, degree and associated risks with the wide range of compounds; while the comparatively low level of public consciousness requires solution (Wee & Aris, 2019).

Particularly plasticizers pollution from plastics has been extensively reported since 2007 and had attracted growing interest from the regulating authorities in addition to scientific community owing to their severe poisonousness, endocrine disrupting consequences, and prevailing occurrences distribution (Lee et al., 2019). The plasticizer compounds are classified as emerging pollutants and endocrine disrupting compounds (EDCs), owing to their structural analogy with internal hormones that can simulate the physiologic actions within the body (Ross Brown et al., 2019). Notably amongst the plasticizers, Bisphenol A has been regularly found in the surface water, groundwater, as well as seawater in several regions of the globe (Pelch et al., 2019), predominantly because of the inadequate removal by conventional treatment facilities (Im & Löffler, 2016) and leaching from plastics as well as microplastics made from BPA (Wei et al., 2019).

Since 1957, there has been industrial utilization of Bisphenol A in the production of various items such as food storage, sports health care products, polycarbonate plastics, as well as electronic equipment for numerous applications (Li et al., 2019). In 2017, the global economy of BPA was valued to be 8.15 million tons and the estimated global gross output was 7.10 million tons (Li et al., 2019). Despite that, reports have indicated that BPA presents visible estrogen damaging action as well as susceptible to leach out of products, and consequently trigger direct vulnerability to humans thereby generated increasing general concern (Zheng et al., 2018). In the light of this, BPA was outlawed in several nations including Malaysia in the last few years, particularly its usage in infant food storage items as well as beverage materials (Lee et al., 2019).

The Malaysian Ministry of Health has outlawed the manufacture, importation, and adverts for sell or sale of any feeding bottles comprising Bisphenol A (BPA) (Malaysia Ministry of Health, 2017). Since some regional governments sternly controlled the utilizations as well as applications of BPA, Bisphenol S and Bisphenol F has been exploited as an alternate to BPA (Wang et al., 2019). The outstanding thermal stability as well as mechanical performance of BPS-aided products (Fang et al., 2020), have yielded the manufacture of approximately 1000–10,000 tons of this product in several European nations (ECHA, 2015), as the principal BPA substitute (Choi & Lee, 2017). BPA and its analogous may navigate into the environment in various ways.

Drinking water supply system typically comprised of three components: a drinking water source (raw water that to be treated), a drinking water treatment facility, and a drinkable water supply (the distributed treated water). In most countries, surface river water is the primary route for raw water intake by drinking water treatment facilities for purification and subsequent delivery of drinking water. BPA infiltrate the raw waterbodies via nonpoint routes (such as leachate and runoff) as well as point sources including discharges from both treated and untreated (Simazaki et al., 2015).

The drinking water purification facility serves as the final line of protection against chemical vulnerability in drinking water source (between consumers and the environment). Unfavourably, inadequate and inconsistent BPA removal via conventioanl biological treatment systems arises given that the system has not been designed for BPA removal, thereby resulting to BPA lading in the drinking water supply (Conley et al., 2017).

Correspondingly, several treatment techniques have been applied to eliminate BPA from both wastewater and potable water in order to enhance the efficiency of traditional treatment approaches. These techniques include enzymatic degradation, membrane technology, ozonation, oxidation processes, adsorption, photocatalytic degradation, and membrane bioreactor (MBR) systems (Gadupudi et al., 2019; Sanguanpak et al., 2019). Although these technologies have proven to be promising alternatives for BPA removal, yet, most of the methods are faced with associated drawbacks in tackling these persistent BPA contaminants, resulting to difficulties in the provision of secure and safe water supplies and therefore not suitable for large scale treatment of such persistent pollutants.

A considerable series of studies have demonstrated the drawbacks of these treatment technologies which includes exorbitant rate, lengthy treatment-duration, large operational footprint, particularly the generation of noxious by-products, toxic sludge, concentrated residues, complex procedures, and high operation and maintenance costs which reduces their applicability (Ho et al., 2017a; Khanzada et al., 2020). Also the relatively advanced hybrid treatment techniques are challenging to expand to upgrade the removal efficiency of conventional drinking water treatment facilities, with the process sustainability and cost efficiency are also of serious concern (Rodriguez-Narvaez et al., 2017).

On the other hand adsorption treatment technique is an efficient, environmental friendly with superior contaminants removal potential among the water purification techniques based on the simplicity of design and operation as well as insensitivity to noxious compounds (Adeleke et al., 2019). Also, membrane technology is suitable for providing quality treated permeate owing to its facile operation, simple to scale up, lowered chemical usage, low energy demand, and excellent separation capacity (Hu et al., 2015). Besides, the benefit of membrane treatment technique is environmentally benign to the humans and environment (Chen et al., 2012), resources recovery with little/or zero odor, treatment process within a shorter time, as well as smaller footprints (Liao et al., 2018). However, susceptibility to fouling deposit formation is the main impediment of this technology (Ghani et al., 2018). Fouling deposit is the resulted drawback from the gentle buildup of the particulates and colloids on the membrane surface and pores walls (Judd, 2016). This impact usually draw excessive working energy and increased operating costs which rendered the treatment technique unproductive for industrial utilization (Krzeminski et al, 2017). Though several nanomaterials has been utilized to mitigate membrane fouling, yet the application of the most stable iron oxides i.e. hematite nanoparticles as a nanofiller to address the problem of membrane fouling and sustainable BPA removal from drinking water remain unresolved.

The application of hematite nanoparticles to subdue membrane fouling is a novel approach that has the potential to significantly improve the efficiency and sustainability of membrane filtration processes. Hematite nanoparticles have unique physicochemical properties that make them highly effective in preventing membrane fouling, which is a major problem in many water treatment applications.

The key novelty of this approach lies in the fact that hematite nanoparticles can be easily synthesized using low-cost and environmentally friendly methods, making them a cost-effective and sustainable alternative to traditional anti-fouling agents. Additionally, the small size and high surface area of hematite nanoparticles make them highly effective in adsorbing and removing foulants from the surface of the membrane, preventing them from accumulating and clogging the membrane pores.

Moreover, hematite nanoparticles are stable under a wide range of environmental conditions and are not toxic, which makes them suitable for use in water treatment applications. The use of hematite nanoparticles to prevent membrane fouling can also extend the lifespan of the membrane, reducing the need for frequent replacement and thereby reducing operational costs. Hence, the use of hematite nanoparticles to subdue membrane fouling is a novel and promising approach that has the potential to significantly improve the efficiency, sustainability, and cost-effectiveness of membrane filtration processes. This approach can have important implications for various water treatment applications, including desalination, wastewater treatment, and drinking water purification.

Against this background, this study focuses on the development of a novel, efficient and cost-effective method for removing Bisphenol A (BPA) from drinking water. The study proposes the use of a novel hybrid system that combines a natural biosorbent made from palm kernel shells with a nanocomposite membrane filtration technology. The palm kernel shell biosorbent acts as a pre-treatment method for the water, removing a significant portion of the BPA compounds, while the nanocomposite membrane filtration further purifies the water by removing the remaining BPA compounds.

The use of a natural biosorbent material such as palm kernel shells is an innovative approach that has several advantages over traditional synthetic materials, such as cost-effectiveness, biodegradability, and sustainability. The combination of the biosorbent with a nanocomposite membrane filtration technology also allows for higher removal efficiencies, making the system more efficient than traditional treatment techniques.

Therefore, this study presents a new and innovative solution for removing BPA from drinking water that has the potential to be implemented on a larger scale to improve the quality of drinking water and protect public health. This research contributes to the development of more sustainable and efficient water treatment methods, which is crucial for addressing the growing concerns over water pollution and the safety of drinking water.

1.2 Problem Statement

The presence of Bisphenol A (BPA) in drinking water has become a significant health concern due to its potential adverse effects on human health. BPA is a frequently used chemical compound in the production of plastics, and it can leach into water sources, posing a potential risk to human health (Wang et al., 2019). BPA is being considered in this study since it's the most widely produced chemical as well as the most predominant and persistent bisphenol compound that exhibit higher endocrine-disrupting potency than most synthetic EDCs (Ohore & Songhe, 2019). The persistent detection of BPA necessitated the need to explore efficient water purification technologies to further enhance the remediation of the concentration level. The conventional biological water treatment processes are often not efficient in removing BPA from drinking water, and there is a need for innovative and cost-effective methods for BPA removal. The application of palm kernel shell biochar as a biosorbent for the remediation of BPA from drinking water is still lacking.

Malaysia, being the second largest palm oil-producing country in the world, generates over 4 million tons of palm kernel shells (PKS) every year. With such a large volume of waste, there is a need to explore ways to preserve the environment (Azunna, 2019). Under exploitation of palm kernel shells in Malaysia is a significant problem that needs attention. Despite the abundant supply of palm kernel shells as a byproduct of the palm oil industry, they are not being fully utilized and could contributes to environmental issues as the shells are often discarded through burning or left to decompose, which can lead to water pollution (Yahayu et al., 2018). Therefore, this study explore the potential of PKS biochar as a biosorbent to promote sustainable and environmentally friendly practices.

In the quest to remediate BPA from water using adsorption technology, activated carbons is widely utilized for the adsorption of BPA. For instance, Koduru et al. (2016) uses goethite/activated carbon composite to remove BPA from water. The BPA adsorption capacity of 59.60 mg/g was recorded. Martín-Lara et al. (2020) examines the BPA adsorption from synthetic solutions by a commercial activated carbon, AC-40, in batch system. The maximum adsorption capacity of 94.34 mg/g obtained. However, the application of activated carbon is constrained as a result of huge cost. Also, the regeneration of activated carbons after adsorption process give rise to a loss of carbon and the reclaimed sorbent may have a considerably reduced sorption ability compared with the raw activated carbon.

Biochar despite having excellent unique properties, including increased surface functional groups, porous structure, profuse specific surface area, eco-friendly, low-cost, abundant inherent mineral components, elevated cation exchange capacity, and efficient in the removal of various hydrophobic and hydrophilic organic pollutants owing to its better aromaticity and hydrophobicity (Ahmed et al., 2017). However, the small size of biochar particles and its lower density create challenges in terms of its ability to be reused, separated, and recovered after adsorbing pollutants. These drawbacks makes its regeneration, separation, and recovery more problematic after adsorption, which could undermine its recycling capacity as well as industrial applications. To subdue these drawbacks, this study synthesize a novel PKS magnetic biochar via physical technique using high energy ball milling and the implantation of iron oxide (Fe_3O_4 and Fe_2O_3) using co-precipitation method to enhance its physicochemical properties.

Coupled with the fact that the major constraint in applying membrane technologies is frequent fouling, high input energy due to the excessive transmembrane (TMP). One of the main promising benefits of using hematite nanoparticles is their potential to enhance the antifouling properties of the membrane. By modifying the surface of the membrane with hematite nanoparticles, the membrane becomes more resistant to fouling by organic and inorganic contaminants, which can improve the overall performance and lifespan of the membrane.

Essentially, nanocomposite membrane blended with iron oxide nanoparticles will be fabricated and incorporated into the integrated system to further enhance the contaminants removal efficiency and mitigate the propensity of membrane fouling.

The development of a reliable and efficient method for BPA removal from drinking water is crucial for protecting public health and ensuring access to safe drinking water. The proposed hybrid system offers a unique and innovative approach that has the potential to overcome the limitations of traditional treatment methods and provide a cost-effective and sustainable solution for BPA removal. Therefore, this study addresses an important problem in the field of water treatment and has significant implications for public health and environmental sustainability.

1.3 Objectives

The main aim of this research is to develop facile continuous flow process of sorptionmembrane separation system for removing BPA from drinking water. To achieve this, the specific objectives of this study are:

1. To synthesize novel magnetic biochar derived from palm kernel shell and investigate the removal of Bisphenol A from Aqueous Solution using batch adsorption system.

2. To synthesize and functionalize hematite nanoparticles as a precursor material for the fabrication of flat sheet nanocomposite membrane.

3. To formulate, fabricate and characterize hollow fibre PVDF-PEG membrane at different dosages of Fe_2O_3 nanoparticles.

4. To design, fabricate and evaluate the overall performance of the integrated system of the adsorption-membrane under continuous flow process for the removal of BPA from drinking water.

1.4 Research Scope and Limitations

In this research, an integrated system comprising magnetic palm kernel shell biosorbent and nanocomposite membrane separation for decontaminating and removing BPA from drinking water was developed.

This study focuses on the application of a hybrid system comprising palm kernel shell biosorbent and hematite nanocomposite membrane filtration for the removal of Bisphenol A (BPA) from drinking water. The study aims to investigate the efficiency of the hybrid system in removing BPA from water.

The study is highly relevant due to the widespread use of BPA in the production of plastic containers and food packaging, which can lead to contamination of water sources. BPA is a known endocrine disruptor, and exposure to it has been linked to various health problems, such as developmental and reproductive disorders, and carcinogenic disorders. Therefore, the development of efficient methods for removing BPA from water is of utmost importance for public health.

The use of palm kernel shell biosorbent and hematite nanocomposite membrane filtration as a hybrid system is particularly relevant because it offers a sustainable and cost-effective approach to water treatment. Palm kernel shell is a low-cost and abundant agricultural waste material that can be used as a biosorbent for removing pollutants from water. Hematite nanocomposite membrane filtration is a promising technology that offers high removal efficiency and selectivity for water treatment applications.

Therefore, the study has significant relevance for the development of sustainable and effective methods for removing BPA from drinking water, which can have far-reaching impacts on public health and environmental sustainability.

1.5 Thesis outline

The thesis comprises of five chapters: Chapter 1 provides the study background, problem statement and existing knowledge-gap, objectives, as well as scope of the study. Chapter 2 covers the comprehensive review on the occurrences and sources, adverse effects, nature and classifications of EDCs. It also gives a detailed review of various treatment techniques for removing EDCs from different water media, including the advance treatment technologies including advanced oxidation processes, adsorption, ozonation, enzymatic degradation, membrane bioreactor system, and hybrid system. The chapter also provides a state-of-art review on the rejection of EDCs using membrane technologies, membrane materials, EDCs removal mechanism during membrane processes, factors affecting membrane processes, and membrane fouling. Fouling mitigation strategies and nanomaterials and membrane technology are also adequately elucidated in the chapter. Chapter 3 present the system design, materials/equipment, experimental layout, and setup. The pictures of the equipment and analysers were all described. The analytical methods and statistical tools used were also reported in this section. Chapter 4 shows the results obtained from each of the experiment conducted. The discussion of the results, adsorbent and membrane characterizations, significant of the experimental factors were all provided in this chapter. Lastly, Chapter 5 highlight the recap of the studies, the significant findings, suggestions as well as recommendations.

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