REVIEW ARTICLE

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POTENTIAL OF FUNCTIONALISED CELLULOSE FROM OIL PALM BIOMASS AS NITROGEN AND PHOSPHORUS-BASED NUTRIENT ADSORBENT - A REVIEW

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ABSTRACT

By the year 2020, oil palm biomass in Malaysia is projected to reach between 85 to 110 million tonnes per year. Instead of disposing off such a massive amount of biomass as waste, the oil palm biomass could be converted into value-added products. Since lignocellulosic materials could be a suitable adsorbent for nitrogen and phosphorus-based nutrients from aquaculture effluent based on studies conducted by other countries, it would be an excellent opportunity to monetise oil palm biomass for a similar purpose as well. There are many well-established extraction methods introduced by researchers. However, only a handful of the extraction method involved the use of green chemicals. This article provides a review of the extraction and modification for oil palm biomass towards becoming a potential adsorbent for nitrogen and phosphorus-based nutrients.

Keywords: adsorption, cellulose, nitrate, oil palm biomass, phosphate.

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INTRODUCTION

Malaysia is the second-largest palm oil producer globally, following behind Indonesia (Abdullah and Sulaiman, 2013; Kushairi *et al.*, 2019). Oil palm was introduced to Malaya in the year 1871 and the first commercial oil palm plantation started in the year 1917 at Tennamarran Estate in Selangor, Malaya (Awalludin *et al.*, 2015; Onoja *et al.*, 2019). Since then, oil palm has been planted extensively in Malaysia until present as Malaysia's tropical weather suits the required condition to plant the oil palm trees (Kamil and Omar, 2016). Subsequently, the amount of biomass from oil palm also inadvertently increases with the extensive planting of oil palm. By the year 2020, oil palm biomass in Malaysia is projected to reach between 85 to 110 million tonnes per year (Malaysian-German Chamber of Commerce and Industry, 2017). Generally, oil palm generates six types of biomass, namely oil palm fronds (OPF), oil palm trunk (OPT), oil palm empty fruit bunch (OPEFB), fresh-pressed fibre, oil palm shell, and palm oil mill effluent (POME) (Abdullah and Gerhauser, 2008; Shuit *et al.*, 2009; Umar *et al.*, 2013; Zakaria *et al.*, 2014; Lamaming *et al.*, 2017; Mohd Rasli *et al.*, 2017; Mohtar *et al.*, 2017). According to Onoja *et al.* (2019), 75% of the oil palm biomass is from OPF and OPT while the other biomass covers the remaining 25%. Comparing OPF and OPT, OPF has higher cellulose content with 60% of the cellulose is in the basal portion, and 40% in the leaf (Malaysia Innovation Agency, 2013). However, currently, usage of OPF is limited to a few industries such

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as for steam production, bio-sugar production and fertiliser (Malaysia Innovation Agency, 2013). Rather than treating it as a common agricultural waste, the high cellulose content can be extracted and isolated for modification into a useful adsorbent.

On the other hand, other than oil palm production, Malaysia covers 1% of the world's seafood supply (Mohamed Omar, 2017b). Here is where the aquaculture industry plays a role where numerous aquaculture industrial zones were reserved for farmed fishes for the purpose of satisfying the global seafood demand (FAO, 2016). Aquaculture in Malaysia is categorised into marine and freshwater, whereby freshwater fishes are highly in demand and provide the most job opportunities compared to marine aquaculture (Mohamed Omar, 2017a). However, the downside of freshwater aquaculture is that the water used to farm fishes has to be replaced once every several months due to excessive nutrients content. Nutrients such as nitrate and phosphate are the main source of eutrophication condition (Cripps and Bergheim, 2000; Wang *et al.*, 2010; Castine *et al.*, 2013; Hokkanen *et al.*, 2016; Kim *et al.*, 2016; Yin *et al.*, 2018; Pan *et al.*, 2018; Wu *et al.*, 2019; Karthikeyan *et al.*, 2019; Lee *et al.*, 2019). It can disrupt the fishes' respiratory system and decrease the dissolved oxygen in the water (Kioussis *et al.*, 2000; Anirudhan *et al.*, 2006; Ng *et al.*, 2018). Other than aquaculture effluent, the sources of nitrate and phosphate nutrients in running water are livestock manure (Yin *et al.*, 2017) and excessive use of fertiliser in industries (Keränen *et al.*, 2013). These would eventually cause the nitrate and phosphate nutrients to leach into receiving water bodies and contaminating it. Overdose of phosphate nutrients can also cause disorder in the growth of seaweed and aquatic plants (Kioussis *et al.*, 2000; Anirudhan *et al.*, 2012; Turcios and Papenbrock, 2014). Over the concentration threshold of nitrate anion nutrients can result in methemoglobinaemia 'baby blue' syndrome in infants, formation of carcinogenic nitrosamines, and nitrosamides upon human consumption (Hamoudi and Belkacemi, 2013). Hence, proper effluent treatment is required to reduce the concentration of both nitrate and phosphate nutrients in the aquaculture effluent before it can be released to receiving water bodies or reused back into aquaculture.

Various treatments based on biological (Lin *et al.*, 2002; Akinbile and Yusoff, 2012) and physicochemical (Kioussis *et al.*, 2000; Orlando *et al.*, 2002; Wang *et al.*, 2007; Anirudhan and Senan, 2011; Keränen *et al.*, 2013; Sehaqui *et al.*, 2016; Fan and Zhang, 2018; Qiao *et al.*, 2019; Stjepanović *et al.*, 2019; Shojaipour *et al.*, 2020) methods have been studied for the adsorption of nutrients from wastewater. However, the adsorption method using biomass must be brought to attention as agriculture waste is relatively cheap, and it serves great flexibility in the aquaculture industry. Typically, activated carbon was used as an adsorbent but due to the increasing cost, many ventured into cheaper alternative solutions to adsorb the nutrients (Stjepanović *et al.*, 2019; Yu *et al.*, 2019).

Therefore, this article aims to review the current trend of adsorption of the nitrate and phosphate nutrients and delve into the potential of functionalised cellulose from oil palm biomass as nitrate and phosphate nutrients adsorbent.

ADSORPTION OF NITRATE AND PHOSPHATE

The adsorption of nitrate and phosphate nutrients from aquaculture effluent has been studied using different lignocellulosic materials and water treatments. *Table 1* shows a summary of the study on the adsorption of nitrate and phosphate anion nutrients previously conducted by other researchers. Based on the studies, the adsorption of the nutrients can generally be conducted at ambient conditions. Biological methods would require a longer duration (between weeks to months) to obtain significant results. Meanwhile, adsorption of the nutrients via adsorption method can be accomplished within hours. These studies accentuate the efficiency of the adsorption method for adsorption of the nitrogen and phosphorus-based nutrients.

The modification of polymer hydrogel was created to have positive charge ends to attract the negatively charged nitrate and phosphate by Kioussis *et al.* (2000). A hydrogel is a three-dimensional polymer with hydrophilic property. Polymer hydrogel can be made of different polymer composition depending on the desirable properties. Based on Kioussis *et al.* (2000), poly(ally amine) hydrochloride was used to make the polymer hydrogel. It is a water-soluble cationic polymer and can be highly water-swollen hydrogel with chemical crosslinking. The poly(ally amine) hydrochloride polymer hydrogel has pendant primary amino groups where it will selectively attract anion nutrients via electrostatic force and trap the nutrients in the gel network. The equilibrium adsorption of nitrate and phosphate anion nutrients was obtained in 3 hr with a neutral pH of the wastewater. In addition, the hydrogels could potentially be reusable for up to five cycles to achieve maximum adsorption. Shojaipour *et al.* (2020) conducted a study on the efficiency of nitrate adsorption on modified hydrogel with quaternary ammonium salt. The gum tragacanth biopolymer hydrogel was functionalised with N-(trimethylsilyl) imidazole and (3-chloropropyl) trimethoxysilane to adsorb nitrate anion nutrient via ion exchange. The highest adsorption of nitrate anion nutrient obtained was at a neutral pH (pH 7.0) in 20 min. This would allow the hydrogels to be conveniently used without using pH adjuster in wastewater.

TABLE 1. COMPILATION OF MULTIPLE TREATMENT TYPE FOR NITRATE AND PHOSPHATE ANION NUTRIENTS

Type of treatment	Parameter			Recovery/ removal performance	Reference
	Temperature (°C)	pH	Time		
Amine cross-linked tea waste via adsorption	25.0	3.0-10.0	60 min	Nitrate: 136.43 mg g^{-1} Phosphate: 98.73 mg g^{-1}	Qiao *et al.* (2019)
Brewer's spent grain via adsorption	25.0	4.0-10.0	60 min	Nitrate: 22.65 mg g^{-1}	Stjepanović *et al.* (2019)
Constructed wetlands system	24.3-29.6	-	1-7 months	Nitrate: 86.0%-98.0% Phosphate: 32.0%-71.0%	Lin *et al.* (2002)
Corn stalks cellulose via adsorption	25.0	6.80	6 hr	Nitrate: 13.6054 mg g^{-1} Phosphate: 22.8833 mg g^{-1}	Fan and Zhang (2018)
Gum tragacanth carbohydrate produce hydrogel via adsorption	25.0	Neutral	20 min	Nitrate: 98.26%	Shojaipour *et al.* 2020)
Nanofibres from waste pulp residues via adsorption	25.0	5.0-7.0	24 hr	Nitrate: 39.0% Phosphate: 61.0%	Sehaqui *et al.* (2016)
Novel cellulose-based via adsorption	30.0	6.0	6 hr	Phosphate: 99.1%	Anirudhan and Senan (2011)
Pine sawdust, pine bark, spruce bark, birch bark, birch pear via adsorption	25.0	3.0-10.0	5-10 min	Nitrate: 70.0%-86.0%	Keränen *et al.* (2013)
Polymer hydrogels via adsorption	25.0	7.7	3 hr	Nitrate: 50.0% Phosphate: 98.0%	Kioussis *et al.* (2000)
Sugarcane bagasse via adsorption	30.0	6.5	24 hr	Nitrate: 9.0 mg $litre^{-1}$ Phosphate: 4.0 mg $litre^{-1}$	Orlando *et al.* (2002)
Water hyacinth and lettuce via biological method	25.0-35.0	6.72	1-4 weeks	Nitrate: 90.92%, 39.78% Phosphate: 83.59%, 55.90%	Akinbile and Yusoff (2012)
Wheat straw via adsorption	24.0	6.8	150 min	Nitrate: 80%	Wang *et al.* (2007)

Besides hydrogel, lignocellulosic materials such as corn stalks, waste pulp residues, sugarcane bagasse, and wheat straws were studied as an adsorbent for nitrate and phosphate nutrients as well. These lignocellulosic materials were pre-treated to extract and isolate cellulose compound, and it was further modified via cationisation to form a positively charged ends (Orlando *et al.*, 2002; Wang *et al.*, 2007; Keränen *et al.*, 2013; Sehaqui *et al.*, 2016; Fan and Zhang, 2018; Qiao *et al.*, 2019; Stjepanović *et al.*, 2019). These positively charged ends will attract the negatively charged nitrate and phosphate ions present in the aquaculture effluent via electrostatic attraction (Keränen *et al.*, 2013; Qiao *et al.*, 2019; Stjepanović *et al.*, 2019). Electrostatic attraction force creates a strong bond between the positively and negatively charged particles and provides easy desorption by adding used adsorbent into sodium chloride (NaCl) solution (Keränen *et al.*, 2013).

Cationisation of cellulose performed to functionalise for the adsorption of nitrate and phosphate nutrients is commonly achieved via quaternary ammonium salts. This allows ammonium positive ends to be grafted onto the cellulose, which promotes the adsorption of nitrate and phosphate anion nutrients. The adsorption will then be carried out via electrostatic force and ion exchange (with the presence of chlorine anions from the chemical used for quaternary ammonium salts) (Shojaipour *et al.*, 2020).

During the adsorption of nitrate and phosphate anion nutrients, the surrounding parameters play a significant role. The varying parameters such as pH of adsorbate solution, the temperature of adsorbate solution, and contact time between the adsorbent and adsorbate should be monitored to obtain high adsorption capacities of nitrate and phosphate anion nutrients. Studies have shown that pH of aquaculture effluent within the range of 3.0-10.0 would be an optimal range for the adsorption of nitrate and phosphate (Kioussis *et al.*, 2000; Orlando *et al.*, 2002; Wang *et al.*, 2007; Keränen *et al.*, 2013; Sehaqui *et al.*, 2016; Fan and Zhang, 2018; Yin *et al.*, 2018; Karthikeyan *et al.*, 2019; Qiao *et al.*, 2019; Stjepanović *et al.*, 2019; Wu *et al.*, 2019). However, the highest adsorption capacities of the nitrate and phosphate anion nutrients were usually achieved within a pH of 5.0-6.5 (Keränen *et al.*, 2013; Qiao *et al.*, 2019; Stjepanović *et al.*, 2019). This could be because of the dominant presence of chloride (Cl^-) ions on the surface of active sites of the adsorbent when the pH is lower than 3.0, thus, reducing the electrostatic force between nitrate and phosphate anions and the active sites of the adsorbent (Stjepanović *et al.*, 2019). When the pH is higher than 10.0, the competition between OH^- ions and both nitrate and phosphate anions will occur, leading to electrostatic repulsion and lower available active sites on the surface of the adsorbent (Qiao *et al.*, 2019).

The temperature of the solution plays a vital role in the adsorption of nitrate and phosphate anion nutrients as well. As reported by Shojaipour *et al.* (2020), increasing the temperature from 25°C-45°C, the adsorption capacity of nitrate anion nutrients had decreased. The decrease in adsorption capacity is due to the exothermic reaction exerted during the adsorption process (Shojaipour *et al.*, 2020). Thus, with higher temperatures, the electrostatic bond between the nitrate anion and cationic end of adsorbent might be weakened.

The effect of contact time between the adsorbent and the nitrate and phosphate anion nutrients serve as a function to provide sufficient time for nitrate and phosphate anion nutrients to fill up all vacant pores on the surface of the adsorbent. Hence, reaching an equilibrium concentration before the used adsorbent undergoes desorption for reusability purposes. As shown in *Table 1*, constructed wetlands systems (Lin *et al.*, 2002) and biological method of adsorption via water hyacinth and lettuce (Akinbile and Yusoff, 2012) required a longer time to achieve better adsorption capacity of nitrate and phosphate anion nutrients. Although the biological method is an alternatively cheap method, it reacts relatively slow when the effluent contains a high load of nitrate and phosphate anion and low temperature (Stjepanović *et al.*, 2019). The same concept applies to constructed wetland systems where it is dependent on the microbial's reactivity to treat the nitrate and phosphate anion loaded effluent (Lin *et al.*, 2002).

OIL PALM BIOMASS

Since lignocellulosic material is currently in focus to act as an adsorbent, oil palm biomass could be a good representation of lignocellulosic material. In *Figure 1*, the detailed composition of oil palm biomass consists of cellulose, hemicellulose, and lignin. Each type of biomass has its unique characteristic which differs from one another.

Lignin is the aromatic polymer in the lignocellulose components. It has a random substitution arrangement of phenylpropane monomeric units which consist of syringly and guaiacyl *p*-hydroxylphenyl units (Klemm *et al.*, 2005). On the other hand, hemicellulose has different monosaccharide units which are pentose sugar dominant. It can exist in the forms of either homo-polymer or hetero-polymer, and it is alkali-soluble (Malaysia Innovation Agency, 2013; Kumneadklang *et al.*, 2019) Hence, it is the main component in the production of bio-sugar due to its high sugar composition and easy hydrolysis process (Malaysia Innovation Agency, 2013). Meanwhile, cellulose is a linear homopolymer formed from the repeating D-anhydroglucosepyranose (AGU) monomers which are connected by β-(1,4)-glycosidic linkages (Klemm *et al.*, 2005). Each monomer is rotated 180° with respect to one another in the arrangement of the polymer, giving it the crystalline structure. Hence, this structure makes cellulose resistant to acid and enzymatic hydrolysis.

Since oil palm biomass is a major representation of lignocellulosic material in Malaysia with an abundance amount, the cellulose content can be extracted and functionalised to adsorb nitrate and phosphate nutrient ions. This study aids in reducing the amount of oil palm biomass by expanding its product value as well as providing an alternative method in wastewater treatment.

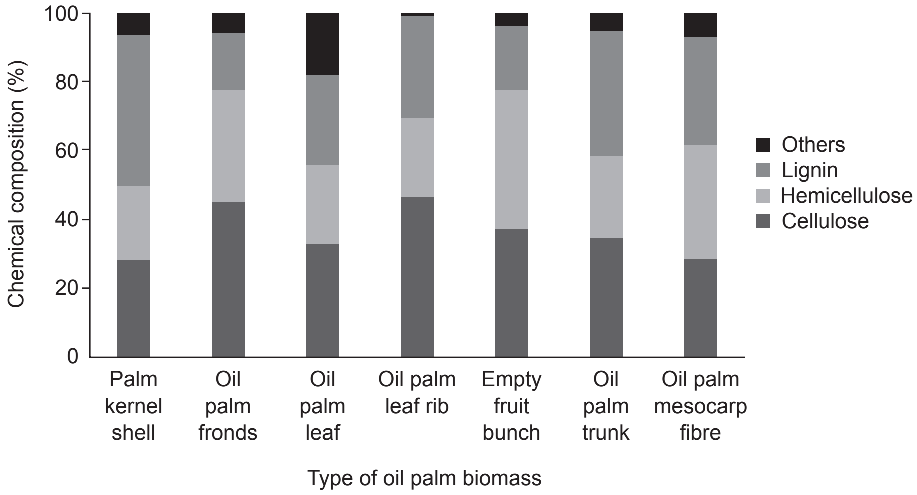

Figure 1. Chemical composition of different types of oil palm biomass (% dry weight).

GENERAL USE OF OIL PALM BIOMASS IN ADSORPTION

For the past years, oil palm biomass was studied to expand its product value (Abdullah and Gerhauser, 2008; Sumathi *et al.*, 2008; Abnisa *et al.*, 2011; Tan and Lee, 2012; Nazir *et al.*, 2013; Zakaria *et al.*, 2014; Awalludin *et al.*, 2015; Lefatshe *et al.*, 2017; Ooi *et al.*, 2017). The expansion of oil palm biomass as an adsorbent was one of the studies which are still ongoing in the current research field. Researchers had been utilising the lignocellulosic material into adsorbing pollutants, especially dyes and heavy metals, in various forms of the adsorbent. *Table 2* shows a summary of the usage of different types of oil palm biomass in adsorption of dye and heavy metal studies through the years.

Oil palm biomass had been mainly used in the study of adsorption on dyes and heavy metals. The oil palm biomass was either used in its isolated cellulose form (Hasnain *et al.*, 2007; Saman *et al.*, 2018), or it was further processed into microcrystalline cellulose (Hussin *et al.*, 2016), activated carbon (Ooi *et al.*, 2017), or biochar (Daneshfozoun *et al.*, 2017). This showed the flexibility of using lignocellulose material, such as oil palm biomass itself, to be an excellent adsorbent for charged ions with or without pre-treatment.

The introduction of oil palm biomass into adsorption started with using oil palm ash. Oil palm ash is the leftover after oil palm biomass was used in burners to produce steam. Essentially, Hasnain *et al.* (2007) discovered that this oil palm ash contains carbonaceous compounds with the presence of pores on them. Therefore, the ash was used to adsorb dyes such as disperse blue and disperse red which are commonly found in textile related industrial wastewater due to colouring of products (Hasnain *et al.*, 2007). Methylene blue was another common adsorbate for study on the adsorption towards oil palm biomass (Hussin *et al.*, 2016; Shanmugarajah *et al.*, 2019). The adsorption of dyes towards oil palm biomass was proven to be feasible under certain parameters. However, the modification of pH was often needed for high adsorption efficiency.

Another form of adsorbate used to study the adsorption capability of oil palm biomass was heavy metals. Iron (III), lead (VI), lead (II), hexavalent chromium (VI), copper (II), zinc (II), nickel (II), and manganese (II) are examples of heavy metals used by previous researchers in their field of study on adsorption of heavy metals on oil palm biomass (Hasnain *et al.*, 2008; Khosravihaftkhany *et al.*, 2013; Daneshfozoun *et al.*, 2017; Saman *et al.*, 2018). The studies indicated good adsorption performance under alkaline pH and with quick equilibrium adsorption contact time. Hence, it is safe to say that the study of adsorption on oil palm biomass with dyes and heavy metals are well established. In addition, as shown in *Table 2*, the tested methylene blue dye favoured adsorption in acidic solution while heavy metals favoured adsorption in alkaline solution. This condition would require an additive to adjust the pH of wastewater prior to the adsorption process, and it could incur additional cost and a more complex operational handling. The concentration of the dye and heavy metals in the wastewater plays a vital role as well, due to the limitation of adsorption capacity of the adsorbent. Thus, usually, the higher the concentration, the lower the percentage removal of the dye and heavy metals. The highest removal percentage (~100%) was mostly achieved in the lowest concentration level (Hasnain *et al.*, 2008). However, none of the studies conducted on the usage of oil palm biomass as adsorbent tested its feasibility for the adsorption of nitrogen and phosphorus based nutrients. Hence, the subsequent sections of this article focus on reviewing methods for the aforementioned purpose.

GREEN EXTRACTION OF CELLULOSE FROM OIL PALM BIOMASS

The extraction of cellulose from oil palm biomass has been well established over the years. Cellulose may come in many forms such as alpha-cellulose, microcrystalline cellulose, and nanocrystalline cellulose. Alpha-cellulose is the main focus in this article as it is quicker to extract from oil palm biomass as compared to the extraction of microcrystalline and nanocrystalline cellulose and separate from adsorbate solution once it is used as an adsorbent. The green method has recently been the highlight amongst researchers due to its environmental friendly advantages such as the usage of environmental friendly chemicals (*i.e.* formic acid and acetic acid) (Nazir *et al.*, 2013). This could eliminate the use of sodium chlorite as a bleaching agent due to its ability to release chlorine ions during the extraction of cellulose from oil palm biomass.

The green method can further be categorised into eco-friendly reagent, ionic liquid, ozonolysis, and hydrothermal pre-treatment (Noorshamsiana *et al.*, 2017). However, ozonolysis and hydrothermal pre-treatment methods are not suitable for the extraction of cellulose from palm oil biomass. This is because ozonolysis generally only removes the high content of lignin neglecting the removal of hemicellulose from the lignocellulosic composition (Noorshamsiana *et al.*, 2017). Meanwhile, hydrothermal pretreatment is mainly used in the extraction of glucose for bio-sugar production and bioethanol production (Malaysia Innovation Agency, 2013). In lieu, only eco-friendly reagent and ionic liquid shall be discussed in this section.

TABLE 2. COMPILATION ON THE STUDY OF OIL PALM BIOMASS AS AN ADSORBENT FOR WATER TREATMENT

Oil palm biomass	Pre-treatment	Pollutant	Type of pollutant	Parameters			Findings	Reference
				pH	Concentration	Time		
Oil palm ash	-	Disperse blue and disperse red	Dye	2.0-5.0	50-250 mg $litre^{-1}$	1-90 min	99% of adsorption was obtained a pH 2.0. Adsorption obeyed Langmuir isotherm and pseudo-second order kinetic.	Hasnain Isa *et al.* (2007)
Oil palm frond	20% active alkaline and 30% sulfidity was used to conduct Kraft Pulping process.Acetic acid, hydrochloric acid and sodium hydrochlorite was used in the extraction of microcrystalline cellulose and alpha-cellulose	Methylene blue	Dye	-	100 mg $litre^{-1}$	-	The microcrystalline cellulose showed significantly higher adsorption capacity compared to the alpha-cellulose.	Hussin *et al.* (2016)
Oil palm empty fruit bunch	NaOH and sodium chlorite were used for bleaching. The bleached sample were further hydrolised using sulphuric acid	Methylene blue	Dye	2.0-11.0	50-300 mg $litre^{-1}$	0-200 min	The highest removal efficiency (~90%) was obtained between pH of 8.0 to 10.0. Adsorption obeyed Langmuir isotherm and pseudo-second order kinetics.	Shanmugarajah *et al.* (2019)
Oil palm fibre	Sulphuric acid with heat	Hexavalent chromium (VI)	Heavy metal	1.5-5.0	20-200 mg $litre^{-1}$	0-6.0 hr	100% of removal efficiency was obtained at pH 1.5. Adsorption obeyed Freundlich and pseudo-second order kinetics.	Hasnain Isa *et al.* (2008)

TABLE 2. COMPILATION ON THE STUDY OF OIL PALM BIOMASS AS AN ADSORBENT FOR WATER TREATMENT (continued)

Oil palm biomass	Pre-treatment	Pollutant	Type of pollutant	Parameters			Findings	Reference
				pH	Concentration	Time		
Oil palm bark, oil palm frond, oil palm leaves	Wash, dry, and sieved at 200-1 400 µm	Iron (III) and Lead (VI) heavy metal ions	Heavy metal	1.5-6.0	500 mg litre^{-1}	0-80 min	The highest removal efficiency was obtained between pH 5.0-6.0, with OPB removing 78%, of iron (III) and lead (VI) respectively. Adsorption obeyed Langmuir and pseudo-second order kinetics.	Khosravihaftkhany *et al.* (2013)
Oil palm empty fruit bunch	Sonicated together with iron (II) oxide	Lead (II), Copper (II), Zinc (II), Nickel (II), Manganese (II)	Heavy metal	3.0-10.0	100-1 000 mg litre^{-1}	0-100 min	The highest removal (~90%) for each heavy metal ions was obtained between pH of 5.0-7.0.	Daneshfozoun *et al.* (2017)
Oil palm trunk	Dewaxed with toluene/ethanol (2/1, v/v), and bleached using sodium hydroxide + hydrogen peroxide	Aurum (III)	Precious metal	3.0-7.0	10-205 mg litre^{-1}	0-1440 min	100% removal was obtained in an acidic condition for low concentration of 10 mg litre^{-1}. However increasing concentration led to lower percentage removal due to the limitation of active site. Adsorption obeyed Langmuir isotherm and pseudo-second kinetics.	Saman *et al.* (2018)
Palm kernel shell	Washed with nitric acid, carbonised and treated with sulphuric acid	Urea	Uremic toxins	-	38 500 µmol	0-120 min	Adsorption of urea increase with increasing temperature of 400°C-600°C due to increasing pore sizes.	Ooi *et al.* (2017)

TABLE 3. COMPILATION ON METHODS OF CELLULOSE EXTRACTION FROM OIL PALM BIOMASS

Type of Biomass	Solvent	Parameter			Findings	Reference
		Temperature (°C)	pH	Time		
Oil palm empty fruit bunch	Fungal and phosphoric acid	-	-	-	Cellulose: 53.81%	Isroi *et al.* (2012)
Oil palm empty fruit bunch	Formic acid, sodium hydroxide, hydrogen peroxide, ethanol	85.0-121.0	7.0-11.0	90 min	Alpha-cellulose: 93.7%	Nazir *et al.* (2013)
Oil palm fronds	Soda-AQ, magnesium sulphate, hydrogen peroxide, acetic acid	70.0-160.0	4.0-5.0	30 min-1 hr	Alpha-cellulose: 96.50%	Dungani *et al.* (2017)
Oil palm frond	Sodium hydroxide, sodium chlorite	80.0	-	2-4 hr	Cellulose: 40.0%	Mohd Rasli *et al.* (2017)
Oil palm mesocarp fibre	Sodium hydroxide, acetic acid, hydrogen peroxide, sulphuric acid	70.0-160.0	-	1-24 hr	Cellulose: 88.8%	Megashah *et al.* (2018)
Oil palm empty fruit bunch					Cellulose: 93.0%	
Oil palm frond					Cellulose: 94.6%	
Oil palm trunk	Ethanol, toluene, sodium chlorite, potassium hydroxide, sulphuric acid	20.0-70.0	4.0-5.0	1-24 hr	Alpha-cellulose: 51.75%	Lamaming *et al.* (2015)
Oil palm trunk and oil palm frond	Sodium hydroxide, sulphuric acid, acetic acid, ethanol, toluene, sodium chlorite	80.0	Neutral	60 min	Alpha-cellulose: 41.55%-64.42%	Lai and Idris (2013)

Table 3 summarises the methods of cellulose extraction from oil palm biomass. Thus far, cellulose extraction from oil palm biomass could be achieved at a minimum yield of 40% using conventional solvents as a bleaching agent. According to Nazir *et al.* (2013), the replacement of sodium chlorite as a bleaching agent by a mixture solution of formic acid with hydrogen peroxide gives a great advantage in reducing the amount of hazardous ions (chloride ions) released. Coupled with steam explosion, this method can soften the lignin and gives better penetration of the bleaching agent to remove lignin and hemicellulose easily. An estimation of 93.7% of alpha-cellulose with 70% crystallinity was obtained using the method. This proves the efficiency of formic acid with hydrogen peroxide when it is coupled with steam explosion method.

Ionic liquid is another effective method in removing lignin and hemicellulose from the lignocellulosic material that originated from oil palm biomass (Noorshamsiana *et al.*, 2017). Ionic liquid is also known as a designers' solvent due to its strong dissolution properties which are made suitable to breakdown complex macromolecules and polymeric materials in the recalcitrant cellulose (Tan and Lee, 2012). According to Mohtar *et al.* (2017), ionic liquid's superior solvency properties came from its capability to disrupt strong linkages between cellulose and lignin via hydrogen bond and dipole bond formation. An added advantage of ionic liquid's method of extraction is the ability to recycle the solvent used for extraction purposes, this method also provides an alternative to replace the usage of harmful organic solvents (Mohtar *et al.*, 2017). However, the solvent used to perform ionic liquid's method is indeed costly. It is also of high viscosity and is usually less effective in removing hemicellulose compared to acid hydrolysis (Jana and Ulla, 2011). Hence, there was a need for further alkaline treatment in the process to improve the yield of cellulose (Mohtar *et al.*, 2017).

MODIFICATION OF CELLULOSE

After most of the alpha-cellulose is extracted, it can be modified chemically to enhance the electrostatic attraction between the adsorbent and adsorbate. Since nitrate and phosphate are negatively charged naturally, it would be of advantage if the adsorbent is positively charged. A modification method proposed by Orlando *et al.* (2002) using epichlorohydrin, dimethylformamide, pyridine, and dimethylamine has inspired many researchers to continue the method of modification and altered to suit different conditions. This method was tested and repeated on a few other lignocellulosic materials such as *Moringa oleifera* hull, lauan sawdust, coconut husk, rice hull, persimmon tealeaf, pine bark sugarcane bagasse (Orlando *et al.*, 2002), banana stem (Anirudhan *et al.*, 2006), wheat straw (Wang *et al.*, 2007), and corn stalk (Fan and Zhang, 2018). The findings obtained from the studies showed encouraging results with the adsorption of 80%-50% for nitrate and up to 90% for phosphate depending on the individual or selective adsorption (Orlando *et al.*, 2002; Anirudhan *et al.*, 2006; Wang *et al.*, 2007; Fan and Zhang, 2018). However, in those studies, there was no proper pre-treatment done to extract the cellulose from the lignocellulosic material. This might eventually affect the attachment of the cations towards the lignocellulosic material. Neither were most of the articles studying the selectivity of nitrate and phosphate nutrient adsorption. In lieu, it was more towards the individual adsorption of nitrate or phosphate nutrient onto the functionalised cellulose of lignocellulosic material.

Aside from the typical cationisation, there were a few studies on creating a neutral charge modification towards commercial cellulose (Anirudhan and Senan, 2011; Anirudhan *et al.*, 2012). Anirudhan and Senan (2011) modified commercial cellulose by grafting it with iron(III) and amino-functionalised poly(glycidylmethacrylate). The modified commercial cellulose showed good adsorption towards phosphate at a pH of 6.0, with a constant agitation of 200 rpm for 2.0 hr of contact time under room temperature of 20°C. These parameters assisted in the adsorption of phosphate, achieving the removal of 99.1%. Although the parameters and result outcome were promising, the modification was conducted onto commercial cellulose and would impose a higher costing price as compared to sourcing it from biomass. Another study conducted by Anirudhan *et al.* (2012) grafted commercial cellulose with epichlorohydrin, and polyethyleneimine with N,N'-methylene bisacrylamide as a crosslinking agent and azetobis isobutyro nitrile as initiator. This method also showed favourable phosphate adsorption with the removal of 99.6% at pH 4.5 for 180 min of contact time. As the desired pH is lower, it is worth noting that the phosphate ions which were predominant in the study were dihydrogen phosphate ($H_2PO_4^-$) ions instead of the typical phosphate (PO_4^{3-}) ions. This is due to the point of zero charge pH of the functionalised cellulose which is pH 4.8. Hence, any pH more than pH 4.8 will cause the surface of the adsorbent to have a negative charge instead of a positive (Anirudhan *et al.*, 2012). They were causing the cationic ends attached to the adsorbent to repel the adsorbate.

CHALLENGES AND FUTURE DIRECTION

The main challenge in the study of adsorption of nitrate and phosphate anion nutrients is the tendency of adsorbent to perform selective adsorption towards both desired nutrients. Thus far, many studies had been conducted to adsorb nitrate and phosphate anion nutrients with a variety of adsorbent material. However, most studies are conducted with either anion nutrient present in the sample solution instead of having both the existences of anion nutrient simultaneously in the sample solution (Keränen *et al.*, 2013; Qiao *et al.*, 2019; Stjepanović *et al.*, 2019). These studies would only provide an ideal situation instead of a real-life application due to other co-existing anion nutrients present which might lower the adsorption capacity of nitrate and phosphate anion nutrients in real-life application.

As for the number of researchers who had studied on the selectivity of anion nutrients in the effluent, the outcome showed that the selectivity adsorption is favourable towards sulphate anion (Orlando *et al.*, 2002; Sehaqui *et al.*, 2016). Meanwhile, in the presence of only nitrate and phosphate anion nutrients in the effluent the outcome contradicts amongst the researchers as in some situation, the nitrate anion is much favourable due to single negative electron needed to be filled (Orlando *et al.*, 2002). Others found that with the requirement of three negative electrons, phosphate anion, it is stronger to attract the cationic ends of the adsorbent (Sehaqui *et al.*, 2016). Therefore, the need of creating an adsorbent which is able to balance the selectivity of adsorption towards nitrate and phosphate anion nutrients in the presence of other anion nutrients would be another challenge in the field.

Another challenging part of this area of study is to functionalise the extracted cellulose to obtain the positively charged ends that fulfil the criteria of green chemistry. This positively charged end enables the cellulose to act as an adsorbent in adsorbing the nitrate and phosphate, which is negatively charged, via electrostatic attraction. Although there are numerous methods introduced to functionalise the extracted cellulose, these functionalisation methods kept focusing on only targeting the hydroxyl group on sixth carbon (C6). Since C6 hydroxyl group is the primary group, it is easier to functionalise it as it is more reactive (Börjesson and Westman, 2015). However, if the functionalisation method can focus even on C2 and C3 hydroxyl groups, it would increase the number of active sites in the adsorbent for nitrate and phosphate anion nutrients.

As for the parameters of adsorption, the pH of effluent is expected to remain in the neutral range, at ambient temperature and the concentration is of the pollutant could vary significantly. Therefore, to enable the application of the modified adsorbent directly by the aquaculture industry, the adsorbent should be robust and able to perform effectively without pH adjustor, water heater/cooler and concentration level indicator.

CONCLUSION

Oil palm biomass is an excellent representation of lignocellulosic material in Malaysia due to its abundant amount produced every year. Therefore, it would be a significant step forward in using the oil palm biomass as an added feature of nitrate and phosphate anion nutrients adsorbent in order to expand its product value. With proper practice of extraction and modification of cellulose using green chemicals into effective adsorbent, it can contribute towards the safety of receiving water bodies from excessive nitrate and phosphate nutrients.

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