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Vegetable oil as organic solvent for wastewater treatment in liquid membrane processes

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ABSTRACT

Stepping into a green paradigm transition era, research works that promote sustainable development while still respecting the general environmental concerns are of great interest to many researchers. This, among other factors, has inspired and driven the research works into the feasibility of replacing the conventional petroleum-based organic solvents with the vegetable oil-based ones as the membrane phase of liquid membrane in wastewater treatment. Confirming the assertion, this paper presents an overview of liquid membrane technology, the conventional petroleum-based organic solvents used as its membrane phase and application of vegetable oil as organic solvent for wastewater treatment in liquid membrane processes. The viability of vegetable oil as organic solvent for wastewater treatment in liquid membrane processes is evaluated and waste vegetable oil as a more sustainable organic solvent is highlighted.

Keywords: Vegetable oil; Organic solvent; Liquid membrane; Wastewater treatment

1. Introduction

Water plays a vital role in sustaining livelihoods and driving all socio-economic development. With the escalating industrialization and urbanization, waterways are contaminated with a wide range of diverse organic (detergent, insecticides, fuel oil, fat, grease, organic and chlorinated solvents, etc.) and inorganic (heavy metals, chemical compounds, acids, bases, etc.) pollutants by means of various industrial activities such as mining, petroleum refineries, paper and pulp, tanning, textile, food, and pesticide processing, electrical and electronic product manufacturing, metal processing and finishing and so forth. This poses a serious threat to the environment as the pollutants discharged into the waterways may cause tremendous immediate and lifelong detrimental impacts on human health and aquatic life.

Numerous physicochemical and biological techniques to treat various pollutants in the wastewater have been developed over the years, among which, chemical precipitation, coagulation–flocculation, flotation, membrane filtration, ion-exchange, adsorption, electrochemical process, aerobic and anaerobic treatment systems are the most widely used ones by various industries [1,2]. Although most of these techniques could achieve fairly high removal efficiency, there is little emphasis on the recovery of the removed pollutants which, if available, is normally carried out in a separate unit by elution with suitable reagents and this incurs additional cost [3]. Recently,

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a technique that removes and recovers solutes in a single unit, namely liquid membrane, has been given a considerable attention by many researchers [4,5]. This is on account of its pronounced advantages such as simultaneous removal and recovery of solutes in a single unit, non-equilibrium mass transfer, high selectivity, high recovery and low energy consumption [6].

Conventionally, liquid membranes use petroleum-based organic solvents, for instance kerosene- [7], toluene- [8], hexane- [9] and chloroform-based [10] ones, as their membrane phase to separate a solute from aqueous solution. These solvents are non-renewable and invariably toxic. Consequently, they are difficult to handle and often result in ecological hazard to the aquatic systems in the case of solvent loss due to entrainment in the aqueous phase [11]. Solvent loss also implies the increase in solvent consumption and, hence, a rise in the material cost since the conventional petroleum-based organic solvents could be inordinately expensive due to the limited resources. Although solvent loss due to entrainment could be minimized in some liquid membrane systems, particularly in the dispersion-free ones, solvent can still be lost by other mechanisms such as solubility in the aqueous phase, volatilization, degradation and loss in crud [11]. Therefore, to counteract the combination of environmental degradation and petroleum depletion with increasing economic development, it is imperative that wastewater is managed according to the principles of sustainable development. This endeavour is possible through the use of greener solvents such as ionic liquids [12] and vegetable oil-based organic solvents [13] in liquid membrane processes. Ionic liquids are generally considered as green solvents owing to their negligible vapour pressure. However, recent research revealed that they may pose different degrees of toxicity to living organisms [12] and most of them remain relatively expensive compared with the conventional petroleum-based organic solvents [14].

This article attempts to review the prospects towards the utilization of vegetable oil as effective organic solvent for wastewater treatment in liquid membrane processes. It provides an overview of liquid membrane and the conventional petroleum-based organic solvents used as its membrane phase. The source and composition of vegetable oil are outlined, and a comprehensive literature that compares the properties between vegetable oil and conventional organic solvent that are essential in solvent selection for liquid membrane processes is summarized. The use of waste vegetable oil as a more sustainable organic solvent is also highlighted and discussed.

2. Liquid membrane

Liquid membrane is a separation system consisting of a liquid film through which selective mass transfer of ions or molecules occurs via permeation and transport processes [15]. Its efficiency and economic advantages have designated it as the optimal solution for separation, recovery and preconcentration in chemical, biotechnology, biomedical and pharmaceutical processes [16].

There are mainly three types of liquid membranes, namely, bulk liquid membrane (BLM), supported liquid membrane (SLM), and emulsion liquid membrane (ELM) [15]. They differ in their configurations and contacts between the feed (F), membrane (M), and stripping (S) phases. Fig. 1 illustrates the schematic diagrams of some typical BLM, SLM, and ELM. In general, BLM has its F and S phases separated by a solid impermeable barrier while those of SLM are separated by the M phase which is immobilized in the pores of a microporous hydrophobic solid support. ELM, on the other hand, consists of water-in-oil emulsions formed by droplets of S phase contained in

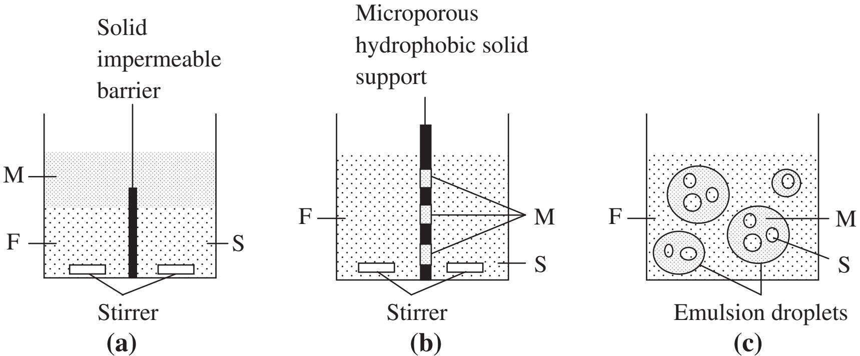

Fig. 1. Schematic diagrams of some typical (a) BLM, (b) SLM and (c) ELM.

the M phase which are suspended or dispersed in the F phase.

A comparison among different types of liquid membrane in the aspects of design, manipulation, solvent inventory, fluxes and membrane stability is presented in Table 1. BLM is the simplest type, easy to manipulate while offering good membrane stability. However, it shows relatively low fluxes due to its small interface area and long transportation path [11]. In addition, the high solvent inventory of BLM tends to increase its material cost and pose an environmental hazard in the case of solvent loss, particularly when the solvent used is toxic. SLM and ELM, on the other hand, are more complicated systems, in which the former requires the impregnation of solvent on a microporous solid support while the latter involves a multistep process [17]. Being a non-dispersive liquid membrane like BLM, SLM is also relatively easy to manipulate. Conversely, ELM, a dispersive liquid membrane which requires the control of many parameters [18], is more difficult to manipulate. An outstanding feature of SLM is its low solvent inventory due to the use of thin sheets of porous supports. Nevertheless, this gain is usually accompanied by a loss in the membrane stability due to the inevitable washing out of solvents from the pores of the support, which is mainly ascribed to lateral shear forces, progressive wetting, static pressure differential and osmotic pressure across the membrane [19]. Similar to BLM, SLM also exhibits relatively low fluxes due to its complex porous structure of the support which is accounted for by a specific tortuosity factor [17]. There is a general belief that fluxes through SLM are larger than those through BLM [11]. Nevertheless, several research workshave reported that this belief is not always borne out by experiments [20]. ELM, on the contrary, provides the highest fluxes as a result of its extremely high interfacial area [21]. However, it often employs a considerable amount of solvents and suffers from poor emulsion stability. The latter is primarily affected by the membrane formulation, technique of emulsion preparation and conditions under which the emulsion is contacted with an external phase [21].

3. Conventional petroleum-based organic solvents

The conventional petroleum-based organic solvents used as the M phase of liquid membrane are typically composed of a carrier and a diluent, where the former functions as an active component which binds and transports pollutants from one phase into another, while the latter helps to control the solvent conditions [11]. Sometimes, a phase modifier is added to improve the phase disengagement and to overcome any emulsion or third phase formation in the aqueous organic systems [22]. Table 2 summarizes some conventional petroleum-based organic solvents composing of carriers, diluents and with or without phase modifiers which are used as the M phase of different types (BLM, SLM and ELM) of liquid membrane to treat various pollutants in wastewater. The pollutants include heavy metals (transition, rare earth and alkali metals), dyes (cationic and anionic), carboxylic and amino acids, aromatic and heterocyclic compounds, polymers, and halogens. They are transported across the M phase of liquid membrane primarily by a variety of phosphorus-, amine- and oxime-based carriers. Other types of carriers such as quinolines, crown ethers, calixarenes and ammonium salts have also been applied. These carriers are dissolved in various aliphatic (kerosene, n-hexane, n-heptane, n-octane, n-decane, n-undecane, n-dodecane, n-heptanol, n-octanol, cyclohexane, dichloromethane, dichloroethane, chloroform, pentanone and cyclohexanone) or aromatic (benzene, toluene, xylene) diluents which, sometimes, are added with phase modifiers such as 1-octanol, 1-decanol and tributylphosphate (TBP) and

Table 1
Comparison among different types of liquid membrane

Aspects	Types of liquid membrane		
	BLM	SLM	ELM
Design	Simple	Complicated	Complicated
Manipulation	Easy	Easy	Difficult
Solvent inventory	High	Low	High
Fluxes	Low	Low	High
Membrane stability	Good	Poor	Poor[a]

[a]Emulsion stability.

Table 2
Conventional petroleum-based organic solvents used as M phase of liquid membrane in wastewater treatment

Organic solvent (M phase of liquid membrane)				Liquid	
Carrier	Diluent	Modifier	Pollutant[c]	Membrane[d]	Reference
Phosphorus-based					
D2EHPA[a]	Hexane	–	Cu(II)	ELM	[50]
	Kerosene	–	Cu(II)	SLM	[6]
	Kerosene	–	Nd(III)	SLM	[51]
	Kerosene, Dichloromethane, Chloroform, 1,2-dichloroethane, n-heptane, n-octane, Benzene, toluene, 4-methyl-2-pentanone	1-Decanol	Cr(III)	SLM	[52]
	Kerosene	–	Cu(II), Ni(II), Zn(II)	BLM	[33]
	Kerosene	–	Ag(I)	ELM	[53]
	Xylene	–	VO_2^+	SLM	[54]
	Hexane	–	Cationic dyes	ELM	[55]
	n-decane	–	L-lysine	ELM	[56]
TBP[a]	Kerosene	–	Cr(VI)	BLM	[57]
	Kerosene	–	Co(II)	ELM	[58]
	Kerosene	–	Cr(III)	ELM	[59]
TOPO[a]	Kerosene	–	Co(II)	ELM	[60]
	Kerosene	–	Phenol	SLM	[42]
	Cyclohexane	–	Cr(VI)	ELM	[61]
	Xylene	–	VO_2^+	SLM	[54]
PC-88A[b]	Kerosene	–	Ce(IV)	SLM	[62]
Cyanex 272[b]	Kerosene	TBP[a]	Co(II), Li(I)	SLM	[63]
Cyanex 301[b]	Chloroform, n-hexane	–	Methylene blue	BLM	[9]
Cyanex 923[b]	Kerosene	–	Phenol	ELM	[64]
Amine-based					
Tri-n-octylamine	Kerosene	–	Co(II), Ni(II)	ELM	[65]
	Kerosene	1-Octanol	Fe(II), Fe(III)	SLM	[7]
	Di-chloroethane	–	Lignosulfonate	BLM	[66]
	Di-chloroethane	–	Lignosulfonate	SLM	[67]
	Di-chloroethane	–	Lignosulfonate	ELM	[68]

(Continued)

Table 2 (*Continued*)

Organic solvent (M phase of liquid membrane)				Liquid	
Carrier	Diluent	Modifier	Pollutant[c]	Membrane[d]	Reference
	Di-chloroethane	–	Lignosulfonate, Hg(II)	SLM	[69]
Triisooctylamine	Kerosene	–	Co(II)	ELM	[70]
Tri-n-dodecylamine	Cyclohexane	–	Ag(I)	SLM	[71]
	Kerosene	–	Remazol red 3BS	ELM	[5]
Triethanolamine	Cyclohexanone	–	Mn(II)	SLM	[72]
Oxime-based					
LIX 860I[b]	Kerosene	–	Ni(II)	SLM	[73]
LIX 84I[b]	Kerosene	–	Cu(II)	ELM	[74]
LIX 984N[b]	Kerosene	–	Cu(II)	BLM	[75]
Miscellaneous					
Quinolines	Kerosene	–	Co(II)	ELM	[76]
Crown ethers	Toluene	–	Ag(I)	SLM	[8]
Calixarenes	Dichloromethane	–	Cr(VI)	BLM	[43]
Ammonium salts	Kerosene	–	Cr(VI)	ELM	[77]
	Kerosene	–	Lactic acid	SLM	[78]
	n-heptane	–	Gibberellic acid	ELM	[79]
Mixed carriers					
DP8R[b]+Acorga M5640[b]	n-paraffin (C_{12}–C_{15})	Fatty ester	Co(II)	SLM	[80]
PC-88A[b]+Cyanex 923[b]	n-dodecane	–	U(VI)	SLM	[81]
Dinonylphenylphosphoric Acid+Cyanex 923[b]+ TBP[a]+TOPO[a]	n-paraffin (C_{12}–C_{14})	–	U(VI)	SLM	[82]
No carrier					
–	n-heptane	–	Crystal violet, Methylene blue	ELM	[83]
–	n-heptane, n-hexane, kerosene	–	Congo red	ELM	[84]
–	n-hexane	–	Bisphenol	ELM	[85]
–	Kerosene	–	Pyridine	ELM	[18]
–	Kerosene	–	Iodine	BLM	[86]
–	n-undecane,		Ethylbenzene,	SLM	[87]
–	n-dodecane, n-heptanol, n-octanol	–	Nitrobenzene		

[a] Abbreviations of carriers: D2EHPA: di-2-ethylhexylphosphoric acid; TBP: tributylphosphate; TOPO: trioctylphosphine oxide.

[b] Active components of carriers: P204: 2-ethylhexyl phosphoric acid mono-2-ethylhexyl ester; Cyanex 272: bis-2,2,4-trimethyl pentyl phosphinic acid; Cyanex 301: bis-2,4,4-tri-

methyl pentyl-dithiophosphinic acid; Cyanex 923: trialkylphosphine oxides; LIX 860I: 5-dodecylsalicylaldoxime; LIX 84I: 2-hydroxy-5-nonyl-acetophenone oxime; LIX-984N: 2-hydroxy-5-nonylacetophenone oxime; DP8R: alkylphosphoric acid; Acorga M5640: salicylaldoxime derivative.

[c] Types of pollutant:

- Heavy metals
- Cu(II): copper(II); Cr(III): chromium(III); Cr(VI): chromium(VI); Co(II): cobalt(II); Nd(III): neodymium(III); Ni(II): nickel(II); Zn(II): zinc(II); Ag(I): argentum(I); VO_2^+: vanadium ion; Li(I): lithium(I); Fe(II): iron(II); Fe(III): iron(III); Hg(II): mercury(II); Mn(II): manganese(II); U(VI): uranium(VI).
- Dyes
- Cationic: Methylene blue, crystal violet; Anionic: Remazol red 3BS, congo red.
- Carboxylic acids: Lactic acid, gibberellic acid.
- Amino acids: L-lysine
- Aromatic compounds: Phenol, bisphenol, ethylbenzene, nitrobenzene
- Heterocyclic compounds: Pyridine
- Polymers: Lignosulfonate
- Halogens: Iodine

[d] Types of liquid membrane: BLM: bulk liquid membrane; SLM: supported liquid membrane; ELM: emulsion liquid membrane.

fatty ester. Attempts to treat some pollutants in wastewater by using mixtures of different types of carriers, as well as without using any carriers in liquid membrane processes have also been reported. The latter is achievable when the pollutants are readily soluble in the diluents according to the "like dissolves like" aphorism [23]. All of these organic solvents used are petroleum-based and, in spite of their efficiency as the M phase of liquid membrane in wastewater treatment, many are toxic and their harms to the environment and human health often far outweigh their benefits to society.

4. Vegetable oil

Vegetable oil is a lipid material derived from an assortment of fruit and seeds of plants such as soybeans, oil palm fruit, palm kernels, rapeseeds, sunflower seeds, cotton seeds, coconuts, corns, peanuts, rice grains and so forth. It is an important commodity produced around the world, mainly for food purposes (80%) and oleochemical industries (14%), as well as for applications in animal feed, pet food and cosmetics (6%) [24]. Being a biological material, vegetable oil is non-toxic, biodegradable and renewable.

Vegetable oil consists almost exclusively of non-polar lipids (>92%) besides having other minor components such as polar lipids (phospholipids and sphingolipids) (<4%), free fatty acids (<2%), unsaponifiable matters (phytosterols, tocopherols and hydrocarbons) (<2%), and trace amounts of heavy metals [24]. The non-polar lipids are composed of triglycerides, diglycerides and monoglycerides which are glycerol molecules with three, two and one long-chain fatty acids attached at the hydroxyl groups via ester linkage. Triglyceride, also called triacylglycerol, is the main constituent (>90%) of nonpolar lipids contained in vegetable oils [24]. It has a wide diversity of structures with different fatty acids attached at the hydroxyl groups, depending on the fatty acid composition of a particular vegetable oil. Table 3 presents the fatty acid composition of some common vegetable oils which include corn, canola (rapeseed), sunflower, soybean and palm oils. All of these oils consist mostly of oleic (18:1) and linoleic (18:2) acids (unsaturated fatty acids), except for palm oil which contains mainly of palmitic (16:0) (unsaturated fatty acid) and oleic acids. These major fatty acids form the basis of the typical triglycerides of corn, canola, sunflower, soybean and palm oils (Table 4). The structure of a typical triglyceride of corn, sunflower, and soybean oils, namely, LLL, where L is linoleic acid (18:2), is illustrated in Fig. 2.

Table 3
Fatty acid composition (wt.%) of corn, canola, sunflower, soybean and palm oils

Vegetable oils						
Fatty acids	C:D[a]	Corn	Canola	Sunflower	Soybean	Palm
Lauric	12:0	⩽0.05-0.3%	⩽0.05%	⩽0.05-0.1%	⩽0.05-0.1%	⩽0.05-0.5%
Myristic	14:0	⩽0.05-0.3%	⩽0.05-0.2%	⩽0.05-0.2%	⩽0.05-0.2%	0.5-2.0%
Palmitic	16:0	8.6-16.5%	2.5-7.0%	5.0-7.6%	8.0-13.5%	39.3-47.5%
Palmitoleic	16:1	⩽0.05-0.5%	⩽0.05-0.6%	⩽0.05-0.3%	⩽0.05-0.2%	⩽0.05-0.6%
Stearic	18:0	⩽0.05-3.3%	0.8-3.0%	2.7-6.5%	2.0-5.4%	3.5-6.0%
Oleic	18:1	20.0-42.2%	51.0-70.0%	14.0-39.4%	17.0-30.0%	36.0-44.0%
Linoleic	18:2	34.0-65.6%	15.0-30.0%	48.3-74.0%	18.0-59.0%	9.0-12.0%
Linolenic	18:3	⩽0.05-2.0%	5.0-14.0%	⩽0.05-0.3%	4.5-11.0%	⩽0.05-0.5%
Arachidic	20:0	0.3-1.0%	0.2-1.2%	0.1-0.5%	0.1-0.6%	⩽0.05-1.0%
Gadoleic	20:1	0.2-0.6%	0.1-4.3%	⩽0.05-0.3%	⩽0.05-0.5%	⩽0.05-0.4%
Eicosadienoic	20:2	⩽0.05-0.1%	⩽0.05-0.1%	⩽0.05%	⩽0.05-0.1%	⩽0.05%
Behenic	22:0	⩽0.05-0.5%	⩽0.05-0.6%	0.3-1.5%	⩽0.05-0.7%	⩽0.05-0.2%
Lignoceric	24:0	⩽0.05-0.5%	⩽0.05-0.3%	⩽0.05-0.5%	⩽0.05-0.5%	⩽0.05%

[a] C: Number of carbon atoms; D: Number of double bonds.
Source: Soon Soon Oil Mill Sdn. Bhd., Malaysia.

Table 4
Typical triglycerides of corn, canola, sunflower, soybean, and palm oils [96,94,95,97,25]

Oil types	Triglycerides[a]
Corn	LLO, LLL, OOL, PLO
Canola	LOO, OOO, LnOO
Sunflower	OLL, OOL, LLL
Soybean	LLL, LLO, LLP, LOO, LOP
Palm	POP, POO, PLO, PLP, PPP

[a] Abbreviation of fatty acids: L = Linoleic acid; Ln = Linolenic acid; O = Oleic acid; P = Palmitic acid.

5. Comparison of properties between vegetable oil and conventional petroleum-based organic solvent

Understanding of the solvent properties like melting point, dielectric constant, Hildebrand solubility parameter, specific gravity, viscosity, flash point, vapour pressure and threshold limit value (TLV) is vital in the selection of a suitable solvent as the M phase of liquid membrane. Table 5 presents the properties of some common vegetable oils (corn, canola, sunflower and soybean oils) and conventional petroleum-based organic solvents (kerosene, n-hexane, toluene and chloroform (diluents from Table 2)). The corresponding properties of water as a reference liquid are also provided. A comparison of properties between vegetable oils and conventional petroleum-based organic solvents is elucidated as follows:

5.1. Melting point

As shown in Table 5, the melting points of vegetable oils and conventional organic solvents are extremely low (<0°C) and, hence, both are liquids at ambient conditions. This liquid state of aggregation of matter is an important feature of solvents applicable

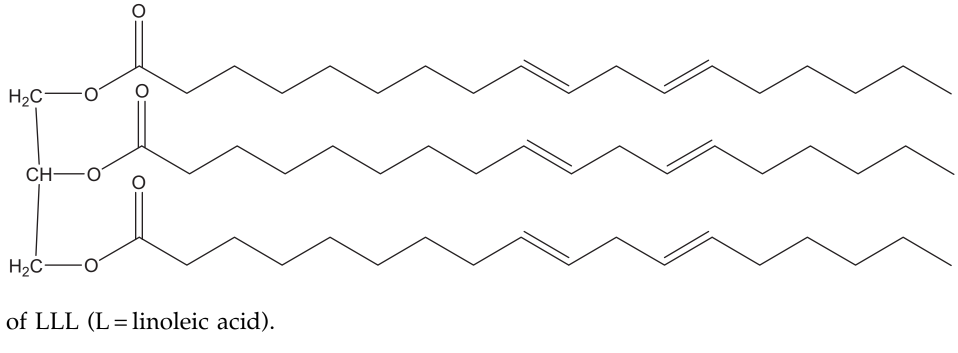

Fig. 2. Structure of LLL (L = linoleic acid).

Table 5
Properties of some vegetable oils and conventional petroleum-based organic solvents

Properties	Vegetable oils				Conventional petroleum-based organic solvents				
	Corn	Canola	Sunflower	Soybean	Kerosene	n-Hexane	Toluene	Chloroform	Water†
T_m @ 1 atm (°C)	-15^{*a}	-9^{b}	-15^{a}	-15^{a}	-51^{c}	-97^{d}	-95^{d}	-64^{d}	0^{d}
ε @ 25 °C	$\sim3^{e}$	$\sim3^{e}$	$\sim3^{e}$	$\sim3^{e}$	1.80^{f}	1.88^{g}	2.38^{g}	4.89^{g}	80^{g}
δ @ 25 °C (MPa)$^{1/2}$	–	–	–	15.8 (333 K)^{h}	15.7^{i}	15.0^{g}	18.8^{g}	19.5^{g}	48^{g}
SG @ 25 °C	0.919^{j}	0.916^{*k}	0.921^{*l}	0.916^{m}	0.823^{n}	0.657^{n}	0.865^{n}	1.47^{n}	1
μ @ 25 °C (mPa.s)	49^{o}	54^{o}	44^{o}	47^{o}	1.64^{n}	0.297^{n}	0.550^{n}	0.530^{n}	0.890^{d}
T_f @ 1 atm (°C)	335^{*j}	283^{*k}	121^{*l}	328^{*m}	50^{*p}	-22^{c}	4.5^{c}	–	–
P_{vap} @ 25 °C (kPa)	–	–	–	0.453^{q}	5.79^{*p}	20.13^{d}	3.78^{d}	26.00^{d}	3.17^{d}
TLV-TWA (mg/L)	–	–	–	–	100,000^{r}	50^{c}	100^{c}	10^{c}	–

T_m = melting point; ε = dielectric constant; δ = Hildebrand solubility parameter; SG = specific gravity; μ = viscosity; T_f = flash point; P_{vap} = vapour pressure; TLV-TWA = TLV-time weighted average; ^{a}[27]; ^{b}[88]; ^{c}[36]; ^{d}[89]; ^{e}[90]; ^{f}[91]; ^{g}[23]; ^{h}[92]; ^{i}[93]; ^{j}[94]; ^{k}[95]; ^{l}[96]; ^{m}[97]; ^{n}[98]; ^{o}[28]; ^{p}[99]; ^{q}[100]; ^{r}[102]; *Average value; †Water is given as a reference liquid.

to liquid membrane processes since they are normally operated under ambient conditions. Palm oil, which has a high melting point (34°C) [25], is a semi-solid at room temperature (25°C) and, hence, can only be used as the M phase of liquid membrane at elevated temperatures under ambient pressure.

5.2. Dielectric constant

The dielectric constant of a solvent provides a rough estimate of the solvent polarity. Generally, solvents with dielectric constants of more than 15 are polar while those with dielectric constants of less than 15 are non-polar [23]. For instance, water, which has a dielectric constant of about 80 at room temperature (Table 5), is a highly polar solvent. Conversely, vegetable oils and conventional organic solvents, which possess much lower dielectric constants (<5) at room temperature (Table 5), are highly nonpolar solvents. The polarity of solvents affects the solubility of solutes. A popular aphorism used for predicting solubility is "like dissolves like", which indicates that polar solvents dissolve polar solutes best and nonpolar solvents dissolve nonpolar solutes best [23]. Following this aphorism, the vegetables oils and conventional organic solvents presented in Table 5 are expected to dissolve non-polar solutes well instead of the polar ones.

5.3. Hildebrand solubility parameter

Hildebrand solubility parameter (δ) provides a numerical estimate of the degree of interaction between materials which can be used to quantify the relative solubility between them [26]. In general, materials with similar values of δ ($\Delta\delta < 4$) are likely to be miscible while those with dissimilar values of δ ($\Delta\delta > 4$) are unlikely to be miscible [23]. At room temperature, water has a δ of 48 (MPa)$^{1/2}$ (Table 5), while vegetable oils and conventional organic solvents have δ ranging from 15 to 20 (MPa)$^{1/2}$ (Table 5). As a result, the vegetable oils and conventional organic solvents are almost completely immiscible with water, but are totally miscible with one another at temperatures above their melting points [23,27]. The immiscibility of organic solvents in water promotes good phase disengagement in aqueous organic systems like liquid membrane.

5.4. Specific gravity

Specific gravity (SG), also called relative density, is the ratio of the density of a substance to that of water at a specific temperature. As can be seen in Table 5, all vegetable oils and conventional petroleum-based organic solvents (except for chloroform) have SG of less than 1 at room temperature and, consequently, float on water. This property provides a good understanding with regard to the configuration of aqueous and organic phases in liquid membrane systems.

5.5. Viscosity

Viscosity measures the thickness or resistance of liquids to flow. Both the vegetable oils and conventional petroleum-based organic solvents follow the Newtonian flow behaviour, that is at a given temperature, a constant viscosity is obtained independent of the force applied [27,28]. In general, vegetable oils are higher in viscosity than the conventional petroleum-based organic solvents (Table 5) which can be ascribed to the intermolecular attractions of the long-chain fatty acids in the vegetable oils [27]. This is a rather undesirable trait in liquid membrane processes as the high viscosity of vegetable oils may impede the transport of solutes across the M phase [29]. To overcome this setback, the viscosity of vegetable oils can be reduced by heating to high temperatures [28,30,31], mixing with suitable viscosity reducers [32] or stirring to minimize the boundary layer resistance (concentration polarization) [33,34]. Alternatively, a liquid membrane system that requires a low solvent inventory such as the thin-sheet SLM can be used to alleviate the transport difficulties stemming from the application of vegetable oils as the M phase [35].

5.6. Flash point

Flash point is the temperature at which the volatiles of liquids can vapourize to form an ignitable mixture in air [27]. Table 5 shows that all vegetable oils have high flash points (>100°C) and, thus, are non-flammable and safe to be used at room temperature. The conventional petroleum-based organic solvents, on the other hand, have much lower flash points, particularly n-hexane and toluene (<5°C) and, thereby, are highly flammable liquids. These flammable solvents pose fire and explosion hazards if they are mishandled or abused. While kerosene is also known for its flammability, chloroform, however, is practically nonflammable [36].

5.7. Vapour pressure

Vapour pressure is the pressure associated with the vapour in equilibrium with a condensed phase at a specific temperature. It measures the tendency of a liquid to vapourize into a gaseous state and, hence, the liquid volatility. At room temperature, vegetable oils are virtually non-volatile due to their low vapour pressures, for instance 0.453 kPa for soybean oil (Table 5). The high vapor pressures of conventional petroleum-based organic solvents, on the other hand, have rendered them volatile at room temperature, with chloroform being the most volatile, followed by n-hexane, kerosene and toluene. This poses a threat of exposure to airborne contaminants when volatile organic compounds are released into the air [36].

5.8. TLV

TLV measures the extent to which a person maybe safely exposed to a hazardous substance without endangering his or her health. It is an estimate based on the known toxicity in humans or animals, as well as the industrial experience [37]. TLV can be expressed in three standard forms: as an 8 h time-weighted average (TLV-TWA), as a 15 min short-term exposure limit (TLV-STEL) and as an instantaneous ceiling limit (TLV-C), among which, TLV-TWA is the most common measure of occupational exposure limit utilized by engineers and safety professionals [38]. The TLV-TWA values of some conventional organic solvents are shown in Table 5. Based on these values, the order of decreasing estimated toxicity of conventional petroleum-based organic solvents is chloroform>n-hexane>toluene>kerosene. Vegetable oils, which are widely acknowledged for their non-toxicity attributes [39], do not have any TLV-TWA values reported in the literatures.

6. Application of vegetable oil as organic solvent for wastewater treatment in liquid membrane processes

In light of the downsides of conventional petroleum-based organic solvents and the favo-rable properties of vegetable oil (Table 5), attempts to apply vegetable oil as the organic M phase of liquid membrane in wastewater treatment are slowly gaining momentum in recent years. Table 6 summarizes three different types of vegetable oil-based organic solvents that have been used as the M phase of liquid membrane by numerous researchers in treating various organic and inorganic pollutants in wastewater. They are vegetable oil alone, vegetable oil loaded with carrier and vegetable oil loaded with carrier and phase modifier.

As shown in Table 6, vegetable oil alone, for instance palm, coconut, sunflower and peanut oils, has been used successfully to transport pollutants like phenol, astacryl golden yellow, rhodamine B, acetic acid and Hg(II) across liquid membrane. These pollutants are generally polar in nature and, thus, do not dissolve in the non-polar vegetable oil readily. However, dissolution of polar pollutants in vegetable oil is possible when they are in their unionized or molecular state. This endeavour can be met by carefully manipulating the experimental conditions of liquid membrane such as the pH of aqueous F phase so that the polar pollutants remain in their unionized or molecular state at all times [40]. Meanwhile, vegetable oil loaded with carrier, namely, coconut oil loaded with tri-n-octylamine or D2EHPA, as well as vegetable oil loaded with carrier and phase modifier, namely, soybean oil loaded with D2EHPA and TBP, have been used effectively to transport pollutants like Hg(II), Cu

Table 6
Vegetable oil-based organic solvents used as M phase of liquid membrane in wastewater treatment

Vegetable oil-based organic solvent				Liquid membrane	Recovery efficiency (%)	Reference
Carrier	Diluent	Modifier	Pollutant			
Vegetable oil alone						
–	Palm oil	–	Phenol*	SLM	100	[40]
–	Palm, Coconut, Sunflower oils	–	Astacryl Golden yellow*	SLM	>90	[41]
–	Palm, Coconut, Sunflower oils	–	Rhodamine B*	SLM	>90	[35]
–	Palm, Coconut, Sunflower, Peanut oils	–	Acetic acid*	SLM	100	[103]
–	Coconut oil	–	Hg(II)*	SLM	~90	[41]
Vegetable oil loaded with carrier						
Tri-n-octylamine	Coconut oil	–	Hg(II)	SLM	95	[41]
D2EHPA	Coconut oil	–	Cu(II)	SLM	>90	[104]
D2EHPA	Coconut oil	–	Methyl violet, Rhodamine B	SLM	>90	[105]
Vegetable oil loaded with carrier and phase modifier						
D2EHPA	Soybean oil	TBP	Cu(II)	BLM	>90	[13,106]

D2EHPA: di-2-ethylhexylphosphoric acid; TBP: tributylphosphate; BLM: bulk liquid membrane; SLM: supported liquid membrane; Hg (II): mercury(II); Cu(II): copper(II).
*Unionized state.

(II), methyl violet and rhodamine B. It was found that while Hg(II) could be transported across liquid membrane by using either vegetable oil alone or vegetable oil loaded with carrier as the M phase, its transport rate was relatively higher in the latter due to the presence of a carrier in the M phase [41]. As yet, application of vegetable oil-based organic solvents as the M phase of liquid membrane is mainly found in both SLM and BLM (Table 6), with the former being more prevalent and, to our knowledge, there has not been any report of its application in ELM. These vegetable oil-based liquid membranes were reported to achieve high recovery efficiencies of up to 100% (Table 6) which are as well as, if not better than, their conventional petroleum-based counterparts [42,43].

7. Waste vegetable oil—a more sustainable organic solvent

With the increasing world population and food demand, the world is currently facing the global food crisis where there are about 1 billion people starving for food in the developing countries [44]. Hence, the use of a food commodity such as vegetable oil in wastewater treatment would exacerbate the food crisis and, thus, is less attractive and should be avoided for future global food security. Moreover, cultivation of terrestrial oleaginous food crops for vegetable oil poses another challenge due to the decreasing amounts of arable land and freshwater [45]. To overcome these challenges, waste vegetable oil could be used as a more sustainable organic solvent for wastewater treatment in liquid membrane processes.

Waste vegetable oil is a waste product generated abundantly by household kitchens, fast-food restaurants and other food processing facilities. McDonald's Malaysia, for instance, is producing approximately 12,000 kg of waste vegetable oil every month [46] which is amounting to a sum of 144,000 kg of waste oil per year. Although some of this waste vegetable oil is recycled into household items like soaps and candles, a major part of it is dumped illegally into rivers and landfills, causing severe environmental pollution [47]. Being a waste product, waste vegetable oil has the additional advantages of not threatening the food security and is cheaper in cost than the fresh unused oil while still featuring green attributes like non-toxic, non-flammable, non-volatile and biodegradable. Therefore, the use of waste vegetable oil as organic solvent for wastewater treatment in liquid membrane processes would contribute towards a cleaner environment while promoting a more sustainable development in liquid membrane processes. It is predicted that waste vegetable oil would take up the same diluent role as the fresh unused oil in transporting pollutants across liquid membrane (Table 6) due to their analogous fatty acid composition and mean molecular weight [48]. However, the higher free fatty acid, glycerol and water contents in waste vegetable oil as a result of oxidation, hydrolytic and cracking reactions that take place during frying [49] might affect its efficiency in removing and recovering pollutants from wastewater. To our knowledge, there has not been any report on the use of waste vegetable oil as organic solvent for wastewater treatment in liquid membrane processes.

8. Conclusions

Conventionally, the membrane phase of liquid membrane is formed by organic solvents derived from petroleum resources and, thus, is toxic, non-renewable and could be extremely expensive due to the limited resources. The prevalence of vegetable oil as a viable organic solvent for wastewater treatment in liquid membrane processes can be evaluated from three aspects: renewability, green features and efficiency. Different types of vegetable oil-based organic solvents that have been used as the membrane phase of liquid membrane in treating wastewater containing pollutants like dyes and heavy metals include vegetable oil alone, vegetable oil loaded with carrier, and vegetable oil loaded with carrier and phase modifier. The various types of vegetable oil that have been applied include palm, coconut, sunflower, peanut and soybean oils. Nevertheless, to safeguard the global food security, waste vegetable oil could be used as a more sustainable organic solvent for wastewater treatment in liquid membrane processes.

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