



A Comprehensive Review on Green Emulsion Liquid Membrane and Its Applicability Towards the Removal of Contaminants from the Aquatic Streams

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Abstract Industrialization and the rise in population have led to the larger utilization of resources which has become the supreme risk for the environment. Different types of pollutants enter the aquatic environment from various sources which create a threat for the aquatic organisms and humans. Many separation techniques like precipitation, adsorption, reactive distillation, ion exchange, electro dialysis, solvent extraction, and ultrafiltration are available, but these techniques have various limitations like the use of excessive and expensive chemicals, high

energy requirement, sludge formation, requirement of utilities in large amount and so. The green emulsion liquid membrane (GELM) is an emerging and promising method that incorporates the traits of ELM for the removal of various pollutants, metal ions, acids, and so on. In the present scenario, much focus has been diverted towards the use of green solvents derived from vegetable and plant origin. These solvents are environmentally friendly and economically viable making the ELM process more reliable. The traditionally used petroleum-based solvents for ELM formation are expensive, toxic, volatile in nature, and are detrimental to the environment. The present study tries to address the recent advancement in the field of GELM. The different factors like concentration of surfactant, carrier, types of diluents, effect of volume ratio of external feed phase to emulsion, agitation speed, effect of internal aqueous phase concentration, and emulsification time play a substantial role in the removal of several pollutants from the aqueous

Highlights

- GELM: A sustainable and reliable advancement in the field of ELM.
- Oils based on vegetable and plant origin are used as green solvents.
- GELM formulation and its transport mechanism are discussed.
- Parametric and statistical studies of GELM.
- Real-time diagnosis of water quality.

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streams through GELM have been discussed in detail. The statistical analysis of the operating variables as executed by different investigators is also mentioned.

Keyword Extraction · Vegetable Oils · Green Solvents · Green Emulsion Liquid Membrane

1 Introduction

Industrial development and booming of population growth are accompanying environmental pollution, especially in developing nations. Rapid evolution of chemical and process industries has led to the greater consumption of resources which has become an utmost threat to the ecosystem. In the era of urbanization, water resources are at high and constant risk of getting polluted. Significant quantities of pollutants such as acids, alkalis, dyes, pesticides, oils, grease, heavy metals, and pharmaceutical compounds are dumped into the freshwater bodies through municipal sewage waste, agricultural waste, industrial waste, landfill leachate, and the waste generated from medical institutions. This creates an ecological imbalance in the environment (Yan et al., 2021; Yaseen et al., 2021). To improve health conditions and to increase the life span of humans and animals, various pharmaceutical active compounds like analgesics, beta-blockers, lipid regulators, anti-inflammatory drugs, and estrogens are been utilised throughout the world (Olasupo & Suah, 2021). The pharmaceuticals compounds and different preservatives like acetaminophen, ibuprofen, ketoprofen, chlorpheniramine, diclofenac, norfloxacin, ethylparaben, methylparaben, and so on are noticed in the water bodies and their adverse effect are reported by the several authors (Ahmad et al., 2019; Dâas & Hamdaoui, 2014; Razo-Lazcano et al., 2018; Kohli et al., 2019a; Muthusaravanan et al., 2019; Kohli et al., 2020a; Shirasangi et al., 2021).

Moreover, low-level discharge of dyes generates numerous adverse effects like prevention of the light and decrease in gas solubility and is also harmful to aquatic life (Fetimi et al., 2021). Fetimi et al. (2021) reported that during the dyeing process, 10–25% of textile dyes are wasted and 2–20% are directly released as the aqueous stream in various water

resources. The US Environment Protection Agency (EPA) observed that heavy metal contamination is a major and a critical issue nowadays (Sujatha et al., 2021a). The occurrence of heavy metals is one of the main reasons for the existence of different diseases like osteoporosis, vomiting, and neurological disorders (Zereshki et al., 2021). With the upward thrust in the financial system and technological enhancement, the electric and digital device production is increasing day by day and is also turning out to be the main aspect of solid waste generation. Printed circuit board is an important component of every electrical and electronic equipment which contains different rare earth metals and valuable metals such as gold, silver, and platinum. Hence recovery of these metals is very important (Zhou et al., 2021).

Numerous techniques such as precipitation, adsorption, reactive distillation, ion exchange, electro dialysis, solvent extraction, and ultrafiltration are being used in the area of separation science and technology, but these methods have some limitations. The precipitation method requires lots of non-eco-friendly chemicals (Ooi et al., 2015). The adsorption process too becomes expensive when pure adsorbents are used (Elsagh et al., 2017). Compounds with low volatility lead to higher energy consumption and also produce high boiling side products in reactive distillation (Kumar et al., 2019a). In the ion exchange method, recovery of product is more difficult, as it highly depends on the resin structure and the reuse of ionic material requires a chemical reagent which is expensive and creates further pollution (Peng & Guo, 2020).

The membrane-based separation processes are considered one of the cleanest and energy-saving processes for the treatment of aqueous as well as gaseous stream contaminants. Also, nowadays the liquid membrane is widely used in different areas like chemistry, chemical engineering, environmental science, and hydrometallurgy (Abbassian & Kargari, 2016). Amid the various membrane-based methods, liquid membrane (LM) has grown into an established unit operation for a diverse range of separations. LM is used in wastewater treatment and industries pertaining to chemical, pharmaceutical, food processing, biotechnology, textile, environmental engineering, pulp, and paper (San Román et al., 2010). LM consists of two phases of the same nature, but dissimilar composition being divided by the third phase having

discrete features. The third phase should be insoluble in the other two phases and is said to be LM (Kislik, 2012). LMs are efficient for the elimination and reclamation of compounds from aquatic streams as they offer merits like low solvent consumption, little operational cost, instant extraction, and stripping in a single unit (Kohli et al., 2021a).

Liquid membranes are mainly classified into three different types: Bulk liquid membrane (BLM), supported liquid membrane (SLM), and emulsion liquid membrane (ELM) (Rouhani et al., 2020). BLM contains the external feed phase and internal aqueous phase which are separated with the help of a third liquid phase (membrane). The BLM is simple and continuous, but has few limitations like having small interfacial area and low mass transfer rate and requiring large amount of solvent and economically not affordable on the industrial scale (Chang, 2016). SLM is prepared by stabilizing the membrane fluid on the solid base. In the SLM, hollow fibers (Kohli et al., 2021b), flat sheets (Zante et al., 2020), and spiral wound (Wang et al., 2020) are most commonly used. SLM offers high interfacial area, low energy consumption, ease of use, and high selectivity but also suffers from limitations like membrane replacement and use of toxic and volatile solvents that make SLM more unstable (Parhi, 2013). Also, recently Sarang et al. (2022) too used the concept of an artificial neural network approach with a pseudo-emulsion hollow fiber strip method for the separation of ethylparaben and diclofenac.

In the current study, the authors have attempted to explore the green emulsion liquid membrane (GELM) technique as another approach for the separation of different metal ions and compounds from the aqueous streams in place of the conventionally used methods. Moreover, the investigation also focuses on sustainable and reliable advancement in the field of ELM. The working principle, transport mechanism, effect of different parameters like concentration of surfactant, carrier, types of diluents, effect of volume ratio of external feed phase to emulsion, agitation speed, effect of internal aqueous phase concentration, influence of emulsification time on the removal of several targeted solutes from the aqueous have been discussed in detail. Moreover, the influence of statistical tools on extraction efficiency and real-time diagnosis of water quality are also elaborated in depth.

2 Emulsion Liquid Membrane

ELM was first used by Li (1968) for the separation of hydrocarbons. ELM is a three-phase dispersion system: innermost phase, intermediate phase, and outer phase. The innermost phase is an internal phase (stripping agent), an intermediate phase is a membrane phase that contains the carrier dissolved in suitable diluents along with surfactant to emulsify the emulsion droplets, and the outer phase is the external feed phase. The external feed phase contains the solute to be separated from the feed solution. The concentration gradient is the driving force for the solute transport through the membrane (Jusoh et al., 2016). Some of the merits of ELM as compared to the other processes are as follows: (1) combines the approach of extraction and stripping in a single step; (2) diffusivity is higher as a smaller emulsion globule provides the larger interfacial area (Teng et al., 2013); (3) ELM could be 40% cheaper than solvent extraction method (Thakur et al., 2014); (4) saving of contacting equipment volume as separate contactors for extraction and stripping process are not required (Ahmad et al., 2011). ELM technique has great potential for a wide variety of applications including the removal, separation, recovery, and purification of solute from dilute streams. Table 1 shows the exhaustive information regarding the surfactant, carrier, diluent, and internal aqueous phase used for the removal of several targeted solutes by the ELM method.

2.1 Green Emulsion Liquid Membrane

For the formulation of ELM, the diluent is a significant component as it decides the total viscosity of the membrane phase and is also responsible for the emulsion stability (Ahmad et al., 2015). Traditionally the solvents from the petroleum feed stocks like kerosene, hexane, heptane, dichloroethane, and toluene are generally used as diluents for the membrane phase preparation in ELM. The petroleum-based diluents are generally used due to their properties like low viscosity, ready availability, and non-polarity, but these diluents also create several problems in the environment as they are non-renewable, non-biodegradable, flammable, difficult to handle and toxic in nature (Ahmad et al., 2016). According to the World health organization, the permitted amount of hydrocarbons in water should

Table 1 Removal of targeted solute by the ELM method

Targeted solute	Surfactant	Carrier	Diluent	Internal aqueous phase	%Extraction efficiency	Reference
Chromium (VI)	Span 80	TOA	Kerosene oil	KOH	92.5	Rajasimman and Karthic (2010)
	Span 80	Aliquat 336	Hexane	NaOH	90	Choudhury et al. (2010)
	Span 80	TOMAC	Kerosene	NaOH	97	Goyal et al. (2011)
Chromium (III)	Span 80	Cyanex 923	Kerosene	NaOH	-	Nosrati et al. (2011)
	LYF	TBP	Commercial kerosene	H ₂ O	99.83	Zhao et al. (2010)
	Span 80	D2EHPA	Kerosene	H ₂ SO ₄	99	Begum et al. (2012)
Cobalt (II)	Span 80	PC-88A	Kerosene	(NH ₄) ₂ S ₂ O ₈	94	García et al. (2013)
	Span 80	D2EHPA	Commercial kerosene	HCl	-	Hachemaoui et al. (2010)
Silver	ECA4360J	TOA	Commercial kerosene	H ₂ SO ₄	99	Kumbasar (2010)
	Span 80	D2EHPA	Sulfonated kerosene	H ₃ PO ₂	> 99.5	Tang et al. (2010)
Copper (II)	Montane 80	Cyanex 302	Industrial solvent (MIPS)	HNO ₃	99	Laki and Kargari, (2016)
	Span 80	D2EHPA	Hexane	H ₂ SO ₄	95	Chiha et al. (2010)
Mercury (II)	Span 80	D2EHPA	Kerosene	H ₂ SO ₄	96	Begum et al. (2012)
	Span 80	D2EHPA	Toluene	CH ₄ N ₂ S + H ₂ SO ₄	98	Gupta et al. (2011)
Cadmium (II)	Span 80	TOA	Kerosene	NH ₃	-	Ahmad et al. (2012)
	Span 80	TOA	Kerosene	NH ₃	~ 100	Ahmad et al. (2014)
Zinc (II)	Span 80	D2EHPA	Kerosene	H ₂ SO ₄	95	Begum et al. (2012)
Bismuth (III)	Triton X-100	D2EHPA	Dichloromethane	H ₂ SO ₄	97	Benyahia et al. (2014)
	Triton X-100	D2EHPA	n-pentanol	H ₂ SO ₄	100	Mokhtari and Pourabdollah (2015)
Arsenic (V)	Span 80	Cyanex 923	Liquid paraffin	Na ₂ SO ₄	60	Mousavi et al. (2012)
	Span 80	2-ethyl hexanol	Commercial kerosene	NaOH	87.5	Srivastava et al. (2017)
Palladium	Span 80	D2EHPA	Kerosene	H ₂ SO ₄	> 90	Othman et al. (2014)
	Span 80	Cyanex 302	Kerosene	CH ₄ N ₂ S + H ₂ SO ₄	~ 100	Noah et al. (2016)
Cerium (IV)	Span 80	D2EHPA	Sulfonated kerosene	HCl	> 98	He et al. (2015)
Neodymium (III)	Span 80	DNPPA + TOPO	Petrofin	H ₂ SO ₄	> 97	Anitha et al. (2015)
Tungsten (VI)	Span 80	Aliquat 336	Hexane	NaOH	80	Lende and Kulkarni (2015)
Manganese	PEG	MDEHPA	Industrial paraffinic solvent (MIPS)	H ₂ SO ₄	93	Laki et al. (2015)
Vanadium (IV)	Span 80	D2EHPA	Sulfonated kerosene	H ₂ SO ₄	87.5	Liu et al. (2017)
Uranium (VI)	Span 80	HTTA	Kerosene	HCl	99.8	Zaheri and Davarkhah (2017)
Thallium	T154	P 507	Aviation kerosene	H ₂ SO ₄	99.76	Yang et al. (2017)

Table 1 (continued)

Targeted solute	Surfactant	Carrier	Diluent	Internal aqueous phase	%Extraction efficiency	Reference
Dysprosium (III)	Span 80	Cyanex 572	Kerosene	HCl	98.99	Raji et al. (2018)
Gadolinium (III)	Span 80	D2EHPA	Commercial kerosene	HNO ₃	99	Davoodi-Nasab et al. (2018)
Europium (III)	Span 80	Cyanex 302	Kerosene	H ₂ SO ₄	> 92	Laguel et al. (2019)
Acetaminophen	Span 80	Aliquat 336	Hexane	KCl	~ 100	Chaouchi and Hamdaoui (2014)
Propylparaben	Span 80	TOPO	Hexane	Na ₂ CO ₃	100	Chaouchi and Hamdaoui (2015)
L-Phenylamine	Span 80	D2EHPA	Sulfonated kerosene	HCl	94.4	Fang et al. (2016)
Ethylparaben	Span 80	TOA	n-heptane	Na ₂ CO ₃	90.26	Kohli et al. (2018)
Ethylparaben	Span 80 + MWCNT	TOA	n-heptane	Na ₂ CO ₃	97.10	Kohli et al. (2019b)
Amoxicillin	Span 80	Aliquat 336	Hexane	NaCl	99.8	Seifollahi and Rahbar-Kelishami (2019)
Methylparaben	Span 80 + Span 20 + MWCNT	TOA	n-heptane	NaOH	100	Shirsangi et al. (2020)
Tetracycline	Span 80	TBP	n-heptane	HCl	> 95	Mohammed et al. (2020a)
Ciprofloxacin	Fe ₂ O ₃ nanoparticles	TBP	n-heptane	HCl	> 98	Mohammed et al. (2020b)
Red 3BS	Span80	TDA	Kerosene	NaCl	100	Othman et al. (2011a)
Red 3BS	Span80	TDA	Kerosene	CH ₄ N ₂ S + NaOH	70	Othman et al. (2011b)
Rhodamine 6G Dye	Span-80	D2EHPA	Kerosene	H ₂ SO ₄	100	Othman et al. (2013)
Bisphenol A	Span 80	-	Hexane	NaOH	~ 100	Dâas and Hamdaoui (2010)
Lignin	OP-4	-	Kerosene	NaOH	97.52	Jiao et al. (2013)
	Span 80	TOA	Kerosene	NaCl	94	Zing-Yi et al. (2014)
Phenol	Span 80	Aliquat 336	Kerosene	NaHCO ₃	95	Ooi et al. (2016)
Pyridine	Span 80	Cyanex 923	Kerosene	NaOH	98.33	Ng et al. (2010)
Pyridine	OP-4	-	Commercial kerosene	HCl	96.5	Peng et al. (2012)
4-Chlorophenol	LK-80	-	Kerosene	NaOH	~ 100	Kargari (2013)
4-Nitrophenol	Span 80	-	Hexane	Na ₂ CO ₃	> 99	Chaouchi and Hamdaoui (2016)
Cyanide	Span 80	TOA	Kerosene	NaOH	95.31	Xue et al. (2016)

be less than 0.05 mg/L but the petroleum-based diluents are water soluble in the range of 10 mg/L (Zereshki et al., 2021). These diluents will have a detrimental effect on the environment if they are discharged into the environment. Due to the limited

resources of petroleum-based diluent, the price is inconsistent and it could affect the total cost for the ELM formation. For economic and environmental considerations, there is a paramount need to find a better substitute for petroleum-based diluents.

Green solvents or vegetable oils like palm oil, corn oil, sunflower oil, coconut oil, mahua oil, and so on have properties like non-toxicity, non-volatility, non-flammability, degradability, and inexpensive and are reusable which makes them better than petroleum-based diluents (Kumar et al., 2018a). In complying with the principle of a greener approach, different vegetable oils have been incorporated in the ELM formulation and the detailed information regarding the removal of targeted solute using green solvent as a membrane phase is mentioned in Table 2. Table 2 also describes the comprehensive details on the surfactant, carrier, diluent, internal aqueous phase, and extraction efficiency reported by the various investigators for the removal of different targeted solute from the feed phase through the GELM system.

2.2 Green Emulsion Liquid Membrane Formulation

GELM process has four different stages: (1) emulsification, (2) dispersion and extraction, (3) settling, and (4) demulsification process. The primary emulsion is prepared by emulsifying the internal phase and membrane phase. This primary emulsion is dispersed into the external feed phase where the solute is extracted. Emulsion and feed solution are allowed to separate in the settling stage. The membrane phase is recovered through demulsification after the extraction of solute (Arabi Ardehali et al., 2020). Demulsification is carried out by two methods: (1) chemical demulsification and (2) physical demulsification. Chemical demulsification is not selected over physical demulsification as after chemical demulsification the oil phase cannot be reused. Electrical, ultrasonication, and thermal treatment are the methods used for physical demulsification (Lin et al., 2016). Figure 1 shows the schematic view of the GELM system.

Table 3 provides the manufacturer details of various components of GELM.

2.3 Transport Mechanism of GELM

The membrane phase in GELM consists of the suitable organic phase of plant origin and an appropriate carrier (extractant) along with a surfactant is dissolved in it. The solute molecules of the feed phase will react with the carrier and the solute-carrier complex is formed at the feed-liquid membrane interface.

At the liquid membrane-stripping side interface, the complex undergoes a reverse reaction removing the solute to the stripping phase as the complex is de-complexed due to its reaction with the stripping phase. The free carrier renews again and diffuses back across the liquid membrane and the cycle repeats (Kohli et al., 2021a). The feed phase will be depleted of the solute molecules and the stripping phase will be enriched with the solute molecules. Figure 2 represents the facilitated transport mechanism of the solute.

3 Effect of Different Parameters on the Removal of Targeted Solute

3.1 Effect of Surfactant Concentration

In ELM, surfactant plays a vital role in the transport rate of solute, break up of emulsion, stability, and swelling (Chakraborty et al., 2010). Surfactant is one of the important constituents of the membrane phase which decreases the interfacial tension between the water and oil by adsorbing at the liquid-liquid interface and helps to form the emulsion (Kumar et al., 2018c; Ting et al., 2022). The concentration of surfactant is an important parameter as studies have indicated that at low surfactant concentration, large emulsion globules are formed which provides a low interfacial area for mass transfer and hence reduces the separation efficiency. Also, low surfactant concentration in ELM is unable to reduce the interfacial tension between water and oil which leads to the separation of water and oil. The emulsion stability is also low at low surfactant concentration as the surfactant is not able to totally cover up the whole internal aqueous phase (Jusoh and Othman, 2017; Zereszki et al., 2018). The rise in surfactant concentration leads to the adsorption of more surfactant on the liquid-liquid interface which tends to enhance the emulsion stability and the strength of the adsorption layer. This reduces the surface tension of the membrane phase producing the smaller emulsion globules which provide a high mass transfer area between the donor and internal phase.

Jusoh and Othman (2017) witnessed less emulsion droplets formation with Span 80 concentration of 5% w/v and 7% w/v. At this high concentration of

Table 2 Extraction of targeted solute by GELM method

Targeted solute	Surfactant	Carrier	Diluent	Internal aqueous phase	Surfactant concentration	Carrier concentration	Internal phase concentration	Extraction efficiency (%)	Reference
Chromium (VI)	Span 80 + Tween 80	TOMAC	Palm oil	NaOH	2.58% wt. Span 80 + 1% wt. Tween 80	0.35% wt	0.1 M	99	Björkegren et al. (2015)
Cadmium (II)	Span 80	Aliquat 336	Corn oil	NH ₄ OH	3% wt	---	0.1 M	98.2	Ahmad et al. (2015)
Chromium (VI)	Span 80	TOMAC	Palm oil: Kerosene (7:3)	NaOH	3% w/v	0.2 M	1.0 M	~100	Othman et al. (2016)
Succinic acid	Span 80 + Tween 80	Amberlite LA2	Palm oil	Na ₂ CO ₃	2% w/v Span 80 + 1% w/v Tween 80	0.7 M	0.01 M	70	Jusoh and Othman (2017)
Cadmium (II)	Span 80	Aliquat 336	Corn oil	NH ₄ OH	3% wt	3% wt	0.1 M	98.9	Ahmad et al. (2017)
Phenol	Span 80	-	Palm oil: Kerosene (7:3)	NaOH	3% w/v	---	0.1 M	83	Othman et al. (2017)
Chromium (VI)	Span 80	Aliquat 336	Sunflower oil	NaOH	---	---	---	99	Davoodi-Nasab et al. (2017)
Copper (II)	---	D2EHPA	Waste vegetable oil	H ₂ SO ₄	---	88 mM	1.5 M	96	Chang (2017)
Chromium (VI)	Span 80 + Tween 80	TOPO	Sunflower oil	Na ₂ CO ₃	---	4%	0.5 M	87.43	Anarakdim et al. (2017)
MB, CV, and MV	Span 80	---	Edible paraffin oil: n-heptane (80:20)	NaOH	5% wt	---	0.04 M	97, 98, and 100 respectively	Zereshtki et al. (2018)
Chromium (VI)	Span 80	TOMAC	Palm oil	NaOH	3% w/v	0.04 M	0.05 M	97	Noah et al. (2018)
Succinic acid	Span 80 + Tween 80	Amberlite LA2	Palm oil	Na ₂ CO ₃	3% w/v Span 80 + 80 + 1% w/v Tween 80	0.05 M	0.5 M	~100	Othman et al. (2018)
Lactic acid	Span 80	TOMAC	RBO:Hexane [70% v/v: 30% v/v]	NaOH	2.66% v/v	0.2% v/v	0.25 M	90	Kumar et al. (2018a)
Lactic acid	Span 80	Aliquat 336	RBO:Hexane [70% v/v: 30% v/v]	NaOH	2.66% v/v	0.2% v/v	0.25 M	95	Kumar et al. (2018b)
Succinic acid	Span 80 + Tween 80	Amberlite LA2	Palm oil	Na ₂ CO ₃	5% w/v Span 80 + 1% w/v Tween 80	0.7 M	0.01 M	71	Jusoh et al. (2019)
Chromium (VI)	Amphiphilic silica nanowires	Aliquat 336	Mahua oil	NaOH	25 mg / 7 mL	0.129% Vol	0.3 M	99.69	Perumal et al. (2019)
Reactive Red 3BS	Span 80	TDA	Palm oil	NaHCO ₃	3% w/v	0.2 M	0.1 M	90	Othman et al. (2019)
Chromium (VI)	Span 80	TDDA	RBO:Hexane [70% v/v: 30% v/v]	NaOH	1.75% v/v	0.75% v/v	0.25 M	97	Kumar et al. (2019b)
Methyl violet 2B	Span 80	---	Sunflower oil	HCl	4.9% wt	---	0.84 M	97	Daraei et al. (2019)

Table 2 (continued)

Targeted solute	Surfactant	Carrier	Diluent	Internal aqueous phase	Surfactant concentration	Carrier concentration	Internal phase concentration	Extraction efficiency (%)	Reference
Phenol	Span 80 + Tween 80	[BMIM] ⁺ [NTf ₂] ⁻	Palm oil: Kerosene (7:3)	NaOH	3% w/v	0.107% w/v	0.1 M	83	Rosly et al. (2019)
Neodim (III)	Span 80	M2EHFA + D2EHFA	Sunflower oil	HNO ₃	1.5% w/v	2% v/v	1.0 M	—	Rajji et al. (2020)
Phenol	Span 80 + Tween 80	—	Palm oil	NaOH	3% w/v	—	0.1 M	83.4	Rosly et al. (2020)
Methyl violet 2B	Span 80	—	Waste cooking oil	HCl	3% wt	—	1.43 M	99.1	Shokri et al. (2020)
Chromium (VI)	PGPR + Tween 80	TOPO	Sunflower oil	Na ₂ CO ₃	4% v/v PGPR + 1% v/v Tween 80	4% v/v	0.5 M	>99	Anarakdim et al. (2020)
Bio-succinic acid	Span 80 + Tween 80	Amberlite LA2	Palm oil	Na ₂ CO ₃	5% w/v Span 80 + 1% w/v Tween 80	0.7 M	1.0 M	~100	Jusoh et al. (2020)
Copper (II)	Span 80	D2EHFA	Sunflower oil	HCl	3.25% wt	104.23 mM	1.44 M	>94	Zereszki et al. (2021)
Arsenic (III)	Span 80	Aliquat 336	Waste cooking oil	NaOH	2.3% v/v	5.6% v/v	0.92 M	99	Sujatha and Rajasimman (2021)
Nickel (II)	Span 80	Cynex 301	Waste cooking oil	H ₂ SO ₄	2.66% v/v	5.2% v/v	0.625 M	98.7	Sujatha et al. (2021a)
Zinc	Span 80	—	Waste cooking oil	H ₂ SO ₄	4% Vol	—	1.61 N	97.4	Rajasimman et al. (2021)
Acetaminophen	Span 80	Aliquat 336	Sunflower oil	NH ₄ OH	6% wt	6% wt	0.1 M	97.73	Zaukiffie et al. (2021)
Lead (II)	Span 80	D2EHFA	Waste cooking oil	H ₂ SO ₄	2.14% v/v	4.6% v/v	2 M	97.39	Sujatha et al. (2021b)
Copper (II)	Span 80	D2EHFA	Sunflower oil	HCl	3.25% wt	104.23 mM	1.44 M	94	Zereszki et al. (2021)
Salicylic acid	Span 20	[TMAm][Cl]	Sunflower oil	NaOH	1% wt	0.2% wt	0.005 M	90.04	Ting et al. (2022)
Vancomycin	Span 80	D2EHFA	Sunflower oil	NaOH	3% wt	0.05 M	0.1 M	100	Daraei et al. (2022)
Cadmium (II)	Span 80	D2EHFA	Waste cooking oil	HCl	1.29% v/v	3.14% v/v	0.93 M	97.40	Sujatha et al. (2022)

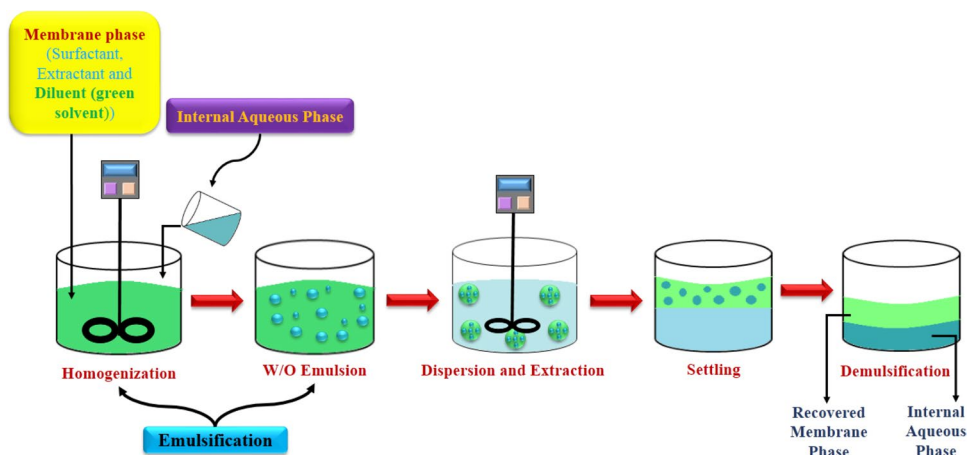


Fig. 1 Schematic diagram of GELM formulation

Table 3 Manufacturer details of various components of GELM

Component	Name	Manufacturer	Reference
Surfactant	Span 80 (Sorbitan Monooleate)	Sigma-Aldrich Co., USA	Othman et al. (2016); Jusoh and Othman (2017)
		Merck (Petaling Jaya, Malaysia)	Björkegren et al. (2015); Ahmad et al. (2015); Ahmad et al. (2017)
Carrier	Tween 80 (Polyoxyethylene sorbitan monooleate)	R & M Chemicals (Petaling Jaya, Malaysia)	Björkegren et al. (2015)
	PGPR (Polyglycerol polyricinoleate)	Sigma-Aldrich Co., USA	Jusoh and Othman (2017)
		Brenntag AG, Germany	Anarakdim et al. (2017)
	Aliquat 336 (Trioctylmethylammonium chloride)	Sigma-Aldrich	Ahmad et al. (2015); Ahmad et al. (2017)
	D2EHPA (Di-2-ethylhexylphosphoric acid)	Acros Organics	Chang (2017)
	TOMAC (Tri-n-octylmethylammonium chloride)	Merck (Petaling Jaya, Malaysia)	Björkegren et al. (2015)
	Amber Lite LA-2	Sigma-Aldrich Co., USA	Othman et al. (2016)
	TOPO (Tri-n-octylphosphine oxide)	Alfa Aesar, Germany	Anarakdim et al. (2017)
	TDA (Tridodecylamine)	Merck	Othman et al. (2019)
	TBP (Tributyl phosphate)	LOBA Chemie, India	Balasubramanian and Venkatesan (2012)
Diluent	[BMIM] ⁺ [PF ₆] ⁻ (1-Butyl-3-methylimidazolium hexafluorophosphate)	Merck	Rosly et al. (2019)
	M2EHPA (Mono-(2-ethylhexyl) ester of phosphoric acid)	Fluka (Hannover, Germany)	Raji et al. (2020)
	Cyanex 301 (Bis (2,4,4 trimethylpentyl) dithiophosphinic acid)	Sigma Aldrich	Sujatha et al. (2021b)
	Diluent	Palm oil	BURUH (Lam Soon Edible Oils)
Sunflower oil		Local Market, Kermanshah, Iran	Daraei et al. (2019)
Rice Bran oil		Ricela Health Food Ltd., India	Kumar et al. (2019b)

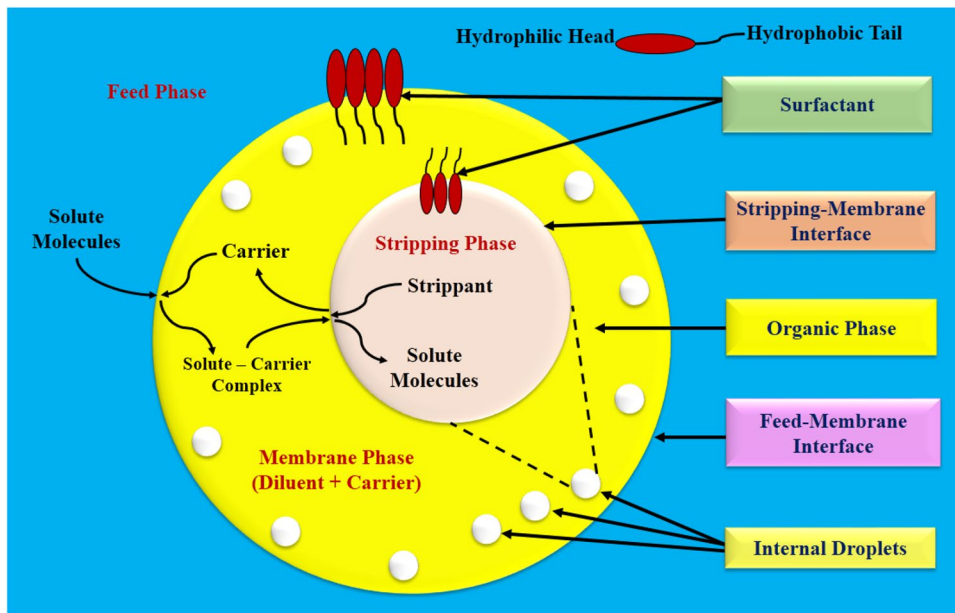


Fig. 2 Facilitated transport mechanism of the solute in GELM

surfactant, the extraction efficiency was reduced due to the increase in the overall viscosity of the membrane phase. This hinders the mass transfer of solute molecules from the feed phase to the stripping phase due to the enhancement in the interfacial resistance and by the reaction with a carrier at the interface. Above a certain limit of concentration, most of the surfactant molecules forms aggregates in the bulk which is referred to as critical micelles concentration (CMC), and these aggregates act as a reservoir for water (Dâas & Hamdaoui, 2010). The reverse micelles promote the transportation of water from the external feed phase to the internal phase which leads to a larger osmotic difference and increases the swelling rate.

Othman et al. (2017) observed an increase in extraction of phenol from 46 to 99% as Span 80 concentration was increased from 1 to 5% w/v with palm oil as the diluent. Noah et al. (2018) observed a reduction in the size of the emulsion globule from 3.70 to 3.14 μm as the surfactant concentration increased from 1 to 3% w/v. At the optimum level of surfactant concentration, no breakage and swelling were observed which indicates the presence of the sufficient number of surfactant molecules. Perumal et al. (2019) too witnessed a rise in the percent extraction of chromium from 94 to 99.5% as the concentration

of amphiphilic silica nanowires was raised from 5 to 25 mg per 7 mL in mahua oil (diluent) respectively. Sujatha et al. (2021a) too observed enhancement in extraction efficiency from 45 to 95% as the surfactant concentration raised from 1 to 2.66% v/v respectively during the removal of chromium (VI).

3.2 Effect of Carrier Concentration

Many investigators have explored the effect of carrier on the separation of different compounds. Carrier is an active compound that binds the compounds at the feed-membrane interface to form the compound-carrier complex. It is seen that a rise in carrier concentration to a certain limit increases the extraction, helps in proper complexation of the compound and a further rise in carrier concentration afterwards does not much affect the extraction efficiency as the membrane phase viscosity rises which increases the membrane phase resistance and hinders the extraction performance.

The viscosity of the membrane phase influences ELM's stability. The rise in carrier concentration raises the viscosity of ELM (Lee, 2011). Kohli et al. (2018) too observed a decrease in the extraction of ethylparaben as the concentration of carrier TOA was raised beyond 3% w/v which happened due to the rise

Table 4 Effect of carrier concentration on extraction in GELM technique

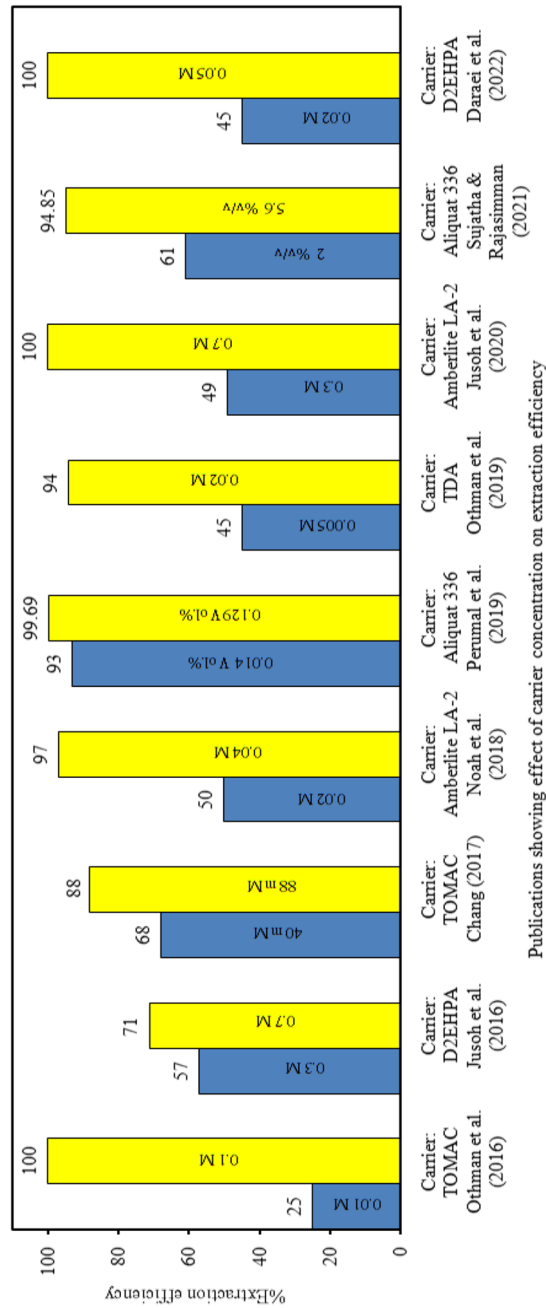
Targeted solute	Surfactant	Stripping phase	Diluent	Carrier	Carrier concentration	%Extraction	Reference
Chromium (VI)	Span 80	NaOH	Palm oil-Kerosene	TOMAC	0.01 M → 0.1 M	25 → 100	Othman et al. (2016)
Copper (II)	—	Na ₂ SO ₄	Waste vegetable oil	D2EHPA	40 mM → 88 mM	68 → 88	Chang (2017)
Chromium (VI)	Span 80	NaOH	Palm oil	TOMAC	0.02 M → 0.04 M	50 → 97	Noah et al. (2018)
Succinic acid	Span 80 + Tween 80	Na ₂ CO ₃	Palm oil	Amberlite LA-2	0.3 M → 0.7 M	57 → 71	Jusoh et al. (2016)
Chromium (VI)	Amphiphilic silica nanowires	NaOH	Mahua oil	Aliquat336	0.014 Vol.% → 0.129 Vol.%	93 → 99.69	Perumal et al. (2019)
Reactive Red 3BS	Span 80	NaHCO ₃	Palm oil	TDA	0.005 M → 0.02 M	45 → 94	Othman et al. (2019)
Bio-succinic acid	Span 80 + Tween 80	Na ₂ CO ₃	Palm oil	Amberlite LA-2	0.3 M → 0.7 M	49 → 100	Jusoh et al. (2020)
Arsenic (III)	Span 80	NaOH	Waste cooking oil	Aliquat 336	2% v/v → 5.6% v/v	61 → 94.85	Sujatha and Rajasimman (2021)
Vancomycin	Span 80	NaOH	Sunflower oil	D2EHPA	0.02 M → 0.05 M	45 → 100	Daraei et al. (2022)

in the viscosity of the membrane phase. Sometimes, low carrier concentration does not enhance the ELM viscosity and results in instability and breakage of ELM (Othman et al., 2016). Moreover, the low concentration of carrier restricts the adequate carrier-complex formation. The osmotic pressure tends to be higher in the organic phase than in the external feed phase at higher carrier concentration (Choudhury et al., 2010; Kumbasar, 2010) and so swelling takes place. The mass transfer resistance increases, and extraction efficiency reduces at high carrier concentration (Jusoh et al., 2016; Othman et al., 2016). So, proper selection and optimum carrier concentration are required for the enhanced performance of an ELM system. Table 4 lists the separation of several targeted solutes using different surfactants, stripping phase, diluent, carrier, and their concentration. Figure 3 describes the impact of different carriers and their concentration on extraction efficiency as investigated by various investigators.

The critical micelles concentration (CMC) is believed to be the benchmark concentration for the surfactant to work efficiently in reducing the interfacial tension. Low CMC has two most important

inferences for the field implementation. Firstly, the low CMC affects the amount of surfactant required to be handled in the field. Secondly, if only low surfactant concentration is required for effective working, then the total cost is also reduced (Abbas et al., 2022). CMC determination carried out by electric conductivity corresponded to the unexpected change in the specific conductance profile (de souza et al., 2021). Ahmad et al. (2012) reported that with non-ionic surfactants, droplet size is further reduced above the CMC during the removal of cadmium. Thus, CMC findings are remarkably important for the emulsion liquid membrane technique.

Solvent or diluent (vegetable oil / seed oil) is one of the key components of GELM. The diluent decides the overall viscosity of the liquid membrane. Jusoh et al. (2017) observed that a rise in carrier concentration does not contribute much towards the enhancement of liquid membrane viscosity, also palm oil viscosity (83 cP) was observed higher than Amberlite LA-2 (18 cP). Lower viscosity with high carrier concentration favours the formation of small emulsion globules during the dispersion process (Jusoh et al., 2020). The higher viscosity of membrane phase



Publications showing effect of carrier concentration on extraction efficiency

provides more stability, but it hinders the mass transfer and provides higher resistance for solute molecules (Zaulkiflee et al., 2021). In the membrane phase, the rise in carrier concentration reduces the interfacial tension which leads to the larger emulsion globule size whereas interfacial tension decreases with increases in the surfactant concentration up to a certain value (Sujatha & Rajasimman, 2021). So, the carrier concentration should be wisely selected to reduce the overall cost of the process and to achieve the finest extraction.

3.3 Effect of Types of Diluents

Diluents play an important role in the preparation of the organic membrane phase. The diluent viscosity should be low as it helps to facilitate the diffusion of the solute-carrier complex. The effect of diluent is significant as both physical and chemical interactions take place between diluent and carrier (Kohli et al., 2020b; Parhi, 2013). Moreover, in the GELM technique the part of the diluent is more substantial as the solvent used should be non-toxic and environmentally benign. The best and most suitable alternative to petroleum-based solvents is vegetable oils. Different vegetable oils like palm oil, corn oil, mahua oil, sunflower oil, rapeseed oil, coconut oil, neem oil, pungai oil, and waste cooking oil have been reported as biodegradable and environmentally benign diluent in the formulation of GELM. Seeds and fruits are good sources of vegetable oils. Vegetable oils contain nonpolar lipids (<92%), polar lipids (<4%), free fatty acids (<2%), and unsaponifiable matter (phytosterols, tocopherols and hydrocarbons) (<2%) (Othman et al., 2019).

As compared to petroleum-based diluents, vegetable oils have higher viscosity due to the intermolecular attractions of long chain fatty acids in vegetable oils and so it increases the mass transfer resistance (Mei et al., 2020; Othman et al., 2019). Many authors have reported that the waste vegetable oil doesn't show good extraction efficiency as compared to the fresh vegetable oil as during frying at a higher temperature, waste vegetable oils generate a large amount of polar, surface active compounds and polymers due to the hydrolytic oxidation and polymerization reaction. Also, in comparison to vegetable oil-based diluents, petroleum-based diluents are non-renewable and expensive (Chang, 2017).

Noah et al. (2018) reported 100% extraction efficiency of chromium using green diluents like palm oil and corn oil and no reduction in extraction efficiency was observed due to the high viscosity of vegetable oil as compared to kerosene. Moreover, Perumal et al. (2019) also reported that high viscous oil like mahua oil provides high stability over low viscous oil like coconut oil and sunflower oil. Rosly et al. (2019) observed that the kerosene-based ELM had lower extraction (37%) efficiency as compared to palm oil-based ELM (77%). The results indicate that aliphatic hydrocarbons like kerosene have lower extraction efficiency due to their aliphatic nature as it contains a long carbon chain that provides nonpolar characteristics. However, palm oil contains oxygen atoms that enable phenol and triglycerides reaction by hydrogen bonding amid the molecules forming phenol triglycerides complex. Also, the phenolic compounds are dissolved due to the hydrophobic nature of palm oil (Rosly et al., 2019). Various vegetable oils such as waste cooking oil, canola oil, sunflower oil, corn oil, rice bran oil, and palm oil were used for the removal of various contaminants which shows the reliability and good capacity of the GELM (Harun et al., 2022; Rajasimman et al., 2021; Ting et al., 2022; Zaulkiflee et al., 2021).

In the last few decades, the worldwide demand for vegetable oil has drastically increased. Palm oil (*Elaeis guineensis*) is one of the most important oil crops in the world (Sakai et al., 2022). The palm oil boom has contributed to financial growth mainly in southeast Asia. Palm oil production is a driver of deforestation which creates air pollution due to the use of fire for land conversion (Ogahara et al., 2022). The conversion of forest into palm oil production has decreased the carbon stocks by over 50% and has enhanced the greenhouse gas emissions by four times as compared to land converted from old rubber plantations (Chiriaco et al., 2022). Due to deforestation, the problems associated with the loss of biodiversity and ecosystem function arise (Qaim et al., 2020). Also, the sheer scale at which palm oil is cultivated can harm the soil and leads to problems concerned to water quality and availability (Mukherjee & Sovacool, 2014).

Few authors have also reported problems associated with the land exploitation impact on native communities' rights and land rights or

Table 5 Effect of treat ratio on %extraction by GELM method

Targeted solute	Diluent	Treat ratio (feed phase: Emulsion)	%Extraction	Reference
Chromium	Palm oil:kerosene (7:3)	2:1 → 3:1	97 → 69	Othman et al. (2016)
Succinic acid	Palm oil	3:1 → 7:1	63 → 40	Jusoh and Othman (2017)
Cadmium	Corn oil	5:1 → 10:1	98.3 → 67.5	Ahmad et al. (2017)
Phenol	Palm oil:kerosene (7:3)	3:1 → 7:1	88 → 83	Othman et al. (2017)
Lactic acid	RBO:hexane [70% (v/v): 30% (v/v)]	2:1 → 4:1	90.5 ± 1.78 → 33.33 ± 1.75	Kumar et al. (2018a)
Succinic acid	Palm oil	3:1 → 5:1	63 → 51	Jusoh et al. (2019)
Bio-succinic acid	Palm oil	3:1 → 7:1	100 → 76	Jusoh et al. (2020)
Vancomycin	Sunflower oil	5:1 → 7:1	100 → 88	Daraei et al. (2022)

on pesticides and their impact on the labourer's health. Socioeconomic issues also include the rights of palm oil labourers in developing countries and support to small farmers to guarantee adequate remuneration and survival (Ruggeri & Samoggia, 2018; Dharmawan et al., 2020).

However, Chiriaco et al. (2022) reported that the cultivation of palm oil contributes positively to the socioeconomic growth of the local public, irrespective of the country of production or approach used, with a substantially positive impact on poverty reduction and economic growth. The production of palm oil is also considered one of the most environmentally sustainable products due to its high productivity, concerning the phenomenon of land grabbing in comparison to the production of other vegetable oils like soy, sunflower, or rapeseed. Also, palm oil is considered the best versatile oil to satisfy the uses of different sectors (Chiriaco et al., 2022).

3.4 Effect of Volume Ratio of External Feed Phase to Emulsion

The volume ratio of the external feed phase to emulsion also known as the treat ratio plays an important role in determining the extraction efficiency. The optimum ratio provides the best extraction efficiency. It is observed that the low treat ratio provides low extraction efficiency as emulsion cannot be dispersed properly and the formation of emulsion globules is reduced due to the osmotic pressure difference (Ahmad et al., 2017). The interfacial surface area decreases and the mass transfer area for extraction are

reduced due to the breakage of the emulsion. Moreover, a high volume of emulsion creates problems in dispersion and increases the total viscosity of ELM (Jusoh and Othman, 2017).

An increase in the treat ratio increases the percent extraction as the osmotic pressure effect is reduced and the emulsion could properly disperse and ease into the feed phase. This leads to the formation of more emulsion globules. The globule size is small and so it can provide more interfacial area for complex formation on the feed-membrane interface (Othman et al., 2017, 2019). Further increment in treat ratio reduces the extraction capacity. The number of emulsion globules available is low and due to this, the interfacial surface area required for the extraction is reduced (Sujatha et al., 2021a; Zereski et al., 2018). Thus, it also reduces the mass transfer of solute ions from the external feed phase to the emulsion. The breakage and rupture found in ELM are more at high ratio due to the large osmotic difference between the external feed phase and emulsion; as less number of emulsion, globules are available for extraction (Jusoh et al., 2020). Table 5 presents the effect of the treat ratio on the %extraction of targeted solute by GELM. Figure 4 shows the significance of the treat ratio on extraction efficiency as examined by many investigators.

3.5 Effect of Agitation Speed

The speed of agitation during the formation of emulsion is crucial. It influences the size of emulsion globules which eventually affects the interfacial mass transfer area, extraction efficacy, and stability

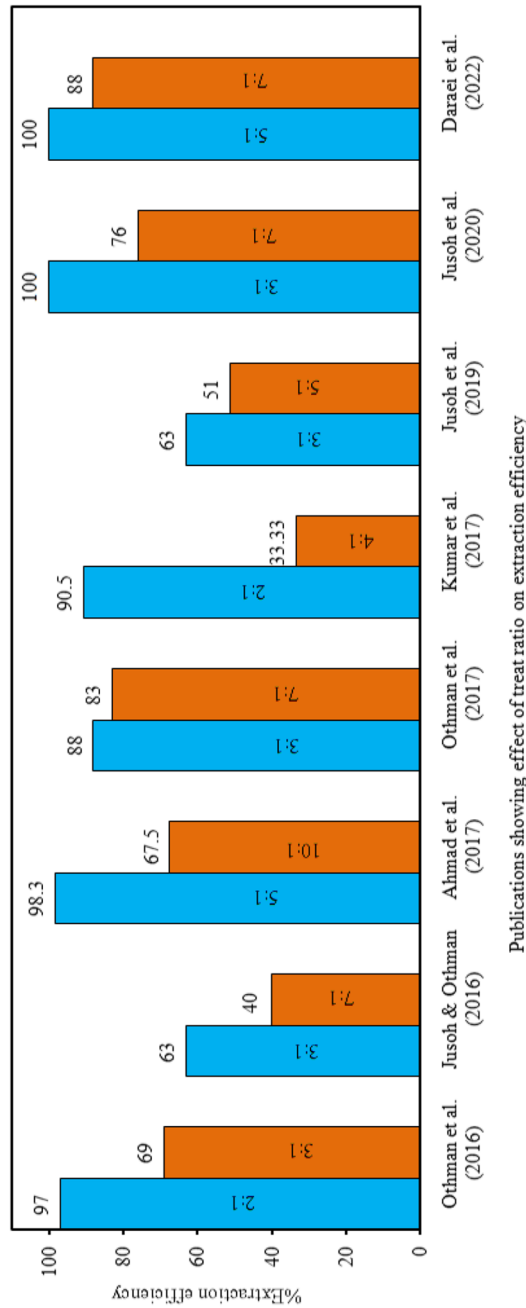


Fig. 4 Publications showing the influence of treat ratio on extraction efficiency

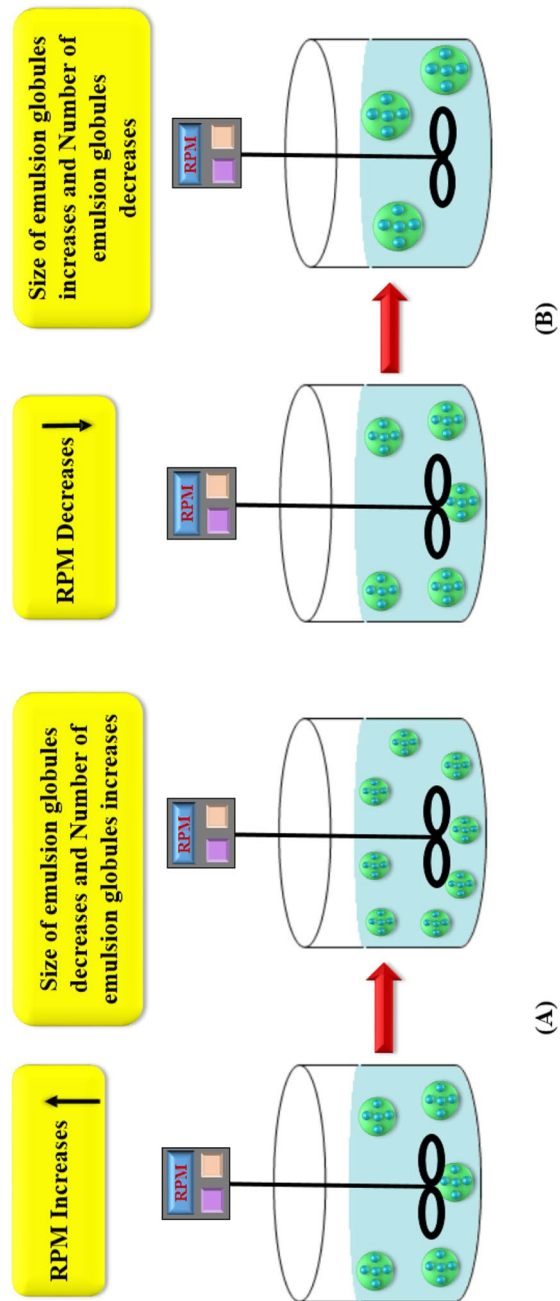


Fig. 5 Effect of agitation speed: **A** Increase in speed. **B** Decrease in speed

Table 6 Effect of internal aqueous phase concentration on the %extraction of solute by GELM technique

Targeted solute	Diluent	Internal aqueous phase	Internal aqueous phase concentration	%Extraction	Reference
Chromium	Palm oil	NaOH	0.5 M → 1.0 M	75 → 93	Othman et al. (2016)
Phenol	Palm oil	NaOH	0.01 M → 0.1 M	90 → 99	Othman et al. (2017)
Lactic acid	Rice bran oil	NaOH	0.18 M → 0.25 M	45.8 → 88.9	Kumar et al. (2018a)
Succinic acid	Palm oil	Na ₂ CO ₃	0.005 M → 0.01 M	55 → 63	Jusoh et al. (2019)
Chromium	Mahua oil	NaOH	0.1 M → 0.3 M	83.74 → 99.69	Perumal et al. (2019)
Chromium	Rice bran oil	NaOH	0.1 M → 0.25 M	63 → 97	Kumar et al. (2019b)
Vancomycin	Sunflower oil	NaOH	0.01 M → 0.1 M	76 → 100	Daraei et al. (2022)

of an ELM system. For a better and uniform dispersion of emulsion droplets in the external feed phase, a comparatively high agitation speed is required (Rosly et al., 2019). At lower agitation speed, the ELM formed is unstable due to the weak and slow dispersion of emulsion droplets into the external feed phase (Rosly et al., 2019; Kumar et al., 2019b). Less shear energy leads to slow or insufficient dispersion (Kumar et al., 2019b). It has been observed that larger emulsion globules tend to coalesce with one another which reduces the extraction efficiency and increases the membrane thickness leading to less mass transfer (Kumar et al., 2019b; Perumal et al., 2019). An increase in the speed of agitation increases the shear forces, which reduces the size of dispersed emulsion droplets and enhances the interfacial area for mass transfer between feed and ELM solution (Kumar et al., 2018a). Figure 5 describes the effect of agitation speed on the formation of emulsion globules.

At the optimum speed of agitation, shear forces hinder the coalescence process between the emulsion droplets and the membrane wall formed is thin. So, the extracted solute needs to travel very less distance to be stripped at the membrane-stripping interface which increases the extraction rate of solute (Ahmad et al., 2017; Othman et al., 2018). The speed of agitation beyond the optimum level provides high shear stress on the fine membrane droplets. The large energy introduced due to such a high speed of agitation leads to swelling where water transports from the external feed phase to the membrane phase and reduces the extraction efficiency (Kumar et al., 2018a; Othman et al., 2016). Also, several investigators have observed that at high speed of agitation, the membrane wall formed is much thinner and so the membrane breakage rate increases due to the rupture

of emulsion globules. This allows the expulsion of extracted solute from the stripping phase to the external feed phase by back diffusion (Othman et al., 2017; Zereshki et al., 2018).

3.6 Effect of Internal Aqueous Phase Concentration

The concentration of the internal aqueous phase (stripping agent) is crucial for the development of emulsion. The internal aqueous phase affects the extraction efficiency, stripping efficiency, and emulsion breakage. The stripping agent has a vital role in the stripping process as the solute is stripped out from the solute-carrier complex which is present in the membrane phase. Othman et al. (2016) observed that the higher stripping rate can be achieved by increasing the internal aqueous phase concentration up to a certain limit only. Cloudiness of stripping solution is observed at low concentration of internal aqueous phase, and this leads to poor removal efficiency. This also indicates the inadequate amount of stripping agent availability in the internal phase which also reduces the stripping efficiency (Shokri et al., 2020). At optimum concentration of stripping agent, the extraction efficiency and stripping efficiency increase as enough number of stripping agent molecules are available for the stripping reaction. So, the carrier is again ready to form the new solute-carrier complex and more solute is being stripped into the internal aqueous phase within less time (Othman et al., 2016). Further rise in the internal aqueous phase concentration reduces the extraction and stripping efficiency and increases the emulsion breakage (Shokri et al., 2020).

The high concentration of the stripping agent has a strong pH gradient which causes a larger difference

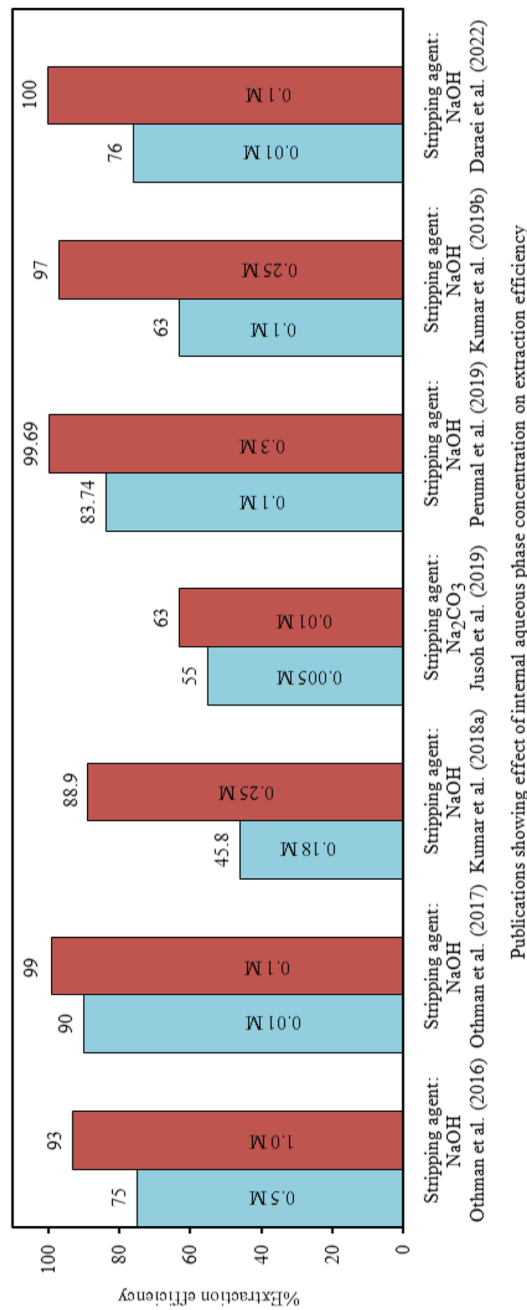


Fig. 6 Publications showing the influence of various stripping agents and their concentration on extraction efficiency

in osmotic pressure (Björkegren et al., 2015; Othman et al., 2016). The higher concentration of the stripping agent creates a larger density difference and increases the overall emulsion viscosity which hinders the mass transfer rate (Perumal et al., 2019). The excess amount of stripping agent molecules reacts (hydrolyzing) with the surfactant reducing the effective number of surfactant molecules and increasing the transport of the aqueous phase (Othman et al., 2017). This in turn leads to emulsion swelling and breakage. The swelling and emulsion breakage rate increases which ultimately destabilizes the emulsion (Zereshki et al., 2018). Table 6 shows the effect of internal aqueous phase concentration on the %extraction in GELM. Figure 6 describes the significance of various stripping agents and their concentration on extraction efficiency as studied by many researchers.

3.7 Effect of Emulsification Time

The internal phase droplet size and emulsion stability directly depend on the emulsification time. Extraction efficiency observed was poor for the shorter emulsification time. This is due to the formation of larger emulsion droplets which provides a smaller interfacial area for the transport of solute molecules (Ahmad et al., 2017). Many authors have observed poor extraction efficiency when the time is not sufficient to generate small emulsion globules. Furthermore, shorter emulsification time tends to the sudden breakage of emulsion. Sufficient time for emulsification is required for the surfactant to migrate and adsorb at the interface. This reduces the interfacial tension and results in small emulsion droplets which lead to the higher extraction of solute and better stability (Rosly et al., 2020).

Kumar et al. (2018c) observed that with the rise in the emulsification time from 10 to 20 min, the stability of GELM (diluent: rice bran oil) increased from 55 ± 2 to 121 ± 2 min respectively. Shokri et al. (2020) also observed the rise in emulsion stability from 20 to 118 min when emulsification time was increased from 3 to 10 min respectively. Exposure of emulsion constituents to the internal shear for a longer duration causes the creation of a large number of fine internal droplets which requires more time to coalesce. Therefore, the emulsion is more stable and the breakage phenomenon is hindered during the dispersion of the emulsion (Othman et al., 2018).

Also, a significant reduction is observed in extraction efficiency when emulsion formation is carried beyond the optimum emulsification time. This happens due to the coalescence of small internal phase droplets produced. Also, the emulsion breakage was observed to take place due to the membrane rupture. The longer emulsification time leads to a more viscous emulsion, which provides more resistance and results in poor extraction efficiency (Othman et al., 2017). Othman et al. (2018) studied the effect of emulsification time (3–9 min) during the removal of succinic acid by using palm oil as a diluent and observed 5 min as the most suitable emulsification time. Noah et al. (2018) observed a rise in emulsion droplets size from 2.67 to 3.60 μm as the emulsification time was raised from 3 to 10 min respectively with an increase in breakage from 10 to 30% while using palm oil (diluent).

3.8 Influence of Statistical Tools on Extraction Efficiency

Experimental design by response surface methodology (RSM) optimizes the process variables and improves the process's effectiveness. RSM consists of mathematical and statistical techniques which are focussed on the fit of a polynomial equation to the experimental data. RSM can be soundly used where a single response or set of responses are affected by many variables. RSM is widely used as it generates large information and evaluates the interaction effect among the variables on the response (Bezerra et al., 2008).

Kumar et al. (2018b) investigated the extraction of lactic acid (LA) from an aqueous solution using green emulsion ionic liquid membrane (GEILM) with the help of a statistical approach comprising of 2-level fractional factorial design (FFD) and BBD (Box-Behnken design). The emulsion consisted of diluents like rice bran oil (70% v/v, natural green solvent) and hexane (30% v/v, organic solvent), surfactant Span 80, Aliquat336 as an ionic liquid, and internal phase reagent NaOH. The parameters namely, LA concentration, NaOH concentration, phase ratio, treat ratio, and stirring speed were screened out using FFD and then RSM was applied for optimization by BBD. BBD presented the design matrix of 42 experiments which were performed separately in triplicates and the average LA extraction efficiency was considered as the response. LA extraction achieved was

$95 \pm 3.5\%$ under the optimum conditions as: LA concentration: 0.03 M, NaOH concentration: 0.25 M, phase ratio: 0.4 v/v, treat ratio: 2.4 v/v and stirring speed: 535 rpm.

Daraei et al. (2019) applied GELM consisting of Span 80 (surfactant), HCl (internal phase), and sunflower oil (diluent) for the removal of methyl violet 2B dye. Plackett–Burman screening design was first used to identify the important factors and then BBD was used for parametric optimization. BBD suggested 15 experimental runs based on different combinations of effective parameters. The model suggested the maximum dye removal as 97.54% with 4.9% wt Span 80, 0.84 M HCl, and equal amounts of GELM and contaminated water.

Kumar and Thakur (2019) studied the lactic acid extraction from an aqueous solution by the use of soybean oil (green solvent) and a synergistic mixture of the extractants: tri-n-octylamine (TOA) and Tri-n-octylmethylammonium chloride (TOMAC). The statistical optimization was carried out with the help of BBD and extraction efficiency achieved was 71.5% under the optimum conditions as: 0.02 M initial LA concentration, 0.5 (v/v) extractant ratio, 28.66% v/v mixed extractants concentration, 2 (v/v) phase ratio, 27 °C temperature, 102 rpm stirring speed and 63 min contact time. BBD provided 62 experimental runs using different combinations of different process factors. It was observed that the synergistic effect of the mixed extractants leads to a higher value of distribution coefficient as compared to the single extractants.

Zereshki et al. (2021) investigated the removal of copper ions from aqueous solutions by GELM. BBD was used for statistical analysis and optimization. BBD provided 29 experiments for four variables namely surfactant (Span 80) concentration, internal phase (HCl) concentration, carrier (D2EHPA) concentration, and internal to organic phase (I/O) ratio. Sunflower oil was used as the solvent. It was observed that more than 94% of Cu(II) ions were extracted at the optimum conditions of 3.25% wt Span 80, 1.44 M HCl, 104.23 mM D2EHPA, and an I/O ratio of 1.

3.9 Real-Time Diagnosis of Water Quality

It has been observed that the existing situation of pollution intensity leads to the rise in a build-up of pollutants in marine waters. This also creates problems in assessing the impact on the ecological system. Varotsos

and Krapivin (2018) proposed the use of geoecological information modelling system (GIMS) for dealing with this problem occurring in the Arctic Basin. GIMS consists of a series of precise models that describes ecological, hydrological, climatic, and hydrochemical processes in Arctic waters. The combination of GIMS with the Arctic basin ecosystem (ABE) model considers a variety of pollutants, like river runoffs, long-range atmospheric transport, and anthropogenic behaviour in the coastal zone. The main aspect of GIMS-ABE lies in the structural liberty of its blocks and the transfer of knowledge among them that occurs through inputs and outputs. Varotsos et al. (2019) also proposed the optical decision-making system for in-situ evaluation of water quality in natural water regions.

Monitoring the quality of water is important for controlling the pollutants which can cause harm to humans and the environment. With the advancement in technology and engineering materials, real-time analysis of water quality is possible. Silva et al. (2022) too discussed the development of low-cost technologies, which can speed up the compilation of data required for the monitoring of physical, chemical, and biological parameters of the water.

4 Concluding Remarks

Conventional techniques used for the separation of targeted solute from the aqueous streams have the disadvantage of high energy consumption, extreme use of chemicals, complicated operating conditions, and high capital and working cost. The liquid membrane on the other side is a more efficient technique as it provides simultaneous extraction and stripping in a solo unit, low consumption of chemicals, and low operational cost. Green emulsion liquid membrane is a type of LM technique that focuses on the removal of targeted solute through a greener approach. Green solvents being non-toxic, non-volatile, reusable, and degradable in nature makes them a suitable solvent to be used in the field of separation science. This saves the environment and also reduces the overall working cost of the system. Parametric studies indicate that several factors like concentration of surfactant, carrier, types of diluents, effect of volume ratio of external feed phase to emulsion, agitation speed, effect of internal aqueous phase concentration, and emulsification time play a substantial role in the removal of targeted solute from the aqueous streams. The statistical tools optimize

the process variables and improve the process effectiveness. A broad scope exists towards the use of greener solvents and more studies can be done in order to explore more solvents of vegetable/plant origin. The GELM approach can further be used for the removal and recovery of several other metal ions and compounds from the aqueous streams.

Author Contribution Akash R. Raval: Conceptualization, methodology, investigation, writing, original draft preparation, and data curation; Himanshu P. Kohli: Visualization, data curation, writing, editing and reviewing; Omprakash K. Mahadwad: Supervision and reviewing.

Data Availability All the data used in the paper are presented in the form of tables and/or figures.

Declarations

Conflict of Interest The authors declare no competing interests.

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