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Parameter Screening of Remazol Black Extraction from Liquid Waste Using Emulsion Liquid Membrane

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ABSTRACT

The textile industry generates substantial liquid waste, primarily containing stable and toxic dye compounds that persist in aquatic environments, posing severe ecological and health risks. Among these, Remazol Black is a commonly used anionic dye known for its resistance to degradation. This study investigates emulsion liquid membrane (ELM) technology, employing waste cooking oil as a sustainable alternative to petroleum-based diluents for extracting Remazol Black dye from wastewater. The ELM process utilizes Span 80 as a surfactant, Aliquat 336 and D2EHPA as carrier agents, and NaOH as a stripping agent to effectively encapsulate and transport dye molecules. Key parameters, including treat ratio, solute concentration, and surfactant concentration, were screened to determine their impact on extraction efficiency. Results indicate that increasing the treat ratio enhances extraction efficiency, reaching up to 96.4% at a 1:2 ratio, while higher solute concentrations further improve dye removal. However, increased surfactant concentration beyond 1% reduced efficiency due to emulsion stability issues. This study highlights the potential of ELM technology with waste-derived materials for effective dye removal, providing a foundation for scalable, environmentally friendly wastewater treatment solutions.

1. Introduction

The rise of consumerism has intensified the demand for "fast fashion", a business model defined by rapid changes in fashion trends and frequent product turnover. This shift has significantly increased textile production and, consequently, textile industry waste, particularly liquid waste laden with dyes, as its primary pollutant [1], [2]. In 2020 [3], approximately 200,000 tons of dyes entered aquatic ecosystems worldwide, posing substantial environmental risks [4]. Reactive dyes like Remazol Black (Reactive Black 5) persist in water bodies. This dye, an anionic and reactive azo compound, resists breakdown due to its chemical stability, causing long-term pollution issues. Widely used in textiles, Remazol Black enhances fabric color durability, yet its resilience allows it to remain in the water, leading to aesthetic and toxicological impacts on ecosystems and human health. Potential human health risks include respiratory and skin irritation and even cellular mutations linked to cancer [3], [5].

The environmental and health risks associated with such stable dyes underscore the critical need for efficient treatment technologies for textile wastewater. Conventional methods, such as adsorption, coagulation [6], filtration [5], [7], [8], and biodegradation [9], each offer specific advantages and drawbacks, often requiring complex configurations that limit scalability [6], [8]. An emerging alternative is the Emulsion Liquid Membrane (ELM) technique, which is highly selective and operates under simpler, energy-efficient conditions compared to traditional methods [10], [11]. ELM utilizes surfactants and carrier agents to encapsulate and transport dye molecules across a membrane, facilitating effective dye separation without side reactions. Additionally, ELM can employ environmentally sustainable materials, such as waste cooking oil, as a diluent, thereby integrating waste reuse into the process.

Emulsion liquid membrane (ELM) is a highly specialized technology for liquid-liquid extraction that has garnered considerable attention in recent years due to its notable advantages [12]–[14]. This technique utilizes the creation of a stable emulsion consisting of an internal dispersed phase and an external continuous phase, where mass transfer occurs through the liquid membrane. The internal phase consists of a stripping agent and, in some cases, a carrier agent [15], [16]. Both of these compounds are responsible for transporting solute from the feed to the inside of the membrane structure, which is why they are referred to as the internal phase. The external phase is alternatively referred to as the feed solution. The ELM technology also involves diluent organic materials in the system.

This study investigates the use of ELM technology, focusing on waste cooking oil as an alternative to conventional petroleum-based diluents [17]–[22], which can be environmentally problematic. Previous studies have highlighted the potential of ELMs for various separation applications, but few have addressed the specific challenges posed by non-petroleum diluents [23], such as membrane stability. This research aims to optimize the operating parameters—including carrier type, dye concentration, surfactant concentration, treat ratio, settling time, and extraction time—to enhance the extraction efficiency of Remazol Black dye from wastewater. Specifically, this study seeks to address the stability concerns of waste cooking oil as a membrane medium and identify the key operational factors that maximize dye extraction.

The parameters examined include the types of carrier agents used (5% w/v D2EHPA; 5% w/v Aliquat 336; or 2.5% D2EHPA + 2.5% w/v Aliquat 336), dye concentration (further referred as solute concentration) (15, 30, 50, 100 ppm), volumetric ratio between membrane and feed solution (further referred as treat ratio) (1:1, 1:2, 1:3), surfactant concentration (1, 3, 5 %w/v), settling time (30 minutes and 18 hours), and extraction time (5, 10, 15 minutes).

This preliminary work offers insights into optimizing ELM for scalable, sustainable application in textile wastewater treatment via identifying key operational parameters influencing the effectiveness of ELM in removing remazol black from textile wastewater, with a focus on improving the stability and sustainability of waste cooking oil as a membrane alternative

2. Research Methodology

2.1. Materials

The diluent utilized in this experiment, waste cooking oil (WCO), was collected from domestic waste and went through simple pretreatment via filtering out solid contaminants. Sodium hydroxide (NaOH) was purchased from Sigma Aldrich Co. and used as the stripping agent. The carrier agents were Aliquat 336 (Loba Chemie PVT.) and D2EHPA (Sigma Aldrich Co.), whereas the surfactant was Span 80 (Sigma Aldrich Co.). Remazol black was dissolved in deionized water to mimic textile waste.

2.2. Procedures

1) *Emulsion Liquid Membrane (ELM) Preparations*

A 0.1 N stripping solution was prepared by dissolving a 0.6 gram NaOH pellet in deionized water to get 150 mL of solution. At the same time, the carrier agent (with variations of 5% w/v D2EHPA, 5% w/v Aliquat 336, or 2.5% w/v D2EHPA + 2.5% w/v Aliquat 336) and Span 80 (with variations of 1, 3, or 5% w/v) were added to a set amount of WCO at 20 mL to create the organic phase. The organic phase was agitated for 10 minutes at 1000 rpm while droplets of stripping solution were introduced continuously. The stripping solution and organic phase had a fixed volumetric ratio of 1:3.

2) *Dye Extraction Process*

Remazol black in different weights was dissolved in deionized water, resulting in a dye-containing solution with varying solute concentrations of 15, 30, 50, and 100 ppm. Each solution, acting as feed, was contacted with the ELM made in step B. A 250 mL beaker containing homogenized ELM was stirred at 300 rpm while feed solution was added in droplets. Five, ten, and fifteen minutes of extraction time were performed. The ELM to feed solution volumetric ratio (known as treat ratio) was varied at 1:1, 1:2, and 1:3. After that, the extracted liquid was separated from the membrane for either 30 minutes or 18 hours (overnight) using a separating funnel, which was held still to facilitate separation by gravitational force. The whole process of extraction and separation was allowed to proceed at room temperature.

The heavier phase, which included extracted feed, went through a UV-Vis Spectrophotometer (Thermo Scientific – Genesys 10 UV) analysis (λ = 595 nm). In contrast, the lighter phase of the used membrane underwent a demulsification procedure.

3) Demulsification Process

The used membrane was placed in a water bath carefully controlled to maintain a temperature of 40 °C for 10 minutes, ensuring a homogenized temperature profile and preventing overheating. The heavier phase, which contained an enriched stripping agent, was separated from the lighter phase, composed of an organic mixture. The separation of the two phases aimed to retrieve the used membrane for further possible cycle(s) and to recover the enriched stripping agent for further analysis. The UV-Vis Spectrophotometer was used to analyze the enriched stripping agent (λ = 595 nm).

4) Emulsion Liquid Membrane (ELM) Performance

Three distinct parameters were used to evaluate the quality of ELM performance. The extraction efficiency evaluates the effectiveness of the ELM in removing solutes, providing a measure of the ELM's functional performance, determined by applying Equation (1)

$$\text{Extraction (\%)} = \frac{C_{eo} - C_e}{C_{eo}} \times 100 \tag{1}$$

On the other hand, the damage that the membrane sustained after use can be measured using membrane stability value, which provides the measure of the stability factor of the membrane, which is a crucial factor in separation longevity, and breakage percentage, which is a value used to assess the ELM structural integrity, as shown in Equation (2) and (3) below

$$\text{Breakage (\%)} = \frac{V_{ei} - V_{ef}}{V_{ei}} \times 100 \tag{2}$$

$$\text{Stability (\%)} = \frac{V_t - V_w}{V_t} \times 100 \tag{3}$$

C_{e0} and C_e represent the feed phase's initial and final solute concentrations, respectively. The emulsion volume before and after the extraction process is defined by each V_{ei} and V_{ef}, while Vt denotes the total emulsion volume, and Vw denotes the separated aqueous phase.

Breakage percentage defines the proportion of emulsion liquid membrane that breaks, allowing the stripping agent to leak into the system. At the same time, the membrane stability value indicates the resistance of each membrane globule to coalesce with one another.

3. Results and Discussion

Six parameters in Table 1 are varied to identify and evaluate crucial factors for remazol black extraction. A literature review was done before the experiment as the base for choosing the values of each parameter.

Table 1. Parameters of the experiment

Parameter	Unit	Value
Carrier	%w/v	5% D2EHPA; 5% Aliquat 336; 2.5% D2EHPA + 2.5% Aliquat 336
Solute	ppm	15; 30; 50; 100
Treat ratio	-	1:1; 1:2; 1:3
Surfactant	%w/v	1; 3; 5
Settling	mins; hrs	30 mins; 18 hrs.
Extraction	mins	5; 10; 15

3.1. Effect of carrier agent

This study examined the effectiveness of different carrier agents—Aliquat 336, D2EHPA, and a combination of the two—on the extraction efficiency and membrane stability in removing Remazol Black dye. The results, as illustrated in Fig 1., demonstrated that using Aliquat 336 alone achieved the highest extraction efficiency, reaching 91.3%, while D2EHPA alone was incompatible with Remazol Black extraction, showing no significant dye removal. The combination of Aliquat 336 and D2EHPA led to a decreased extraction efficiency of 88.8%, indicating that adding D2EHPA detracts from the efficacy of Aliquat 336 in binding dye molecules.

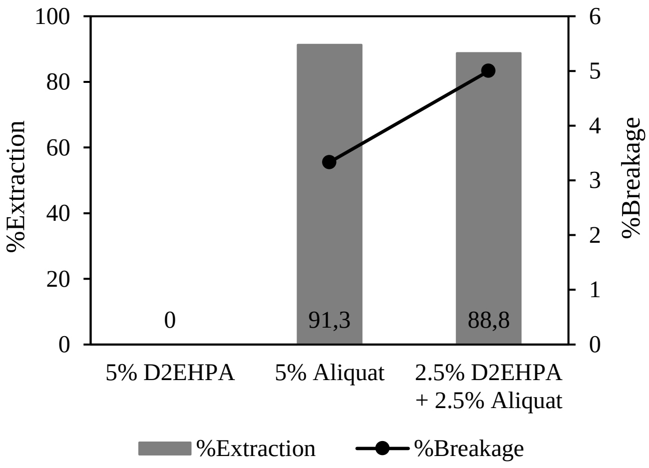

Fig. 1. Effect of different types of carrier agents on extraction efficiency and membrane breakage

Aliquat 336, a quaternary ammonium compound, effectively interacts with the anionic dye due to its positive ionic charge, facilitating complex formation and enhancing the mass transfer of dye molecules across the membrane. This compound also acts as a mass transfer catalyst, promoting efficient solute migration from the feed solution into the internal phase. Conversely, D2EHPA, a phosphoric acid-based carrier, is better suited for cationic dye and metal extraction, making it less effective with the anionic Remazol Black [19], [24]. The detailed mechanism of the mass transfer of remazol black is depicted in Fig. 2.

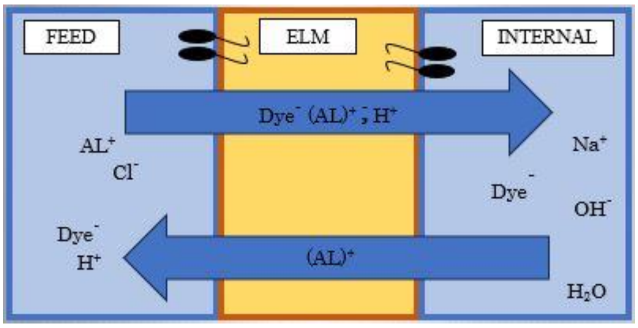

Fig. 2. Mass transfer mechanism for remazol black extraction process using ELM

Dye ions and carrier ions are diffused in the bulk phase of the feed solution and then move toward the feed-membrane interface. Here, at this point, a dye-carrier complex is formed through the exchange of chloride anion from Aliquat 336 with anion from remazol black, following the reaction below:

$$(\mathrm{AL})^{+}\mathrm{Cl}^{-} + \mathrm{Dye}^{-}\mathrm{H}^{+} \rightarrow \mathrm{Dye}^{-}(\mathrm{AL})^{+} + \mathrm{H}^{+} + \mathrm{Cl}^{-} \tag{4}$$

where $(AL)^+Cl^-$ and Dye^-H^+ denote Aliquat 336 and dye solution, respectively. Ionized dye and Aliquat 336 dissociate into AL^+, Cl^-, H^+, and Dye^-. $(AL)^+$ and Dye^- ions formed a complex of dye-carrier ($Dye^-(AL)^+$), while H^+ and Cl^- ions remain unreacted. The dye-carrier complex ($Dye^-(AL)^+$) Proceeds to diffuse into the bulk phase of the internal solution and interact with the stripping agent (NaOH) to release back the dye anion and carrier ion following the reaction below:

$$\mathrm{Dye^-(AL)^+ + H^+ + NaOH \rightarrow Na^+Dye^- + (AL)^+ + H_2O} \quad (5)$$

The released dye anion ($\mathrm{Na^+Dye^-}$) is then trapped in the internal phase, whereas the released carrier ion (AL+) re-enters the bulk phase of the feed solution to extract more remazol black dye. This suggests that remazol black dye extraction can be enhanced with the presence of Aliquat 336.

On the other hand, D2EHPA is a phosphoric acid carrier, which is more compatible with aide cation dye removal. [23], [24] and heavy metal [12]. Additionally, using a combination of Aliquat 336 and D2EHPA increased the membrane's viscosity and density, resulting in larger and less stable emulsion globules, as evidenced by a rise in the breakage percentage from 3.3% (with Aliquat 336 alone) to 5% when D2EHPA was added. This higher viscosity can impede dye transfer, reducing overall extraction efficiency [25] and making the membrane more prone to breaking under processing conditions [12].

3.2. Effect of solute concentration

Varying Remazol Black dye concentrations in the feed solution evaluated the impact of solute concentration on extraction efficiency and membrane stability. As shown in Fig. 3, results show that extraction efficiency also rises as the solute concentration increases, achieving values of 66.7%, 67%, 76.8%, and 79.8% at concentrations of 15, 30, 50, and 100 ppm, respectively. This trend aligns with mass transfer theory, where a more significant concentration gradient between the feed and internal phases drives higher solute migration rates, thus enhancing dye extraction efficiency [25], [26]. However, the increase in efficiency is not linear; the jump in extraction efficiency from 30 ppm to 50 ppm is more pronounced than that from 50 ppm to 100 ppm, suggesting a saturation point around 50 ppm where further concentration increases yield diminishing returns.

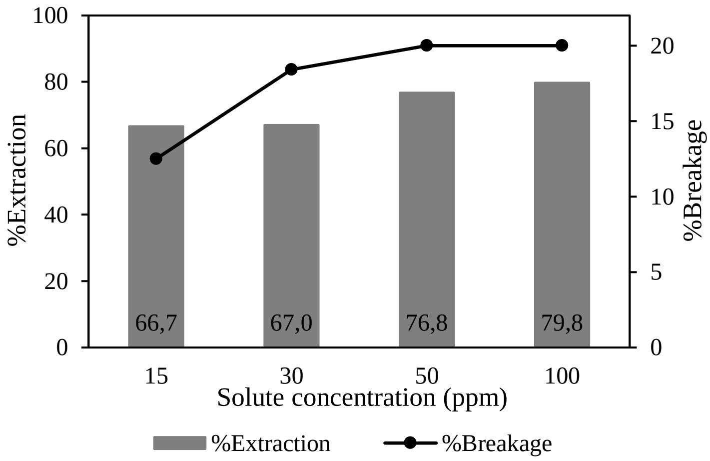

Fig. 3. Effect of solute concentration on extraction efficiency and membrane breakage

Higher solute concentrations also affect membrane stability. As solute concentration rises, feed solution viscosity increases, forming more extensive, less stable emulsion globules. These globules become oversaturated more rapidly, causing structural stress that raises the membrane breakage percentage [12], [27]. For instance, the breakage rate increased from 12.5% at 15 ppm to 18.4% at 30 ppm, leveling off around 50 ppm, indicating a potential capacity limit for solute loading. This observation highlights the importance of optimizing solute concentration to balance high extraction efficiency with membrane stability.

3.3. Effect of treatment ratio

The treat ratio, defined as the volumetric ratio between the emulsion liquid membrane (ELM) and the feed solution, significantly influences the efficiency of dye extraction, as depicted in Fig. 4. Increasing the treat ratio from 1:1 to 1:2 enhanced extraction efficiency from 92% to 96.4%, as a higher ratio provides a more significant driving force for mass transfer [25], allowing more dye molecules to move from the feed solution into the internal phase. However, further increasing the treatment ratio to 1:3 resulted in a drop in efficiency to 91.3%, likely due to excessive water content causing globule swelling, which reduces the interfacial area available for mass transfer [20], [27], [28].

The increase in water content at higher treatment ratios also impacts membrane stability. Excess water causes globules to swell, thinning the membrane walls and making them more prone to breakage [25]. As shown in Fig. 4, this instability was evidenced by a rise in breakage percentage from 3.75% at a 1:1 ratio to 6.67% at a 1:2 ratio, suggesting that optimal treatment ratios are necessary to balance efficient dye extraction and membrane durability. These findings emphasize the need to carefully control the treat ratio to maximize extraction outcomes while preserving membrane integrity.

3.4. Effect of surfactant concentration

Surfactant concentration plays a crucial role in stabilizing the emulsion and enhancing dye extraction by reducing the interfacial tension between the membrane's organic and aqueous phases [26], [28]. In this study, a surfactant concentration of 1% w/v resulted in the highest extraction efficiency of 95.3%, as shown in Fig. 5. However, increasing the surfactant concentration to 3% and 5% w/v lowered the extraction efficiency to 92% and 94.3%, respectively. This decline is likely due to excess surfactant, which causes droplet swelling and increases emulsion viscosity, hindering dye transfer by raising surface resistance [18], [19].

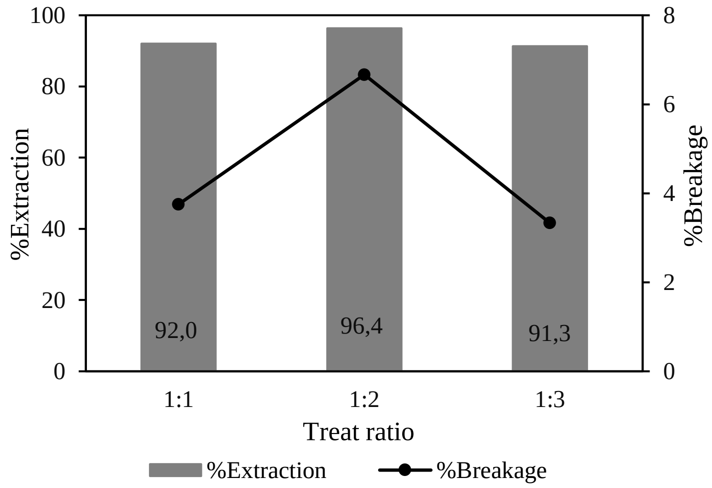

Fig. 4. Effect of treat ratio on extraction efficiency and membrane breakage

Surfactant concentration also affects membrane stability. At lower concentrations (1% w/v), the breakage percentage was higher, around 15%, as insufficient surfactant coverage destabilizes the emulsion. Increasing the surfactant to 3% w/v reduced breakage to 9.2%, but further increase to 5% w/v raised breakage again to 17.5%. Higher surfactant levels can accelerate droplet coalescence, thinning the membrane walls [12] And compromising structural integrity. These findings suggest an optimal surfactant concentration of 1–3% w/v, which balances extraction efficiency with membrane stability, minimizing breakage and ensuring effective dye removal.

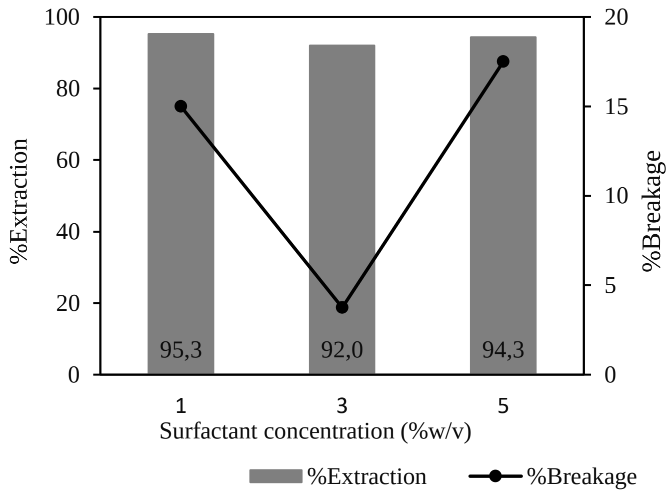

Fig. 5. Effect of surfactant concentration on extraction efficiency and membrane breakage

3.5. Effect of settling time

Settling time, the duration allowed for phase separation after extraction, markedly impacts both extraction efficiency and membrane stability, as depicted in Fig. 6. When the settling time was set to 30 minutes, extraction efficiency was low at 6.3%, likely due to incomplete separation, which prevents complete dye transfer from the emulsion to the stripping phase. This short settling time also correlated with a high breakage percentage of 15%, as the emulsion remained unstable without sufficient time to form a cohesive barrier [29].

In contrast, extending the settling time to 18 hours significantly improved extraction efficiency, achieving 92% while simultaneously reducing membrane breakage to 3.75%. The prolonged settling period stabilizes the emulsion, facilitating thorough separation and reducing emulsion stress, which minimizes breakage. These results underscore the importance of optimal settling times for achieving high extraction efficiency and stable membrane performance, suggesting that overnight settling provides the conditions for effective dye separation.

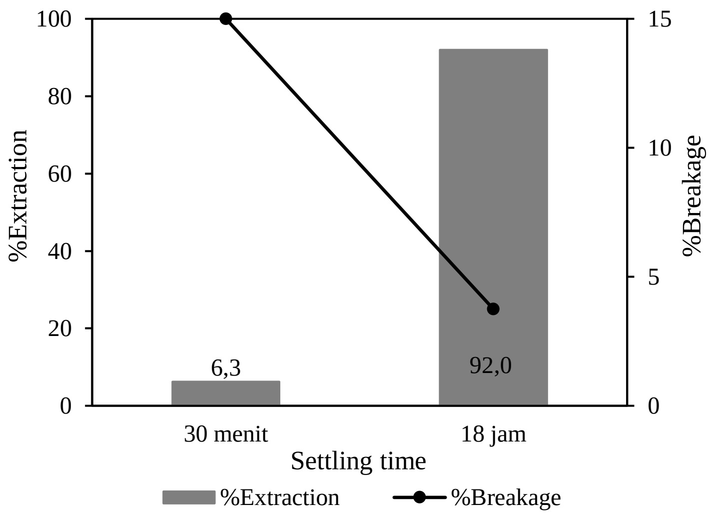

Fig. 6. Effect of settling time on extraction efficiency and membrane breakage

3.6. Effect of extraction time

Extraction time, the duration of contact between the feed solution and the emulsion liquid membrane (ELM), influences both the efficiency of dye removal and membrane stability, as depicted in Fig. 7. In this study, a contact time of 5 minutes yielded the highest extraction efficiency at 79.8%. However, extending the extraction time to 10 and 15 minutes decreased efficiency to 77.8% and

73.7%, respectively. This decline likely results from prolonged stirring, which increases globule collisions, causing them to shrink and reducing the available surface area for dye transfer [28].

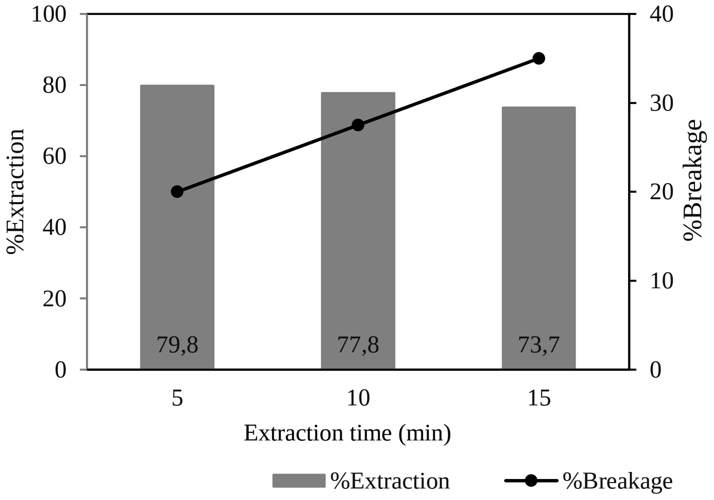

Fig. 7. Effect of extraction time on extraction efficiency and membrane breakage

Extended extraction times also impact membrane stability, with longer durations leading to higher breakage rates. Increased stirring time promotes droplet coalescence, weakening the emulsion structure and causing membrane breakage [12], [19]. These findings indicate that shorter extraction times optimize dye removal efficiency and enhance membrane integrity by minimizing structural stress. Therefore, a 5-minute extraction time is suggested as optimal for balancing effective dye extraction with membrane stability. This shorter extraction time not only helps ensure optimal extraction but is also efficient in terms of time and energy usage.

3.7. Pareto chart

The Pareto chart analysis was employed to identify and prioritize the operational parameters that most significantly impact the extraction efficiency of Remazol Black dye using the emulsion liquid membrane (ELM) technique. As shown in Fig. 8, the chart highlights three primary factors: treat ratio, surfactant concentration, and solute concentration, ranked in descending order of influence based on their Standardized Effect Size (SES). Treat ratio emerged as the most impactful parameter, indicating its critical role in maximizing dye transfer across the membrane. Surfactant concentration and solute concentration were also significant, affecting emulsion stability and mass transfer efficiency, respectively [15], [25].

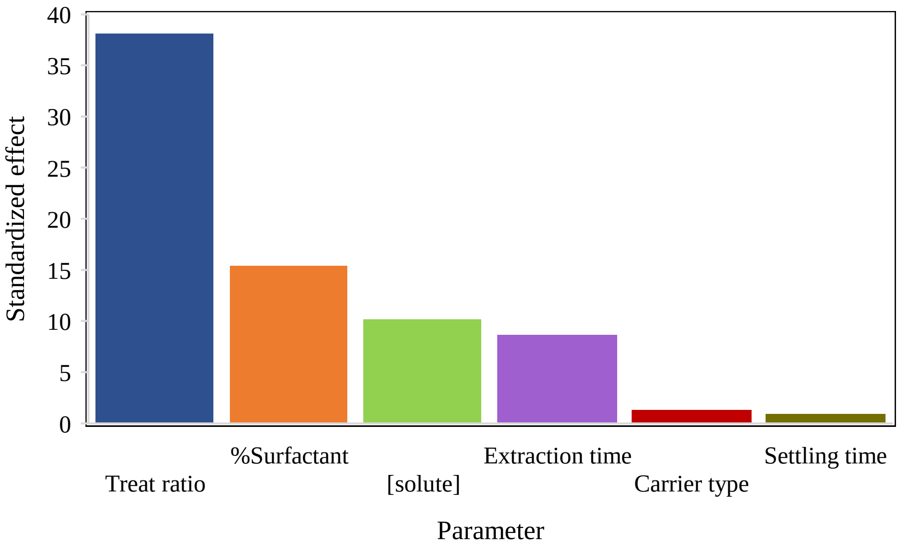

Fig. 8. Pareto chart for remazol black extraction

Parameters such as extraction time, carrier type, and settling time had comparatively minor effects on extraction efficiency. These findings suggest optimizing treat ratio, surfactant concentration, and solute concentration could yield the most significant improvements in ELM performance. This targeted approach to parameter optimization provides a foundation for refining the ELM process to enhance extraction efficiency and scalability for industrial wastewater applications.

4. Conclusion

This study evaluated six critical parameters in the emulsion liquid membrane (ELM) process for extracting Remazol Black dye from wastewater, emphasizing waste cooking oil as an eco-friendly diluent. Pareto chart analysis identified treat ratio, surfactant concentration, and solute concentration as the most influential factors affecting extraction efficiency. Optimal conditions, including a 1:2 treat ratio, 1% surfactant concentration, and higher solute concentrations, achieved 96.4% dye extraction while maintaining membrane stability. These findings underscore ELM technology's potential as an efficient, sustainable solution for industrial dye removal.

Future research should focus on fine-tuning these key parameters through advanced optimization techniques, such as response surface methodology, to maximize extraction efficiency and membrane durability. Further studies could also explore integrating these optimized ELM processes into large-scale treatment systems, enabling cost-effective and scalable industrial applications.

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References

[1] K. Niinimaki, G. Peters, H. Dahlbo, P. Perry, T. Rissanen, and A. Gwilt, "The environmental price of fast fashion," *Nat. Rev. Earth Environ.*, vol. 1, pp. 189–200, Apr 2020, doi: 10.1038/s43017-020-0039-9.

[2] B. Garcia-ortega, J. Galan-cubillo, F. J. Llorens-montes, and B. de M. Molina, "Sufficient consumption as a missing link toward sustainability : The case of fast fashion," *J. Clean. Prod.*, vol. 399, pp. 136678, May 2023, doi: 10.1016/j.jclepro.2023.136678.

[3] M. M. Felista, W. C. Wanyonyi, and G. Ongera, "Adsorption of anionic dye (Reactive black 5) using macadamia seed Husks: Kinetics and equilibrium studies," *Sci. African*, vol. 7, pp. e00283, Mar 2020, doi: 10.1016/j.sciaf.2020.e00283.

[4] N. Apriyani, "Industri batik: Kandungan limbah cair dan metode pengolahannya," *Media Ilm. Tek. Lingkung.*, vol. 3, no. 1, pp. 21–29, Mar 2018, doi: 10.33084/mitl.v3i1.640.

[5] N. Fitriana and M. Rahmayanti, "Aplikasi membran filter keramik untuk menurunkan konsentrasi zat warna Remazol Red dan nilai COD limbah cair batik," *Al Kim.*, vol. 8, no. 2, pp. 159–167, Dec 2020, doi: 10.24252/al-kimia.v8i2.15932.

[6] H. Soleimani, K. Sharafi, J. Amiri Parian, J. Jaafari, and G. Ebrahimzadeh, "Acidic modification of natural stone for Remazol Black B dye adsorption from aqueous solution- central composite design (CCD) and response surface methodology (RSM)," *Heliyon*, vol. 9, no. 4, pp. e14743, Apr 2023, doi: 10.1016/j.heliyon.2023.e14743.

[7] Kiswanto, L. N. Rahayu, and Wintah, "Pengolahan limbah cair batik menggunakan teknologi membran nanofiltrasi di Kota Pekalongan," *J. Litbang Kota Pekalongan*, vol. 17, pp. 72–82, Dec 2019, doi: 10.54911/litbang.v17i0.109.

[8] M. Hidayah, M. Mujiburohman, and N. Hidayati, "Synthesis of polyamide-Al_2O_3 nanocomposite membranes using the nanofiltration phase," in *Mechanical Engineering, Science and Technology International Conference*, 2022, vol. 1, pp. 611–627.

[9] P. S. Komala, Y. Dewilda, and Z. Wulandari, "Biodegradation of azo dye Remazol Black 5 by mono culture bacteria with tempe industrial wastewater as co-substrate," *Int. J. Technol.*, vol. 3, pp. 240–248, 2013, doi: 10.14716/ijtech.v4i3.120.

[10] A. R. Raval, H. P. Kohli, and O. K. Mahadwad, "Application of emulsion liquid membrane for removal of malachite green dye from aqueous solution: Extraction and stability studies," *Chem. Eng. J. Adv.*, vol. 12, pp. 100398, Nov 2022, doi: 10.1016/j.ceja.2022.100398.

[11] M. Wakle and S. Khuntia, "Developments and studies in bio-emulsion liquid membranes (Bio-ELM) synthesis, parameters studies, characterizations, and applications," *J. Water Process Eng.*, vol. 56, pp. 104300, Dec 2023, doi: 10.1016/j.jwpe.2023.104300.

[12] H. K. Admawi and A. A. Mohammed, "A comprehensive review of emulsion liquid membrane for toxic contaminants removal: An overview on emulsion stability and extraction efficiency," *J. Environ. Chem. Eng.*, vol. 11, no. 3, pp. 109936, Jun 2023, doi: 10.1016/j.jece.2023.109936.

[13] A. E. Kostanyan, A. A. Voshkin, V. V Belova, and Y. A. Zakhodyaeva, "Modelling and comparative analysis of different methods of liquid membrane separations," *Membranes (Basel).*, vol. 13, no. 6, pp. 554–567, May 2023, doi: 10.3390/membranes13060554.

[14] A. Kusumastuti, S. Anis, R. Syamwil, and A. L. Ahmad, "Emulsion liquid membrane for textile dyes removal: Extraction process," *J. Phys. Sci.*, vol. 29, pp. 175–184, Jul 2018, doi: 10.21315/jps2018.29.s2.13.

[15] P. Daraei, S. Zereshki, and A. Shokri, "Application of nontoxic green emulsion liquid membrane prepared by sunflower oil for water decolorization : Process optimization by response surface methodology," *J. Ind. Eng. Chem.*, vol. 77, pp. 215–222, Sept 2019, doi: 10.1016/j.jiec.2019.04.039.

[16] A. Shokri, P. Daraei, and S. Zereshki, "Water decolorization using waste cooking oil: An optimized green emulsion liquid membrane by RSM," *J. Water Process Eng.*, vol. 33, pp. 101021, Feb 2020, doi: 10.1016/j.jwpe.2019.101021.

[17] M. M. H. S. Buddin, N. D. S. M. N. Salizan, A. L. Ahmad, O. S. J. Elham, and A. R. Rashidi, "Water-in-oil-in-water (W/O/W) emulsion instability in emulsion liquid membrane: membrane breakage," *J. Phys. Conf. Ser.*, vol. 1349, no. 1, pp. 012106, Nov 2019, doi: 10.1088/1742-6596/1349/1/012106.

[18] S. A. Parbat, B. A. Bhanvase, and S. H. Sonawane, "Investigation on liquid emulsion membrane (LEM) prepared with hydrodynamic cavitation process for cobalt (II) extraction from wastewater," *Sep. Purif. Technol.*, vol. 237, Apr 2020, doi: 10.1016/j.seppur.2019.116385.

[19] M. B. Rosly, N. Jusoh, N. Othman, H. A. Rahman, N. F. M. Noah, and R. N. Raja Sulaiman, "Synergism of Aliquat336-D2EHPA as carrier on the selectivity of organic compound dyes extraction via emulsion liquid membrane process," *Sep. Purif. Technol.*, vol. 239, pp. 116527, May 2020, doi: 10.1016/j.seppur.2020.116527.

[20] P. Chhaganlal, B. Shailesh, A. Ghodke, and B. Apparao, "Intensified hydrodynamic cavitation - based process for the production of liquid emulsion membrane (LEM) for the extraction of chromium (VI) ions," *Int. J. Environ. Res.*, vol. 15, no. 2, pp. 313–320, Feb 2021, doi: 10.1007/s41742-021-00322-4.

[21] M. Djenouhat, F. Bendebane, L. Bahloul, M. E. H. Samar, and F. Ismail, "Optimization of methylene blue removal by stable emulsified liquid membrane using plackett–burman and box–behnken designs of experiments," *R. Soc. Open Sci.*, vol. 5, no. 2, Feb 2018, doi: 10.1098/rsos.171220.

[22] M. Hazarel, Z. Mohd, A. L. Ahmad, and L. Rajandram, "Emulsion liquid membrane screening for ibuprofen removal from aqueous solution," *J. Phys. Sci.*, vol. 33, no. 1, pp. 109–122, Apr 2022, doi: 10.21315/jps2022.33.1.8.

[23] H. Purnama, N. M. Jannah, and N. Hidayati, "Removal of rhodamine B from liquid waste using used cooking oil solvent in emulsion liquid membranes (ELM)," *Eksergi*, vol. 21, no. 2, pp. 94–102, Apr 2024.

[24] H. W. Khan, A. A. M. Elgharbawy, M. A. Bustam, M. Goto, and M. Moniruzzaman, "Vegetable oil-ionic liquid-based emulsion liquid membrane for the removal of lactic acid from aqueous streams: emulsion size, membrane breakage, and stability study," *ACS Omega*, vol. 7, no. 36, pp. 32176–32183, Aug 2022, doi: 10.1021/acsomega.2c03425.

[25] N. Othman, R. N. Raja Sulaiman, H. A. Rahman, N. F. M. Noah, N. Jusoh, and M. Idroas, "Simultaneous extraction and enrichment of reactive dye using green emulsion liquid membrane system," *Environ. Technol. (United Kingdom)*, vol. 40, no. 4, pp. 1–20, Jan 2018, doi: 10.1080/09593330.2018.1424258.

[26] C. Das, M. Rungta, G. Arya, S. DasGupta, and S. De, "Removal of dyes and their mixtures from aqueous solution using liquid emulsion membrane," *J. Hazard. Mater.*, vol. 159, no. 2–3, pp. 365–371, Nov 2008, doi: 10.1016/j.jhazmat.2008.02.027.

[27] N. Othman, O. Zing-Yi, N. Harruddin, R. Norimie, N. Jusoh, and S. Nazrah Zailani, "Carrier assisted emulsion liquid membrane process for recovery of basic dye from wastewater using continuous extractor," *J. Teknol.*, vol. 67, no. 2, pp. 69–74, Feb 2014, doi: 10.11113/jt.v67.2739.

[28] N. Othman, O. Z. Yi, S. N. Zailani, E. Z. Zulkifli, and S. Subramaniam, "Extraction of rhodamine 6G dye from liquid waste solution: study on emulsion liquid membrane stability performance and recovery," *Sep. Sci. Technol.*, vol. 48, no. 8, pp. 1177–1183, Apr 2013, doi: 10.1080/01496395.2012.731123.

[29] M. Muhammad, H. Shah, A. L. Ahmad, A. Tasneem, A. Khalil, and S. W. Puasa, "A review of demulsification technique and mechanism for emulsion liquid membrane applications," *J. Dispers. Sci. Technol.*, vol. 0, no. 0, pp. 1–18, Nov 2020, doi: 10.1080/01932691.2020.1845962.