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Research Article

Performance of Poly-BADGE 2:1 Based Polymer Inclusion Membrane for Selective Transport of Malachite Green: Optimization and Stability Studies

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Abstract

Malachite green (MG) is a persistent and toxic triphenylmethane dye. This dye is widely used in textile industries, contributing to significant contamination of aquatic environments. The development of selective and efficient separation methods for MG therefore remains an important challenge. In this study, MG transport was investigated using a polymer inclusion membrane (PIM) incorporating poly-bisphenol A diglycidyl ether (Poly-BADGE 2:1) as a carrier. The effects of key operational parameters, as well as the presence of competing metal ions (i.e., Pb(II) and Cu(II)), were systematically evaluated. The membrane was prepared using Poly-BADGE 2:1 as the carrier, polyvinyl chloride as the base polymer, and dibenzyl ether as the plasticizer. MG concentration was determined by UV–vis spectrophotometry at 617 nm, while membrane characteristics before and after transport were analyzed using Scanning Electron Microscopy (SEM) and Fourier-Transform Infrared (FTIR) instruments. The results showed that MG transport reached 90.90% under optimal conditions (source phase pH 7, receiving phase 1.25 M HNO_3, membrane thickness T_{54}, carrier concentration 0.09 M, and transport time of 24 h). In the presence of Pb(II) and Cu(II) ions, the transport efficiency decreased to 85.67%, compared to 91.32% without metal ions, suggesting competitive interactions at the carrier sites. These findings demonstrate that Poly-BADGE-based PIMs provide an effective and selective system for MG separation, with good structural stability and promising potential for wastewater treatment applications.

Keywords: malachite green; membrane stability; Poly-BADGE; polymer inclusion membrane; selective separation

1. INTRODUCTION

Environmental pollution from textile wastewater remains a critical issue in many developing countries, including Indonesia [1]. A major contributor to this problem is the widespread use of synthetic dyes such as malachite green (MG), which are highly persistent, toxic, and resistent to natural degradation processes [2]. The presence of MG in aquatic system not only causes visible discoloration, but also poses serious risks to aquatic organisms and human health due to its carcinogenic nature [3]. Therefore, the removal and selective separation of MG from industrial wastewater has become an important challenge in environmental chemistry.

Various treatment methods have been explored for dye removal, including adsorption [4], coagulation [5], membrane [6], and photodegradation [7]. However, these approaches often suffer from inherent limitations such as incomplete removal, high operational costs, and difficulties in material regeneration [8,9]. In this context, polymer inclusion membrane (PIM) technology has emerged as a promising alternative due to its high selectivity and ability to facilitate the transport of specific ionic species, including MG [10] and phenol [11] through carrier-mediated mechanisms.

The performance of PIM system is strongly governed by the properties of the carrier, which directly influence transport efficiency and membrane stability [12]. A variety of carriers have been reported for dye and organic compound transport, such as Co-EDVB [13], Co-EDAF [14], and Co-EEGDMA [15]. Nevertheless, many of these systems still exhibit limited long-term stability and declining performance upon repeated use [16]. Furthermore, the relationship between carrier structure, composition ratio, and transport behavior of cationic dyes remains insufficiently understood [17]-[20].

Poly-Bisphenol A Diglisidyl Ether (abbreviated as Poly-BADGE) is an epoxy-based material containing reactive functional groups capable of

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Table 1. Composition of PIM membrane components used in membrane preparation.

Membrane	Poly-BADGE 2:1 (g)	PVC (g)	DBE (g)	Total (g)
T_{27} (Thin)	0.0270	0.0864	0.1556	0.2700
T_{54} (Medium)	0.0540	0.1728	0.3132	0.5400
T_{108} (Thick)	0.1080	0.3456	0.6264	1.0800

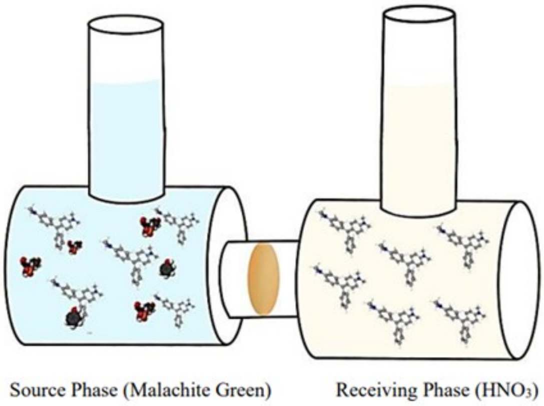

Figure 1. Schematic of the MG transport system.

interacting with cationic dye molecules through hydrogen bonding and electrostatic interactions [21]. Previous studies have demonstrated that the composition ratio of Poly-BADGE influences membrane stability and transport efficiency, particulary in phenol transport system (e.g., 1:1 and 3:1 mass ratio) [22]. However, the application of Poly-BADGE with a 2:1 composition ratio for MG transport in PIM systems has not been investigated. In addition, studies that explicitly correlate operating parameters (such as source phase pH, receiving phase concentration, membrane thickness, and transport time) with membrane physicochemical properties remain limited.

In this study, a Poly-BADGE 2:1-based PIM system was developed to address these gaps. The work focuses on optimizing MG transport conditions, evaluating membrane stability, and elucidating the relationship between membrane structure and transport performance. The findings are expected to contribute to the rational design of more efficient and stable PIM system for selective dye separation in wastewater treatment.

2. MATERIALS AND METHODS

2.1. Materials

The materials used in this study are Poly-BADGE 2:1 (Sigma-Aldrich) as the carrier, MG (Merck), polyvinyl chloride (PVC, Sigma-Aldrich), dibenzyl ether (DBE, Sigma-Aldrich), tetrahydrofuran (THF, Merck), nitric acid (Merck), lead(II) carbonate, and copper(II) sulfate pentahydrate (Merck). Distilled and deionized water are prepared using a GFL distillation unit.

2.2. Methods

2.2.1. PIM Membrane Preparation

PIM membranes were prepared according to the composition listed in Table 1. The membrane components Poly-BADGE 2:1 as the carrier, PVC as the base polymer, and DBE as the plasticizer, were dissolved in 10 mL of THF. The mixture was stirred for 30–60 min until a homogeneous solution was obtained. The solution was then cast into a cylindrical mold (4.5 cm diameter) and allowed to evaporate at room temperature for 72 h. The resulting membranes were designated as T_{27}, T_{54}, and T_{108} based on the total mass of the components, corresponding to thin, medium, and thick membranes, respectively. MG transport experiments were conducted using a two-compartment transport cell separated by the PIM membrane (Figure 1). Each compartment had a diameter of 5 cm, with an effective membrane area of 3.5 cm in diameter and a working volume of 50 mL.

2.2.2. PIM Membrane Characterization

Prior to use, the membranes were dried and weighed using an analytical balance. Membrane thickness was measured at multiple points using a micrometer, an the results were expressed as mean ± standard deviation (n = 3). Functional groups were analyzed using Fourier-transform infrared spectroscopy (FTIR) (Thermo Fisher Scientific type Nicolet iS50-AS), while surface morphology was characterized using scanning electron microscopy (SEM) (Hitachi type FlexSEM 1000 II-Japan). MG concentrations in both source and receiving phases were determined using UV-vis spectrophotometry at 617 nm.

2.2.3. MG Transport Process

The transport performance of MG through the PIM membrane was evaluated by optimizing several operational parameters, including source phase pH, receiving phase concentration, membrane thickness, and carrier concentration.

2.2.3.1. Effect of Source Phase pH

The pH of the MG solution in the source phase was assessed by varying its pH at 5, 6, 7, 8, and 9, while the receiving phase consisted of 50 mL of 1 M HNO_3.

2.2.3.2. Effect of Receiving Phase Concentration

The concentration of HNO_3 in the receiving phase was determined by varying the concentration of HNO_3 to 0.50; 0.75; 1.00; 1.25; and 1.50 M, while the source phase pH was maintained at its optimal value.

2.2.3.3. Effect of Membrane Thickness

Membrane thickness plays an important role in determining the transport efficiency of malachite green in PIM system, as it influences both diffusion resistance and membrane stability. To evaluate this effect, three membranes with different compositions were prepared, resulting in varying thicknesses classified as thin (T_{27}), medium (T_{54}), and thick (T_{108}). These membranes were then used to examine how thickness affects MG transport performance, while the source phase pH and HNO_3 concentration in the receiving phase were kept at their previously optimized values.

2.2.3.4. Effect of Carrier Concentration

The concentrations of polyvinyl chloride and dibenzyl ether were kept constant. The carrier concentration in the membrane solution was varied according to the composition shown in Table 2, ranging from 0.06 to 0.10 M Poly-BADGE 2:1. This variation was carried out to evaluate the effect of carrier concentration on MG complexation and transport efficiency. All transport experiments were performed under the previously optimized conditions.

2.2.3.5. Effect of Competing Metals Ions

This experiment was conducted to evaluate the selectivity of the carrier in transporting MG in the presence of competing metal ions in the source phase. These ions may interfere with the formation of carrier MG complexes during the transport process. Artificial wastewater solutions were prepared consisting of:

1. MG + Pb(II),
2. MG + Cu(II), and
3. MG + Pb(II) + Cu(II),

with each component at a concentration of 25 ppm. All experiments were carried out under the previously optimized conditions to assess the ability of the Poly-BADGE 2:1 based PIM membrane to selectively transport MG in the presence of

Table 2. Variations in the concentration of carriers

Concentration (M)	Poly-BADGE 2:1 (g)	PVC (g)	DBE (g)	Total (g)
0.06	0.0067	0.1728	0.3132	0.5261
0.07	0.0134	0.1728	0.3132	0.5346
0.08	0.0201	0.1728	0.3132	0.5400
0.09	0.0267	0.1728	0.3132	0.5462
0.10	0.0334	0.1728	0.3132	0.5529

Figure 2. PIM membrane with Poly-BADGE 2:1 carrier.

interfering ions.

For each transport experiment, the membrane was placed between the two compartments of the transport cell, and the system was sealed with parafilm. The solution were stirred using a magnetic stir bar for 24 h at room temperature. After the transport process, the membrane was removed and dried for 24 h, and its mass was recorded to evaluate any changes. MG concentrations in both the source and receiving phase were determined using UV-Vis spectrophotometry at 617 nm. Transport performance was evaluated in terms of removal percentage (%Removal) and transport efficiency (% Transport), calculated using the following Equations 1 and 2.

$$\%Removal = \frac{C_0 - C_t}{C_0} \times 100\% \quad (1)$$

$$\%Transport = \frac{C_r}{C_0} \times 100\% \quad (2)$$

where C_0 is the initial MG concentration in the source phase (ppm), C_t is the MG concentration at time t, and C_r is the MG concentration in the receiving phase. All experiments were performed in triplicate (n = 3), and the results are reported as mean ± standard deviation. Error bars in all figures represent the standard deviation of independent measurements. The membrane was further characterized using FTIR and SEM to examine changes in its chemical structure and surface morphology after the transport process.

3. RESULTS AND DISCUSSIONS

3.1. PIM Membrane

The prepared PIM membrane consisted PVC as the base polymer, which provides mechanical strength and prevents carrier leakage, and DBE as a plasticizer, which enhances membrane flexibility and stability. The incorporation of DBE also facilitates the formation of a homogeneous membrane phase, thereby improving the overall transport properties [23]-[25]. The physical appearance of the prepared PIM membrane is shown in Figure 2. Three membrane types (T_{27}, T_{54}, and T_{108}) were deliberately prepared with different total component masses to obtain membranes with distinct thicknesses for subsequent transport study. The inclusion of membrane weight and thickness data is important to highlight the physical differences among the prepared membranes before performance evaluation.

Membrane weight represents the total amount of its constituents, including PVC as the base polymer, DBE as plasticizer, and Poly-BADGE 2:1 as the carrier. An increase in membrane weight corresponds to a higher loading of these components, which may lead to a greater number of available active sites within the membrane matrix. This variation is expected to affect diffusion resistance, carrier solute interactions, and ultimately the transport efficiency of MG across the membrane. Table 3 provides a structural reference linking membrane composition and physical properties to the analysis of membrane thickness effects. The T_{27}, T_{54}, and T_{108} membranes correspond to samples prepared using different total masses of membrane components.

The physical characteristics of the prepared PIM membranes, including membrane weight and thickness before the transport experiments, are presented in Table 3. All measurements were performed in triplicate to ensure reproducibility and reliability. Membrane weight increased with increasing thickness from T_{27} to T_{108}. In addition, the calculated standard deviation (SD) and relative standard deviation (%RSD) values were low for all samples. The %RSD values were below 0.1%, indicating a high level of reproducibility and uniformity in the membrane preparation process.

3.2. Transport of MG

The transport behavior of MG across the Poly-BADGE 2:1 based PIM was evaluated using % Removal and %Transport values. These parameters provide a basis for examining the influence of operational variables on membrane performance and transport efficiency.

3.2.1. Effect of Malachite Green pH

pH variation is one of the optimization parameters in this study, because pH is a factor that can encourage diffusion in the membrane, namely the difference in proton concentration between the source phase and the receiving phase. The presence of this pH difference indicates that the number of protons in the source phase will increase. The results obtained show that pH has a strong effect on %removal shown in Figure 3. The results of this study showed that the %removal of MG at pH 5, 6, 7, 8, and 9 was 76.52%, 81.12%, 85.32%, 82.03%, and 77.97%, respectively. The results show that the optimal conditions with the largest amount of MG concentration transported are shown at pH 7. The optimum transport observed at pH 7 can be attributed to the predominance of the cationic form of MG^+ under near-neutral conditions. In this form, MG interacts effectively with the carrier in the membrane through electrostatic interactions, π–π interactions between aromatic structures, and possible hydrogen bonding with functional groups in the membrane matrix [26]. These interactions contribute to enhanced transport efficiency. At more acidic condition (pH 5 – 6), the transport efficiency decreases, indicating less favorable interactions between MG and the carrier. In this system, optimal transport occurs when the target compound is present under near-neutral pH conditions.

Kiswandono et al. [10] examined the separation of MG using polyeugenol-carrying compounds, an optimal pH of pH 7 with a %removal of 85.63% was obtained. Another study by Abdul-Halim et al. [27] revealed that studied the separation of dyes using the PIM system with Aliquat 336 carriers, the optimal pH of the donor solution was obtained in the pH range of 6-7. If the pH is too low (too acidic), the carrier will undergo excessive protonation, reducing its ionic binding ability to MG^+. As a result, transport efficiency decreases due to the formation of weak interactions between MG and carriers. In contrast, at pH too high (alkaline), the MG structure becomes unstable or degrades, thus degrading its ability to bind to carriers within the membrane [14]. In addition, alkaline conditions can also cause depronation or conformational changes in some carriers. Therefore, neutral pH is the optimal condition because it is able to maintain the stability of the MG structure while maintaining the effectiveness of the performance of carriers in PIM [10][28].

Table 3. Total weight of membrane components and physical characteristics of PIM membranes before transport (mean ± SD, n = 3).

Parameter	T_{27}	T_{54}	T_{108}
Total Component Weight (g)	0.2700	0.5400	1.0800
Membrane Weight (g)	0.2623 ± 0.0003	0.5293 ± 0.0002	0.9832 ± 0.0001
%RSD	0.0959	0.0393	0.0117
Membrane Thickness (mm)	0.1460 ± 0.0001	0.3120 ± 0.0001	0.6840 ± 0.0001
%RSD	0.0395	0.0321	0.0084

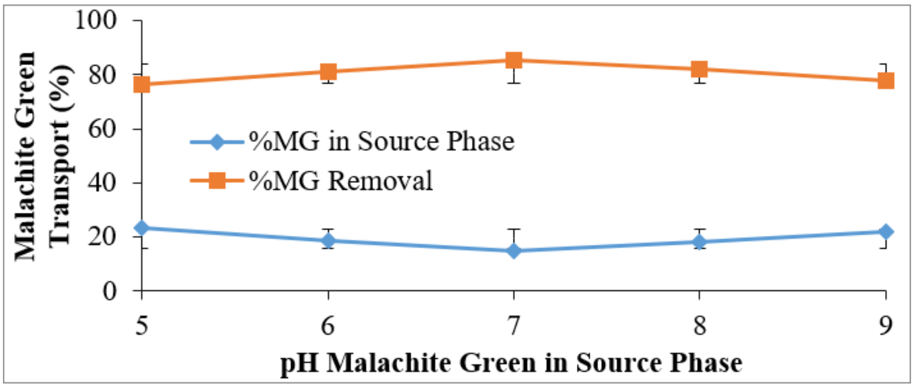

Figure 3. Effect of pH in the source phase on transported MG concentrations.

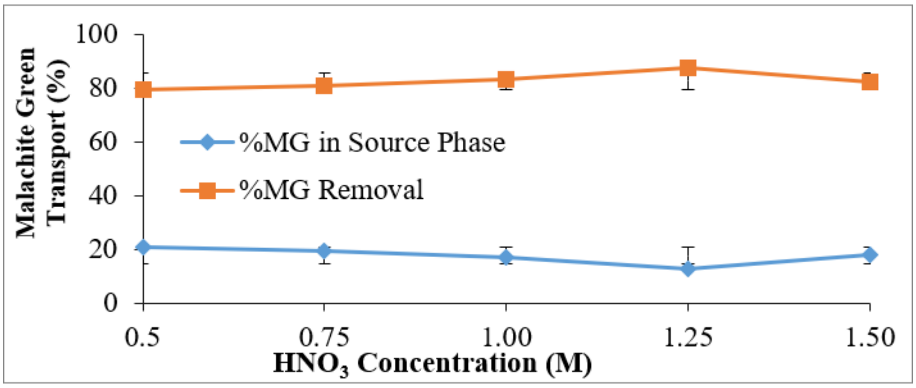

Figure 4. Effect of HNO_3 concentration in the recipient phase on the concentration of transported MG.

3.2.2. Effect of HNO_3 Concentration

The concentration of HNO_3 in the receiver phase can also affect the MG transport process. This study was conducted at the optimum MG pH, namely pH 7 for 24 h with the concentration of MG used in the source phase of 25 ppm. The results of the study on the effect of HNO_3 concentration in the receiving phase on MG transport can be seen in Figure 4. MG transport can occur due to an interaction between MG and carriers that can push MG to the receiving phase [10]. The concentration of HNO_3 of 0.50, 0.75, 1.00, 1.25, and 1.50 M results with %removal of 79.23%, 80.69%, 83.08%, 87.32%, and 82.02%, resepectively. The optimal condition was obtained from the concentration of HNO_3 1.25 M. Judging from the %removal, there was a very significant increase and decrease. This behavior can be attributed to the dissociation of HNO_3 into hydronium ions (H_3O^+) and nitrate ions (NO_3^-) in the receiving phase. The presence of H_3O^+ creates a strongly acidic environment that facilitates the release of MG from the membrane carrier into the receiving phase, thereby maintaining the driving force for transport. Meanwhile, NO_3^- ions help maintain electroneutrality and may stabilize the cationic MG species in the receiving phase. It can be caused because the active sites of the membrane undergoes leaching (components that are lost to the receiving phase) as a result of which the membrane has a reduced active sites so that it cannot transport the target compound to the receiving phase perfectly [29].

3.2.3. Effect of PIM Membrane Thickness

The thickness of the PIM membrane used can affect the process of MG transport. The thickness of a membrane can be influenced by several factors, one of which is that based on the variation in the total weight of the components that make up the membrane will cause differences in the thickness of the membrane produced [14]. Variations in membrane thickness are carried out in the hope that the thicker the membrane used, the total number of its constituent components namely carriers (Poly-BADGE 2:1), DBE as a plasticizer, and polyvinyl chloride will also increase. An increase in the number of these components is assumed to increase the number of active sites of the –OH group of carriers in the membrane structure. With the increase in the number of active sites, the chance of interaction between carriers and MG will be greater, so this indicates the efficiency of the transport process and the percentage of MG that successfully moves to the receiving phase. The results of the study on the effect of PIM membrane thickness on the MG transport process can be seen in Figure 5.

The results showed that the graph results of the membrane thickness with 24-hour transport experienced a very significant increase and decrease in %removal. PIM T_{27} membrane (thin) produced 80.47% %removal, T_{54} membrane (normal) 88.52% and T_{108} membrane (thick) 81.18%. Optimum conditions are obtained on the T_{54} membrane (normal). This is because the number of MG molecules interacting with the carrier is very maximum, and the resulting membrane is neither too thick nor thin. Membrane thickness T_{27} (thin), this membrane has a very thin thickness compared to T_{54} and T_{108} membranes because the number of carriers possessed is small, so it is possible that competition between MG to interact with carriers is less than optimal. Meanwhile, in the T_{108} membrane (thick) the levels of MG transported in the recipient phase were also less. This is because the resulting membrane is thicker and stiffer because it has a larger total of components, so that the process of transporting MG through the membrane becomes difficult or obstructed and results in less optimal MG transportation. Based on this study, it can be concluded that thinner membrane components will be difficult to transport MG because they have fewer total membrane components so that the resulting %removal is less effective.

Plasticizers are an important component in the manufacture of PIM, which serves to improve the flexibility, elasticity, and homogeneity of membranes. With plasticizers, the membrane structure becomes softer and more flexible, making it easier for solutes such as dyes to diffuse across the membrane. If the amount of plasticizer added exceeds the optimal composition, the membrane structure can become too soft or even oily, causing the pores of the membrane to become clogged and resulting in a decrease in mechanical stability and selectivity of the membrane. This condition can cause carriers to dissolve (exit) from the membrane matrix, thereby reducing separation efficiency due to inhibition of MG to diffuse and accelerate

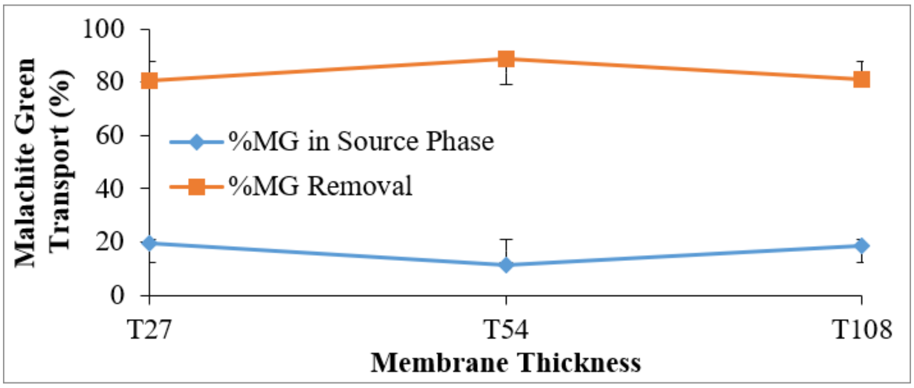

Figure 5. Effect of membrane thickness on transported MG concentrations.

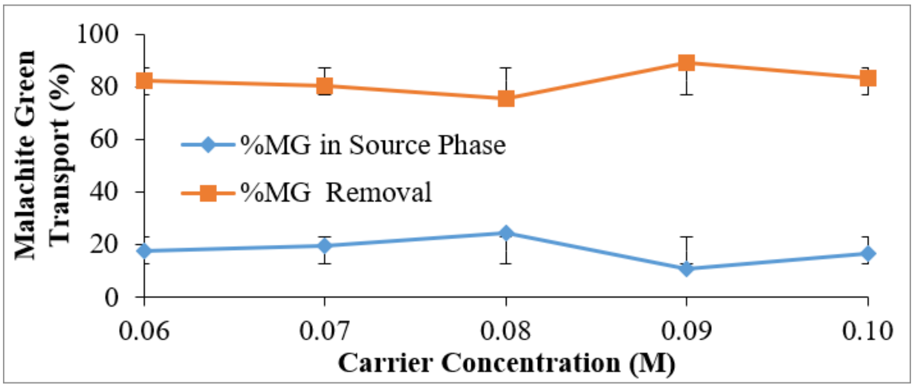

Figure 6. Effect of carrier concentration on transported MG concentration.

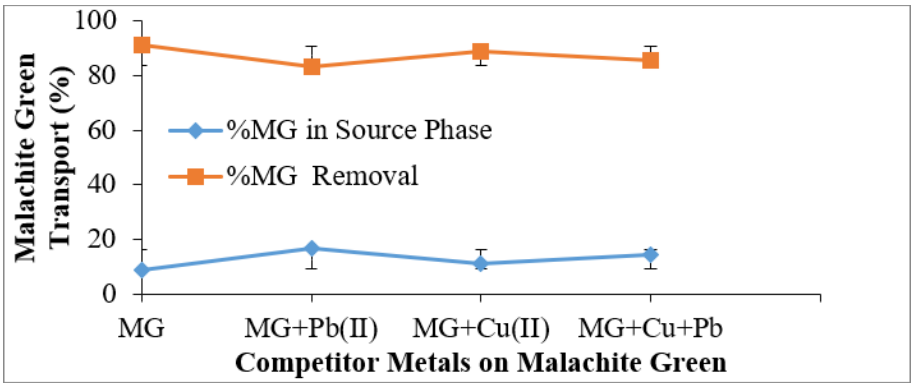

Figure 7. MG concentration in artificial waste with Cu(II) and Pb(II) metals as competitors.

membrane degradation. A small amount of plasticizer can lower the viscosity of the membrane. This means that a small amount of plasticizer will have a low viscosity and conversely a large amount of plasticizer will increase the viscosity.

3.2.4. Effect of Carrier Concentration

The concentration of carriers in this study plays an important role and is a determining factor in the transport process of MG (as a target compound). The higher the concentration of carriers in the transport process, this results in the amount of MG transported the larger. The results of the study on the effect of the concentration of carriers on the MG transport process can be seen in Figure 6. It presents the effect of carrier concentration on the transport efficiency of MG through the PIM membrane. The membranes containing 0.06 M, 0.07 M, 0.08 M, 0.09 M, and 0.10 M Poly-BADGE 2:1 carriers produced %removal values of 82.34%, 80.16%, 75.71%, 89.31%, and 83.24%, respectively. Based on these results, the optimal MG transport was observed at a carrier concentration of 0.09 M.

In general, increasing the carrier concentration increases the availability of active –OH groups in the membrane, which enhances the interaction between the carrier and MG molecules through hydrogen bonding and π–π interactions. However, the transport efficiency does not increase linearly with carrier concentration because the distribution and mobility of carrier molecules within the membrane matrix also influence the transport process. At carrier concentrations of 0.07–0.08 M, a slight decrease in %removal was observed. This decrease may be related to the non-uniform distribution of carrier molecules within the polymer matrix at intermediate loading levels. Under these conditions, partial clustering of carrier molecules may occur, which can reduce the accessibility of active –OH sites and increase the local diffusion resistance for MG molecules moving through the membrane phase. As a result, the transport efficiency temporarily decreases.

When the carrier concentration reaches 0.09 M, the distribution of carrier molecules within the membrane matrix becomes more effective, allowing better interaction between MG molecules and the carrier. At this concentration, a balance between the availability of active sites and the molecular mobility within the membrane phase is achieved, resulting in the highest transport efficiency. However, further increasing the carrier concentration to 0.10 M leads to a decrease in % removal. This behavior may be attributed to excessive carrier loading, which can increase the microviscosity of the membrane matrix and promote aggregation or overlapping of active sites. Consequently, the diffusion of MG through the membrane becomes more restricted, leading to a reduction in the overall transport efficiency.

3.2.5. The Effect of Competitor Metals on MG Transport

The study of MG transport competition on artificial waste was carried out with the aim of determining the influence of metals on the concentration of MG transported to the recipient phase and to determine the selectivity value of the PIM membrane in transporting MG. The study of the competition of MG transportation on artificial waste is one of the applications in efforts to reduce dye waste in the aquatic environment, especially

MG.

These Pb(II) and Cu(II) metals will be assumed to be metals found in liquid waste. The results of the study on the effect of the presence of competing metals Pb(II) and Cu(II) with UV-Vis spectrophotometers on MG transport, can be seen in Figure 7.

Figure 7 shows a decrease in the concentration of MG (%removal) that is transported to the recipient phase due to the presence of metal added in the source phase. The concentration of MG transported in the membrane phase to the receiving phase (% removal) containing the MG control solution was greater at 91.32% compared to MG added with metal. Based on the results obtained, the presence of competing metals, namely Cu(II) and Pb(II) metals, can affect the MG transport process so that the number of MG transported becomes less effective. A complexation reaction may occur between Pb(II), Cu(II), and the carrier on the membrane when interacting with the source phase that has a low pH, meaning that the presence of competing metals results in a tendency to form complexes with carriers. The presence of metal competitors that form complexes with carriers precludes the interaction of the active group –OH in the carriers present in the membrane.

The decrease in MG transport efficiency in the presence of Pb(II) and Cu(II) ions can be attributed to a competitive interference mechanism at the carrier active sites. Poly-BADGE 2:1 contains hydroxyl (–OH) and ether functional groups which serve as coordination sites for cationic species. In the absence of metal ions, these active sites preferentially interact with MG^+ through hydrogen bonding and electrostatic interactions, facilitating its transport across the membrane.

However, Pb^{2+} and Cu^{2+} ions exhibit strong affinity toward oxygen-containing functional groups, leading to the formation of metal–carrier complexes within the membrane phase. This coordination interaction reduces the availability of active –OH sites for MG complexation, resulting in a competitive occupation of the carrier. Consequently, the formation of MG–carrier complexes decreases, leading to a lower MG transport efficiency.

This phenomenon confirms that the presence of divalent metal ions induces competitive interference by blocking active carrier sites and limiting the selectivity of the PIM system toward MG transport.

3.2.6. PIM Membrane Characterization

The FTIR and SEM analyses were performed to evaluate the physicochemical characteristics of the PIM membrane before and after MG transport under the optimal experimental conditions. The FTIR spectroscopy was used to identify the

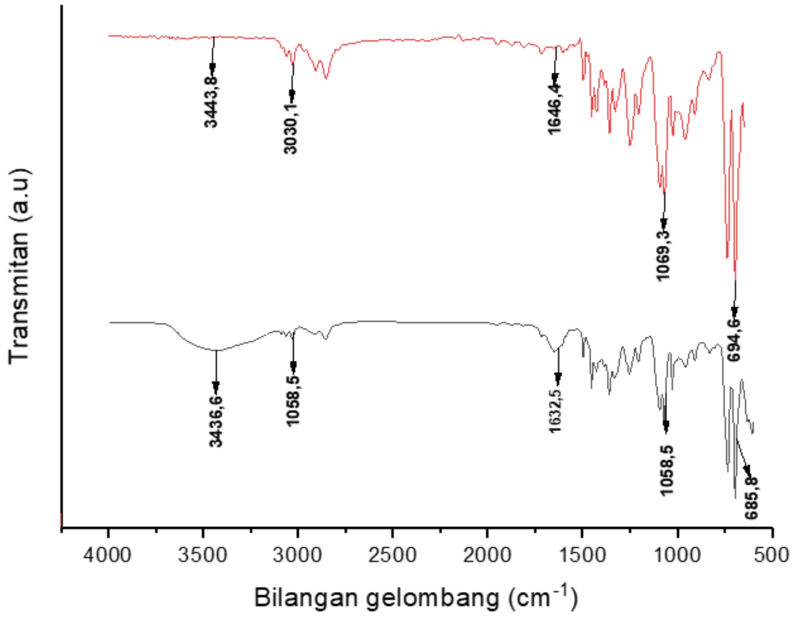

Figure 8. Comparison of FTIR spectra of PIM membrane (a) before transport and (b) after 24 h MG transport.

Table 4. Characteristic FTIR absorption bands of Poly-BADGE 2:1 PIM membrane before and after MG transport.

	Number of Waves(cm^{-1})	
Gugus Fungsi	**Membrane Before Transport**	**Membrane After Transport**
Alcohol (hydroxyl) (-OH)	3443.8	3436.6
Stretch Csp^3-H ($-CH_3$)	3030.1	3026.5
Aromatic ring (C=C)	1646.4	1632.5
Stretch (C-O)	1069.3	1058.5
Aromatic ring (C-H)	694.6	685.8

functional groups present in the Poly-BADGE 2:1-based PIM membrane and to determine possible structural changes occurring during the transport process. The FTIR spectra of the membrane before and after MG transport are presented in Figure 8.

The FTIR spectra show several characteristic absorption bands corresponding to the functional groups present in the membrane matrix. These include the hydroxyl group (–OH), aliphatic Csp^3–H stretching ($–CH_3$), aromatic C=C stretching, C–O stretching, and aromatic C–H vibrations. The presence of these peaks confirms that the main functional groups of the Poly-BADGE-based PIM membrane remain present both before and after the transport process. A detailed comparison of the wavenumbers of the PIM membrane before and after MG transport is presented in Table 4.

The FTIR results indicate slight shifts in several characteristic absorption bands after the MG transport process. The O–H stretching vibration associated with the hydroxyl groups of the Poly-BADGE carrier appears at 3443.8 cm^{-1} before transport and shifts to 3436.6 cm^{-1} after transport. A shift toward a lower wavenumber is commonly attributed to hydrogen bonding interactions, as hydrogen bond formation weakens the O–H bond and decreases the vibrational frequency observed in the FTIR spectrum [30].

This observation suggests the formation of hydrogen bonding interactions between the –OH groups of the Poly-BADGE carrier and the nitrogen -containing functional groups of MG during the transport process. Minor shifts were also observed in other absorption bands, including the Csp^3–H stretching vibration and the aromatic C=C stretching vibration, indicating slight changes in the chemical environment of the membrane matrix after MG transport. Overall, the observed spectral shifts suggest that MG transport through the Poly-BADGE-based PIM membrane involves intermolecular interactions between the carrier and MG molecules. The absence of new absorption bands further indicates that no significant chemical degradation or structural modification of the membrane occurred during the transport experiment.

The loss of active sites on the membrane after transport can be seen from the results of PIM characterization using SEM. PIM membrane characterization using SEM is carried out to obtain information about the physical properties of the material, especially the morphology of the surface of the material being tested. Characterization using SEM serves to look at the morphology of the PIM membrane before and after transport. The PIM membrane used is a membrane containing a Poly-BADGE 2:1 carrier before and after MG transport under optimal conditions. The results of the characterization of the surface morphology of the PIM membrane after transport using SEM can be seen in Figure 9.

Figures 9(a)–9(d) are presented at identical magnifications to enable direct morphological comparison before and after the transport process. Figures 9(a)–9(b) show the surface morphology of the PIM membrane before transport. In the morphology of the membrane before transport with magnification of 500× and 2500×, it can be seen that the membrane surface is still covered by plasticizer as the liquid medium, while in Figures 9 (c)–9(d) reveal an uneven surface and there are pores. This is because the PIM membrane is a porous liquid membrane, the pores that are visible on the surface of the membrane after transport are formed due to the presence of membrane components that are lost during the transport process (leaching). The use of membranes for transport allows the components that make up the membrane leaching. This leaching membrane component results in the surface of the PIM membrane becoming porous. Leaching components can come from carriers, base polymers or plasticizers. This is reinforced by the difference in membrane weight before and after transport through weighing.

SEM images further reveal the formation of surface pores after transport, which is attributed to partial leaching of membrane components such as plasticizer and carrier under acidic receiving phase conditions. However, the membrane does not exhibit cracking or catastrophic structural collapse, indicating that the overall integrity of the membrane matrix is preserved. These observations suggest that MG transport induces moderate physical changes but does not compromise the functional stability of the membrane, supporting its suitability for prolonged or repeated use. Overall, the physicochemical characterization results help explain the separation performance observed in the transport experiments discussed in the previous sections. The FTIR analysis confirms the presence of hydroxyl functional groups in the Poly-BADGE carrier, which can interact with MG molecules through hydrogen bonding and π–π interactions, thereby facilitating the transport process. In addition, SEM observations reveal morphological changes in the membrane after the transport experiment, including the formation of pores due to partial component leaching. These structural features may influence the diffusion pathway of MG through the membrane phase and contribute to the transport efficiency observed under the optimized experimental conditions. These structural and chemical characteristics therefore support the efficient MG transport observed under the optimized experimental conditions.

3.2.7. Performance Comparison with Previously Reported PIM Systems

To evaluate the practical significance of the developed membrane system, the transport performance of the Poly-BADGE 2:1–based PIM

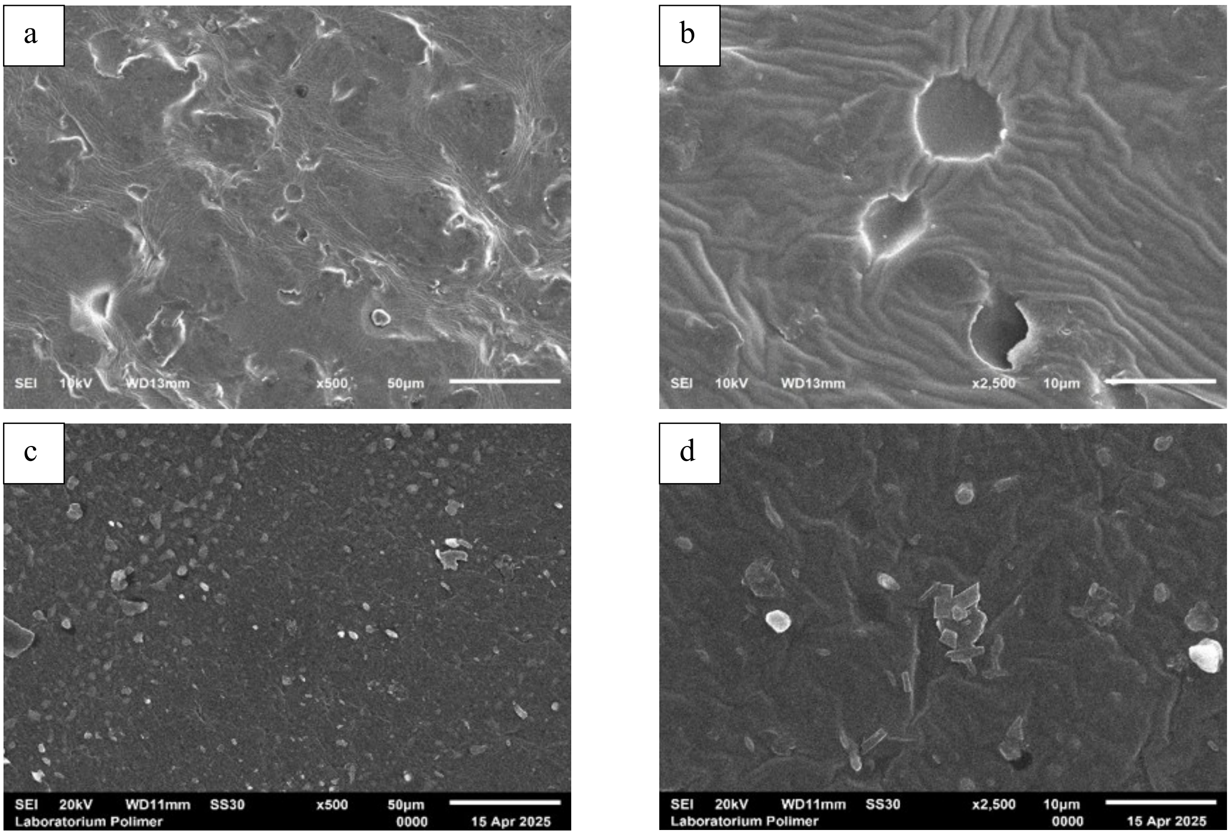

Figure 9. SEM images of Poly-BADGE 2:1–based PIM membrane before transport at (a) 500× and (b) 2500× magnification. Membrane after transport at (c) 500× and (d) 2500× magnification.

Table 5. A comparison of the performance of carriers based PIM

Carrier/ System	Target Dye	% Removal	Optimum Condition	Stability/Reusability	Ref.
Aliquat 336–PIM	MG	~80–85%	pH 6–7	Moderate	[23]
Polyeugenol–PIM	MG	85.63%	pH 7	Limited reuse	[10]
Co-EDVB–PIM	Phenol	~88%	pH 7	Moderate	[13]
Poly-BADGE 2:1–PIM	MG	90.90%	pH 7; 1.25 M HNO_3	Good stability	This study

was compared with previously reported carrier systems for malachite green and related cationic dyes. Such comparison is essential to position the developed membrane within the current state-of-the-art PIM technologies and to clarify its relative advantages and limitations. The comparison presented in Table 5 highlights differences in removal efficiency, operating conditions, and membrane stability among various PIM configurations.

Compared to previously reported PIM systems for MG and similar cationic dyes, the Poly-BADGE 2:1-based PIM developed in this study demonstrates several advantages. The system exhibits higher transport efficiency under mild operating conditions, strong carrier–dye interaction through hydroxyl functional groups, and good membrane integrity after the transport process as confirmed by FTIR and SEM analyses. Unlike conventional quaternary ammonium or polyeugenol-based carriers, Poly-BADGE 2:1 provides multiple active sites for hydrogen bonding and coordination, leading to enhanced selectivity toward MG. These characteristics highlight the potential of Poly-BADGE-based PIM as a promising alternative for dye separation in wastewater treatment. This improvement suggests that the synergistic interaction between membrane thickness, carrier loading, and hydrogen-bonding capability plays a critical role in enhancing MG transport efficiency. Overall, these findings confirm that the engineered Poly-BADGE-based PIM not only performs effectively under optimized laboratory conditions but also shows strong potential for practical application in selective dye separation processes.

4. CONCLUSIONS

This research successfully developed a PIM system based on Poly-BADGE 2:1 as a carrier for the selective transport of MG. The optimization results showed that the optimal conditions were obtained at the source phase pH 7, the HNO_3 concentration of the receiver phase was 1.25 M, the membrane thickness was T54, the concentration of carriers was 0.09 M, and the transport time was 24 hours with a transport efficiency of 90.90%. The membrane showed good stability and selectivity despite the presence of competing metal ions Pb(II) and Cu(II), with the transport efficiency decreasing slightly to 85.67%. FTIR and SEM analysis confirmed the interaction between the active –OH groups in Poly-BADGE and MG as well as changes in membrane morphology after transport due to leaching of some membrane components. Overall, the results of this study confirm that PIM based on Poly-BADGE 2:1 has high potential as an effective, selective, and environmentally friendly separation system for the removal of cationic dyes such as MG from synthetic and actual wastewater. Future work should include cyclic transport experiments to evaluate the long-term stability and reusability of the Poly-BADGE-based PIM system.

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Conflicts of Interest

The authors declare no conflict of interest.

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DECLARATION OF GENERATIVE AI

During the preparation of this manuscript, the authors used only generative AI intelligence and AI -assisted technology in manuscript preparation for grammar correction and language editing. However, all scientific content, data interpretation, and conclusions were developed and verified by all authors. The authors are solely responsible for the content of the manuscript.

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