

RESEARCH ARTICLE

Aging Dynamics of Bio-Coagulants: Implications for Oily Wastewater Treatment Efficiency

Najmuddin Mohd Ramli^{1,2,3}, Puteri Nur Ain Daim¹, Syahirah Syazwani Mohd Tarmizi², Mohd Najib Razali^{3*}

¹ College of Engineering, Universiti Malaysia Pahang Al-Sultan Abdullah, Lebu Persiaran Tun Khalil Yaakob, 26300 Gambang, Pahang, Malaysia.

² MNR Multitech Sdn Bhd, K02 Ground Floor, Kompleks UMP Holdings, Lebu Persiaran Tun Khalil Yaakob, 26300 Gambang, Pahang, Malaysia.

³ Faculty of Chemical and Process Engineering Technology, Universiti Malaysia Pahang Al-Sultan Abdullah, Lebu Persiaran Tun Khalil Yaakob, 26300 Gambang, Pahang, Malaysia.

ABSTRACT - Inadequate treatment of oily wastewater discharge can contribute to an increase in the levels of both biological oxygen demand (BOD) and chemical oxygen demand (COD) in the water. Various studies have highlighted the effectiveness of natural bio-coagulants in particular chitosan as an alternative to the chemical coagulants available in the market. However, minute studies have been reported in discussing the correlation between the aging effect of the bio-coagulant and its performance. Thus, this paper presents research on the aging effect of bio-coagulants for treating oily wastewater. In this study, G-Treat was used as the bio-coagulant, which was produced from chitosan powder, acetic acid, and deionized water. The experimental procedure started by filling 500 mL of oily wastewater into a beaker. Next, 10 wt. % of G-Treat was added into the beaker for the jar test, which was mixed for 30 min at 150 rpm, followed by 2 hours settling time. Then, the mixture was separated using filter paper. The analysis of oily wastewater characteristics was conducted using five types of analyses, which are pH, chemical oxygen demand (COD), biological oxygen demand (BOD), total suspended solids (TSS), and oil and grease (O&G) under the parameters of days (0–7 days), weeks (0–6 weeks), and months (0–3 months). From the result, the best performance of G-Treat was achieved at the optimum parameters during week 2, with 33.16%, 65.75%, 35.73%, 28.58%, and 0% removal of pH, COD, BOD, TSS, and O&G, respectively. In conclusion, the studied bio-coagulant demonstrated higher removal of pH, COD, BOD, TSS, and O&G at optimum parameters, and eventually, the level of effectiveness will drop.

ARTICLE HISTORY

Received : 3rd Mar. 2023
Revised : 14th Dec. 2023
Accepted : 14th Dec. 2023
Published : 30th Dec. 2023

KEYWORDS

Oily wastewater
Bio-coagulant
Chitosan
G-Treat
Wastewater treatment

1.0 INTRODUCTION

Oily wastewater is one of the main pollutants emitted into the water by industries and domestic sewage [1]. Emulsions are widely used as emulsified coolants in metal processing, and also as lubricating oil in power plants, coolants, or lubricants. Usually, this type of oily wastewater will be drained or treated until it has lost its efficiency. Under the waste category of Malaysia Environmental Quality (Scheduled Waste) Regulation 2005, this waste can be categorized as oil and hydrocarbon. The scheduled waste code for this type of oily wastewater is SW314, which can be described as oil or sludge from an oil refinery plant maintenance operation [2]. Oil and hydrocarbon type of scheduled waste is in the top four waste categories produced yearly [3]. Therefore, it is important to treat the waste to avoid it from getting to the top of the list of inappropriate disposals.

Throughout the year, advancements in wastewater treatment technologies have been developed. One of them is using coagulation-flocculation to treat wastewater. Chemical treatment is typically conducted through coagulation-flocculation [4]–[6]. Most of the chemicals used as coagulants in these processes can cause health problems. The toxicity of the treated oily wastewater increases due to the reaction between colloidal particles in the oily wastewater and the chemical coagulant [7], [8]. Therefore, this study examines the aging effect of bio-coagulants for treating oily wastewater. Oily wastewater has been proven to be dangerous for the environment. Adsorption can be considered the most advanced and efficient method for residual treatment by selectively removing organic compounds. It is practical to create a coagulant or technology that is both efficient and environmentally beneficial for treating oily wastewater before it is discharged [9]. Oily wastewater contains a significant amount of emulsified oil, which is very difficult to separate from aqueous media.

In Malaysia, most of the industrial waste is transferred to Kualiti Alam Sdn. Bhd. for treatment. Being a developing country is no exception to the global trend of sewage sludge volume that increases annually. Industrial sludge disposal

*CORRESPONDING AUTHOR | Mohd Najib Razali | ✉ najibrazali@umpsa.edu.my

will necessitate additional post-processing. According to statistics, Malaysia produced roughly 3 Mt of sewage sludge each year, with the amount increasing each year [10]. Hence, almost RM 7 billion is spent every year to treat wastewater [11]. As a company spends a lot of money on wastewater treatment, it is likely to have a considerably less expensive treatment facility with high-performance treatment. Coagulation-flocculation offers various advantages for treating industrial wastewater. However, it has been proven in various studies that chemicals (e.g., aluminium salts, acrylamides) that are used in the process can cause health problems. During the treatment procedure, artificial polymers and other unwanted substances could react with other components [12]. Conventional chemicals used for coagulation include aluminium or iron-based salts. The most widely used are metal-based coagulants because of their low cost, wide availability, and high removal efficiencies. However, metal-based water purification coagulants can have drawbacks. They can change the pH of the water because they deplete alkalinity, resulting in the addition of lime or soda ash to maintain the pH levels. They also cause the production of large quantities of sludge and the presence of residue metals in the sludge [13]–[16]. Research on a biological approach that acts as a coagulant has been proven to be efficient in treating oily wastewater. The performance of a coagulant is influenced by the characteristics of oily wastewater, which is dependent on the method utilized, as each approach has its advantages and disadvantages [17]. Therefore, this research was conducted to determine the optimum efficiency of bio-coagulants to treat oily wastewater after storage.

The objective of this research is to study the aging effect of bio-coagulants for treating wastewater treatment. In meeting this objective, several scopes of work have been identified, which are characterizing and analysing wastewater samples before and after treatment in reducing pH, chemical oxygen demand (COD), biological oxygen demand (BOD), total suspended solids (TSS), and oil and grease (O&G); characterizing the aging effect of bio-coagulant in terms of pH, viscosity, Fourier transform infrared (FTIR) absorbance, and ultraviolet-visible (UV-Vis) absorbance; and comparing the storage time efficiency and aging factor of bio-coagulant in reducing pH, COD, BOD, TSS, and O&G.

In this research, G-Treat was used as a bio-coagulant to treat oily wastewater. G-Treat was produced using three materials: chitosan, acetic acid, and distilled water. Chitosan is derived from the exoskeletons of a marine invertebrate (i.e., prawn). It is a white amorphous solid suggested as a coagulant due to its biodegradable properties. As this polymer is only soluble in dilute acidic solutions, many of its applications are limited [18]–[20]. Chitosan are hydrophilic chemicals that are not harmful to humans and are easily obtainable. Numerous polymers that are found in nature naturally have cationic qualities that may be altered to create cationic polyelectrolytes that are flocculants in solid-liquid separations [21], [22]. Chitosan is widely regarded as safe for health since it is non-toxic like most natural polymers. Chitosan is a cost-effective, highly biodegradable method of treating water at the point of use. They also are unlikely to create treated water with an excessive pH (beyond the WHO recommended range of 6.5 to 8.5) [23], [24].

The aging of chitosan plays a critical role in determining the effect of oily wastewater treatment. Chemical aging is an important and complex process, which involves the loss of volatiles and oxidation. When chitosan is subjected to high temperatures, such as during manufacturing, transportation, and laying of the mixture, the oxidation and volatilization processes, which are slow at ambient temperature, are accelerated. These processes are irreversible. Following the compaction of the mixture, oxygen diffuses rapidly through linked air gaps, which is known as the oxidation process [25].

2.0 METHODS AND MATERIAL

2.1 Experimental Materials

Approximately 30 L of oily wastewater was collected from the waste of a cafeteria at Universiti Malaysia Pahang Al-Sultan Abdullah (UMPSA). The sample was collected from the cafeteria, as oily wastewater is released into the oil trap. The oily wastewater emits a strong odour and dark colour. So, this sample has been taken as an effort to treat the oily wastewater by using G-Treat. The samples were kept in a jerry can, sealed tightly, and labeled before being transferred to the laboratory. The samples collected were refrigerated at around 4 °C to avoid the occurrence of bioactivities. Analysis was conducted within 24 hours of the collection of the sample. About 9.5 L of G-Treat was produced in the laboratory, and analysis was performed within 24 hours of the production of the bio-coagulant.

2.2 Experimental Procedures

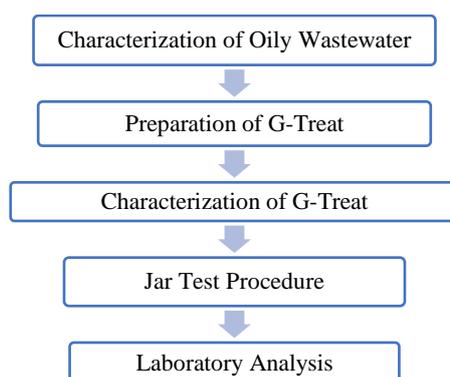


Figure 1. Flow Chart of the Experimental Procedure

2.3 Characterisation of Oily Wastewater

The characterisation of oily wastewater was carried out in the MNR Lab, UMP Holdings and Central Lab, UMP. pH, FTIR spectroscopy, viscosity, and UV-Vis spectroscopy were carried out to characterize the oily wastewater. The methods are based on the Examination Manual of Water and Wastewater [30]. Table 1 summarizes the analysis methods.

Table 1. Analysis methods for oil

Analysis	Test Method	Equipment
pH	ASTM D1293-18 [26]	Seven Easy pH, Mettler-Toledo
FTIR	ASTM D7889-13 [27]	Nicolet Summit FTIR Spectrometer
Viscosity	ASTM D445-21 [28]	HK-265 Kinematic Viscosity Apparatus
UV-Vis	ASTM D2800-12 [29]	Hach Model DR/2400 and HACH Reactor

The pH meter and electrodes are calibrated using at least two buffer solutions that have pH values close to the expected sample pH. The sample is measured under strictly controlled procedures and techniques [26].

The FTIR has established the characteristic infrared spectroscopic signals associated with key in-service monitoring characteristics. This technique provides property values based on examining each characteristic's infrared spectroscopic signature. The essence of this method is to capture the fundamental chemical patterns linked to each characteristic for in-service fluid analysis using a self-contained field device and coupled wipe-clean transmission cell [27].

The viscosity test measures the time taken for a set volume of liquid to flow by gravity through the capillary of a calibrated viscometer under a reproducible driving head and at a closely regulated known temperature. The kinematic viscosity (measured value) is the product of the measured flow time and the viscometer's calibration constant. Two such measurements are required to calculate a kinematic viscosity result that is the average of two acceptable measured values [28].

The UV-VIS analytical method is based on rapidly saponifying the oil with methanolic sodium hydroxide followed by boiling the soaps with BF₃-methanol in the same vessel to quantitatively convert the fatty acids to methyl esters. The methyl esters are separated from the mixture by adding a saturated salt solution [29].

2.4 Preparation of G-Treat

Chitosan powder was used as a starting material in preparing G-Treat. Chitosan powder was activated by diluting acetic acid to produce a gel-like substance. A ratio of 7:2:91 for chitosan: acetic acid: deionized water was used to produce a certain chitosan concentration. 35 mL of acetic acid was added into 455 mL of deionized water. Then, 10 g of chitosan powder was added slowly into the stirring solution and stirred for 15 minutes. Finally, 500 mL of prepared G-Treat was stored in a glass jar for further use. This ratio is based on the data from previous research [18]–[20].

2.5 Characterisation of G-Treat

The characterisation of G-Treat was carried out in the MNR Lab, UMP Holdings and Central Lab, UMP. pH, FTIR spectroscopy, viscosity, and UV-Vis spectroscopy were carried out to characterize G-Treat. The methods are based on the Examination Manual of Water and Wastewater [30].

Table 2. Analysis methods for G-Treat characterisation

Analysis	Test Method	Equipment
pH	ASTM D1293-18	Seven Easy pH, Mettler-Toledo
FTIR	ASTM D7889-13	Nicolet Summit FTIR Spectrophotometer
Viscosity	ASTM D445-21	HK-265 Kinematic Viscosity Apparatus
UV-Vis	ASTM D2800-12	Hach Model DR/2400 and HACH Reactor

2.6 Jar Test Procedure

A jar test apparatus was used in this experiment to treat oily wastewater. A batch test was conducted with a series of three beakers, each having a stirring device. Each jar contains 500 mL of oily wastewater and 10 wt. % of G-Treat. The experiment was conducted at 150 rpm for 30 minutes and then settled for 2 hours. The experiment was repeated 18 times based on the parameters (0–7 days, 0–6 weeks, and 0–3 months) [31].

2.7 Laboratory Analysis

The analysis of the treated sample was carried out in the MNR Lab, UMP Holdings. The analysis of pH, COD, BOD, TSS, and O&G was conducted. These methods are based on the Examination Manual of Water and Wastewater [30]. Table 3 summarizes the analysis methods.

Table 3. Analysis methods for laboratory analysis

Analysis	Test Method	Equipment
pH	ASTM D1293-18	Seven Easy pH, Mettler-Toledo
COD	ASTM D1252-06 (2012)	Hach Model DR/2400 and HACH Reactor
BOD	ASTM D1252-60	Yellow Springs Model 5010 and BOD Incubator
TSS	ASTM D5907-18	Laboratory apparatus
O&G	ASTM D3921	Laboratory apparatus

3.0 RESULTS AND DISCUSSION

3.1 Characterisation of Wastewater Samples Before and After Treatment

Characterization and analysis of wastewater samples before and after treatment were done to study the effectiveness of the bio-coagulant in reducing pH, COD, BOD, TSS, and O&G. Each analysis was conducted for three different parameters: days, weeks, and months. The characterization of oily wastewater samples was conducted first before a further investigation was done to understand the nature and behaviour of oily wastewater samples. The sample was compared with Standard A and Standard B of the water quality standard limits set by the Department of Environmental Malaysia (DOE) [32]. The comparison will provide a guideline for choosing the most optimum efficiency to treat oily wastewater after being stored.

Table 4. Characterisation of wastewater samples

Analysis	Unit	Water Quality Standards		Oily Wastewater Sample
		Standard A	Standard B	
pH Value	-	6.0-9.9	5.5-9.0	4.59
COD	mg/L	50	100	5,105.5
BOD5	mg/L	20	40	196.5
TSS	mg/L	50	100	466.7
O&G	mg/L	1.0	10	10

Table 4 lists the values for the analyzed oily wastewater samples before the treatment. Based on Table 4, Standard A under the water quality standard is more rigid compared to Standard B. This is because Standard A is applicable as a regulation at water catchment areas, which includes the area upstream of the surface or above the sub-surface of waterways, mainly directed to the community and for human use.

Figure 2 shows the pH values before and after treatment of wastewater samples under three parameters (days, weeks, and months). The pH values before treatment are higher than after treatment with day 0 (D-0), week 2 (W-2), and month 0 (M-0) recorded the lowest values out of all days, weeks, and months, respectively. The pH of the sample became more acidic due to the presence of acetic acid as one of the components in producing G-Treat. pH is the concentration of hydrogen ions in a solution [33]. From the result, the lower the pH, the lower the hydrogen ion concentration. The initial pH value was higher than after treatment. When chitosan is introduced into a strong acidic environment, it can produce de-emulsification, which causes the droplet size to grow. Thus, the adsorption of oil may be effective [34], [35].

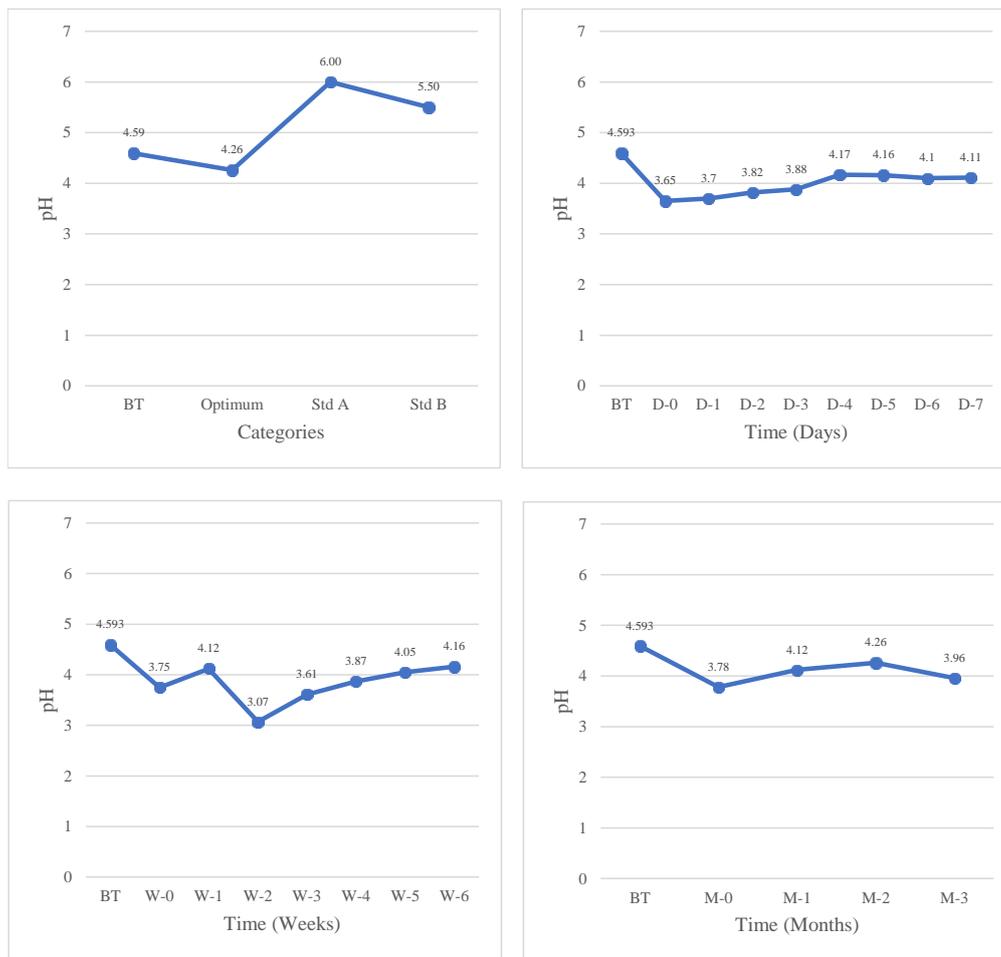


Figure 2. pH Values Before and After Treatment. BT: Before Treatment, D: Day, W: Week, M: Month.

Figure 3 presents the COD values before and after treatment of wastewater samples under three parameters (days, weeks, and months). The COD values before treatment are higher than after treatment with day 7 (D-7), week 1 (W-1), and month 2 (M-2) recorded the lowest values out of all days, weeks, and months, respectively. The COD test is used to determine the quantity of oxygen needed to oxidize organic matter in wastewater samples by chemical oxidation [36]. Based on Figure 3, it can be concluded that the higher the COD value, the higher the oxygen stripping capacity of the waste when it is discharged into receiving water. The COD value decreased significantly from day 1 until day 7 and became inconsistent throughout the weeks until month 3. The dosage of the coagulant may affect the COD value within the specified time. The high percentage of COD removals in the oily wastewater was due to the high charge density of chitosan and this property helped to occur rapid destabilization of the particles in less amount of chitosan [37].

Figure 4 shows the BOD5 values before and after treatment of wastewater samples under three parameters (days, weeks, and months). The BOD5 values before treatment are lower than after treatment with day 3 (D-3) and month 0 (M-0) recorded the highest values out of all days and months, respectively. Furthermore, the BOD5 value before treatment is lower than after treatment, except for week 2 (W-2). The BOD test is done to measure the amount of dissolved oxygen required by aerobic microorganisms to determine organic matter present in a sample of water at a certain temperature [38]. The trend shown above is slightly different from the data from a previous study that highlighted the usage of chitosan can help to reduce the BOD5 by up to 92% [39]. Based on this research it can be concluded that the dosage of chitosan in the G-Treat must be further enhanced to improve the performance in terms of BOD reduction. Even though the BOD values in all ranges of time are higher than Standard A and Standard B but are still within the optimum range.

Figure 5 presents the TSS values before and after the treatment of wastewater samples under three parameters (days, weeks, and months). The TSS values before treatment are higher than after treatment with day 0 (D-0), day 5 (D-5), day 6 (D-6), week 0 (W-0), week 6 (W-6), and month 2 (M-2) recorded the lowest values for the respective parameter. The TSS test measures the total concentration of suspended solids in waste [40]. Based on Figure 5, it can be concluded that a lower TSS value indicates lower solid generation due to a decrease in BOD loading. Figure 6 shows the O&G values before and after treatment of wastewater samples under three parameters (days, weeks, and months). The difference in O&G values is smaller before and after treatment. Thus, it is considered insignificant.

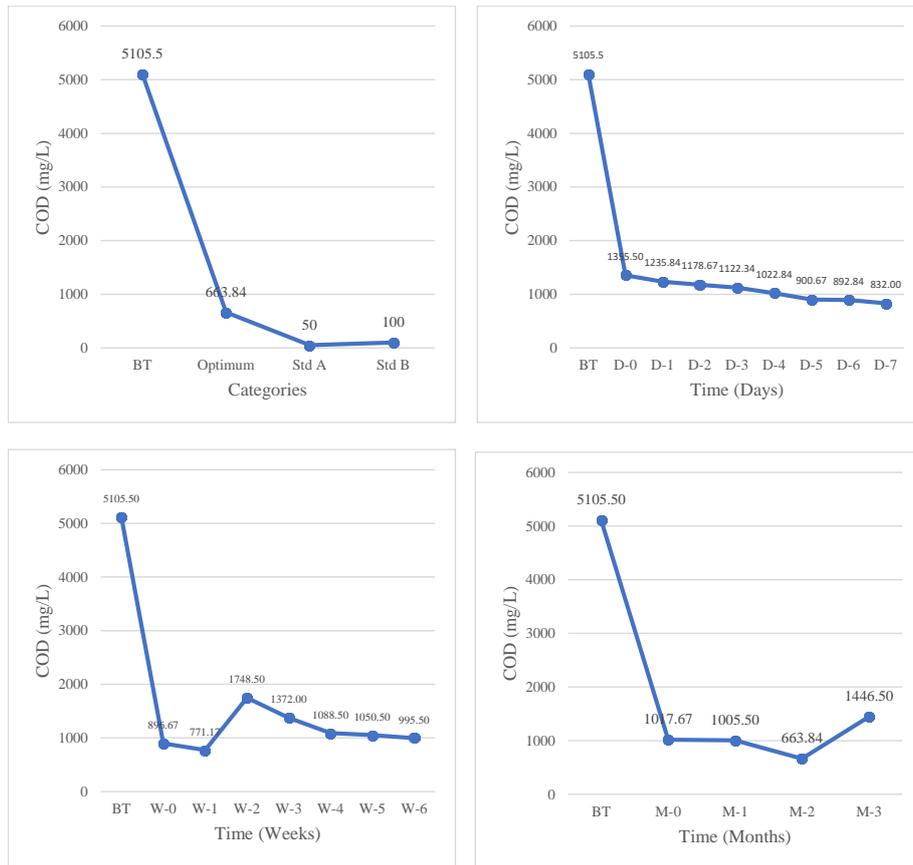


Figure 3. COD Values Before and After Treatment. BT: Before Treatment, D: Day, W: Week, M: Month

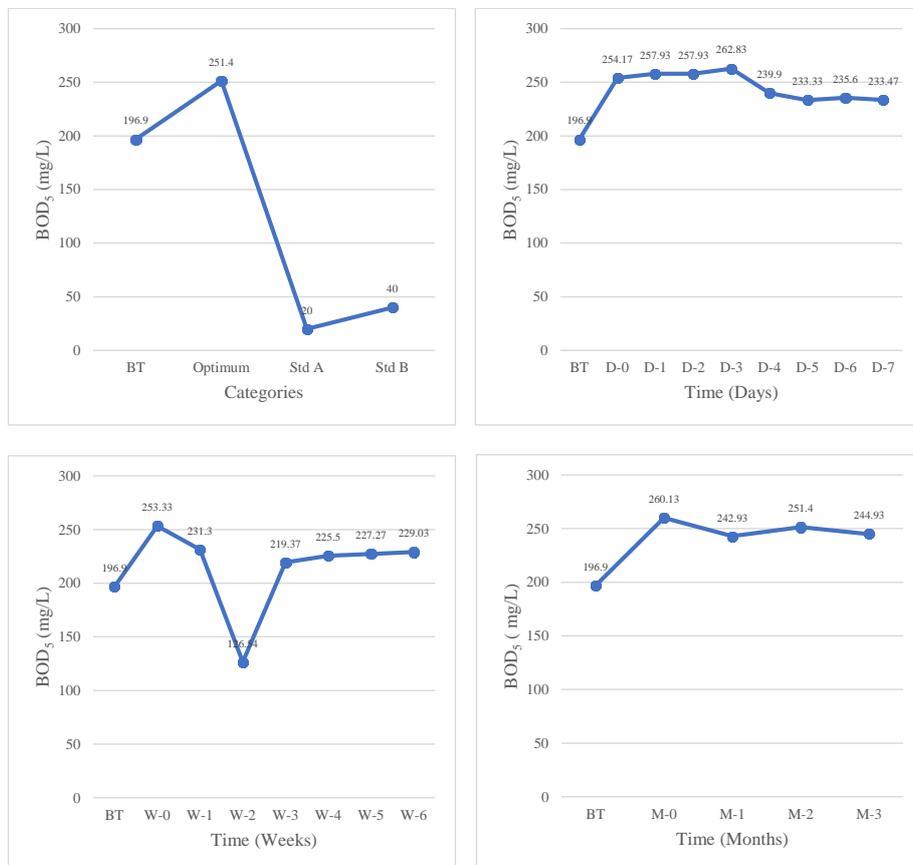


Figure 4. BOD₅ Values Before and After Treatment. BT: Before Treatment, D: Day, W: Week, M: Month

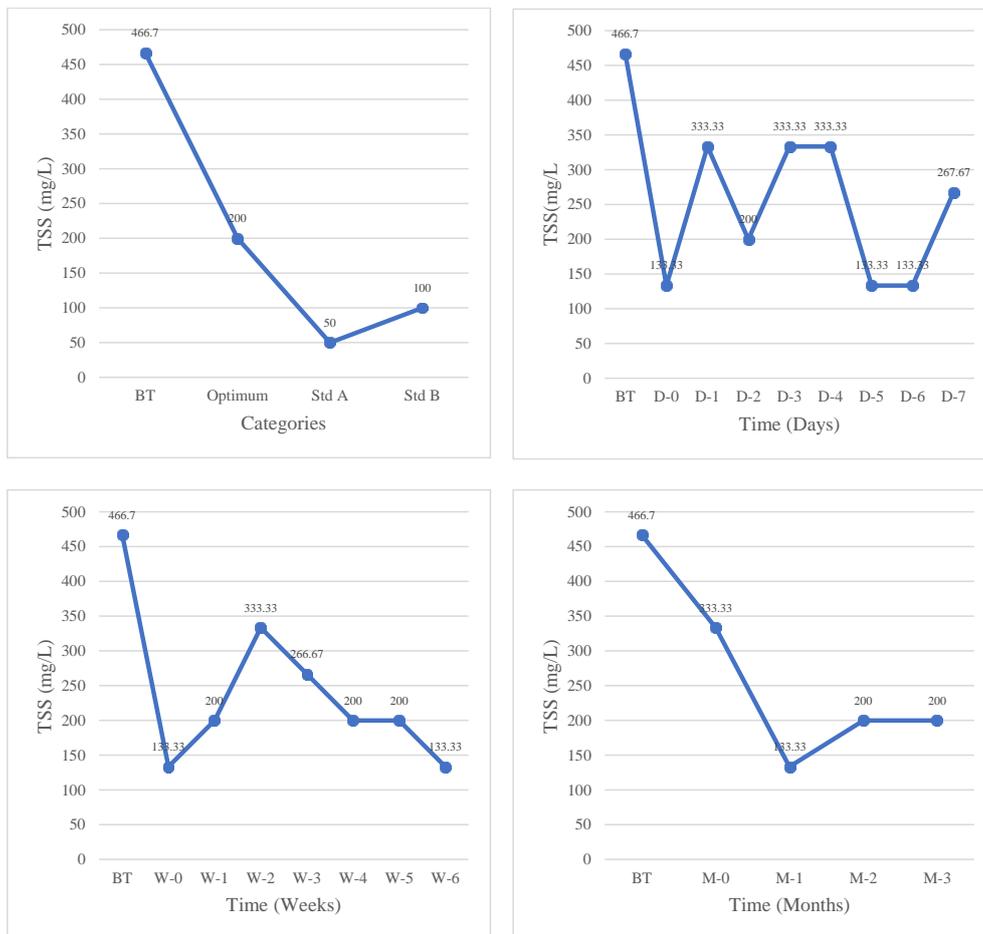


Figure 5. TSS Values Before and After Treatment. BT: Before Treatment, D: Day, W: Week, M: Month

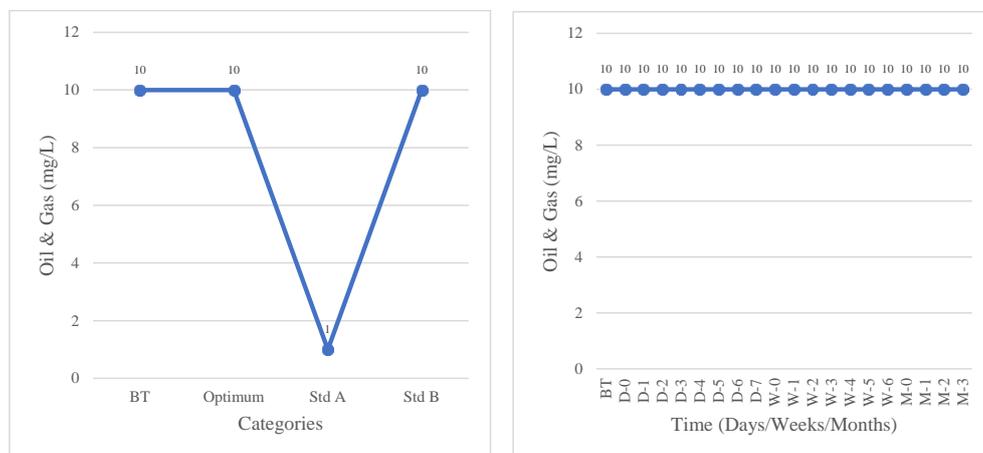


Figure 6. Oil and grease values before and after treatment. BT: Before Treatment, D: Day, W: Week, M: Month

3.2 Characterisation of Aging Effect on G-Treat

Characterisation of the aging effect on G-Treat is important to be conducted before adding the bio-coagulant to the oily wastewater sample to determine the level of effectiveness of the bio-coagulant in reducing pH, COD, BOD, TSS, and O&G. In this experiment, pH, FTIR spectroscopy, viscosity, and UV-Vis spectroscopy were used to understand the nature and behavior of the bio-coagulant. The aging effect or period effect on G-Treat was investigated to evaluate the outcome or performance of G-Treat in a certain period.

Figure 7 depicts the aging effect of bio-coagulant on pH values under three parameters (days, weeks, and months). The figure shows an increment of pH values under days, an increment of pH values from week 0 (W-0) until week 2 (W-2) and dropped subsequently. The figure also indicates an increment of pH values until month 2 (M-2) and dropped subsequently. pH plays a major role in coagulant-particle interaction for optimal neutralization and agglomeration in flocculation [41]. With an increase in pH, the species become charged, resulting in a change in the mechanism.

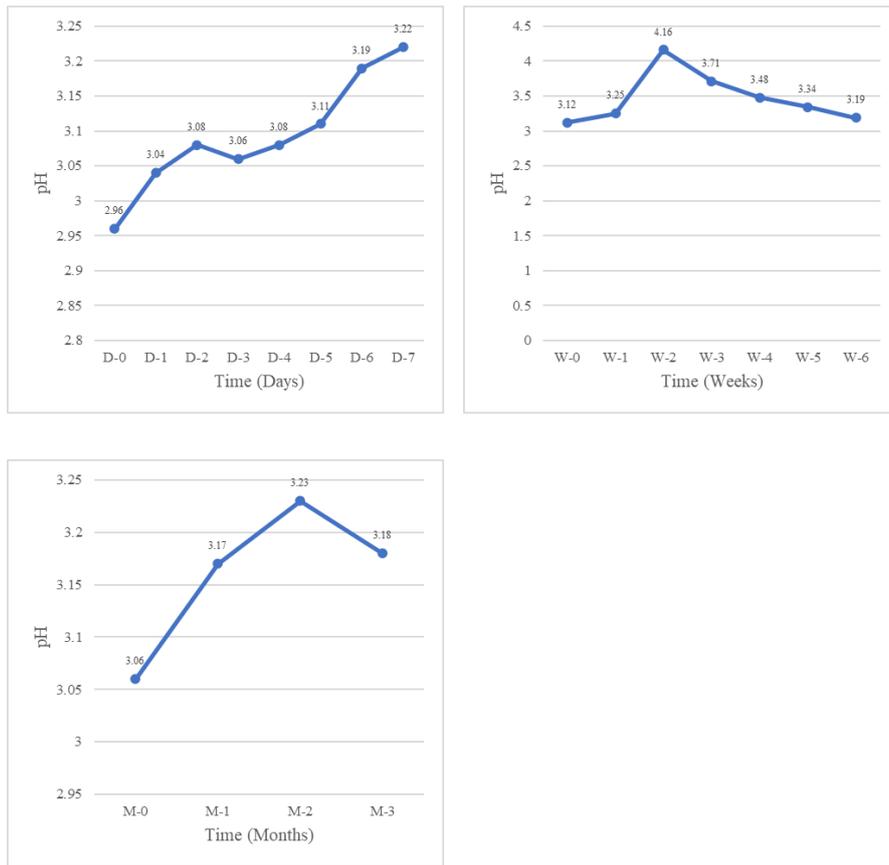


Figure 7. Aging Effect of Bio-coagulant on pH Values. D: Day, W: Week, M: Month

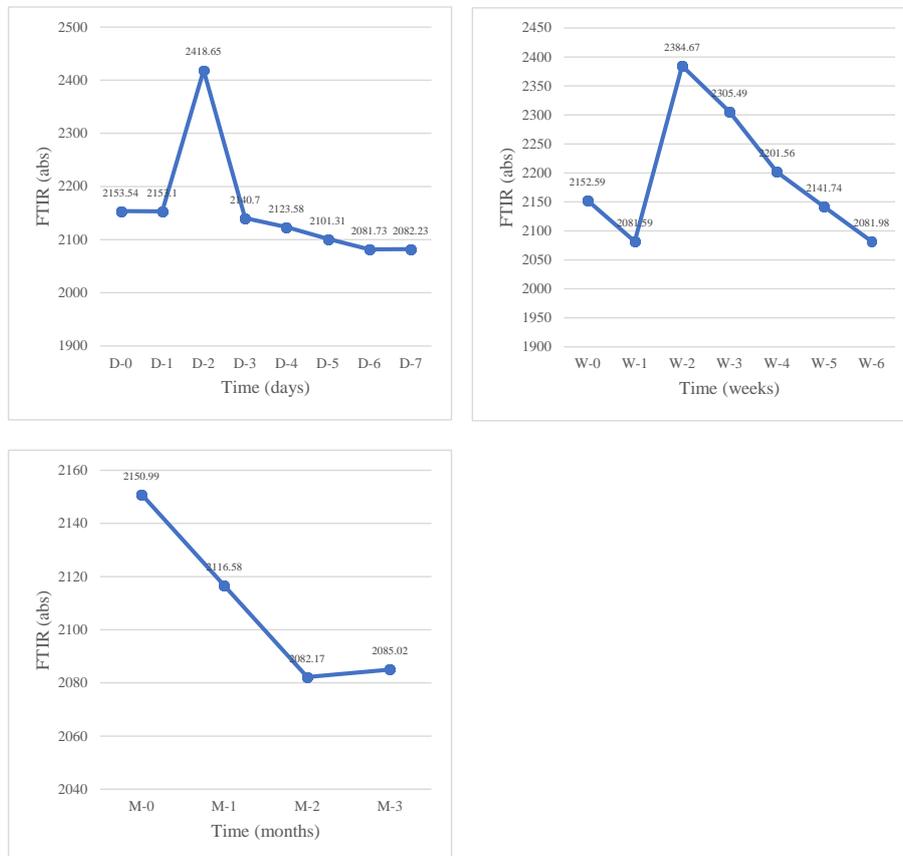


Figure 8. Aging Effect of Bio-coagulant on FTIR Absorbance. D: Day, W: Week, M: Month

Figure 8 illustrates the aging effect of bio-coagulant on FTIR absorbance under three parameters (days, weeks, and months). The figure shows a decrement of FTIR absorbance except on day 2 (D-2), a decrement of FTIR absorbance on week 1 (W-1), followed by an increment of FTIR absorbance on week 2 (W-2) and dropped subsequently, and a decrement of FTIR absorbance until month 2 (M-2) and increased subsequently. The range of wavelengths in the infrared region absorbed by a material is measured via FTIR analysis [42]. From the result, the absorbance of the bio-coagulant decreased over time due to the aging effect.

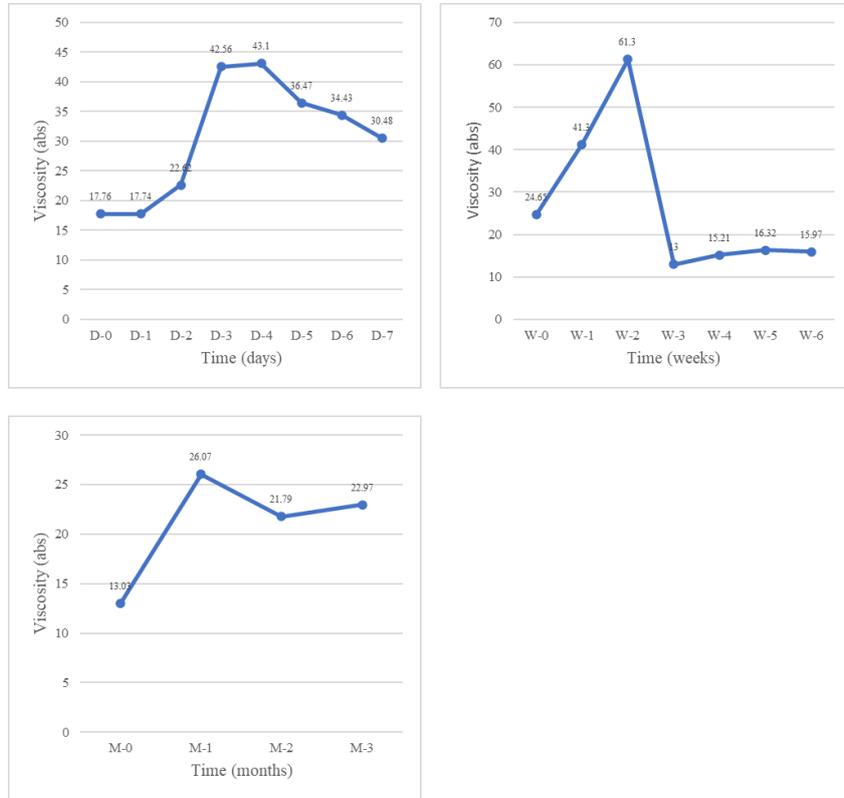


Figure 9. Aging Effect of Bio-coagulant on Viscosity. D: Day, W: Week, M: Month

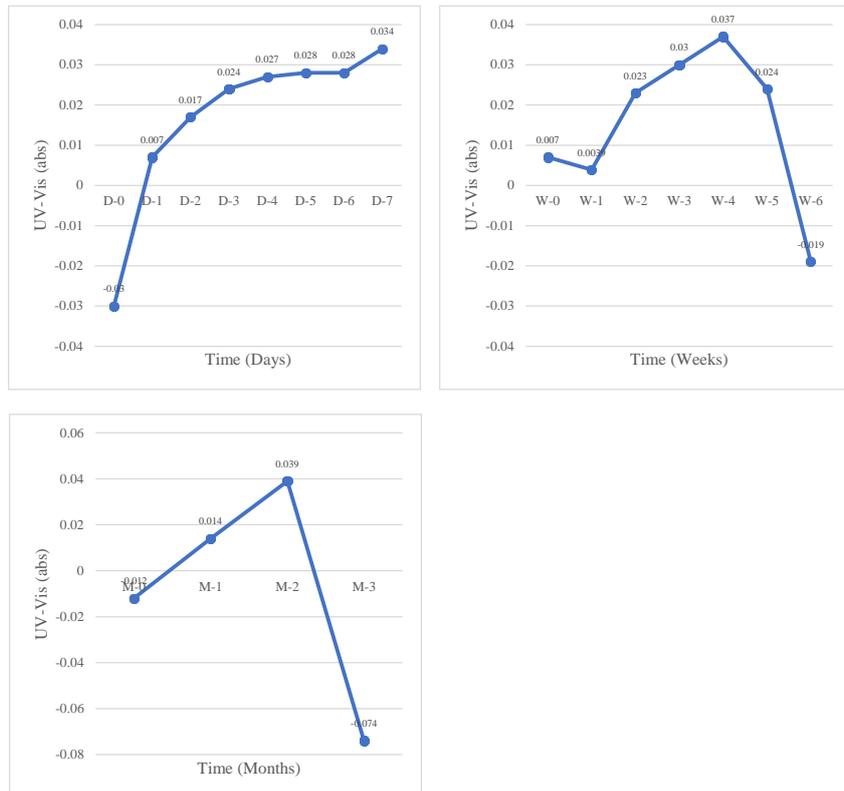


Figure 10. Aging Effect of Bio-coagulant on UV-Vis Absorbance. D: Day, W: Week, M: Month

Figure 9 depicts the aging effect of bio-coagulant on viscosity under three parameters (days, weeks, and months). The figure shows an increment of viscosity until day 4 (D-4) and dropped subsequently. Meanwhile, the viscosity reached its peak on week 2 (W-2) and dropped subsequently, and an increment until month 1 (M-1) before the viscosity fluctuated. Viscosity increases with molecule length; therefore, as charge density rises, polyelectrolyte chains are stretched further due to increased electrostatic repulsion between charged units [43].

Figure 10 illustrates the aging effect of bio-coagulant on UV-Vis absorbance under three parameters (days, weeks, and months). The figure shows an increment of UV-Vis absorbance under days, whereas the UV-Vis absorbance dropped in week 1 (W-1), rose again, and then dropped subsequently. The figure also shows an increment until month 2 (M-2) before the UV-Vis absorbance dropped subsequently. Spectral absorption is a measurement of the amount of ultraviolet or visible light absorbed by a water sample of a given path length. Absorption is directly proportional to the concentration of matter in the sample [44]. From the result, it can be concluded that the higher the value of UV-Vis absorbance, the higher the light absorption of G-Treat.

3.3 Efficiency of Storage Time and Aging Effect in Treating Wastewater Samples

As the bio-coagulant tends to lose its efficiency over time, it is important to determine the efficiency of storage time and the aging effect in treating wastewater samples in reducing pH, COD, BOD, TSS, and O&G.

Figure 11 shows the storage time efficiency and aging factor of bio-coagulant in reducing pH values under three parameters (days, weeks, and months). The figure shows that day 0 (D-0) is the most efficient day at 20.53%, week 2 (W-2) is the most efficient week at 33.16%, and month 0 (M-0) is the most efficient month at 17.70%. From the result, the efficiency of pH removal decreased over time, resulting in the declining performance of bio-coagulants in treating pH. It was observed that the trends for all parameters were almost identical but with different percentages of reduction for particular pH. From the jar test experiment, the curves for the graphs were in a U-shape form for the COD level and turbidity level versus pH.

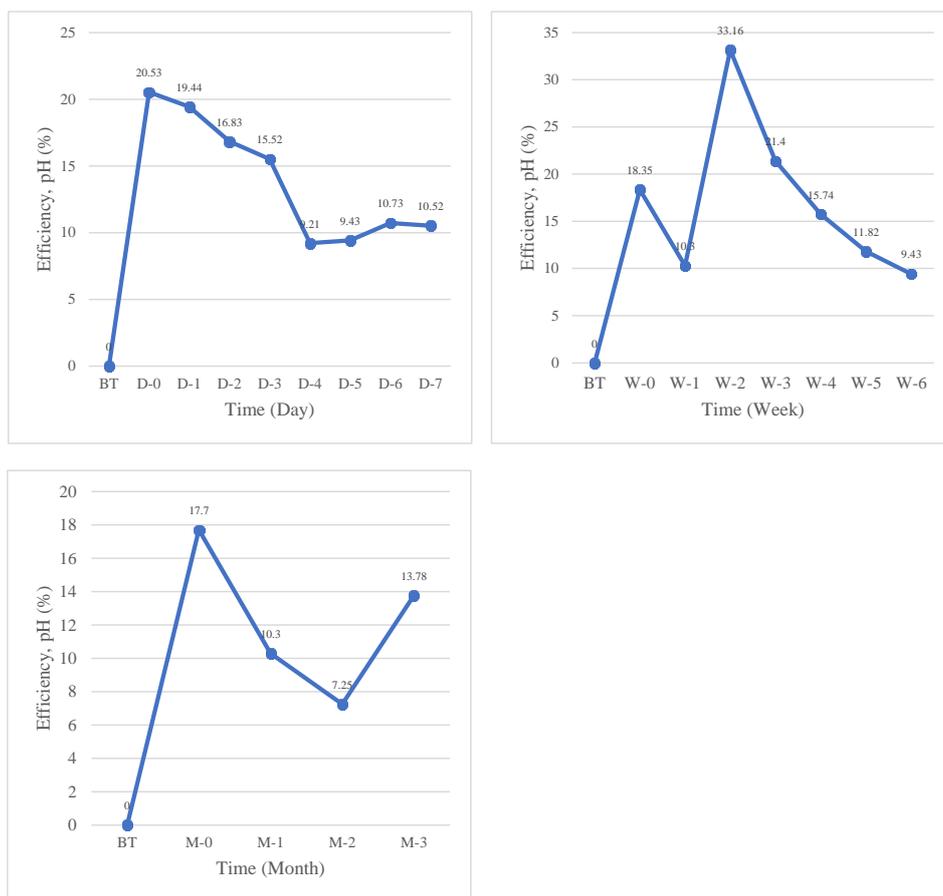


Figure 11. Storage Time Efficiency and Aging Factor of Bio-coagulant in Reducing pH. D: Day, W: Week, M: Month

Figure 12 presents the storage time efficiency and aging factor of bio-coagulant in reducing COD under three parameters (days, weeks, and months). The figure shows that day 7 (D-7) is the most efficient day at 83.70%, week 1 (W-1) is the most efficient week at 84.90%, and month 2 (M-2) is the most efficient month at 87.00%. From the result, it can be concluded that the longer the storage period, the efficiency of the bio-coagulant in reducing COD increases. The performance of bio-coagulant in reducing COD increased until a certain period (M-3) where it eventually dropped.

Figure 13 shows the storage time efficiency and aging factor of bio-coagulant in reducing BOD₅ under three parameters (days, weeks, and months). The figure shows that day 5 (D-5) is the most efficient at -18.50%, week 2 (W-2) is the most efficient week at 35.73%, and month 1 (M-1) is the most efficient month at -23.38%. From the result, it can be concluded that all BOD₅ values are considered at unfavorable conditions as the efficiency reached negative values, except for week 2 (W-2) [45]. The efficiency of the bio-coagulant in reducing BOD dropped after it reached the optimum time.

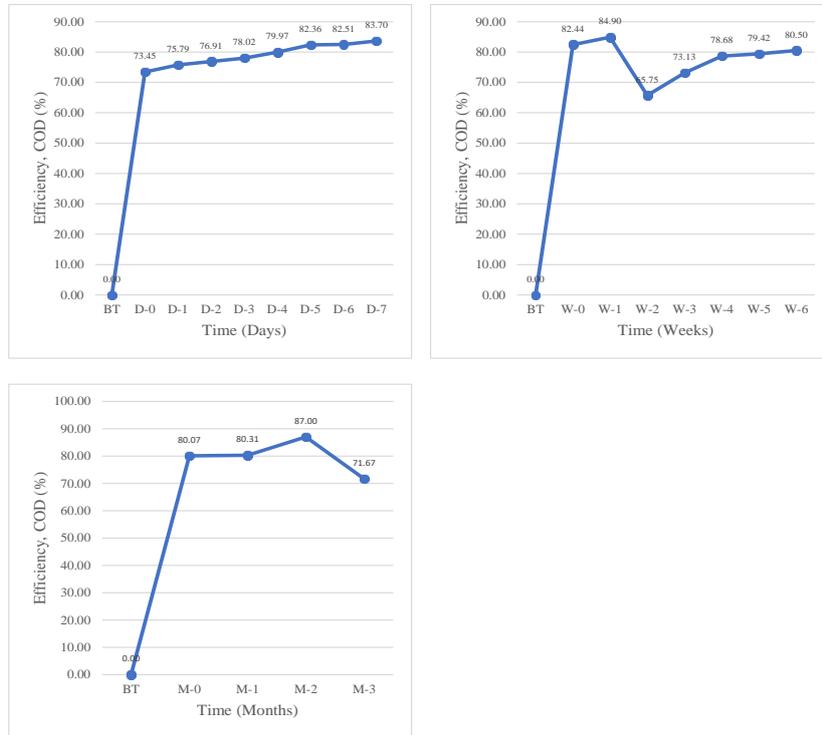


Figure 12. Storage Time Efficiency and Aging Factor of Bio-coagulant in Reducing COD. D: Day, W: Week, M: Month

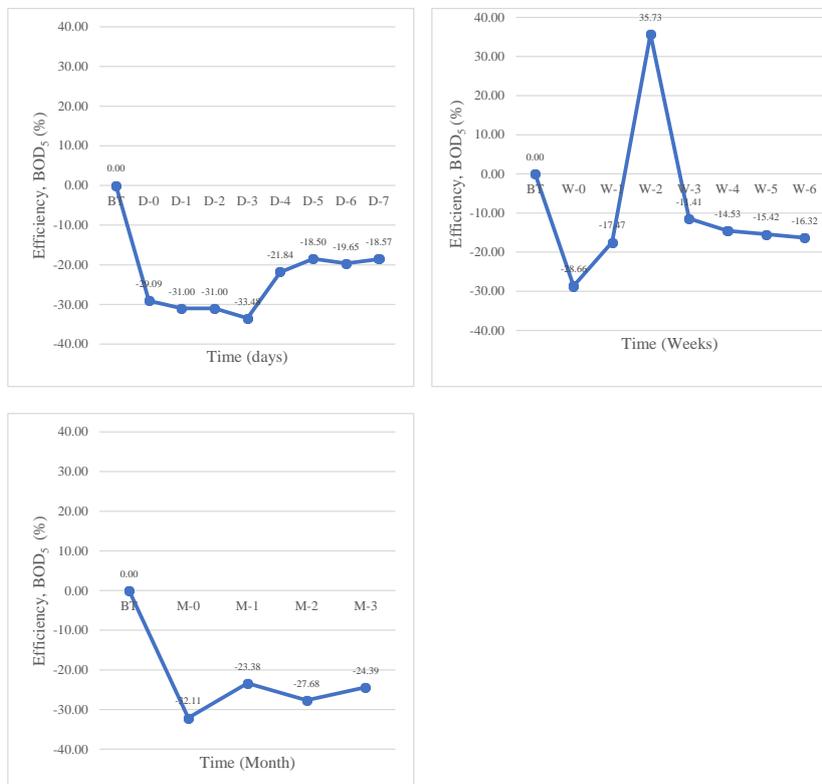


Figure 13: Storage Time Efficiency and Aging Factor of Bio-coagulant in Reducing BOD₅. BT: Before Treatment, D: Day, W: Week, M: Month

Figure 14 presents the storage time efficiency and aging factor of bio-coagulant in reducing TSS under three parameters (days, weeks, and months). The figure shows that day 0 (D-0), day 5 (D-5), and day 6 (D-6) are the most efficient days at 71.43%, week 0 (W-0) and week 6 (W-6) are the most efficient weeks at 71.43%, and month 1 (M-1) is the most efficient month at 71.43%. From the result, it can be concluded that the bio-coagulant efficiency depends on the absorption of the amount of sample during the heating and drying procedure. The more the bio-coagulant dried, the higher the efficiency of storage time and aging factor of the bio-coagulant. The performance of bio-coagulant drops if TSS is not removed properly, causing a high concentration of TSS in the sample [46]. Figure 15 shows the storage time efficiency and aging factor of bio-coagulant in reducing O&G under three parameters (days, weeks, and months). The efficiency of O&G is at 0% due to the small difference; hence, it is considered insignificant.

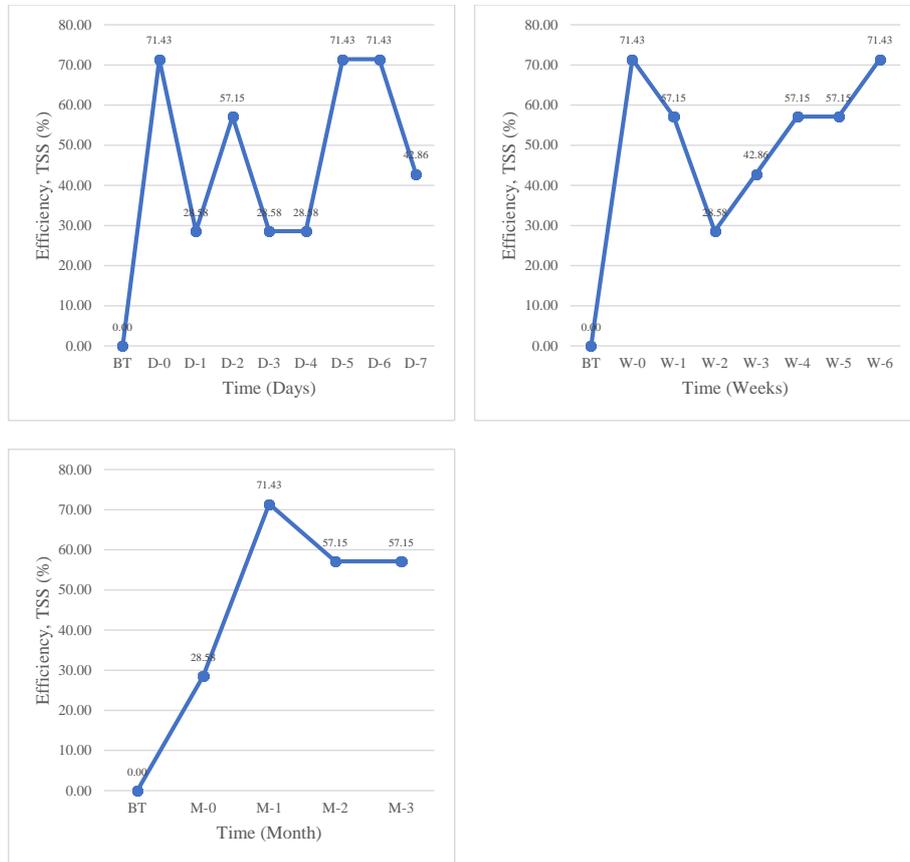


Figure 14. Storage Time Efficiency and Aging Factor of Bio-coagulant in Reducing TSS. BT: Before Treatment, D: Day, W: Week, M: Month

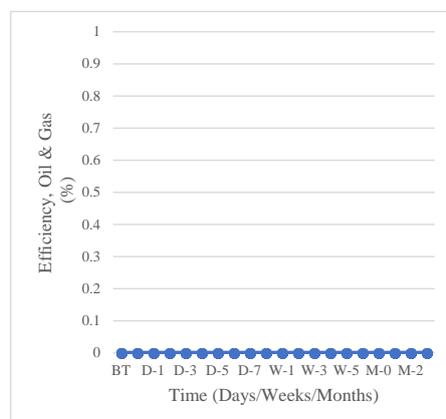


Figure 15. Storage Time Efficiency and Aging Factor of Bio-coagulant in Reducing O&G. BT: Before Treatment, D: Day, W: Week, M: Month

4.0 CONCLUSION

The present study of the aging effect of bio-coagulants for oily wastewater treatment can determine the optimum conditions of bio-coagulants in treating oily wastewater. Treating oily wastewater using bio-coagulants is a good approach for protecting nature and also human health. Despite being a good solution for treating oily wastewater, it is unavoidable that bio-coagulants have a time limit, unlike chemical-based coagulants. Based on the results, the treatment of wastewater using G-Treat was the most effective on week 2 (W-2) and the level of effectiveness of G-Treat dropped subsequently. This is because the aging factor of bio-coagulant causes the performance of G-Treat to drop in treating oily wastewater. Hence, it is recommended to treat oily wastewater at the optimum time of bio-coagulant for optimum efficiency.

5.0 CONFLICT OF INTEREST

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

6.0 AUTHORS CONTRIBUTION

Najmuddin Mohd Ramli: Investigation, Writing-original draft, Visualisation. **Mohd Najib Razali:** Conceptualisation, Methodology, Writing-review & editing, Project administration, Funding acquisition. **Puteri Nur Ain Daim:** Methodology, Resources. **Syahirah Syazwani Mohd Tarmizi:** Validation, Data curation, Resources.

7.0 ACKNOWLEDGEMENTS

The authors wish to express their gratitude and appreciation for the financial support from MNR Multitech Sdn Bhd and the support from the Faculty of Chemical and Process Engineering Technology, Universiti Malaysia Pahang Al-Sultan Abdullah, Malaysia.

8.0 REFERENCES

- [1] K. Abuhasel, M. Kchaou, M. Alquraish, Y. Munusamy, and Y. T. Jeng, "Oilywastewater treatment: Overview of conventional and modern methods, challenges, and future opportunities," *Water (Switzerland)*, vol. 13, no. 7, Apr. 2021, doi: 10.3390/w13070980.
- [2] Federal Subsidiary Legislation, "Environmental Quality Scheduled Wastes Regulations 2005," 2005.
- [3] N. Che Jamin and N. Z. Mahmood, "Scheduled Waste Management in Malaysia: An Overview," *Adv Mat Res*, vol. 1113, pp. 841–846, Jul. 2015, doi: 10.4028/www.scientific.net/amr.1113.841.
- [4] L. Yu, M. Han, and F. He, "A review of treating oily wastewater," *Arabian Journal of Chemistry*, vol. 10, Elsevier B.V., pp. S1913–S1922, May 01, 2017. doi: 10.1016/j.arabjc.2013.07.020.
- [5] A. D. M. de Medeiros, C. J. G. da Silva Junior, J. D. P. de Amorim, I. J. B. Durval, A. F. de Santana Costa, and L. A. Sarubbo, "Oily Wastewater Treatment: Methods, Challenges, and Trends," *Processes*, vol. 10, no. 4, MDPI, Apr. 01, 2022. doi: 10.3390/pr10040743.
- [6] S. B. Kurniawan *et al.*, "Challenges and opportunities of biocoagulant/bioflocculant application for drinking water and wastewater treatment and its potential for sludge recovery," *International Journal of Environmental Research and Public Health*, vol. 17, no. 24, MDPI AG, pp. 1–33, Dec. 02, 2020. doi: 10.3390/ijerph17249312.
- [7] A. I. Adetunji and A. O. Olaniran, "Treatment of industrial oily wastewater by advanced technologies: a review," *Applied Water Science*, vol. 11, no. 6, Springer Science and Business Media Deutschland GmbH, Jun. 01, 2021. doi: 10.1007/s13201-021-01430-4.
- [8] M. Almutairi, "Evaluate the effectiveness technology for the treatment of oily wastewater," *J Water Health*, vol. 20, no. 8, pp. 1171–1187, Aug. 2022, doi: 10.2166/wh.2022.089.
- [9] N. B. Singh, G. Nagpal, S. Agrawal, and Rachna, "Water purification by using Adsorbents: A Review," *Environmental Technology and Innovation*, vol. 11, Elsevier B.V., pp. 187–240, Aug. 01, 2018. doi: 10.1016/j.eti.2018.05.006.
- [10] L. M. Quan *et al.*, "Review of the application of gasification and combustion technology and waste-to-energy technologies in sewage sludge treatment," *Fuel*, vol. 316, May 2022, doi: 10.1016/j.fuel.2022.123199.
- [11] Baharudin Mohamad, "Ministry Of Economy Department Of Statistics Malaysia Annual Economic Statistics 2022 Water Supply; Sewerage, Waste Management & Remediation Activities," 2023.
- [12] C. Zhao *et al.*, "Application of coagulation/flocculation in oily wastewater treatment: A review," *Science of the Total Environment*, vol. 765, Elsevier B.V., Apr. 15, 2021. doi: 10.1016/j.scitotenv.2020.142795.
- [13] R. S. Al-Wasify, S. R. Hamed, and S. Ragab, "Assessing the potential of de-oiled peanut (*Arachis hypogea*) seeds for surface water treatment: A sustainable alternative to chemical coagulants," *Egypt J Aquat Res*, vol. 49, no. 3, pp. 297–302, Sep. 2023, doi: 10.1016/j.ejar.2023.04.003.
- [14] K. O. Iwuozor, "Prospects and Challenges of Using Coagulation-Flocculation Method in the Treatment of Effluents," *Advanced Journal of Chemistry-Section A*, vol. 2, no. 2, pp. 105–127, 2019, [Online]. Available: <http://ajchem-a.com>
- [15] M. T. F. de Souza *et al.*, "Extraction and use of *Cereus peruvianus* cactus mucilage in the treatment of textile effluents," *J Taiwan Inst Chem Eng*, vol. 67, pp. 174–183, Oct. 2016, doi: 10.1016/j.jtice.2016.07.009.

- [16] D. Diver, I. Nhapi, and W. R. Ruziwa, "The potential and constraints of replacing conventional chemical coagulants with natural plant extracts in water and wastewater treatment," *Environmental Advances*, vol. 13. Elsevier Ltd, Oct. 01, 2023. doi: 10.1016/j.envadv.2023.100421.
- [17] R. M. El-taweel *et al.*, "A review of coagulation explaining its definition, mechanism, coagulant types, and optimization models; RSM, and ANN," *Current Research in Green and Sustainable Chemistry*, vol. 6. Elsevier B.V., Jan. 01, 2023. doi: 10.1016/j.crgsc.2023.100358.
- [18] M. Najib Razali, A. Alhammadi, M. Musa, M. Khairul Nizam, and A. Ermafiqka Anuar, "Comparison Of Natural Adsorbent For Emulsified Wastewater Treatment," *IOP Conf Ser Mater Sci Eng*, vol. 1092, no. 1, p. 012020, Mar. 2021, doi: 10.1088/1757-899x/1092/1/012020.
- [19] M. N. Razali, A. E. Anuar, M. Musa, and N. Mohd Ramli, "Remediation of Oil and Gas Sewage Effluents Using Physical Treatments," *Journal of Chemical Engineering and Industrial Biotechnology*, vol. 6, no. 1, pp. 12–19, Sep. 2020, doi: 10.15282/jceib.v6i1.4417.
- [20] M. T. Karia, A. H. Haziq, N. M. Ramli, M. K. N. M. Zuhan, and M. N. Razali, "Remediation of aquaculture effluents using physical treatment," *Mater Today Proc*, vol. 57, pp. 1196–1201, Jan. 2022, doi: 10.1016/j.matpr.2021.10.386.
- [21] A. Patchaiyappan and S. P. Devipriya, "Application of plant-based natural coagulants in water treatment," in *Cost Effective Technologies for Solid Waste and Wastewater Treatment*, Elsevier, 2021, pp. 51–58. doi: 10.1016/B978-0-12-822933-0.00012-7.
- [22] M. Sillanpää, M. C. Ncibi, A. Matilainen, and M. Vepsäläinen, "Removal of natural organic matter in drinking water treatment by coagulation: A comprehensive review," *Chemosphere*, vol. 190. Elsevier Ltd, pp. 54–71, 2018. doi: 10.1016/j.chemosphere.2017.09.113.
- [23] D. S. Duttagupta, V. M. Jadhav, and V. J. Kadam, "Chitosan: A Propitious Biopolymer for Drug Delivery," *Curr Drug Deliv*, vol. 2, pp. 369–381, 2015. doi: 10.2174/1567201812666150310151657
- [24] E. K. Tetteh and S. Rathilal, "Application of Organic Coagulants in Water and Wastewater Treatment," *Organic Polymer*, pp. 1–18, 2019. doi: http://dx.doi.org/10.5772/intechopen.84556
- [25] L. Lamanna *et al.*, "Oil-Water Emulsion Flocculation through Chitosan Desolubilization Driven by pH Variation," *ACS Omega*, vol. 8, no. 23, pp. 20708–20713, Jun. 2023, doi: 10.1021/acsomega.3c01257.
- [26] ASTM International D1293-18, "Standard Test Methods for pH of Water 1," West Conshohocken, 2018. doi: 10.1520/D1293-18.
- [27] ASTM International D7889-13, "Standard Test Method for Field Determination of In-Service Fluid Properties Using IR Spectroscopy 1," West Conshohocken, 2013. doi: 10.1520/D7889-13.
- [28] ASTM International D445-21, "Designation: D445 – 21 Standard Test Method for Kinematic Viscosity of Transparent and Opaque Liquids (and Calculation of Dynamic Viscosity) 1," West Conshohocken, 1998. doi: 10.1520/D0445-21.
- [29] ASTM International D2800-92, "Standard Test Method for Preparation of Methyl Esters From Oils for Determination of Fatty Acid Composition by Gas-Liquid Chromatography," 1998.
- [30] Arnold E. Greenberg; Lenore S. Clesceri; Andrew D. Eaton; Mary Ann H. Franson, *Standard Methods For The Examination of Water and Wastewater*, 18 th Edition. Washington: American Public Health Association, 1992.
- [31] A. J. León-Luque, C. L. Barajas, and C. A. Peña-Guzmán, "Determination of the Optimal Dosage of Aluminum Sulfate in the Coagulation-Flocculation Process Using an Artificial Neural Network," *International Journal of Environmental Science and Development*, vol. 7, no. 5, pp. 346–350, 2016, doi: 10.7763/IJESD.2016.V7.797.
- [32] H. R. Lim, C. M. Choo, C. H. Chong, and V. L. Wong, "Optimization studies for water defluoridation with two-stage coagulation processes using new industrial-based chemical coagulants," *Journal of Water Process Engineering*, vol. 42, Aug. 2021, doi: 10.1016/j.jwpe.2021.102179.
- [33] H. Zhang *et al.*, "The function of doping nitrogen on removing fluoride with decomposing La-MOF-NH₂: Density functional theory calculation and experiments," *J Environ Sci (China)*, vol. 135, pp. 118–129, Jan. 2024, doi: 10.1016/j.jes.2023.01.015.
- [34] S. M. Khumalo, B. F. Bakare, E. K. Tetteh, and S. Rathilal, "Application of Response Surface Methodology on Brewery Wastewater Treatment Using Chitosan as a Coagulant," *Water (Switzerland)*, vol. 15, no. 6, Mar. 2023, doi: 10.3390/w15061176.
- [35] W. Du, J. Guo, H. Li, and Y. Gao, "Heterogeneously Modified Cellulose Nanocrystals-Stabilized Pickering Emulsion: Preparation and Their Template Application for the Creation of PS Microspheres with Amino-Rich Surfaces," *ACS Sustain Chem Eng*, vol. 5, no. 9, pp. 7514–7523, Sep. 2017, doi: 10.1021/acssuschemeng.7b00375.
- [36] B. A. Lasaki, P. Maurer, H. Schönberger, and E. P. Alvarez, "Empowering municipal wastewater treatment: Enhancing particulate organic carbon removal via chemical advanced primary treatment," *Environ Technol Innov*, vol. 32, p. 103436, Nov. 2023, doi: 10.1016/j.eti.2023.103436.
- [37] H. A. Abdullah and A. J. Jaedel, "Turbidity, Color and Chemical Oxygen Demand Removals from Synthetic Textile Wastewater Using Chitosan as a Coagulant," in *IOP Conference Series: Materials Science and Engineering*, Institute of Physics Publishing, Aug. 2019. doi: 10.1088/1757-899X/584/1/012016.
- [38] R. S. M. A. Hamdon, A. Salem, H. G. I. Ahmed, and M. M. H. ElZahar, "Use of Chitosan for Enhancing the Process of Surface Water Purification in Egypt," *International Journal of Environmental Science and Development*, vol. 13, no. 2, pp. 26–34, Apr. 2022, doi: 10.18178/ijesd.2022.13.2.1368.

- [39] B. Belgis, "Industrial Application of Chitosan as Promising Material for Wastewater Purification: A Review," *CSID Journal of Infrastructure Development*, vol. 3, no. 1, p. 51, May 2020, doi: 10.32783/csid-jid.v3i1.92.
- [40] B. A. Lasaki, P. Maurer, H. Schönberger, and E. P. Alvarez, "Empowering municipal wastewater treatment: Enhancing particulate organic carbon removal via chemical advanced primary treatment," *Environ Technol Innov*, vol. 32, p. 103436, Nov. 2023, doi: 10.1016/j.eti.2023.103436.
- [41] B. Koul, N. Bhat, M. Abubakar, M. Mishra, A. P. Arukha, and D. Yadav, "Application of Natural Coagulants in Water Treatment: A Sustainable Alternative to Chemicals," *Water (Switzerland)*, vol. 14, no. 22. MDPI, Nov. 01, 2022. doi: 10.3390/w14223751.
- [42] M. M. H. Elzahar and M. Bassyouni, "Removal of direct dyes from wastewater using chitosan and polyacrylamide blends," *Sci Rep*, vol. 13, no. 1, Dec. 2023, doi: 10.1038/s41598-023-42960-y.
- [43] S. Botelho da Silva, M. Krolicka, L. A. M. van den Broek, A. E. Frissen, and C. G. Boeriu, "Water-soluble chitosan derivatives and pH-responsive hydrogels by selective C-6 oxidation mediated by TEMPO-laccase redox system," *Carbohydr Polym*, vol. 186, pp. 299–309, Apr. 2018, doi: 10.1016/j.carbpol.2018.01.050.
- [44] A. Radzevičius, M. Dapkienė, N. Sabienė, and J. Dzięcioł, "A rapid uv/vis spectrophotometric method for the water quality monitoring at on-farm root vegetable pack houses," *Applied Sciences (Switzerland)*, vol. 10, no. 24, pp. 1–15, Dec. 2020, doi: 10.3390/app10249072.
- [45] A. Al-Sayed, G. K. Hassan, M. T. Al-Shemy, and F. A. El-gohary, "Effect of organic loading rates on the performance of membrane bioreactor for wastewater treatment behaviours, fouling, and economic cost," *Sci Rep*, vol. 13, no. 1, Dec. 2023, doi: 10.1038/s41598-023-42876-7.
- [46] N. M. Daud, S. R. S. Abdullah, H. A. Hasan, A. R. Othman, and N. 'Izzati Ismail, "Coagulation-flocculation treatment for batik effluent as a baseline study for the upcoming application of green coagulants/flocculants towards sustainable batik industry," *Heliyon*, vol. 9, no. 6, Jun. 2023, doi: 10.1016/j.heliyon.2023.e17284.