



The effects of the natural coagulant *Moringa oleifera* and alum in wastewater treatment at the Bandar Abbas Oil Refinery

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Abstract

Background: The refining process generates large volumes of wastewater containing a variety of chemical contaminants. The use of natural substitutes in treating wastewater which have fewer harmful effects is considered an effective step towards protecting the environment and sustaining the development of these industries. This study focused on the use of *Moringa oleifera* and alum at the Wastewater Unit at Bandar Abbas Refinery.

Methods: This study was performed in 2014 in a laboratory using jar apparatus. These experiments were conducted in batch system and effective parameters including pH, coagulant dose and contact time were investigated on the wastewater obtained from Bandar Abbas Oil Refinery.

Results: The jar test experiment showed that *M. oleifera* at 70 mg/L, optimum temperature, pH, and mixing speed could remove 38.60% of chemical oxygen demand (COD), 63.70% of turbidity, and 62.05% of total suspended solids (TSS). Also, alum at 40 mg/L removed COD, turbidity, and TSS by 51.72%, 92.16%, and 85.26% respectively from the refinery wastewater. Moreover, when *M. oleifera* and alum was used together with a 2:1 dosage ratio (alum at 80 mg/L and *M. oleifera* at 70 mg/L), they will remove COD, turbidity, and TSS by up to 50.41%, 86.14%, and 81.52% respectively.

Conclusion: The use of *M. oleifera* as a natural coagulant is important in treating refinery wastewater not only from an environmental but also an economic point of view.

Keywords: *Moringa oleifera*, Alum, Bandar Abbas Refinery, Industrial wastewater

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Introduction

Rapid population growth and industrial development in the countries of the world are degrading the environment through the uncontrolled growth of urbanization and industrialization, as well as contributing to the destruction of natural habitats. The growing population and the environmental deterioration pose the challenge of sustained development without environmental damage. Nowadays, chemical compounds in industrial production is increasingly used and many of these substances have found their way into nature with adverse impacts, which can gradually destroy the environment (1). For example, the vast development of hydrocarbon-processing industries and the extensive consumption of petroleum and gas derivatives in most big industries have increased the danger of oil pollution in the natural environment (2). One of the problems with chemical industries is the presence of effluents containing emulsifiable petroleum products which

pollute the environment enormously (3).

Wastewater from the petroleum industry contains organic and inorganic compounds, dissolved and suspended solids (4). The methods of treating wastewater include activated carbon adsorption, oxidation, chemical coagulation/flocculation, electrochemical methods, membrane techniques, and biological treatment processes, which are frequently used to treat textile effluents (5). In the process of wastewater treatment, especially during the flocculation and coagulation processes, various chemical materials are used. The effectiveness of these chemicals as coagulants is well recognized; however, there are some disadvantages in connection with using these coagulants, such as the cost of the process, the operation, and the maintenance and production of large volumes of sludge (6). In this regard, the use of substitute materials with fewer harmful effects can be a step taken to protect the environment and human health (7). Compounds extracted from plant and animal



tissues are among these substitute materials (8). The use of these natural compounds as coagulant aids in some cases also is cost-effective (9).

The utilization of natural herbal substances in water and wastewater treatment is not a new idea (10). Natural biopolymers having applications in the water and wastewater industries include starches (potato, wheat, corn, and rice starches), sodium alginate, *Moringa oleifera*, calcium alginate, chitosan, tannin, natural gums and xanthan (7).

Moringa oleifera habitats originate from around the Dead Sea and extend in scattered regions along the Red Sea coast to northern Somalia and around the Arabian Peninsula, up to the mouth of the Persian Gulf states, United Arab Emirates, Saudi Arabia, and Palestine (11). *M. oleifera* grows in arid, semi-arid, and humid areas and has been named a "miracle tree" owing to its several benefits (12). Javanshir first published *M. oleifera* in Iranian flora in 1993. Major distribution areas of the species include Mt Bashagerd in Nikshahr through to south Jazmorian (11). Sutherland et al have suggested that *M. oleifera* seeds do not constitute a serious hazard to human health (13). Their use in new technologies of wastewater treatment is, therefore, very promising. The advantages of using *M. oleifera* are: a positive cost-effectiveness, water treated without extreme pH, and a high level of biodegradability (14). These advantages are especially the case in rural areas.

The use of *M. oleifera* for wastewater treatment is perhaps one of its more interesting usages. Although there are many previous papers investigating its utilization as a natural adsorbent for special pollutant removal (12), the seeds of this tropical tree have a high amount of proteins that act like cationic polyelectrolytes once they are added to raw water (15).

Moringa oleifera seeds are also used as a primary coagulant in wastewater treatment due to the presence of a water-soluble cationic coagulant protein able to reduce the turbidity, chemical oxygen demand (COD), and Total dissolved solids (TDS) of the wastewater treated. *M. oleifera* has been shown to be one of the most effective uses as a primary coagulant for water and wastewater treatment (7,16-21).

The aim of this study was to compare the efficiency of *M. oleifera* seed and alum in total suspended solids (TSS), COD, and turbidity removal in the wastewater unit at Bandar Abbas Refinery as one method for treating wastewater in the petroleum industries in developing countries.

Methods

Sampling

In this study, the coagulation performance of *M. oleifera* and alum in treating wastewater at Bandar Abbas Refinery was investigated.

First, wastewater samples were taken daily from the outlet of the equalization pool near the inlet into the dissolved air flotation (DAF) pool of the refinery for the duration of one month. Table 1 presents the four parameters studied and their mean values. All measurement methods of the parameters were based on the standards for water and

wastewater experiments (22).

Preparing *Moringa oleifera*

Moringa oleifera seeds were obtained from Busher province (Herbarium number 53054) and *Moringa* powder and the extract was prepared and kept at room temperature. To prepare the powder, the seeds were placed in an oven at 35°C for 5 hours to make sure they were dry. The seeds were dehusked by hand, and then turned into a soft powder with a particle size of about 600 µm using a moulinex grinder. One gram of the powder was added to one liter of doubled distilled water and mixed using a stirrer for 3 minutes at 120 rpm. The extract was then passed through 0.45-micron filter paper to obtain a homogeneous solution free of suspended materials. This solution was freshly prepared each time it was used in water treatment, and kept in a cool place with a maximum temperature of 20°C to prevent changes in its pH and viscosity (23).

Measurement methods and laboratory equipment

For the jar test, 1 L of the wastewater was poured in beakers and quickly mixed using a stirrer at a high speed (120 rpm) for 1 minute, and then mixed slowly at 40 rpm for 20 minutes, followed by the precipitation process which took 30 minutes (24). To measure turbidity, a turbidity meter and the standard solutions 1 NTU, 10 NTU, and 100 NTU were employed by using the ASTM method. TSS was measured based on the SM 2540D method by using a vacuum oven with an Erlenmeyer flask (Oven-Vacuum Erlen), and COD measurements were carried out based on the ASTM DI252-06 method by using vials manufactured by reputable companies and a COD reactor (25-27). All experiments were performed at the laboratory of Bandar Abbas Oil Refinery Company.

Methodology

The jar test was performed first with a constant concentration of wastewater and varying concentrations of *M. oleifera* (10, 20, 40, 60, 70, and 80 mg/L) to determine the optimum dosage.

The pH test was then carried out at the pH values of 5, 5.5, 6, 6.5, 7, 7.5, 8, 8.5 and 9 to find the optimum pH value. Considering the weather conditions in Bandar Abbas, the jar test was performed at the optimum values of *M. oleifera* and pH at 5 different temperatures (20, 25, 30, 35, and 40°C) to determine the optimum temperature for

Table 1. Qualitative characteristics of the wastewater obtained from the equalization pool at the refinery

Parameter	Textile effluent	
	Range	Average ± SE
COD (mg/L)	390-440	435 ± 18.40
TSS (mg/L)	85-120	95 ± 8.08
Turbidity (NTU)	29-41	37 ± 3.57
pH	4.5-9	8.7 ± 0.67

Abbreviations: COD, chemical oxygen demand; TSS, total suspended solids; SE, standard error.

Table 2. The range of parameters examined in this study

pH	Temperature (°C)	Rapid mix (rpm)	Dosage of <i>Moringa oleifera</i> (mg/L)	Dosage of Alum (mg/L)
5, 5.5, 6, 6.5, 7, 7.5, 8, 8.5, 9	20, 25, 30, 35, 40	220, 260, 300	10, 20, 40, 60, 70, 80	40

wastewater treatment.

In the next stage, rapid mixing was performed at three different speeds (220, 260, and 300 rpm) for one minute and the results were compared to determine the effects of mixing speed on treatment efficiency (Table 2).

Considering the fact that, at present, the Bandar Abbas Oil Refinery Company uses alum at 40 mg/L for the inoculation in its wastewater treatment, the experiment was repeated at optimum values to combine speed, temperature, and pH with alum at 40 mg/L to determine treatment efficiency.

Finally, the experiment was repeated at the optimum values to combine speed, temperature, and pH using the optimum concentration of alum and *M. oleifera* with the ratios of 1:2, 1:1, and 2:1, respectively.

Results

In this study, the efficiency of coagulant materials *M. oleifera*, alum, and the simultaneous use of alum and *M. oleifera* in the removal of turbidity, TSS, and COD was examined. The results of the jar test at various *M. oleifera* concentrations (10, 20, 40, 60, 70, and 80 mg/L) are presented in Figure 1. The optimum concentration of *M. oleifera* is 70 mg/L, which reduced the COD of wastewater by 17.24% (from 435 to 360 mg/L), turbidity by 52.43% (from 37 to 19.4 NTU), and TSS by 46.31% (from 95 to 44 mg/L).

Figure 2 shows the effects of various pH values at the constant *M. oleifera* concentration of 70 mg/L. As seen in this figure, the optimum pH is 6 where COD, turbidity, and TSS decreased by 39.78%, 62.16%, and 61.05% respectively.

In the jar test with *M. oleifera* concentration at 70 mg/L and pH 6 at 5 different temperatures (20, 25, 30, 35, and 40 °C), the optimum temperature for removal efficiency of

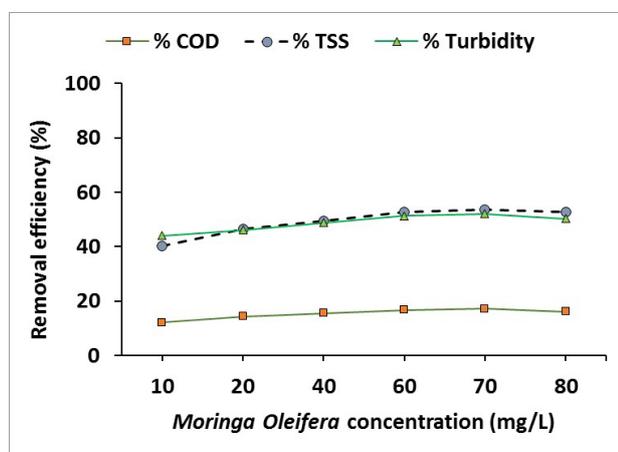


Figure 1. The effects of various *Moringa oleifera* concentrations on the efficiency of the removal of COD, turbidity, and TSS from petroleum refinery wastewater.

20 °C was obtained (Figure 3). Considering the results of the jar test at the three levels of rapid mixing, the removal efficiency at the mixing speed of 300 rpm was the highest. This removed COD, turbidity, and TSS by 38.60%, 63.70%, and 62.05% respectively (Figure 4).

After obtaining the optimal condition of the natural coagulant *M. oleifera* (pH: 6, temperature: 20 °C, and mixing speed: 300 rpm), in the next stage the experiment was performed with the chemical compound alum at 40 mg/L. As seen in Figure 5, alum removed COD, turbidity, and TSS from the refinery wastewater with a higher efficiency than *M. oleifera* (51.72%, 92.16%, and 85.26% respectively).

Finally, an experiment was performed under optimum conditions (constant pH: 6, constant temperature: 20 °C, and mixing speed: 300 rpm) in which mixtures of the two coagulants alum and *M. oleifera* were used. The re-

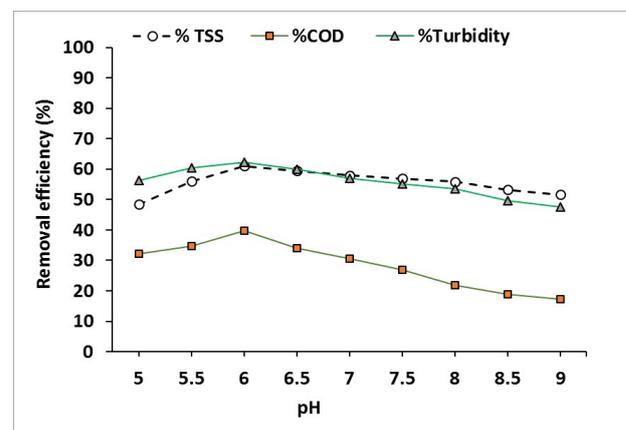


Figure 2. The effects of various pH values with a constant *Moringa oleifera* concentration of 70 mg/L on the efficiency of the removal of COD, turbidity, and TSS from petroleum refinery wastewater.

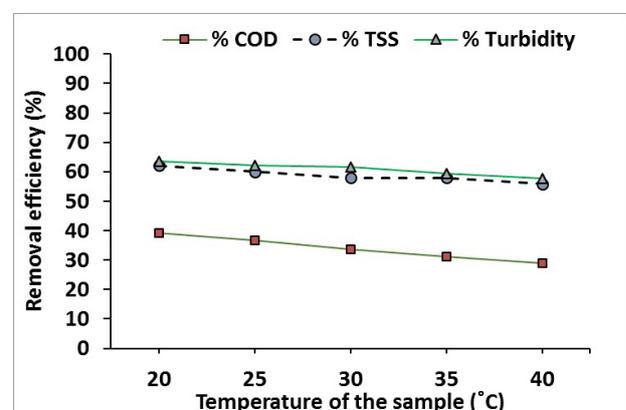


Figure 3. The effects of various temperatures with a constant *Moringa oleifera* concentration of 70 mg/L and pH value of 6 on the efficiency of the removal of COD, turbidity, and TSS from petroleum refinery wastewater.

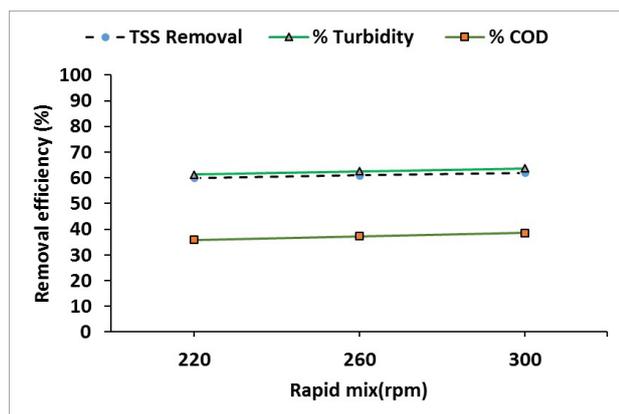


Figure 4. The effects of various mixing speeds with a constant *Moringa oleifera* concentration of 70 mg/L, constant pH value of 6, and constant temperature of 20°C on the efficiency of the removal of COD, turbidity, and TSS from petroleum refinery wastewater.

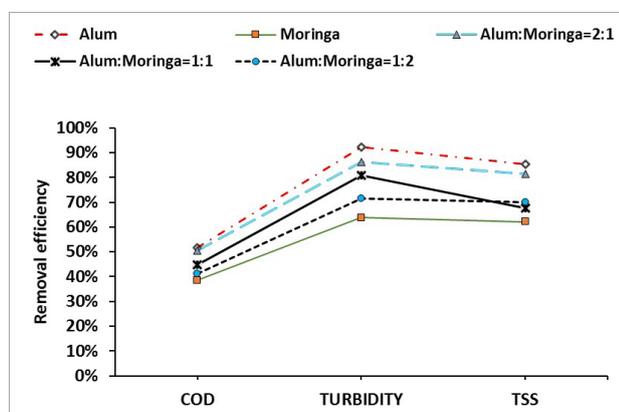


Figure 5. Efficiencies of alum and *Moringa oleifera* on the efficiency of the removal of COD, turbidity, and TSS from petroleum refinery wastewater.

sults showed that, if they are used with the optimal ratio of 2:1 (alum at 80 mg/L and *M. oleifera* at 70 mg/L), they will remove turbidity, COD, and TSS by up to 86.14%, 50.41%, and 81.52% respectively. Moreover, a mixture in the optimal ratio of 1:1 (alum at 40 mg/L and *M. oleifera* at 70 mg/L) removed the three parameters by 80.71% for turbidity, 44.70% for COD, and 67.50% for TSS, while a mixture in the ratio of 1:2 (40 mg/L alum and 140 mg/L *M. oleifera*) reduced the parameters by lower percentages (Figure 5).

Discussion

The results of this research indicated that *M. oleifera* has a good removal efficiency turbidity, COD, and TSS by 63.70%, 38.60%, and 62.05% respectively. Under optimum conditions (pH: 6, temperature: 20°C, and *M. oleifera* dosage: 70 mg/L) it can improve the quality of the refinery wastewater.

Figure 1 showed the effects of various *M. oleifera* dosage on the efficiency of the removal of COD, turbidity, and TSS. An analysis of variance (ANOVA) analysis was car-

ried out to evaluate the significance of the difference between the mean of measured values. This statistical analysis did not confirm any significant difference for all parameters ($P=0.086$, $P=0.064$, and $P=0.069$ respectively for COD, TSS and turbidity).

The evaluation of pH effectiveness in the removal of COD, turbidity, and TSS due to the addition of *M. oleifera* is presented in Figure 2 and shows that the maximum removal occurred with pH in the range 5–6.5 (turbidity, COD and TSS reduces below 82%). The results were in line with some previous research studies (28). The basic amino acids present in the protein of *M. oleifera* would act as a proton acceptor and result in the release of hydroxyl functional group making the solution basic. This accounted for the basic pH values observed for *M. oleifera* treatments compared with alum treatments. These results also require high-tech equipment to ensure the preciseness of the alum dosage.

Both coagulants have high turbidity removal values ($\geq 60\%$). The optimal dosage of *M. oleifera* was 70 mg/L, obtaining a reduction of turbidity of 63.70%. The mechanism of coagulation with the seeds of *M. oleifera* consists of absorption and neutralization of the colloidal positive charges that attract the negatively charged impurities in water (29). *M. oleifera* reduced the turbidity of abattoir wastewater from 218.4 NTU to 68 NTU representing a 68.86% reduction (30).

It was found that *M. oleifera* seed extract at 10–30 mg/L and pH of 6–8 removed more than 90% of turbidity (31). Also, another study revealed that the removal of turbidity was 50%–87.5% and 12%–62.5% for chitosan and coagulant protein of *M. oleifera* respectively (20), which was almost equal to the result obtained in the experiments performed on refinery wastewater (a 42% reduction in turbidity by alum). In addition, in other researches at optimum concentrations, alum, poly aluminum chloride (PAC), and *M. oleifera* removed turbidity by 99.5%, 99%, and 99.7% respectively (19). In these studies, it was found that natural biopolymer *M. oleifera* could be used to remove coagulum and turbidity. Also, similar results from highly contaminated waters were reported (31).

In the research that was performed at the optimum dose, pH value, and temperature, *M. oleifera* was used as a natural coagulant on synthetic samples containing turbidity of 40–200 NTU. It was found that the optimum *M. oleifera* dosage was 30–55 mg/L at pH value of 6.5, and temperatures below 15°C (21). It was reported that overall COD removal was 50% at both 50 and 100 mg/L of *M. oleifera* dosage. When 50 and 100mg/L seed dosages were applied in combination with 10 mg/L of alum, COD removal increased to 58 and 64% respectively (14).

These results are compatible with those found in the study carried out with samples taken from the wastewater unit of Bandar Abbas Refinery (where the optimum dosage, pH, and temperature were found to be 70 mg/L, 6, and 20°C respectively). Muthuraman and Sasikala showed that among 3 natural coagulants (*M. oleifera*, *Strychnos potatorum* and *Proteus vulgaris*), *M. oleifera* seed extracts

had the highest performance in turbidity removal (99%) at pH 7 using 250 mg/L *M. oleifera* (7). Javid et al showed that only ferric chloride and aluminum sulphate among the chemical coagulants (aluminum sulphate, ferric chloride, PAC and lime) were highly efficient in the removal of turbidity. Turbidity removal using aluminum sulfate within the pH range 4.5–5 was 99.33% (turbidity reduced from 166 to 1.1 NTU). In the process of ozonation to wastewater of chemical treatment with aluminum sulphate within the above range of time, COD was reduced from 22500 mg/L to 4800 mg/L (6). De Paula et al showed that with a combination of coagulant $Al_2(SO_4)_3$ and *M. oleifera* more than 90% of turbidity was removed, and a ratio of 20:80 (w/w) was obtained for *M. oleifera* and $Al_2(SO_4)_3$ and their results are in agreement with those of the present research (32). Abedini and Alipour indicated that under optimized absorption conditions (pH: 5, sorbent: 1.5 g, temperature: 45°C, Cd concentration: 2 ppm and exposure time: 180 minutes), the removal rate of Cd was 80% by using *M. oleifera* seed powder (33).

Muralimohan et al showed that the 50:50 blended coagulant *M. oleifera* and alum has a higher removal efficiency with respect to turbidity, TSS, TDS and COD; while the result of this study was showed in the ratio of 1:2 alum and *M. oleifera* the removal efficiency was optimal (34).

Although alum is considerably more efficient than *M. oleifera* in removing these parameters (COD, turbidity, and TSS reduced by 51.72%, 92.16%, and 85.26% respectively), the economic and environmental aspects of its use must be considered. Therefore, replacing alum (the material used in the wastewater treatment unit at the refinery in Bandar Abbas) with a natural biopolymer such as *M. oleifera*, or utilizing these two together, can be useful and effective. Moreover, the use of natural coagulants can substantially save money on the cost of chemical materials and reduce to a minimum the sludge produced and, hence, require less disposal. Some natural compounds have coagulant properties, and learning about these properties and understanding their exact mechanisms of action can lead to their correct application.

Conclusion

Considering the fact that *M. oleifera* coagulum can be locally produced, its use in water purification should be encouraged. This is likely to reduce the high cost of the current water treatment processes. It is recommended that a combination of alum and *M. oleifera* in different proportions be investigated to establish their effectiveness in treating wastewater. From the economic point of view, since *M. oleifera* trees grow naturally in the deprived region of Beshagard in Hormozgan province, the promotion of its cultivation can help the economy of the local communities, in addition to expanding the plant cover in the region. This valuable tree is found in about one million hectares of the lands in southern Iranian provinces (Sistan and Baluchestan, Hormozgan, and Fars provinces) and, with planning, one million hectares of grade 3 rangelands in these provinces can be annually turned into valuable

land by planting *Moringa* trees. In this regard, industrial units such as refineries and petrochemical complexes are required to abide by regulations related to the protection of the environment, while the use of a natural biopolymer could substantially help to protect the environment as these biopolymers are biodegradable.

Suggestions

1. Natural biopolymers such as *M. oleifera* should be used, singly or in combination with another suitable coagulant, in other wastewater treatment units in the petroleum, gas, and petrochemical industries, and in urban wastewater treatment.
2. Other natural biopolymers, including chitosan, tannin, starches, etc., should be utilized as coagulants in the petroleum, gas, and petrochemical industries and to treat urban wastewater.
3. Research should be conducted on the optimum use of sludge resulting from the utilization of biopolymers.
4. Various biopolymers should be used as coagulants and coagulant aids in the Iranian water and wastewater treatment industries.

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Ethical issues

It is confirmed that this manuscript is the original work of the authors. It has not been published, nor is it under review by another journal, and it is not being submitted for publication elsewhere.

Competing interests

We affirm that this article is the original work of the authors and they have no conflict of interest to declare.

Authors' contribution

All authors were involved in all stages of the article. On behalf of the co-authors, the corresponding author bears full responsibility for this submission.

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