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Effectiveness of salt-extracted freeze-dried *Moringa oleifera* as a coagulant

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ABSTRACT

The effectiveness of freeze-dried *Moringa oleifera* (*MO*) seeds powder in the coagulation of synthetic wastewater was studied. Three modes of extraction were introduced, namely distilled water-extracted freeze-dried *M. oleifera* (FD-DW *MO*), salt-extracted freeze-dried *M. oleifera* (FD-KCl *MO*), and distilled water-extracted freeze-dried *M. oleifera* with salt addition (FD-DW *MO* + KCl). Jar test was used to evaluate the effectiveness of the extracted freeze-dried powder, in the coagulation of a synthetic municipal wastewater. The results show that FD-DW *MO* at an optimal dosage of 132 mg/l, increases the turbidity removal efficiency to nearly 93% as compared to distilled water-extracted *MO* which gave 76% removal at an optimal dosage of 300 mg/l. However, the comparison of FD-KCl *MO* with non-FD-KCl *MO* show that the freeze-drying neither improves the efficiency of salt-extracted *M. oleifera* nor the optimal dosage. On the other hand, adding KCl to the FD-DW *MO* in treating high turbidity synthetic wastewater at 200 ± 5 NTU improved the optimal dosage from 40 mg/l (without adding KCl) to 10 mg/l corresponding to 91% removal efficiency. As a conclusion, the freeze-drying process did not improve the coagulation efficiency with salt-extracted *MO* using KCL. But using the salt (KCL) solution for dilution of FD-DW *MO* causes the optimal dosage to decrease from 40 mg/l (using distilled water for dilution) to 10 mg/l. This is associated with a slight decrease in the efficiency from 92.73 to 91.32%.

Keywords: Moringa oleifera; Extraction; Freeze-drying; Coagulation; Turbidity

1. Introduction

Water quality management to protect public health and minimize the cost of water supply becomes a concern to overcome the decline in the quality of drinking water sources [1]. Surface water, which is a source of water supply, generally contains suspended and colloidal solids from land erosion, decaying vegetation, micro-organisms, and color-producing

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compounds [2]. Hence, the removal of these organic and inorganic materials from raw water is essential before it can be disinfected for human consumption.

In water treatment works, this purification stage is normally achieved by the application of chemical coagulants which change the water from a liquid to semi-solid state. This is usually followed by flocculation, the process of gentle and continuous stirring of coagulated water, which encourages the formation of "flocs" through the aggregation of the minute particles present in the water. Flocs can be easily removed by settling or filtration.

In some developing countries and due to high cost and low availability, the application of chemical coagulants in water and wastewater treatments is not affordable [3]. In addition, recent studies have pointed out several serious drawbacks of using aluminum salt such as Alzheimer's disease and similar health-related problems associated with residual aluminum in treated water [1]. Beside that, production of large sludge volumes and the reaction of alum with natural alkalinity of water result in pH reduction and low coagulation efficiency of cold waters are the main shortcomings of aluminum salt [4].

Ferric salts and synthetic polymers have also been used as coagulants but with limited success because of the same disadvantages found in using aluminum salts [5]. Therefore, it is desirable that other cost-effective and more environmentally acceptable alternative coagulants to be developed in order to supplement or replace alum, ferric salt, and synthetic polymers. In this context, natural coagulants present a viable alternative because they are biodegradable and are presumed safe for human health.

A water soluble extract of the dry seeds of *Moringa oleifera* (*MO*) is one of these natural coagulants [4]. Numerous laboratory studies have so far shown that *MO* seeds possess effective coagulation properties [5,6] and they are not toxic to humans or animals. As a coagulant, the crude water extract of *MO* seeds compares favorable with alum, and its use as an effective coagulant, which is recommended for use in developing countries to reduce the exorbitant cost of water treatment [7].

However, recent studies found that the highest turbidity removal for surface water was obtained when using *MO* which was kept for three days or less and its efficiency decreased when using *MO* stock solution stored for longer period [8].

In recent years, there has been an increasing amount of literature on improving extraction of active ingredient in *MO* seeds powder. Some of these studies carried out using different solutions for extraction similar to the work done by Okuda et al. [9,10] and Ghebremichael et al. [11] or in the extraction techniques conducted by Mohammad et al. [12] and Noor et al. [13].

The main objective of this study is to evaluate the effectiveness of 1.0 mol/l salt-extracted freeze-dried *MO* extract in coagulation of synthetic wastewater. *MO* seeds powder will be extracted with distilled water and salt solution. The filtrate or extract will be used for freeze-drying process and act as a coagulant. Three extraction modes were used in this study, namely distilled water-extracted freeze-dried *MO* (FD DW *MO*), salt-extracted freeze-dried *MO* (FD KCl *MO*), and distilled water-extracted freeze-dried *MO* used with salt addition (FD DW*MO* + KCl). Synthetic wastewater prepared from the kaolin was treated with the three above-studied extraction modes, and their effectiveness in turbidity removal were compared. Parameters, such as the residual turbidity of the treated sample and the dosage required, were used to evaluate the efficiency of each extraction method.

2. Materials and methods

2.1. *Collection of* MO *seeds*

In this study, the *MO* seeds, which were used as natural coagulant, were collected from University Putra Malaysia farm. Only dried *MO* seeds or dry pods in brownish color were collected. Good quality seeds of *MO*, which were not rotten or infected with diseases, were used. The seeds were dried in the oven for 24 h at 50˚C. The winged seed cover was shelled and the kernel was ground to fine powder with a domestic food blender.

2.2. *Preparation of saturated* MO *stock solution*

In order to compare the effectiveness of freeze-dried and non-freeze-dried *MO* extracted with different solutions (distilled water and potassium chloride (KCl) salt solution), two saturated *MO* stock solutions were prepared. Basically, the method for the preparation was almost the same where 20% solid content of stock solution or 200,000 mg/l stock solution is prepared with different solutions extraction.

2.2.1. *Distilled water extraction*

Fifteen grams of *MO* fine powder were added to 50 ml distilled water in order to prepare a saturated stock solution. The mixture was blended for two minutes to allow the extraction of the active ingredient. The suspension was then filtered through muslin cloths and the filtrate was made up to 75 ml to get a saturated stock solution. The saturated *MO* stock solution was then used for freeze-drying process and to run the non-freeze-dried jar test after 20 times of dilution.

2.2.2. *Potassium chloride (KCl) solution extraction*

Fifteen grams of *MO* fine powder were added to 50 ml of 1.0 mol/l KCl salt solution to prepare a saturated stock solution. The mixture was blended for two minutes to allow the extraction of the active ingredient. The suspension was then filtered through muslin cloths and the filtrate was made up to 75 ml to give a saturated stock solution. The saturated *MO* stock solution was then used for freeze-drying process and to run the non-freeze-dried jar test after 20 times of dilution.

2.3. *Preparation of* MO *stock solution*

2.3.1. *Non-freeze-dried*

A 10 ml of the saturated *MO* stock solution was measured using a pipette and placed in a beaker contain 190 ml of distilled water or 1.0 mol/l potassium chloride, KCl salt solution to give a stock solution of 10,000 mg/l of distilled water or salt extraction. This dilution of 10,000 mg/l stock solution was then used as coagulant. Different series of concentrations were prepared from the stock solution by adding certain volume of distilled water into measured stock solution. Eq. (1) was used to obtain different concentrations of stock solution.

$$C_1 \times V_1 = C_0 \times V_0 \qquad (1)$$

where C_1 = required concentration, C_0 = stock solution concentration, V_1 = required volume, and V_0 = stock solution volume

The measured stock solution was used for jar test trials which were conducted to determine optimum dosage of *MO* on high strength synthetic wastewater.

2.3.2. *Freeze-dried*

There were three modes of extraction prepared from the freeze-dried *M. oleifera*. The preparation of stock solution is as below:

(a) Distilled water extracted and dilute with distilled water (FD DW *MO*).

A 500 mg of distilled water-extracted freeze-dried *MO* extract was placed in a beaker containing 50 ml of distilled water in order to get a stock solution of 10,000 mg/l. The stock solution was used in the coagulation process. Eq. (1) was used to obtain the desired concentration of the coagulant to run the jar test and to determine optimum dosage of *MO* for high-strength synthetic wastewater.

(b) Distilled water extracted and diluted with 1.0 mol/l potassium chloride, KCl, (FD DW *MO* + KCl).

A 500 mg of distilled water-extracted freeze-dried *MO* extract was placed in a beaker containing 50 ml of 1.0 mol/l KCl salt solution in order to get a stock solution of 10,000 mg/l. This stock solution was used in the coagulation process. Eq. (1) was used to obtain the desired concentration of the coagulant and to run the jar test to determine optimum dosage of *MO* for high-strength synthetic wastewater.

(c) 1.0 mol/l potassium chloride, KCl extracted and diluted with distilled water, (FD KCl *MO* + DW).

A 500 mg of 1.0 mol/l KCl salt solution-extracted freeze-dried *MO* extract was placed in a beaker that contains 50 ml of distilled water in order to get a stock solution of 10,000 mg/l which was used in the coagulation process. Eq. (1) was used to obtain the desired concentration of the coagulant to run the jar test to determine optimum dosage of *MO* for high-strength synthetic wastewater.

2.4. *Preparation of freeze-dried* MO *extract*

As mentioned earlier, two different solutions for extractions were used in the freeze-drying process, so two different types of freeze-dried *MO* extract were produced, namely freeze-dried *MO* extracted with distilled water and freeze-dried *MO* extracted with 1.0 M potassium chloride, KCl salt solution.

The same preparation approach was used to produce two different extraction freeze-dried *MO* extract. Generally, freeze-drying process involves two stages which are overnight prefreeze and freeze-drying. Two milliliters of the prepared saturated *MO* stock solution were measured using a pipette and placed in a COD vial. The number of vials to be sent for freeze-drying process depends on the extracted volume. Normally, around 49 ml filtrate was obtained from the extraction and about 24 vials were prepared. The vials with cap are then sent for frozen overnight (which is also known as "prefreeze" at −35℃ in a freezer). Then the vials without cap were sent for freeze-drying in a Benchtop manifold freeze-dryer. Manifold freeze-dryers are usually used when drying a large amount of small containers and the product will be used in a short period of time. The samples were freeze-dried for 24 h at temperature between −50 and −80℃, and the pressure applied is around 100 mbar but the freeze-dryer was operated at ambience temperature.

2.5. Storage of freeze-dried and non-freeze-dried MO

The freeze-dried and non-freeze-dried *MO* seed powders were stored in the refrigerator at 4˚C after placing them into several small containers, sealed to keep them airtight and wrapped with aluminum foil. The purpose of dividing the powder into several containers was to minimize the risk of the powder being spoiled. Sealing and wrapping of the containers with aluminum foil was to minimize the affect of air, humidity, and sunlight on the the powder.

2.6. Preparation of water sample

Experiments were carried out on synthetic wastewater with turbidity of approximately 200 NTU and which was prepared by adding (Laguna Clay, USA) into distilled water. Ten grams of kaolin were added to 1 l of distilled water. The suspension was stirred slowly at 20 rpm for one hour in a jar test for uniform dispersion of kaolin particle. The suspension was then left for 24 h to allow complete kaolin hydration. The kaolin suspension was used as stock solution for the preparation of samples used for coagulation test.

2.7. Coagulation test

Experiments were carried out to optimize three different modes of extraction for freeze-dried *MO* extract, which intend to be used for coagulation of high-strength synthetic wastewater.

Coagulation efficiency for freeze-dried and non-freeze-dried *MO* was verified by using jar test which consists of rapid mixing, slow mixing, and sedimentation process. In this study, Stuart scientific flocculator SW1 which enables six beakers to be agitated simultaneously was used for coagulation activity. In each run, six beakers were filled with 500 ml of water sample and agitated at pre-selected rapid mixing intensity as shown in Table 1. During rapid mixing, coagulant dosage was added into each beaker simultaneously. After rapid mixing, the pre-selected intensity of slow mixing was quickly established and subsequently the beakers were left for the sedimentation phase to take place. In this study, the operating parameters, such as rapid mixing intensity (rpm) and duration, slow mixing intensity (rpm), and duration and settling time, were fixed as shown in Table 1.

Table 1
Operating parameters used to run the jar test [8]

Mixing type	Speed (rpm)	Time (min)
Rapid mix	100	4
Slow mix	40	25

After settling, 20 ml of the sample was taken from the middle of each beaker using a pipette and then placed in a turbidity vial for measurement using turbidimeter (HACH, model 2100P). For determination of optimum dosage, different amounts of *MO* stock solutions were added into the beakers and the amount that gave the lowest turbidity was the optimum dosage for that particular sample.

3. Results and discussion

3.1. Physical characteristic of freeze-dried extract

Table 2 shows the yield and physical characteristic of the freeze-dried *MO* extract for distilled water extracted and potassium chloride, KCl salt solution extracted. From the observation, it is found that with the same amount of raw *MO* seed powder (15 g) used for freeze-drying process, the yield production of freeze-dried extract was different for each type of extraction. Table 2 shows that the yield production of the freeze-dried extract for salt solution extraction was 26.6% higher compared to the distilled water extraction. The collected freeze-dried extracts were about 5.76 and 4.55 g for salt solution extraction and distilled water extraction, respectively. The result is in agreement with the finding of Okuda et al. [9] which stated that salt addition improves extraction efficiency of the active component *MO* seeds powder. The improvement is due to the loosening up of protein associations leading to more soluble solution.

On the other hand, the physical observation of the extract textures also differs between these two extraction solutions. It is found that the freeze-dried extract from distilled water extraction looks fine and soft, while the freeze-dried extract from salt solution extraction was dry, coarse, and rough.

The moisture contents for salt extraction and distilled water extraction were recorded as 6.84% and 7.32%, respectively, which are found in agreement with the observation of the freeze-dried extract.

3.2. Coagulation activity of raw MO (non-freeze-dried)

The coagulation activity of non-freeze-dried *MO* seeds powder using different concentrations and two different solutions for extraction (distilled water and KCl salt solution) are shown in Figs. 1 and 2.

Fig. 1 shows the optimum dosages of non-freeze-dried distilled water-extracted *MO* were 300 mg/l for 10,000 mg/l stock solution and 30 mg/l for 200,000 mg/l stock solution with removal efficiency of 76.04% and

Table 2
Physical characteristic of freeze-dried *MO* seed powder

Parameters	Distilled water extraction	1.0 mol/l KCl salt solution extraction
Extraction time, min	2	2
Freeze-drying duration, h	24	24
Raw *MO* powder weight, g	15	15
After freeze-dried		
FD powder, g	4.5559	9.2992
Impurities	0.006	3.5352
FD *MO* powder, g	4.5499	5.7640
Therefore		
1 g FD *MO* powder =	3.2968 g Raw *MO* powder	2.6024 g Raw *MO* powder
1 g raw *MO* powder =	0.3033 g FD *MO* powder	0.3843 g FD *MO* powder
Moisture content	7.32	6.84
Extract texture	Fine and soft	Dry and coarse

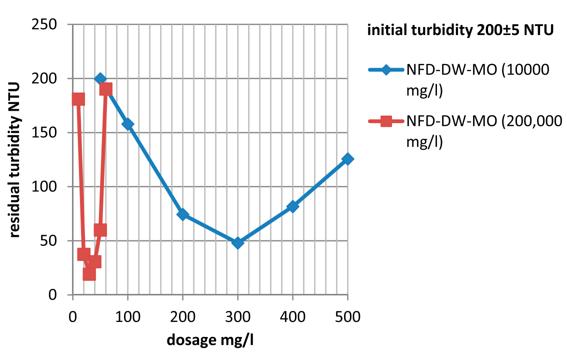

Fig. 1. Coagulation activity of non-freeze-dried distilled water-extracted *M. oleifera* seeds.

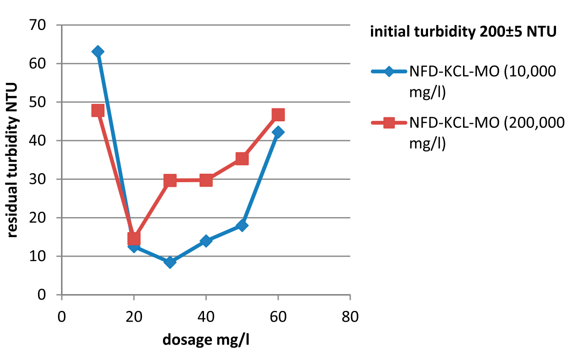

Fig. 2. Coagulation activity of non-freeze-dried KCl-extracted *M. oleifera* seeds.

90.42%, respectively. The optimum dosage was found in agreement with the finding of Katayon et al. [8] which reported that the optimum dosage of *MO* for water sample with initial turbidity of 100–200 NTU is 300 mg/l. However, the comparison of the optimum dosage and removal efficiency of different stock solution concentrations show that if the concentration is high, the optimum dosage is low, and the removal efficiency is higher. This is because, for high concentration, there is a high active ingredient in the extract.

Fig. 2 shows that the optimum dosages of non-freeze-dried KCl-extracted *MO* on turbidity removal prepared from 10,000 and 200,000 mg/l stock solution were 30 mg/l and 20 mg/l, respectively. These optimal dosages were associated with 95.78 and 92.72% removal efficiencies.

Figs. 1 and 2 show that the coagulation activity using different extraction solutions gave the superiority to the salt solution compared to distilled water. This is in agreement with findings of Okuda et al. [9,10] and Mohammed et al. [12] which showed that using salt as an extract solution will enhance the extraction of active ingredients in *MO* seeds powder.

3.3. *Coagulating high-strength synthetic wastewater using freeze-dried and non-freeze-dried* MO

The coagulation activity of freeze-dried *MO* using different extraction solution was examined and the results are shown in Figs. 3 and 4.

Fig. 3 shows the coagulation activity of freeze-dried and non-freeze-dried distilled water-extracted *MO* seeds. The results revealed that freeze-dried distilled water-extracted *MO* seeds achieve better coagulation activity compared to the non-freeze-dried distilled water-extracted *MO* seeds with an optimum

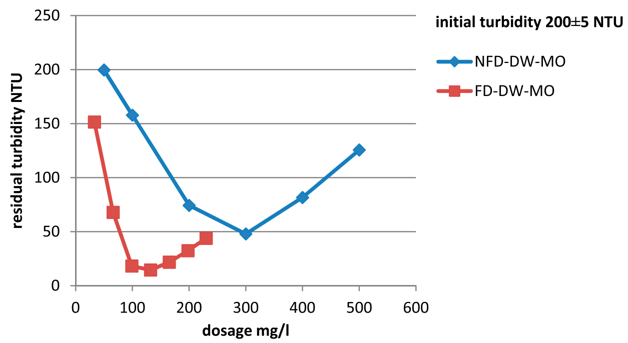

Fig. 3. Coagulation activity of freeze-dried and non-freeze-dried distilled water-extracted *M. oleifera* seeds.

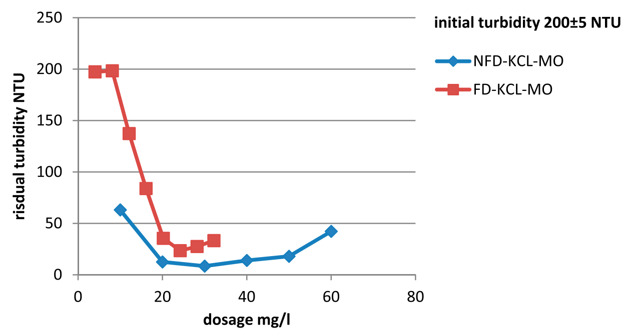

Fig. 4. Coagulation activity of freeze-dried and non-freeze-dried KCl-extracted *M. oleifera* seeds.

dosage of 132 and 300 mg/l and removal efficiencies of 93% and 76%, respectively.

Fig. 4 shows the coagulation activity of freeze-dried and non-freeze-dried KCl-extracted *MO* seeds. It shows that freeze-drying process does not enhance the coagulation activity of the extracted *MO* seeds as the optimum dosage slightly decrease from 30 to 24.2 mg/l, and the removal efficiency decreased from 95.78 to 88.27% for non-freeze-dried KCl-extracted *MO* seeds and freeze-dried KCl-extracted *MO* seeds, respectively.

3.4. Coagulating high-strength synthetic wastewater using freeze-dried distilled water-extracted MO *seeds powder diluted by different solutions*

Fig. 5 shows the results of coagulation activity of freeze-dried distilled water-extracted *MO* seeds powder diluted by different solutions namely distilled water and KCL solution. The results indicate that, using the salt solution in dilution, the freeze-dried powder affects the coagulation in terms of achieved optimal dosage. Fig. 5 shows that the optimal dosage was 40 mg/l for that diluted with distilled water compared with 10 mg/l for that diluted with KCL

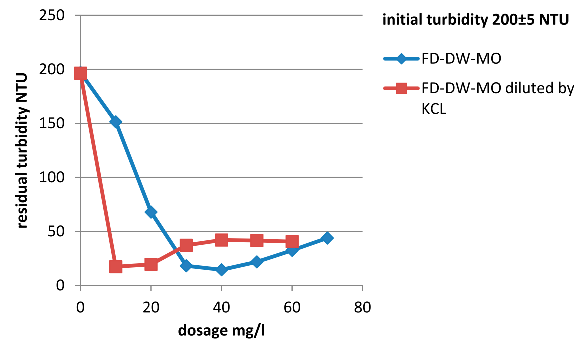

Fig. 5. Coagulation activity of freeze-dried distilled water-extracted *M. oleifera* seeds powder diluted by different solutions.

solution. However, no significant differences were found in terms of achieved removal efficiencies which were 92.73% and 91.32%, respectively. These results suggest that using the salt solution in dilution enhances the mechanisms of coagulation since it is helping in bridging [9,10,12,13].

4. Conclusions

In this investigation, the aim was to assess the coagulation activity of salt-extracted freeze-dried *MO* in treating synthetic wastewater. This study has shown that the freeze-drying process enhance the coagulation activity of *MO* in terms of achieved optimal dosage compared to non-freeze-dried *MO*. However, the removal efficiency significantly improved for distilled water-extracted freeze-dried MO compared with the removal efficiency of salt-extracted freeze-dried *MO* which was decreased by 7.5%.

Using salt solution to dilute the distilled water-extracted freeze-dried *MO* is significantly lower the optimal dosage by 75% and gave almost the same removal efficiency.

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