Desalination and Water Treatment
www.deswater.com
51 (2013) 1941–1946
February
1944-3994/1944-3986 © 2013 Desalination Publications. All rights reserved
doi: 10.1080/19443994.2012.715435
Taylor & Francis
Taylor & Francis Group

Coagulation activity of spray dried salt extracted *Moringa oleifera*

Thamer Ahmad Mohammad[a,*], Ezzuldin Hasan Mohamed[a], Megat Johari Megat Mohd Noor[b], Abdul Halim Ghazali[a]

[a]*Faculty of Engineering, Department of Civil Engineering, Universiti Putra Malaysia, 43400 UPM, Serdang, Selangor, Malaysia*
Tel. +60 3 89466352; Fax: +60 3 86567129; email: thamer@eng.upm.edu.my
[b]*Malaysia Japan International Institute of Technology, Universiti Tecknologi Malaysia, Malaysia*

Received 8 February 2012; Accepted 18 July 2012

ABSTRACT

A study on the effectiveness of salt extracted *Moringa oleifera* seeds as a coagulant for turbidity removal in water treatment process under different operational parameters (configurations) of spray drying, storage, and packaging conditions is presented. The operational parameters were inlet temperature, outlet temperature, and pumping rates. The coagulation activity was studied for the spray dried salt extracted *M. oleifera* which is stored under two different conditions, room temperature (29˚C) and refrigerator (3˚C), and two different packaging forms, closed container and vacuum packed. Eight configurations for operational parameters were used in this study. The results show that for inlet temperature below 115˚C the spray drying process failed to produce dry powder. For the successful configurations, there was no significant difference in coagulation activity between different configurations stored at the same storage and packaging conditions. However, the comparison of the coagulation activity of the spray dried *M. oleifera* with the same packaging conditions (closed container or vacuum packing) under different storage conditions (room temperature 29˚C or refrigerator 3˚C) revealed that there was a significant difference between them; the coagulation activity of spray dried *M. oleifera* stored at room temperature was significantly better than refrigerator during the study duration. On the other hand, the packaging condition (closed container or vacuum packing) does not affect the coagulation activity of spray dried *M. oleifera* stored at the same temperature.

Keywords: Moringa oleifera; Coagulation; Extraction; Spray drying; Storage

1. Introduction

Finding safe drinking water in many countries becomes a national problem. Sustainable water supply and treatment systems with low cost, robust, and minimal maintenance and operation skills are needed. One of the important water treatment processes is the turbidity removal, which could be achieved by using various types of coagulants [1]. Several coagulants are widely used in conventional water treatment processes and can be classified into inorganic, synthetic organic polymer, and natural coagulants [2]. Alum (aluminum sulfate) is the most widely used in coagulation [3]. However, recent studies have indicated various serious drawbacks of using aluminum salts, such causing

*Corresponding author.

Presented at the International Conference on Desalination for the Environment, Clean Water and Energy, European Desalination Society, 23–26 April 2012, Barcelona, Spain

Alzheimer's disease and resulting in large sludge volumes [4]. Several studies were carried out to introduce the natural coagulants produced or extracted from animals and micro-organisms in order to overcome the problems associated with the use of chemical coagulants [5–7]. Recently, there is an interest in using natural coagulants even though they had been used before the synthetic coagulants were used in water treatment [8–10]. *Moringa oleifera* seeds can be used as a natural coagulant to replace the synthetic coagulants in water treatments [3,4,11–15]. In recent years, there has been an increasing interest in enchaining extraction of the active agent in *M. oleifera* seeds as a natural coagulant. For example, Ndabigengesere et al. [14] carried out a study on *M. oleifera* seed powder to determine the active agent in the seed and the mechanism of coagulation. The study shows that the active agents are dimeric cationic proteins of molecular weight of 13 kDa isoelectric points range between 10 and 11. The adsorption and neutralization of the colloidal charges were reported to be the mechanism of the coagulation.

Okuda et al. [2,3,15] carried out a number of experiments on developing the extraction method of active components from *M. oleifera* seeds by using salt solutions rather than water. The study reported that the salt extracted *M. oleifera* seeds coagulant showed better coagulation activity with dosages 7.4 times lower than that resulted from the distilled water extracted *M. oleifera*. On the other hand, the purification and isolation of the active component from an aqueous salt extraction was not a protein, polysaccharide, or lipid, but an organic polyelectrolyte with molecular weight of about 3.0 kDa. This implies that the water and salt extract may be of different nature. The coagulation efficiency of *M. oleifera* decreases with an increase in storage duration [1]. On the other hand, other studies have also shown that the extracting active component using a salt solution to increase the coagulation efficiency [3,15].

In this study, the spray drying technique was used to extract the effective coagulant in the *M. oleifera* seed salt solution; the effects of storage condition and duration on the coagulation activity of spray dried salt extracted *M. oleifera* seeds were investigated.

2. Materials and methods

2.1. Collection of M. oleifera

M. oleifera dry pods were collected from the trees located at the campus of University Putra Malaysia. Good quality pods with good quality seeds were selected and used for *M. oleifera* seed extraction. After every collection, the wing seeds were dried in the oven for 24 h at 50˚C then kept in desiccators until enough amount of *M. oleifera* seeds were collected.

2.2. Preparation of synthetic turbid water

In all coagulation experiments, samples of turbid water were prepared by adding kaolin (R&M Chemicals) into tap water [16]. Kaolin of 10 g was added to one liter of tap water. This suspension was stirred using stirrer (R1333, IKA WORKS Malaysia) and a mixer (IKA RW 20 n S2, IKA WORKS, Malaysia) at 120 rpm for 1 h for uniform dispersion of kaolin particle and stood for 24 h to allow for complete hydration of particles. This suspension was used as stock solution. The initial turbidity used in all experiments was 200 ± 5 NTU.

2.3. Preparation of spray dried M. oleifera seed extracts

The winged seed cover was shelled just before the extraction. The kernel was grounded to a fine powder by using domestic blender model National Deluxe Family Kitchen Model (MJ-C85N, Japan). The fine powder was then packed into closed containers and kept in the refrigerator until the required amount of fine powder was collected. *M. oleifera* seed fine powder of 210 g was suspended in 700 ml of 1 M (NaCl) solution and the suspension was stirred using the blender for 5 min to extract the active component. The suspension was then filtered using muslin cloth and the filtrate was referred as stock solution for spray drying process.

2.4. Preparation of raw M. oleifera seed distilled water extracts and salt solution extracts

Two *M. oleifera* seed extracts were prepared, one by using distilled water as an extract agent and the other by using 1 M (NaCl) solution. The winged seed cover was shelled just before the extraction. The kernel was ground to a fine powder by using domestic blender model National Deluxe Family Kitchen (MJ-C85N, Japan). Fine powder (2.5 g) was placed in the blender added with 250 ml of distilled water to prepare a stock solution of 10,000 mg/l in concentration for the *M. oleifera* distilled water extract. The same amount of the fine *M. oleifera* seed powder was used with 250 ml of 1 M (NaCl) solution to prepare 10,000 mg/l stock solution for the *M. oleifera* salt extract. The blender was operated for 1 min to allow the extraction of active components and the suspension was then filtered through muslin cloth and the filtrate was referred as the stock solution of 10,000 mg/l [1]. Different volumes

were taken from the solution by using the equation below to obtain the required concentration (dosages).

$$C_1 * V_1 = C_0 * V_0 \quad (1)$$

where C_0 is the stock solution concentration, V_0 is the stock solution volume, C_1 is the required concentration, and V_1 is the required volume.

The different dosages were calculated and applied to the coagulation experiments (jar test) for optimizing of raw *M. oleifera* seed extract.

2.5. Packaging and storing of spray dried salt extracted M. oleifera seeds powders

After spray drying process, the resulted powder was packed into closed container and stored in the refrigerator at 3˚C. After running the first coagulation test as week 0 for the spray dried *M. oleifera*, the resulted powder was packed into two different packaging forms (closed container and vacuum packed), and then stored in two different storage conditions (room temperature (29˚C) and refrigerator at 3˚C).

All packaged forms were covered with aluminum foil. The vacuum packaging apparatus (MASTER-PACK, NEW DIAMONED VAC, USA) was used for packaging. The coagulation activity tests were done under all packaging and storing conditions mentioned above every week starting from week 0 up to week 6.

2.6. Coagulation experiment

Jar tester (BIBBY Stuart Scientific, UK) was used to carry out the coagulation test. The jar test consist of three stages: rapid mixing, slow mixing, and settling. Synthetic turbid water (300 ml) was filled in a 500 ml beaker placed on the slot in the jar tester. During the rapid mixing, the coagulant dosages were added into the beaker simultaneously by turning the dosing tubes which fixed in clips that made for this purpose. The time was controlled by stopwatch. After slow mixing, the beakers were carefully removed from the jar tester slots and placed in a safe place for particles settlings. Jar test was repeated three times for each sample under different storage and packaging conditions and the average was taken as a result. In this study, rapid and slow mixing speeds and time were fixed as recommended by Katayon et al. [17]. Table 1 shows the mixing speed and time.

After a settling time of 30 min, the samples were taken from the beakers. The samples were collected at a depth of 1 cm from each backer's surface using a pipette. The turbidity of the samples was measured by using Turbidimeter (HACH, Model 2100AN, USA).

Table 1
Operating variables used for jar test

Rapid mix velocity (rpm)	100
Rapid mix duration (min)	4
Slow mix velocity (rpm)	40
Slow mix duration (min)	25
Settling time (min)	30

2.7. Optimizing of raw M. oleifera

Jar test was used for optimizing the coagulation activity of the two different *M. oleifera* seed extracts (distilled water extract and salt extract). The 300 ml of synthetic turbid water of initial turbidity 200 ± 5 NTU was filled into 500 ml beaker then placed on the slot in a jar tester. The jar test operation parameters are shown in Table 1. The dosages used for the first trail were 50, 100, 200, 300, 400, and 500 mg/l, while the dosages for the second trail were 20, 30, 40, 60, 80, and 100 mg/l.

2.8. Coagulation activity of spray dried salt extracted M. oleifera

Jar test was conducted using spray dried salt extracted *M. oleifera* as a natural coagulant. An analytical balance (model PRECISA 2100 AN, Swiss) was used to get 0.3 g of the spray dried salt extracted *M. oleifera* to prepare the stock solution for coagulation test. A 200 ml distilled water was used to get stock solution with concentration of 1,500 mg/l. Six beakers of 500 ml in volume were filled with 300 ml synthetic turbid water of initial turbidity 200 ± 5 NTU placed on the slot in a jar tester and during rapid mixing, the coagulant dosage was added into each beaker simultaneously, the dosages of 8, 12, 16, 24, 32, and 40 mg/l were applied.

2.9. Statistical analysis

One way ANOVA test with Tukey's HSD test were executed to confirm the significance of variance between the means of residual turbidity of spray dried *M. oleifera* in different conditions by using SPSS statistical software (version 16) were used for this purpose. Statistical analysis shows that the confidence interval was 95% ($P < 0.05$).

3. Results and discussion

The coagulation activity of different extraction methods used is shown below by comparing the results of the optimizations of the raw *M. oleifera* distilled water extracted with *M. oleifera* salt extracted and spray dried salt extracted *M. oleifera* with salt extracted *M. oleifera*.

3.1. Optimizing of raw M. oleifera distilled water extracted/salt extracted

Results of optimization for both raw *M. oleifera* distilled water extracted and salt extracted are shown in Fig. 1. The optimal dosage was found to be 150 mg/l for water extracted *M. oleifera* with 90.7% removal efficiency achieved. While a 30 mg/l corresponds to 95.3% removal efficiency for salt extracted *M. oleifera*. This is in agreement with the findings of Muyibi and Evison [18] and Okuda et al. [3]. The salt ions increased the solubility of the active component in *M. oleifera* seed powder [15]. Dosage increment does not improve the turbidity removal. The large dose will eventually lead to the saturation of polymer bridge sites and cause restabilization of particles due to an insufficient number of particles needed to form inter-particle bridges [18].

3.2. Coagulation performance of spray dried salt extracted M. oleifera and salt extracted M. oleifera

To compare the effect of spray drying process on the coagulation activity of *M. oleifera* seed salt extracted; Fig. 2 shows the residual turbidity achieved of spray dried salt extracted *M. oleifera* and salt extracted *M. oleifera* seeds. The optimization results show that the residual turbidity achieved by spray dried salt extracted *M. oleifera* was 17.89 NTU (91.05% removal efficiency) with optimal dosage 16 mg/l. On the other hand, the salt extracted *M. oleifera* gave lower residual turbidity 9.4 NTU (95.3% removal efficiency) with optimal dosage 30 mg/l. The use of salt in extraction enhances the solubility of active gradi-

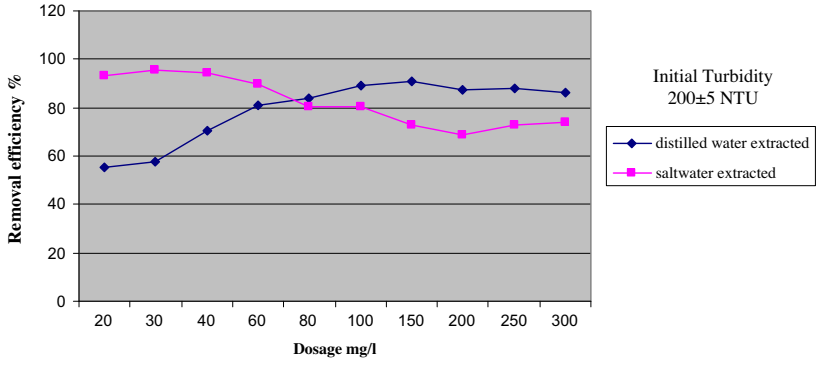

Fig. 1. Average residual turbidity of raw *Moringa oleifera* distilled water extracted/salt extracted.

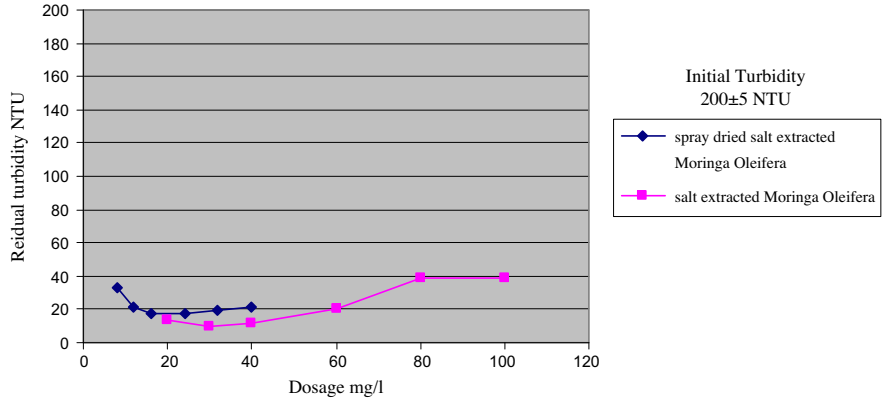

Fig. 2. Spray dried salt extracted *Moringa oleifera* and salt extracted *Moringa oleifera*.

ents in the *M. oleifera* seed powder (as a result of salting-in mechanism) as reported by Okuda et al. [15]. The results indicate that the spray drying process affects some of the active gradients so that the removal efficiency of spray dried salt extracted *M. oleifera* was higher than salt extracted *M. oleifera* (Fig. 2). However, in term of optimal dosage amount, it is clear that spray dried salt extracted *M. oleifera* achieved the optimal dosage with half of the optimal dosage of salt extracted *M. oleifera*. This reflects the advantage of spray drying process in purification of the active gradients of *M. oleifera* seed powder.

3.3. Coagulation performance of spray dried salt extracted *M. oleifera* under different conditions

The spray dried salt extracted *M. oleifera* was packed in two different packaging conditions, closed container and vacuum packed, and stored in two different storage conditions, room temperature (29˚C) and refrigerator (3˚C). Coagulation performance of the spray dried salt extracted *M. oleifera* under different packaging forms (closed and vacuum packed) and storage conditions (room temperature (29˚C) and refrigerator (3˚C)) were presented below.

3.4. Effect of packaging and storage condition

Coagulation tests were done and residual turbidity for different conditions was measured and results are shown in Fig. 3. The results show that residual turbidity were found to be as 37.6–16.4 NTU (81–91.8% removal efficiency) for closed container at room temperature, 41–18.4 NTU (79.4–90.78% removal efficiency) for closed container in refrigerator, 30.45–16.35 NTU (84.77–91.8% removal efficiency) for vacuum packed at room temperature, and 36.76–19.03 NTU (81.6–90.44% removal efficiency) for vacuum packed in refrigerator. Comparison between the means of residual turbidity of treated water with spray dried *M. oleifera* kept in closed container in room temperature and in refrigerator shows that there were no significant ($p < 0.05$) differences between them, same result also found for the comparison between spray dried *M. oleifera* vacuum packed stored in room temperature and that stored in refrigerator.

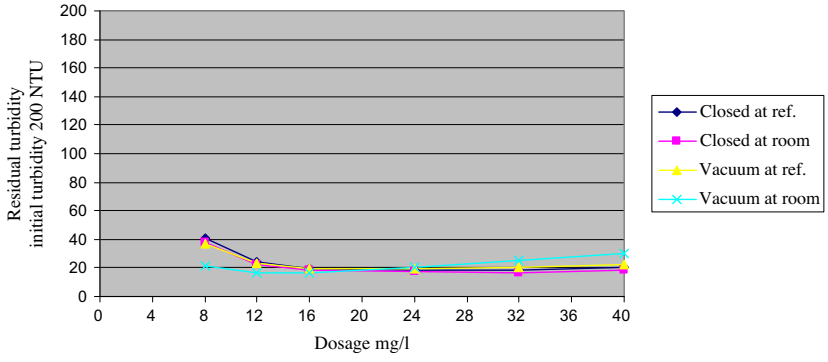

Fig. 3. The average residual turbidity at different packaging and storage conditions.

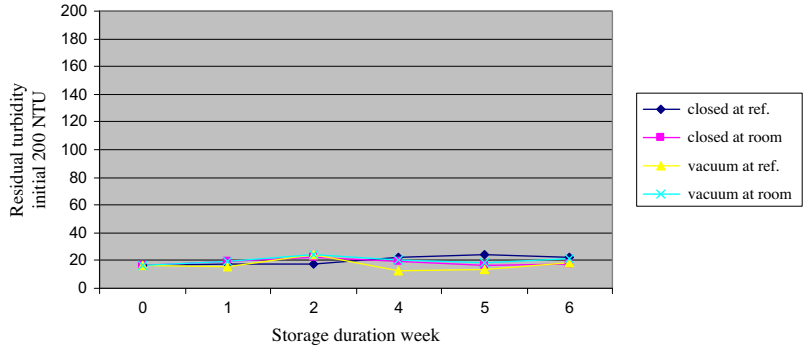

Fig. 4. The average residual turbidity at optimal dose 16 mg/l for different weeks.

Fig. 3 shows that the optimum dosage was between 12–24 mg/l for all studied conditions which found in agreement with the result reported by [3]. The salting-in mechanism in protein leading to increasing protein solubility and as a result increasing it is coagulation activity [3].

Fig. 4 shows the same trend in the value of the residual turbidity with respect to the dosages except the vacuum packed stored at room temperature which shows an increase in residual turbidity for dosages higher than 24 mg/l. Means that coagulation activity of spray dried *M. oleifera* vacuum packed stored at room temperature decreased in a high rate compared to the other storing and packaging conditions. This is due to the fast deterioration of spray dried *M. oleifera* kept in vacuum packed at room temperature.

3.5. Effect of storage duration

Comparing the residual turbidity achieved for different durations under different storage and packaging conditions, the increase of storage duration from 0 to 6 weeks was associated with an increase in residual turbidity. This was found in agreement with results reported by Katayon et al. [1,16]. The statistical analysis shows that the differences in the mean residual turbidity achieved for different storage and packaging conditions were insignificant ($p < 0.05$). The average deterioration in residual turbidity achieved was 16–20 NTU because of the short storage duration, which is in agreement with the results of freeze-dried *M. oleifera* reported by Katayon et al. [16].

4. Conclusion

The salt extracted *M. oleifera* achieved high coagulation activity with the dosage five times lower than that achieved by *M. oleifera* distilled water extracted.

The optimal dosage using spray dried salt extracted *M. oleifera* was 50% lower than the optimal dosage of nonspray dried *M. oleifera*. However, the residual turbidity achieved by nonspray dried was higher than spray dried one. There was no significant difference in the coagulation activity of spray dried salt extracted *M. oleifera* of different packaging and storage conditions. The residual turbidity of spray dried salt extracted *M. oleifera* increased insignificantly with the increase in the duration.

References

[1] S. Katayon, M.J.M.M. Noor, M. Asma, L.A.A. Ghani, A.M. Thamer, I. Azni, J. Ahmad, B.C. Khor, A.M. Suleyman, Effects of storage conditions of *Moringa oleifera* seeds on its performance in coagulation, Bioresour. Technol. 97(13) (2006) 1455–1460.

[2] T. Okuda, A.U. Baes, W. Nishijima, M. Okada, Coagulation mechanism of salt solution-extracted active component in *Moringa oleifera* seeds, Water Res. 35(3) (2001) 830–834.

[3] T. Okuda, A.U. Baes, W. Nishijima, M. Okada, Improvement of extraction method of coagulation active components from *Moringa oleifera* seed, Water Res. 33(15) (1999) 3373–3378.

[4] A. Ndabigengesere, K.S. Narasiah, Quality of water treated by coagulation using *Moringa oleifera* seeds, Water Res. 32(3) (1998) 781–791.

[5] H. Ganjidoust, K. Tatsumi, T. Yamagishi, R.N. Gholian, Effect of synthetic and natural coagulant on lignin removal from pulp and paper wastewater, Water Sci. Technol. 35(2–3) (1997) 291–296.

[6] S. Kawamura, Effectiveness of natural polyelectrolytes in water treatment, J. Am. Water Works Assoc. 83 (1991) 43–50.

[7] S.H. Lee, S.O. Lee, K.L. Jang, T.H. Lee, Microbial flocculant from Arcuadendron SP-49, Biotechnol. Lett. 17 (1995) 95–100.

[8] S.A.A. Jahn, Using Moringa seeds as coagulants in developing countries, J. Am. Water Works Assoc. JAWWA 80(6) (1988) 43–50.

[9] S.A.A. Jahn, Proper use of African coagulants for rural water supply: Research in the Sudan and a guide for new projects, Deutsche Gesellschaft fur Technische Zusammenarbeit Publishing Company, ISBN: 3880853053, 1986.

[10] R.K. Prasad, Color removal from distillery spent wash through coagulation using *Moringa oleifera* seeds: Use of optimum response surface methodology, J. Hazard. Mater. 165(1–3) (2009) 804–811.

[11] S. Bhatia, Z. Othman, A.L. Ahmad, Coagulation–flocculation process for POME treatment using *Moringa oleifera* seeds extract: Optimization studies, Chem. Eng. J. 133(1–3) (2007) 205–212.

[12] A. Diaz, N. Rincon, A. Escorihuela, N. Fernandez, E. Chacin, C.F. Forster, A preliminary evaluation of turbidity removal by natural coagulants indigenous to Venezuela, Process Biochem. 35(3–4) (1999) 391–395.

[13] S.A. Muyibi, L.M. Evison, Optimizing physical parameters affecting coagulation of turbid water with *Morninga oleifera* seeds, Water Res. 29(12) (1995) 2689–2695.

[14] A. Ndabigengesere, K.S. Narasiah, B.G. Talbot, Active agents and mechanism of coagulation of turbid waters using *Moringa oleifera*, Water Res. 29(2) (1995) 703–710.

[15] T. Okuda, A.U. Baes, W. Nishijima, M. Okada, Isolation and characterization of coagulant extracted from *Moringa oleifera* seed by salt solution, Water Res. 35(2) (2001) 405–410.

[16] S. Katayon, S. Ng, M. Johari, L. Ghani, Preservation of coagulation efficiency of *Moringa oleifera*, a natural coagulant, Biotechnol. Bioprocess Eng. 11(6) (2006) 489–495.

[17] S. Katayon, M. Megat Johari, M. Asma, A. Thamer, L. Abdul Ghani, A. Idris, S. Muyibi, M. Aminuddin, B. Khor, Effects of storage duration and temperature of *Moringa oleifera* stock solution on its performance in coagulation, Int. J. Eng. Technol. 1(2) (2004) 146–151.

[18] S.A. Muyibi, L.M. Evison, *Moringa oleifera* seeds for softening hardwater, Water Res. 29(4) (1995) 1099–1104.