Optimization of Water Treatment Parameters using Processed *Moringa oleifera* As a Natural Coagulant for Low Turbidity Water

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Abstract— *Moringa oleifera* is a natural coagulant which can be used for water treatment in tropical developing countries. This study presents the results of production of natural coagulant from processed *Moringa oleifera* with simple technique (oil extraction, salt extraction, and microfiltration with 0.2 µm). The optimization study on physical factors was carried out for coagulation-flocculation process. Low initial turbidity water samples (synthetic and river water) were used with turbidity less than 50 Nephelometric Turbidity Units (NTU). The response surface methodology (RSM) was used, and the analysis of variance (ANOVA) was performed to validate the developed regression model. The residual turbidity obtained was 4.514 NTU for synthetic water and 1.598 NTU for river water by applying the optimum conditions of 40 rpm (low speed), mixing time of 41 minutes, and *Moringa oleifera* dosage of 0.75 mg/L.

Keywords- ***Moringa oleifera, optimization, coagulation, response surface methodology, low turbidity.***

I. Introduction

The conventional method for water treatment is used worldwide in the water treatment industry before consuming the water [1]. The chemicals used in water treatment are with high cost that is increasing annually which is difficult for developing countries to cope with [2]. The Lime is needed in water industry for pH adjustment when the Alum used with high concentration [3] which is an extra cost for water treatment. The natural product of processed *Moringa oleifera* consider as an alternative for Alum in water treatment industry. Therefore, it was used in this study and the optimization of water treatment parameters was performed to find the optimum conditions. The traditional experimental method, one factor at a time approach, is not giving the relationships among independent factors and dependant factors besides that it consume more time and energy. Furthermore, since the results are valid only under fixed experimental conditions, the prediction for other conditions is uncertain [4]. Therefore, design of experiment (DOE) was used in this study to get better relationships between all the parameters and the response with a minimum number of experiments. The design of experiments was done based on response surface methodology (RSM), which gives the lowest number of experiments to be carried out. RSM quantifies relationships among one or more measured responses and the vital input factors [5, 6].

Multiple effects of physical parameters on a coagulation of turbid water with *Moringa oleifera* was performed [7], the selected operating variables were used to establish the relationship between the physical factors and their effect on coagulation with *Moringa oleifera*. The residual turbidity was equal or less than 10 NTU, no oil extraction took place in the research. A similar study was conducted on the velocity gradient parameters which showed high effect on flocculation process [8]. Other studies about parameters affecting the use of *Moringa oleifera* in water treatment was carried out by [9, 10].

The optimal design for the turbidity removal is a very important aspect in the development of jar test in water treatment process. In this research work, the optimization of operating conditions was carried out for the use of flocculating bio-active constituents in coagulation of turbid water for potable use. The dependent variable is the residual turbidity (NTU), and the independent variables chosen were: i) the high speed (rpm); ii) high speed mixing time (min); iii) low speed (rpm); iv) low speed mixing time (min); and v) processed *Moringa olefeira* dosage (mg/L). Statistical optimization was conducted by using central composite design (CCD) with five factors and three levels. The experimental data were analyzed using statistical software [5] to develop the factorial regression model for determining the optimum conditions.

II. Materials & Methods

A. Preparation of Moringa oleifera seeds

Moringa oleifera pods were collected from Serdang, Selangor Darul Ehsan, Malaysia. The dry brown colour pods were chosen [11]. The seeds were cleaned from the coat and wings, then the seeds were grinded by using domestic blender (National, MX-896TM), and sieving the powder using 250 µm sieve.

B. Oil Extraction

The oil in *Moringa oleifera* seed powder was extracted using hexane. Weighing of 10 gm *Moringa oleifera* seed powder and locate it in the thimbles of the electro thermal soxhlet extraction chamber (Electro Thermal Soxhlet apparatus, ROSS, UK). Hexane with volume of 170 ml was added in the volumetric flask of the soxhlet, then evaporating of hexane through three cycles each for 30 min till the hexane become colorless [12]. The *Moringa oleifera* cake residue was used in this research; the oil percentage was 35% w/w. One kilogram of *Moringa oleifera* seeds was defatted and kept at room temperature 23±2ºC to be used throughout the study.

C. Salt extraction

Salt extraction was carried out by using 1 Molar NaCl to extract the bioactive constituents from *Moringa oleifera* cake residue. The extraction was done for 30 minutes using the magnetic stirrer (ERLA, ERLA Technologies (M) Sdn. Bhd. Malaysia) according to [13]. The salt extracted sample was filtered with Whatman filter paper #1, the clear solution was injected to the microfiltration cartridge [14].

D. Cross flow filtration

The microfiltration was proposed to be used in this study for bioactive constituent's separation [14]. The organic load was reduced by microfiltration.
The process flow chart for producing natural coagulant from *Moringa oleifera* seeds is shown in Fig. 1.

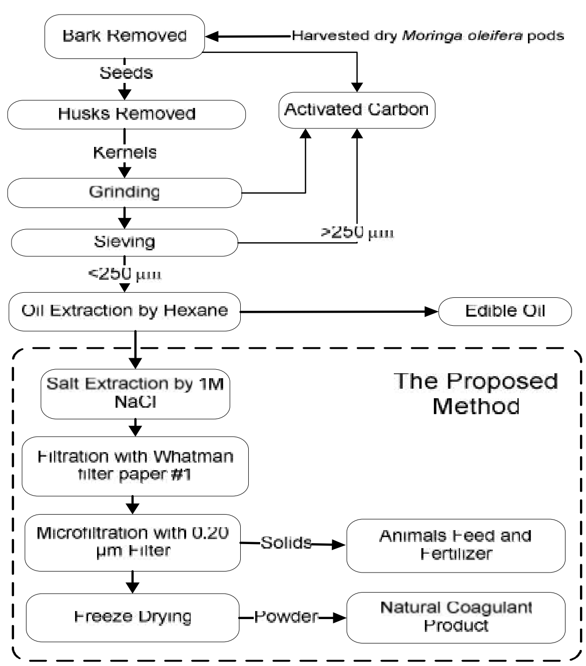


Figure 1: Process flow chart

E. Coagulation activity tests

To monitor the performance of the processed *Moringa oleifera* seed in water treatment by measuring turbidity removal, the jar test was performed as followed:

- Synthetic Water (Kaolin Suspension) Preparation.
  The synthetic water was prepared according to [7, 11, 13, and 14].
- Jar Test
  The processed *Moringa oleifera* was tested by performing jar test to monitor the coagulation activity, following same parameters as [12, 14].

F. Optimization

- Preliminary Optimization
  Five parameters were chosen for this optimization. The range of high speed was (100, 125, and 150 rpm), the high speed mixing time was (2, 4, and 6 min), low speed (20, 40, and 60 rpm) within mixing time of (15, 25, and 35 min), and the processed *Moringa oleifera* seeds dosage was (0.25, 0.75, and 1.25 mg/L). Thirty eight experiments were carried out according to the RSM design.

- Second Optimization
  Three parameters were chosen for this optimization according to the results obtained from preliminary optimization. The low speed (40, 60, and 80 rpm) within mixing time of (20, 40, and 60 min) and the processed *Moringa oleifera* seeds dosage was (0.25, 0.75, and 1.25 mg/L). Twenty experiments were done according to RSM design.

- Jar Test
  The samples for both optimization studies were applied to synthetic water for optimization. Then, both synthetic and river water were applied for verification of results. The synthetic water was prepared as mentioned in section E, and the river water samples were collected from Sungai Pusu which goes through International Islamic University Malaysia (IIUM) campus.

III. RESULTS AND DISCUSSION

A. Preliminary Optimization

To get a good relationship between the variable and response, RSM was used. The jar test experimental work was carried out with six centre points to fix the dosage of processed *Moringa oleifera* and the results for the points are shown in Fig. 2.

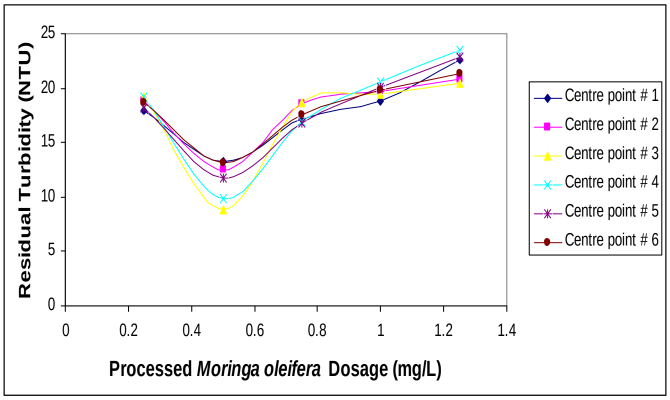


Figure 2: Jar test results for center points

The processed *Moringa oleifera* dosage ranges have been chosen based on preliminary experiments. It was found from the preliminary study that the best dosage was 0.50 mg/L.of coagulant added to get the optimum residual turbidity as shown in Fig. 2. The central point (zero levels) chosen for experimental design was according to [12]. The factorial design with five factors and six replicates at central points were employed to fit the second order factorial model, which indicated thirty eight experiments are required.

According to ANOVA (Table I), the factors B, C, D, and E were highly significant with value of $p < 0.01$ while the factor A was significant at $p < 0.05$. The Model F-value of 25.95 implies the model is significant. There is only a 0.01% chance that a "Model F-Value" this large could occur due to noise. Values of "Prob > F" less than 0.05 indicate model terms are significant. In this case A, B, C, D, E, AB, AE, BD, BE, DE, ABD, ACE, ADE, BCE, BDE, ABCD, BCDE are significant model terms. The "Curvature F-value" of 195.52 implies there is significant curvature (as measured by difference between the average of the centre points and the average of the factorial points) in the design space. There is only a 0.01% chance that a "Curvature F-value" this large could occur due to noise. The "Lack of Fit F-value" of 1.59 implies the Lack of Fit is not significant relative to the pure error. There is a 31.15% chance that a "Lack of Fit F-value" this large could occur due to noise.

The "Predicted R-Squared" of 0.7080 is not as close to the "Adjusted R-Squared" of 0.9474. This may indicate a large block effect or a possible problem with the model or data. Therfore, it is important to consider model reduction.
It was noticed that the most solutions indicate using high speed between 100-132 rpm and the best solution selected was by using high speed of 102 rpm. With respect to the high speed mixing time the best solution was by using 5.94 min and for most of the solutions, the chosen time was around six min.

TABLE I. ANOVA FOR REGRESSION MODEL

Source	Sum of Squares	DF	Mean Square	F Value	Prob > F	
Model	240.0844	26	9.234014423	25.95495065	< 0.0001	significant
A	3.25125	1	3.25125	9.138607484	0.0128	
B	24.32531	1	24.3253125	68.37354336	< 0.0001	
C	44.88781	1	44.8878125	126.1705803	< 0.0001	
D	45.36281	1	45.3628125	127.5057094	< 0.0001	
E	18.605	1	18.605	52.29489957	< 0.0001	
AB	11.40031	1	11.4003125	32.04397728	0.0002	
AC	1.320313	1	1.3203125	3.711131932	0.0829	
AD	0.262813	1	0.2628125	0.738712889	0.4102	
AE	3.25125	1	3.25125	9.138607484	0.0128	
BC	0.78125	1	0.78125	2.195936054	0.1692	
BD	13.26125	1	13.26125	37.27469696	0.0001	
BE	3.577813	1	3.5778125	10.05650875	0.0100	
CD	1.71125	1	1.71125	4.809978333	0.0531	
CE	1.240313	1	1.2403125	3.48626808	0.0914	
DE	3.315313	1	3.3153125	9.31867424	0.0122	
ABC	0.36125	1	0.36125	1.015400832	0.3374	
ABD	2.31125	1	2.31125	6.496457223	0.0289	
ACD	1.20125	1	1.20125	3.376471277	0.0960	
ACE	10.46531	1	10.4653125	29.41588101	0.0003	
ADE	17.55281	1	17.5528125	49.33741289	< 0.0001	
BCD	0.052813	1	0.0528125	0.148445277	0.7081	
BCE	3.38	1	3.38	9.500497746	0.0116	
BDE	7.41125	1	7.41125	20.83152779	0.0010	
CDE	0.91125	1	0.91125	2.561339814	0.1406	
ABCD	12.37531	1	12.3753125	34.78450548	0.0002	
BCDE	7.507813	1	7.5078125	21.10294548	0.0010	
Curvature	69.56055	1	69.56054825	195.5206603	< 0.0001	significant
Residual	3.557708	10	0.355770833			
Lack of Fit	2.184375	5	0.436875	1.59056432	0.3115	not significant
Pure Error	1.373333	5	0.274666667			
Cor Total	313.2026	37				

First Regression model equation

Design-Expert 6.0.8 software [5] was used to evaluate the coefficients of the model equation and their statistical significance. The factorial regression model for the residual turbidity (NTU) in term of symbol factors are given in (1).

Residual Turbidity = - 14.51875 + 0.16878 A + 0.61164 B + 6.68906 C + 1.60750D + 19.68750 E - 4.10625E-003 AB - 0.043656 AC - 0.010331 AD - 0.031125 AE - 0.23439 BC - 0.031625 BD - 0.33094 BE - 0.35531 CD + 0.46875 CE - 1.45250 DE + 1.66094E-003 ABC + 1.95000E-004 ABD + 2.10000E-003 ACD - 0.022875 ACE + 5.92500E-003 ADE + 9.48828E -003 BCD + 0.044297 BCE + 0.014500 BDE + 0.11375 CDE - 6.21875E-005 ABCD - 2.42188E-003 BCDE (1)

where:
A: High speed (rpm), B: Low speed (rpm), C: High speed mixing time (min), D: Low speed mixing time (min), E: Processed *Moringa oleifera* dosage (mg/L).

Low speed is important here as the residual turbidity is decreased by increasing the flocculation speed. At the same time, the mixing time is important too, as the residual turbidity is decreased by increasing the time, and this means that it is of great importance to give enough time to the floc to agglomerate. The regression values are plotted showed the relationship between the actual and predicted residual turbidity. The "Predicted R-Squared" of 0.7080 is not as close to the "Adjusted R-Squared" of 0.9474. This indicats a large block effect or a possible problem with the model or data. Therfore, it is important to consider model reduction. The insignificant terms AC, AD, BC, CD, CE, ABC, ACD, BCD, AND CDE were excluded from the regression model, but there was no improvment in R^2.

Since there is no other research done regarding using water treatment parameters optimization by using processed *Moringa oleifera* by microfiltration method to be used for comparison with this study. Therefore, a second optimization study was carried out.

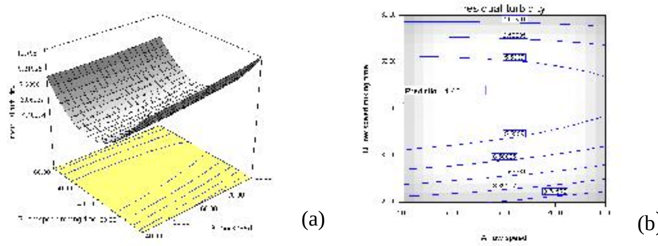
B. Second optimization

The high speed and the high speed mixing time are excluded and not considered in this optimization because most of the solutions given by RSM showed that the high speed were within the value of 100 – 132 rpm. However, best results of residual turbidity was with high speed of 100 rpm, which was then fixed. The high speed mixing time was 6 minutes for most of the solutions, which was fixed as a rapid mixing parameter chosen in the regression model. The jar test results showed that the minimum residual turbidity was achieved at high speed of 100 rpm and 6 minutes high speed mixing time. The analysis of variance for regression model was shown in Table II.

TABLE II. ANOVA FOR REGRESSION MODEL

Source	Squares	DF	Square	Value	Prob > F	
Model	304.8058	9	33.86731	235.7515104	< 0.0001	significant
A	1.681	1	1.681	11.7014982	0.0065	
B	6.4	1	6.4	44.55061779	< 0.0001	
C	2.601	1	2.601	18.10564951	0.0017	
A2	0.046151	1	0.046151	0.321255676	0.5834	
B2	48.03865	1	48.03865	334.3986814	< 0.0001	
C2	49.1949	1	49.1949	342.447377	< 0.0001	
AB	0.845	1	0.845	5.882073755	0.0357	
AC	2	1	2	13.92206806	0.0039	
BC	1.805	1	1.805	12.56466642	0.0053	
Residual	1.436568	10	0.143657			
Lack of Fit	1.054485	5	0.210897	2.759829483	0.1447	not significant
Pure Error	0.382083	5	0.076417			
Cor Total	306.2424	19				

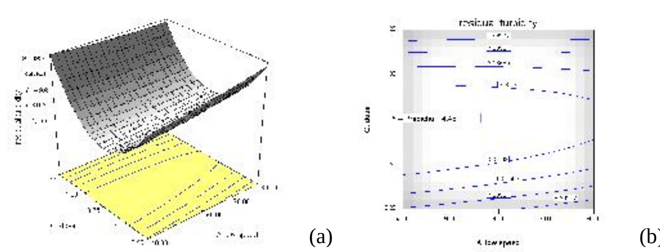
The solution for the regression model was only one solution with a low speed of 40 rpm, mixing time 41.28 minutes, and the processed *Moringa oleifera* dosage was 0.69 mg/L. The residual turbidity was 4.47547 and the desirability was 0.983902, using synthetic water with initial turbidity of 32.6 NTU. The regression value showed the relationship between the actual and predicted residual turbidity with R^2 value of 99.81%. The 3D surface and contour plot in Fig. 3, shows the reduction in turbidity increases with low speed mixing time between 30-50 minutes which is about the same by increasing the low speed. The response surface in Fig. 3 shows a curvature and this indicates that the interaction effect of mixing time on residual turbidity is greatly pronounced as confirmed by the significant test. The low speed is a very important factor as the low speed is for flocculation process and the flocculation depends on the characteristics of the particles as well as the fluid mixing conditions. In flocculation, mixing is used to move particles around so the particles can get in contact with each other after chemistry (coagulant) has been used to neutralize their natural repulsion to each other [15]. The flocculation of small particles caused by diffusion and the rate of flocculation are relative to the rate at which particles diffuse toward one another. Thus, for small particles (less than 0.1 µm), the primary mechanism of aggregation is Brownian motion, or micro scale flocculation. As aggregation of small particles proceeds, larger particles are formed and after a short time, microflocs ranging in size from 1 to about 100 µm are formed [16].



(a) (b)

Figure 3: The interaction between low speed and mixing time. (a) 3D surface response, (b) Contour plots.

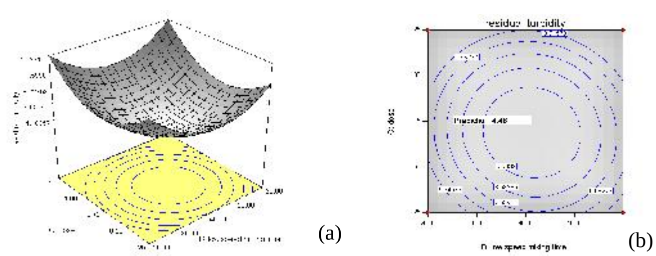
Fig. 4 shows the effect of low speed and dosage in 3D surface response and contour plots. The plot shows that the dose of 0.5 – 1.00 mg/L reduced the residual turbidity. Thus, the dosage is highly significant and affecting the residual turbidity to a certain point where the higher dosages will produce higher residual turbidity again due to destabilization.



(a) (b)

Figure 4: The interaction between low speed and dose. (a) 3D surface response, (b) Contour plots.

Fig. 5 shows the result of contribution of the interaction effect between the low speed mixing time and dosage. It can be seen that the residual turbidity decreased with low speed mixing time between 30-50 minutes when dose was between 0.5-1.00 mg/L.



(a) (b)

Figure 5: The interaction between low speed mixing time and dose. (a) 3D surface response, (b) Contour plots.

Fig 3, 4, and 5 indicated that the lowest residual turbidity is obtained at the intersection of zero levels of the other two factors, because the 3D surface and contour plot of the RSM was drawn as a function of two factors at a time, while the other two factors were fixed at zero level.

Second Regression model equation

The response (residual turbidity) was correlated with the three variables studied using the quadratic model and the coefficients of the model equation, The quadratic regression model for the residual turbidity (NTU) in terms of symbol factors is given in (2).

$$\text{Residual Turbidity} = +29.06375 + 0.051636\,A - 0.86278\,B - 23.25727\,C + 0.010449\,B^2 + 16.91818\,C^2 - 8.12500E\text{-}004\,AB - 0.050000\,AC + 0.047500\,BC \quad (2)$$

where:

A: Low speed (rpm), B: Low speed mixing time (min), and C: Processed *Moringa oleifera* dosage (mg/L).

Statistical analysis gives several comparative measures for the model selection. Ignoring the aliased model, the quadratic model seems to be the best based on low standard deviation of 0.38 and high R^2 statistics of 0.9981. This is also evident from the value of to y=x showing that the prediction of experimental data is satisfactory.

C. Verification of the Model

More experiments were performed for residual turbidity (NTU) by using the numerical optimization of the Design-Expert 6.0.8 software based on the model proposed, to verify the model developed. The optimum parameters were used to conduct the verification experiments. The results of experiments were compared the predicted results according to the model. The minimum values of residual turbidity were selected as an optimum values. The results demonstrated that the model prediction from (2), the residual turbidity agreed reasonably well with the experimental data. Thus, the optimum values of the process variables were: low speed 40 rpm; mixing time 41 minutes; and dosage of 0.75 mg/L. The residual turbidity was 4.5398 NTU by calculations (predicted) and an average of 4.514 NTU by experimental work at the optimum conditions. Other correlation is represented in Table III for synthetic and Table IV for actual river water samples treated with the same product of processed *Moringa oleifera*. The adequacy of the model was verified effectively by the validation of experimental data. Additionally, the optimum results showed that the residual turbidity was 4.514 NTU for synthetic water and 1.598 NTU for river water. The difference in results between using synthetic water and river water is due to that the river water represents the actual water which contains all the negative charged components while synthetic water is not including all the contents of actual water. The results showed the high performance of processed *Moringa oleifera* seeds compared to the study [9], which showed that only 50% reduction in turbidity for low turbidity water of 40 NTU was achieved. This was due to using *Moringa oleifera* seeds without removing oil and other un-active components from the seeds.

TABLE III. VALIDATION OF THE RESIDUAL TURBIDITY USING SYNTHETIC WATER*

Low speed (rpm)	Mixing time (min)	Dosage (mg/L)	Predicted residual turbidity from model (NTU)	Experiments					Mean value (NTU)
				1	2	3	4	5	
40	41	0.75	4.5398	4.56	4.62	4.51	4.49	4.39	4.514
40	40	0.60	4.6308	4.65	4.69	4.72	4.61	4.59	4.652
40	45	0.75	4.6956	4.72	4.65	4.74	4.78	4.70	4.714

*Initial turbidity 32.6 NTU, high speed 100 rpm, mixing time 6 minutes, settling time 60 minutes.

TABLE IV. VALIDATION OF THE RESIDUAL TURBIDITY USING RIVER WATER*

Low speed (rpm)	Mixing time (min)	Dosage (mg/L)	Predicted residual turbidity from model (NTU)	Experiments					Mean value (NTU)
				1	2	3	4	5	
40	41	0.75	4.5398	1.41	1.76	1.45	1.52	1.90	1.598

*Initial turbidity 32.4 NTU, high speed 100 rpm, mixing time 6 minutes, settling time 60 minutes

IV. CONCLUSION

It is the first time to produce *Moringa oleifera* as coagulant/flocculant by microfiltration technique. This study showed the possibility of producing natural coagulant with high efficiency by very simple and straightforward process (oil extraction; salt extraction; and micro filtration with 0.2 µm). The very low residual turbidity < 5 NTU was achieved by treatment of low initial turbidity surface waters with very low dosage of processed *Moringa oleifera*.

ACKNOWLEDGMENT

The authors are grateful to the Ministry of Science Technology and Innovation, Malaysia. For funding the Project (IRPA 09-02-08-10002 EAR).

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