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Optimization of process conditions in water treatment through coagulation diagrams, using *Moringa oleifera* Lam and aluminium sulphate

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ABSTRACT

The construction of coagulation diagrams is provided to obtain colour, turbidity and compounds with absorption at $UV_{254\ nm}$ removal by associating the coagulants *Moringa oleifera* Lam and aluminium sulphate to define the optimal conditions of the process in water treatment. Experiments were performed in jar test with several coagulant dosages and coagulation pH. Optimal conditions for raw water with high colour/turbidity were obtained at pH values ranging from 7.5 to 8. After filtration, three dosages of association aluminium sulphate—*M. oleifera*, 25 ppm/300 ppm; 20 ppm/350 ppm and 15 ppm/400 ppm, performed efficient removal of evaluated parameters, namely 97.7% plus for colour and turbidity and 68.2% for compounds with absorption at $UV_{254\ nm}$. Results showed that the use of coagulation diagram is useful since it provides development of tests in optimal conditions, depending on water initial characteristics. The association of coagulants aluminium sulphate—*M. oleifera* may be considered an alternative technique for conventional water treatment.

Keywords: Water treatment; Coagulation diagrams; Association of coagulants; *Moringa oleifera* Lam; Aluminium sulphate

1. Introduction

Currently, many people worldwide still use surface water or water from unprotected wells [1], without any treatment whatsoever.

The use of natural coagulants, that is, more accessible materials to underserved populations, may reduce problems associated with non-potable water consumption. Highly promising natural coagulants for water purification are the concern of many researchers, because of their abundant sources, low price, environment-friendly, multifunctional and biodegradable nature [2].

Moringa oleifera Lam, one of the most promising natural coagulants [3–5], belongs to the family

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Moringaceae and comprises only one genus (*Moringa*) and 14 species [6].

When the seeds of *M. oleifera* are crashed and added to water, the protein produces positive charges and acts like magnet by attracting predominantly negatively charged particles such as clay and toxic particles. Under proper agitation, the bound particles grow in size to form flocculates which are left to settle by gravity. The above stated accounts for the effectiveness of *M. oleifera* as a coagulant for raw water purification [4].

Although studies have been conducted to determine potential risks associated with the use of *M. oleifera* seeds, no evidence has so far been found that the seeds cause any side effects on human populations especially, since only low dosages are needed for water treatment [7].

M. oleifera does not alter significantly pH and alkalinity of water after treatment and does not cause corrosion problems. Furthermore, the sludge produced during the coagulation process with *M. oleifera* is not only innocuous, but also produces one-fourth to one-fifth of the sludge when compared with alum [6,8].

The use of the *M. oleifera* seeds may be considered a sort of viable partial replacement for other chemical products used for water treatment [9]. One of the above-mentioned products is aluminium sulphate, the chemical coagulant widely used in Brazil, due to its high efficiency removal of suspended solids and low acquisition cost [3].

The association of *M. oleifera* seed solution with aluminium sulphate reduces the amount of alum in water treatment and gives good flocculation results [10].

The use of mathematical tools to define the best conditions from a set of variables is extremely important to obtain optimized conditions of the process. Kim et al. [11] claim that the use of the coagulation diagram is important for planning, analysis and interpretation of results obtained in tests performed in jar test and may define the coagulant dosages and pH conditions for turbidity removal.

Coagulation diagram consists in plotting all the rates of parameter removal, such as colour and turbidity, obtained in the performed tests, while varying pH coagulation and coagulant dosage, and also in obtaining ranges where removal percentages are considerable enough to allow choosing the study points [12]. The coagulation diagrams reveal pH coagulation in the abscissa axis and coagulant dosage in the ordinate axis.

The current study proposes the construction of coagulation diagrams to obtain efficient removal ranges for the parameters colour, turbidity and compounds with absorption at $UV_{254\,nm}$, using the coagulants *M. oleifera* and aluminium sulphate to define optimized process conditions in water treatment.

2. Materials and methods

Raw water was collected from the Pirapó River basin by the Paraná Waterworks Company (Sanepar) and its characterization was determined for the following quality parameters: colour and turbidity (HACH DR/2010 spectrophotometer), compounds with absorption at $UV_{254\,nm}$ (LS Logen Scientific—AL-PAX spectrophotometer), total dissolved solids [13], pH (Digimed DM-2 pH metre), alkalinity and volatile acidity, total hardness (EDTA titrimetric method), total coliforms and *Escherichia coli* (3 M Petrifilm plates).

2.1. Preparation of coagulants

A concentration of 1% wv^{-1} was employed for the preparation of *M. oleifera* standard solution, or rather, 100 mL of distilled water was added for every 1 g of seed pulp. The latter was crushed in a blender, agitated for 30 min and vacuum filtered in qualitative paper [3,12]. This solution was prepared on the day of the assay.

For the preparation of a standard solution of aluminium sulphate with 1% concentration, 10.3 g of the chemical coagulant (97% purity) was dissolved in distilled water and the volume was completed up to 1 L [12].

2.2. Coagulation/flocculation process

The coagulation/flocculation tests were carried out in jar test, Milan—Model JT 101/6 of six jars, with rotation regulator of mixer shafts, in duplicate. Water temperature was maintained at 25.0 ± 3.0°C in all experiments [12].

The experimental conditions for the coagulation/flocculation process were rapid mixing gradient (183.5 s^{-1}), rapid mixing time (3 min), slow mixing gradient (5.8 s^{-1}), slow mixing time (15 min) and settling time (90 min). The above rates were based on experimental studies in order to determine the process operational conditions.

Further, pH coagulation rates were corrected at 4, 5, 6, 7 and 8 values with 25% sodium hydroxide solution (NaOH) and 98% sulphuric acid (H_2SO_4) of analytical grade, for the construction of the coagulation diagrams.

Table 1 shows the dosages of the coagulants aluminium sulphate and *M. oleifera*, based on previous

Table 1
Dosages of the coagulants in the association of aluminium sulphate and *M. oleifera* used in the coagulation/flocculation process for the construction of coagulation diagrams

Point	1	2	3	4	5	6	7	8	9	10	11	12
Aluminium sulphate (mgL^{-1})	55	50	45	40	35	30	25	20	15	10	5	0
M. oleifera (mgL^{-1})	0	50	100	150	200	250	300	350	400	450	500	550

studies by Madrona et al. [3] and Valverde et al. [12], by adding the solution of chemical coagulant, followed by the natural coagulant.

After coagulation/flocculation completion and settling, a sample was collected and the parameters colour, turbidity and compounds with absorption at $UV_{254\,nm}$ were evaluated, according to the methodology described above.

2.3. Coagulation diagrams construction

Coagulation diagrams were built with the program 3DField 2.7.0.0., using the results from the experiments. Study points were chosen to obtain optimal operational conditions for the raw water. The filtration step was included to verify whether the quality parameters were in accordance with the Environmental Protection Agency (EPA) [14].

2.4. Filtration step

A gravity filter was constructed so that the thickness of the filtration layers was proportional to the filter of Sanepar, Maringá City, Paraná State, Brazil. A filter was obtained with the following thicknesses: 7.5 cm of anthracite coal with specific size from 0.9 to 1.5 mm; 4.2 cm of sand with specific size from 0.45 to 0.5 mm; 1.3 cm of rolled pebbles $\frac{1}{8}$–$^{1}/_{16}$´´; 1.3 cm of rolled pebbles ¼–$\frac{1}{8}$´´; 1.3 cm of rolled pebble ½–¼´´. The total height of the filter was 16.5 cm, with 15 cm diameter.

The flow rate was approximately 100 $mLmin^{-1}$ and the retention time of water in the filter was approximately 30 s.

3. Results and discussion

Table 2 shows the characterization results of raw water in the coagulation/flocculation, sedimentation and filtration tests.

Figs. 1–3 show the coagulation diagrams with the association of coagulants, taking into account the removal efficiency of colour, turbidity and compounds with absorption at $UV_{254\,nm}$, obtained after coagulation/flocculation and sedimentation tests.

Table 2
Quality parameters of raw water

Quality parameter	Raw water
Colour (uH[a])	413
Turbidity (NTU[b])	179
$UV_{254\,nm}$ (cm^{-1})	0.2301
TDS[c] (mgL^{-1})	1,228
pH	7.8
Alkalinity (mg $CaCO_3L^{-1}$)	62
Volatile acidity (mg CH_3COOHL^{-1})	30
Total hardness (mg $CaCO_3L^{-1}$)	23
Total coliforms (CFU [d])	1,500
E. coli (CFU[d])	700

[a]uH: Hazen unit (mg Pt–CoL^{-1}).
[b]NTU: nephelometric turbidity unit.
[c]TDS: total dissolved solids.
[d]CFU: colony forming units per 100 mL.

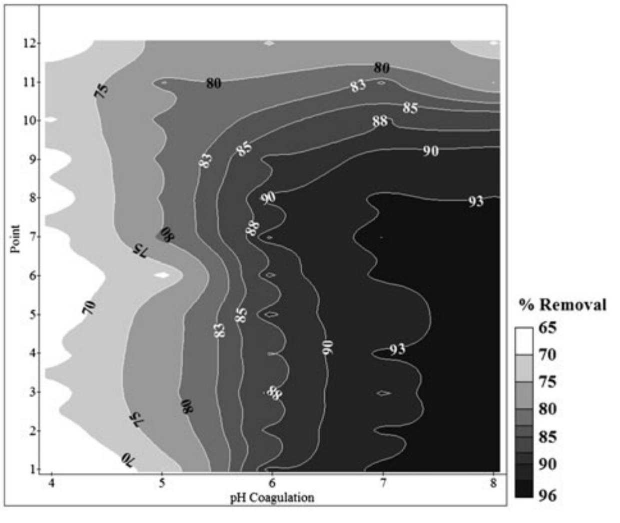

Fig. 1. Coagulation diagram with curves of colour removal after coagulation/flocculation and sedimentation tests with the association of coagulants aluminium sulphate—*M. oleifera* in the pH range from 4 to 8.

Although Fig. 1 shows that a 65–80% removal was obtained by *M. oleifera* (point 12), the association of coagulants provided the best efficient colour removal.

Amagloh and Benang [4] recommended the association of coagulants to obtain optimum results in cases of low removal efficiency. Even in small amounts, the chemical coagulant enhanced the removal of quality parameters [12].

The evaluation of the coagulation diagram containing curves of colour removal (Fig. 1) showed over 80% removal by coagulants association (points 2–11) for the entire pH range above 5.5 and in all dosages studied. Bests results were obtained at pH values above 7, where removals above 93% were obtained up to point 8 (20 ppm of aluminium sulphate standard solution/350 ppm of *M. oleifera*). Similarly, the same removal efficiency was observed when aluminium sulphate was applied as the primary coagulant (point 1). It would actually be a great advantage in terms of reducing residual aluminium [15].

The coagulation diagram containing curves of turbidity removal (Fig. 2) revealed over 90% removal for coagulation pH range between 6.5 and 8 in point 4 (40 ppm of aluminium sulphate—150 ppm of *M. oleifera*). There was no change in removals obtained at pH values higher than 5.5, with 85% removal, from point 7 to 10.

Pritchard et al. [5] achieved a 60% turbidity removal for raw water with initial turbidity of 175 NTU at pH 7.5 and, with 150 ppm standard solution of *M. oleifera* added to the process. As shown at point 4 (40 ppm of aluminium sulphate—150 ppm of *M. oleifera*), a removal above 95% was obtained with similar pH and initial turbidity conditions. It may be underscored that the association between *M. oleifera* and chemical coagulant improved significantly the process efficiency.

According to Westphal et al. [16], optimal dosage of aluminium sulphate was 25 ppm, when raw water with turbidity around 180 NTU was used. In the current study, the association of coagulants caused 90% and 80% removal in colour and turbidity, respectively, with a dosage of 15 ppm of aluminium sulphate plus 400 ppm of *M. oleifera* (point 9), at pH values above 7. Consequently, there was a decrease in the consumption of the chemical coagulant used for the clarification of raw water.

In the case of compounds with absorption at $UV_{254\,nm}$, it may be observed that good percentage removals were found with pH 7 throughout the concentration range.

The coagulation diagrams indicated the optimized areas of colour, turbidity and compounds with absorption at $UV_{254\,nm}$ removal, taking into consideration the quality of raw water, according to the experimental conditions under analysis. The working pH range was chosen between 7.5 and 8, since the coagulation diagrams (Figs. 1–3) showed satisfactory removal results under these conditions.

Consequently, six points of study were defined, as shown in Table 3. They were based on the dosages lower than 30 ppm aluminium sulphate, because it was found in the results obtained that the use of

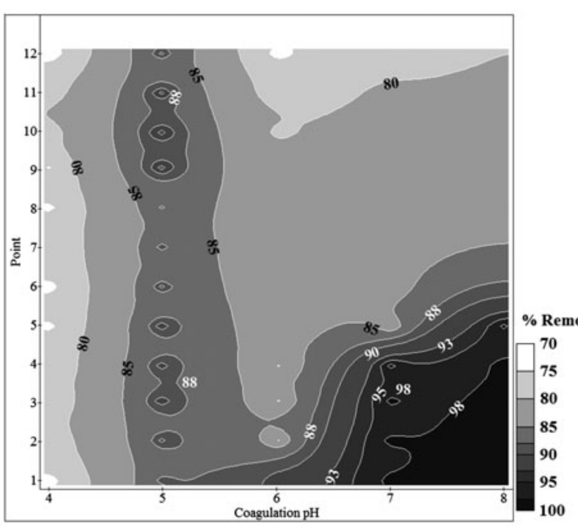

Fig. 2. Coagulation diagram with curves of turbidity removal after coagulation/flocculation and sedimentation tests with the association of coagulants aluminium sulphate—*M. oleifera* in the pH range from 4 to 8.

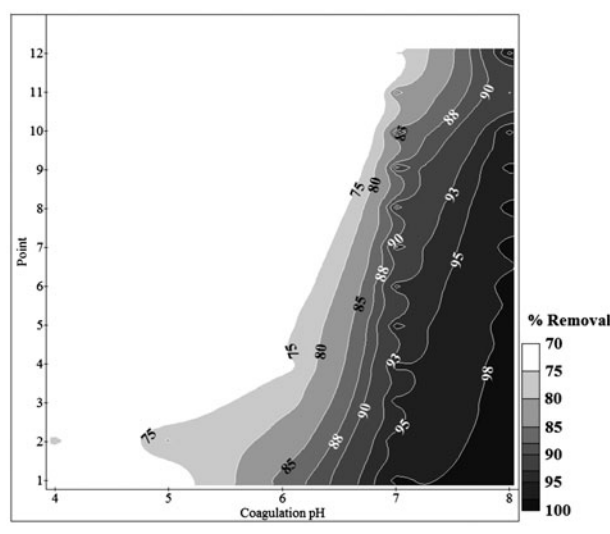

Fig. 3. Coagulation diagram with curves of compounds with absorption in $UV_{254\,nm}$ removal after coagulation/flocculation and sedimentation tests with the association of coagulants aluminium sulphate—*M. oleifera* in the pH range from 4 to 8.

Table 3
Points of study selected as pre-treatment for filtration process

Points of study	Dosage (mgL^{-1}) Aluminium sulphate	*M. oleifera*	Removal range Colour (%)	Turbidity (%)	Compounds with absorption in $UV_{254\,nm}$ (%)
6	30	250	93–96	85–88	95–98
7	25	300	93–96	80–85	95–98
8	20	350	93–96	80–85	93–98
9	15	400	90–93	80–85	93–98
10	10	450	85–88	80–85	90–98
11	5	500	75–80	80–85	88–93

Table 4
Quality of water treated by the association of coagulants after the coagulation/flocculation, sedimentation and filtration process

Quality parameters	Treated water—points of study 6	7	8	9	10	11	Values tolerated by EPA (2009)
Colour (uH)	16	8	10	9	34	30	15
Turbidity (NTU)	8	4	3	2	16	14	5[a]
$UV_{254\,nm}$ (cm^{-1})	–	0.082	0.069	0.065	–	–	–
TDS (mgL^{-1})	–	89	41	105	–	–	500
pH	–	7.1	7.1	7.2	–	–	6.5–8.5
Alkalinity (mg $CaCO_3L^{-1}$)	–	50	50	48	–	–	–
Volatile acidity (mg CH_3COOHL^{-1})	–	22	30	22	–	–	–
Total hardness (mg $CaCO_3L^{-1}$)	–	20.50	20.25	23.50	–	–	–
Total coliforms (CFU)	–	1,190	1,060	1,125	–	–	Absence
E. coli (CFU)	–	Absence	Absence	Absence	–	–	Absence

[a]Considering alternative technique.

M. oleifera could lead to a 50% reduction in the use of inorganic chemicals, according to Abaliwano et al. [8], and on the removal ranges of three parameters evaluated: colour, turbidity and compounds with absorption at $UV_{254\,nm.}$

The filtration step was added to further reduce the residual amount of each parameter evaluated. Table 4 shows results for the treated water characterization by means of the association of coagulants after the coagulation/flocculation, sedimentation and filtration process.

Three study points (7, 8 and 9) were investigated in more detail by analysing all the parameters listed in Table 4, since colour and turbidity rates with 97.5% removals were considered acceptable by the EPA [14]. These points correspond to the dosages of aluminium sulphate—*M. oleifera* standard solution: 25 ppm/300 ppm; 20 ppm/350 ppm and 15 ppm/400 ppm, which account for more than 97.7% removal of colour and turbidity and 68.2% removal of compounds with absorption at $UV_{254\,nm}$.

All other physicochemical parameters evaluated in the study to produce potable water under operational conditions had rates which met the specifications of the EPA [14].

Despite having occurred a significant decrease in microbial load, the presence of total coliforms can still be perceived. It was ascertained the absence of *E. coli*, which had been already observed by Fatombi et al. [17]. Abaliwano et al. [8] and Mangale Sapana et al. [18] suggest that this can be an indication of bactericidal activity of *M. oleifera.* Chlorination should be performed so that the treated water would meet quality standards.

4. Conclusions

Current tests showed that the coagulation diagram is useful to determine the working conditions in coagulation/flocculation and sedimentation process, because it enabled the selection of better operational conditions among the experimental conditions tested.

By means of the combined coagulation/flocculation, sedimentation and filtration processes, it was possible to obtain drinking water, using dosages of aluminium sulphate/*M. oleifera* solution: 25 ppm/300 ppm; 20 ppm/350 ppm and 15 ppm/400 ppm, which resulted in more than 97.7% removal of colour and turbidity and 68.2% removal of compounds with absorption at $UV_{254\,nm}$.

Nevertheless, it is important to perform the disinfection step in order to assure the potability of water. *M. oleifera* associated with aluminium sulphate contributed towards a decrease in the amount of chemical agent used for water treatment. It might actually be an advantage and a promising step towards the improvement of water treatment process.

References

[1] UNICEF/WHO, Progress on drinking water and sanitation: 2012 update, WHO Library, New York, NY, 2012.

[2] M. Asrafuzzaman, A.N.M. Fakhruddin, M.A. Hossain, Reduction of turbidity of water using locally available natural coagulants, ISRN Microbiol. 2011 (2011) 1–6. doi:10.5402/2011/632189.

[3] G.S. Madrona, G.B. Serpelloni, A.M.S. Vieira, L. Nishi, K.C. Cardoso, R. Bergamasco, Study of the effect of saline solution on the extraction of the *Moringa oleifera* seed's active component for water treatment, Water Air Soil Pollut. 211 (2010) 409–415, doi:10.1007/s11270-009-0309-0.

[4] F.K. Amagloh, A. Benang, Effectiveness of *Moringa oleifera* seed as coagulant for water purification, Afr. J. Agricult. Res. 4 (2009) 119–123.

[5] M. Pritchard, T. Craven, T. Mkandawire, A.S. Edmondson, J.G. O'Neill, A comparison between *Moringa oleifera* and chemical coagulants in the purification of drinking water —An alternative sustainable solution for developing countries, Phys. Chem. Earth 35 (2010) 798–805, doi:10.016/j.pce.2010.07.014.

[6] C.P. Pise, D.S.A. Halkude, Blend of natural and chemical coagulant for removal of turbidity in water, IJCIET 3 (2012) 188–197.

[7] D. Schwarz, Water clarification using *Moringa oleifera*, Technical Information W1e, Gate Information Service, 2000, pp. 1–7.

[8] J.K. Abaliwano, K.A. Ghebremichael, G.L. Amy, Application of the purified *Moringa oleifera* coagulant for surface water treatment, WaterMill Working Paper Series, 5 (2008) 1–19.

[9] M. Awad, H. Wang, F. Li, Preliminary study on combined use of *Moringa* seeds extract and PAC for water treatment, Res. J. Recent Sci. 2 (2013) 52–55.

[10] N.E. Nwaiwu, A.A. Bello, Effect of *Moringa oleifera*-alum ratios on surface water treatment in north east Nigeria, Res. J. Appl. Sci. Eng. Technol. 3 (2011) 505–512.

[11] S.H. Kim, B. H. Moon, H. I. Lee, Effects of pH and dosage on pollutant removal and floc structure during coagulation, Microchem. J. 68 (2001) 197–203, doi:10.1016/S0026-265X(00)00146-6.

[12] K.C. Valverde, L.C.K. Moraes, M.C. Bongiovani, F.P. Camacho, R. Bergamasco, Coagulation diagrams using the *Moringa oleifera* Lam and the aluminium sulphate, aiming the removal of color and turbidity of water, Acta Sci. Technol. 5 (2013) 485–489, doi:10.4025/actascitechnol.v35i3.12268.

[13] APHA—American Public Health Association, Standard Methods for the Examination for Water and Wastewater, nineteenth ed., American Public Health Association, Washington, DC, 1995.

[14] EPA—Environmental Protection Agency, USA, National Primary Drinking Water Regulations, Baltimore, MD, 2009.

[15] M. Suleyman, A. Alfugara, Treatment of surface water with *Moringa oleifera* seed extract and alum—A comparative study using a pilot scale water treatment plant, Intern. J. Environ. Studies 60 (2003) 617–626.

[16] V.L.H. Westphal, W. Dias, R. Hickel, Teste dos Jarros, in Química Tecnológica Geral B (mimeo), ENQ/UFSC, Florianópolis, 1982.

[17] K.J. Fatombi, T.A. Ahoyo, O. Nonfodji, T. Aminou, Physico-chemical and bacterial characteristics of groundwater and surface water quality in the lagbe town: Treatment essays with *Moringa oleifera* seeds, J. Water Resour. Prot. 04 (2012) 1001–1008, doi:10.4236/jwarp.2012.412116.

[18] M. Mangale Sapana, S.G. Chonde, P.D. Raut, Use of *Moringa oleifera* (drumstick) seed as natural absorbent and an antimicrobial agent for ground water treatment, Res. J. Recent Sci. 1 (2012) 31–40.