Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2019.23255

143 (2019) 184–191
March

Exploring potential of pearl millet *(Pennisetum glaucum)* and black-eyed pea *(Vigna unguiculata* subsp. *unguiculata)* as bio-coagulants for water treatment

Ghulam Hussain*, Sajjad Haydar

Institute of Environmental Engineering & Research (IEER), University of Engineering & Technology (UET), Lahore, Pakistan, emails: ghussain@uet.edu.pk (G. Hussain), sajjad@uet.edu.pk (S. Haydar)

Received 29 April 2018; Accepted 9 October 2018

ABSTRACT

Plant-based natural coagulants can act as potential alternatives to most of the commonly used chemical coagulants. Not only plant-based coagulants have been found to be eco-friendly but also they can generally be classified as non-toxic and produce less sludge in comparison with chemical coagulants. In this study, the coagulation potential of crude extract from seeds of Pearl millet *(Pennisetum glaucum)* and Black-eyed pea *(Vigna unguiculata subsp. unguiculata)* was evaluated using lab-prepared samples having initial turbidity of 200 ± 5 NTU. Response surface method was used for the design of experiments while statistical analysis was also applied on the experimental results obtained. The obtained results revealed that pearl millet and black-eyed pea can effectively remove turbidity and produce water with a turbidity of less than 5 NTU. The statistical analysis revealed that pH was a more significant parameter as compared with coagulant dose. Pearl millet and black-eyed pea performed best in a pH range of 2–4 and 4–6, respectively. Fourier transform infrared spectroscopy and scanning electron microscopy were employed on coagulants and flocs formed. Mechanism of coagulation for pearl millet was found to be adsorption due to the carboxyl group and adsorption due to hydrogen bonding for black-eyed pea.

Keywords: Coagulation; Plant-based coagulants; Optimization; Coagulation mechanism

---

1. Introduction

The processes of coagulation–flocculation–sedimentation are essential steps in the treatment of surface waters. Conventionally, chemical coagulants, such as alum and different salts of iron have been used to carry out coagulation [1], with alum being the most commonly used coagulant in public water supplies [2,3]. However, the odd associated with alum is the presence of residual aluminum in the treated water. Various studies show that residual aluminum may be a causal factor leading to the occurrence of Alzheimer's disease [4–6]. In addition, alum produces a large amount of sludge [7] that poses a number of subsequent handling, treatment and disposal issues. Furthermore, it also alters the pH of the treated water. Meanwhile, the chemical cost may also become a problem where there is an issue of resource constraint [3].

Due to the aforementioned issues with chemical coagulants, the natural plant-based coagulants may serve as a suitable alternative for water treatment. Plant-based coagulants have long been recognized for their effectiveness in traditional water treatment [2,8,9]. In comparison with chemical coagulants, plant-based coagulants are eco-friendly [2,10] and generally toxin free [11]. Sludge produced by natural coagulants is much smaller in quantity as compared with the amount of sludge generated by chemical coagulants [12]. Lower the volume of sludge generated, lower will be the costs associated with its handling and disposal. Also, natural coagulants do not consume alkalinity hence pH adjustment costs can be avoided or circumvented during water treatment. Along with other advantages such as being non-toxic and less expensive, natural coagulants provide a sustainable water treatment method. Such low-cost and sustainable water

---

* Corresponding author.

treatment solutions are of special significance and interest for developing countries.

Various plant-based coagulants have remained the focus of studies in water treatment. These include *Moringa oleifera* seeds, nirmali seeds, tannin and *Opuntia ficus-indica* cactus [13–15]. Coagulation activity has also been reported for other plant-based coagulants such as common bean (*Phaseolus vulgaris*) [16], red bean (*Phaseolus vulgaris* sp.), red maize and sugar maize (*Zea mays* sp.) [17], *Jatropha curcas* and Guar gum [18], algal alginate [19], *Enteromorpha* extract (green algae) [20], *Plantago major* [21] and walnut shell [22] and *Opuntia stricta* [23]. Recent studies on vegetables and legumes have provided insights into less commonly known water purifying agents [24]. The suggested ingredient, responsible for coagulation activity in most of the plant-based coagulants, is cationic protein content [25].

Despite several studies pertaining to plant-based coagulants, there is a need to identify new plant-based coagulants with a focus not only on their water purification potential but also on their widespread availability and the potential for marketability. Above in view, the objective of this study was to explore the potential of pearl millet (*Pennisetum glaucum)* and black-eyed pea *(Vigna unguiculata* subsp. *unguiculata)* as natural coagulants for water treatment. This study is an effort to contribute, some new plant-based coagulants to the scientific data base, which can be used as economical and sustainable alternatives for water treatment.

Pearl millet (*Pennisetum glaucum)* is an important cereal crop of the semi-arid regions of the world, primarily Asia and Africa. It has been grown in Indian sub-continent and Africa since pre-historic times [26]. Pearl millet is adapted to grow in low rainfall, low soil fertility and high temperatures, making it suitable for be grown in tough environments. It is generally used as a pasture crop or as a food crop in some of the poorest countries in the world [27]. As reported in literature, plant-based coagulants are generally proteins or polysaccharides [15]. Pearl millet contains significant amount of proteins (11.6 g/100 g) and starch polysaccharides [28] making it a potential coagulant for water purification. Black-eyed pea *(Vigna unguiculata* subsp. *unguiculata)* is cultivated in all tropical areas and temperate climates. Its seeds are used as food while its leaves and young pods are used as fodder [29]. Even though the first domestication of this specie occurred in Africa, it is currently widely grown in many countries of Asia, Europe and some parts of United States [30]. Black-eyed pea seeds can contain up to 25% protein and a significant amount of starch polysaccharides [31], hence its extract can be turned into a potential bio-coagulant.

2. Materials and methods

2.1. Preparation of crude extracts of coagulants

Seeds of pearl millet and black-eyed pea were obtained from the local market. Seeds were washed with distilled water, sun-dried for 2 d and finally oven-dried at 103°C for 24 h. After drying, the moisture content was 13.5% and 18.3% for seeds of pearl millet and black-eyed pea, respectively. Dried seeds were ground to a fine powder using electric kitchen grinder (Cambridge GC-5026). The powder was sieved through mesh no. 40 (opening size 0.42 mm) to achieve particles of size smaller than 0.42 mm. The sieved particles were dissolved in distilled water and agitated by using a magnetic stirrer for 30 min. The suspension was then passed through Whatman filter paper No. 42 and the filtrate, referred to as crude extract, was used as coagulant stock solution.

2.2. Preparation of colloidal suspensions

Colloidal suspensions were prepared by adding kaolin particles to the tap water. 10 g of laboratory grade kaolin was added to 1 L of water and the suspension was stirred for 30 min using magnetic stirrer and then was allowed to settle for 24 h. The supernatant was carefully collected and stored in a plastic bucket. The kaolin suspension was diluted using tap water to produce initial turbidity of 200 ± 5 NTU and was used as a raw sample for the coagulation process.

2.3. Coagulation–flocculation–sedimentation

Phipps and Bird PB-900 jar tester was used to simulate coagulation–flocculation–sedimentation in the laboratory. Prepared colloidal suspension was poured into the 1 L beaker. pH was adjusted to the desired level using 1 M NaOH and 1 M HCl solutions, and the desired coagulant dose was added to each beaker.

Samples, after coagulation–flocculation–sedimentation, were withdrawn from sampling ports and analyzed for pH and residual turbidity. All the experimental runs were conducted in duplicate and mean values are reported. A blank was also run. The summary of the experimental parameters is presented in Table 1.

2.4. Analytical methods

Turbidity of samples was measured using 2100 AN Turbidimeter (Hach, USA) with range 0–10,000 NTU and pH was analyzed by Hach sensION 3 pH meter, with range –2.00 to 14.00 pH and accuracy ≤0.02 pH. To analyze the available functional groups in each coagulant and its coagulation process, Fourier transform infrared (FTIR) spectra of coagulant powder, suspended particles and flocs formed were recorded using Agilent Cary 600 series FTIR Spectrophotometer, make USA, with range 4,000–650. Scanning electron microscopy (SEM) images of the coagulants and flocs formed were obtained using VEGA3 TESCAN, make Czech Republic to observe the morphology of flocs.

2.5. Design of experiments, statistical analysis and optimization

To reduce the experimental runs and optimize the treatment conditions, a response surface method (RSM) design

Table 1
Experimental parameters

pH range	2–10
Dose (mg/L) range	20–100
Rapid mixing	200 rpm for 1 min
Slow mixing	30 rpm for 25 min
Settling (min)	30

was employed. RSM is a well-recognized statistical technique for evaluating and optimizing the results of treatment parameters [32]. The variables selected for design of experiments were pH and coagulant dose whereas, turbidity removal (%) was analyzed as the response. A software package of Design-Expert version 10 was used in this study for the design of experiments and statistical analysis. pH was varied from 2 to 10, while coagulant dose was varied from 20 to 100 mg/L. The design summary is presented in Table 2.

Results of analysis were tested for various models (i.e., linear, quadratic, cubic, etc.). The most suitable model was selected to study the effects of the variables on the response. Analysis of variance (ANOVA) was used to validate, statistically, the results of response and the suggested models. Desirability function available in the software was used for optimization of parameters for a target response.

3. Results and discussion

3.1. Turbidity removal efficiency of pearl millet and black-eyed pea

The experimental runs of both the natural coagulants, at various combinations of factors, and the response, that is, turbidity removal (%) is presented in Table 3.

It can be deduced from Table 3 that maximum removal of turbidity achieved by pearl millet was 99.2% (residual turbidity <2 NTU) at pH = 2 and dose = 80 mg/L, whereas the maximum turbidity removal obtained by black-eyed pea was 97.6% (residual turbidity <5 NTU) at pH = 4 and dose = 20 mg/L. The results of Table 3 were further analyzed by the 3-D response surface plots which are shown in Fig. 1. The surface plots represent the effect of pH and coagulant dose and their interactions on the turbidity removal. The peak of the plot represents the maximum removal and the contours at the bottom show the interaction of variables (pH and dose) and their effect on the response (turbidity removal efficiency).

The surface plot for pearl millet (Fig. 1(a)) showed that turbidity removal was highly dependent on pH and the effect of coagulant dose on turbidity removal was negligible. Maximum turbidity removal, that is, >96% could be obtained at pH range of 2–4 at all dose levels. As impurity particles carry a negative charge [15], it is likely that, at acidic pH range, natural electrolytes from the crude extract of pearl millet, facilitated adsorption. Acidic pH range, however, may limit the suitability of pearl millet as the pH of surface waters is usually near neutral and for drinking water supply it must be in the range of 6.5–8.5 [33]. Nevertheless, owing to its very high coagulation activity, pearl millet could be used, effectively, for treating industrial effluents with acidic pH range.

For Black-eyed pea, the surface plot (Fig. 1(b)) indicated that effect of pH on its coagulation activity was more as compared with coagulant dose. However, the trend of turbidity removal with change in pH was not linear. Turbidity removal was maximum (89%–97%) at pH range of 4–6 and reduces with decrease or increase in pH beyond this range. Hence, black-eyed pea could be, appositely, used for treating surface water with little or no pH adjustments. In this optimized pH range (4–6), lower coagulant doses (20–40 mg/L) produced greater turbidity removal, that is,. >95%. Lower doses would result in lesser costs associated with treatment, hence it is a virtual advantage of black-eyed

Table 3
Design matrix and response (turbidity removal) for pearl millet and black-eyed pea

Experimental run	Factors		Response	
	A: pH	*B*: Dose (mg/L)	Turbidity removal efficiency (%)	
			Pearl millet	Black-eyed pea
1	10	40	83.7	39.5
2	2	60	97.3	55
3	2	40	97.9	65.5
4	10	100	45	43
5	6	100	47.4	77.8
6	10	20	50	47.3
7	4	60	98.4	90.4
8	6	60	39	42.5
9	8	40	38.8	41
10	4	100	57.7	89.5
11	8	100	69	38
12	6	40	39.7	82.8
13	4	40	90	95
14	8	60	38.8	37
15	2	80	99.2	46.6
16	2	100	98.9	48.7
17	4	80	89.5	87
18	6	80	52.3	79.4
19	6	20	48	96.8
20	4	20	82	97.6
21	10	60	38	37.6
22	10	80	48	21.5
23	2	20	96.7	76.3
24	8	80	46	82.9
25	8	20	32.8	64

Table 2
Response surface design summary for experimental runs

Study type		Response surface	Subtype		Randomized	
Design type		User defined	Run		25	
Factor	Name	Type	Minimum	Maximum	Code values	
A	pH	Numeric (discrete)	2	10	–1.000 = 2	1.000 = 10
B	Dose, mg/L	Numeric (discrete)	20	100	–1.000 = 20	1.000 = 100

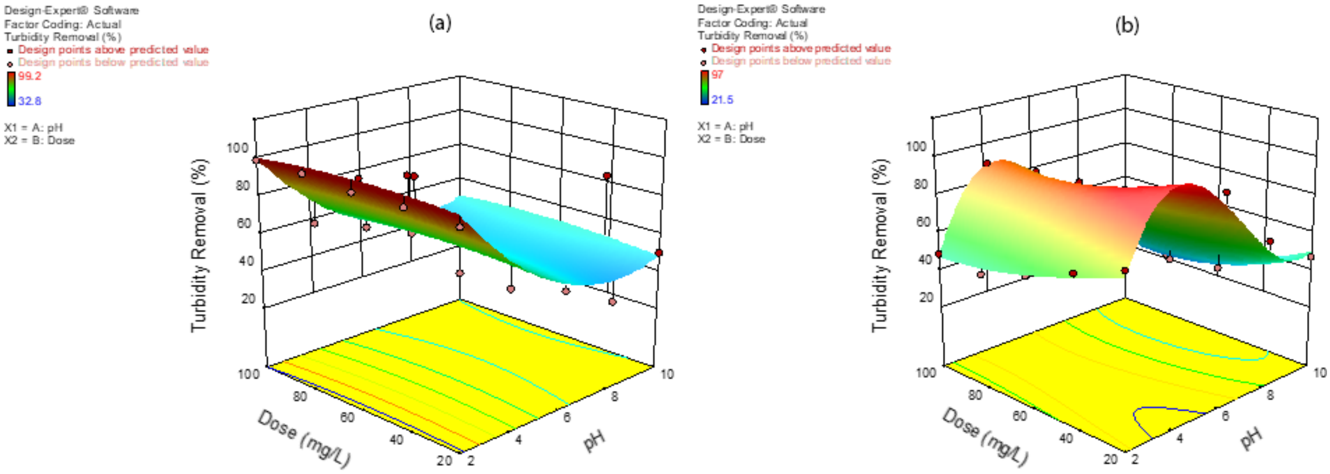

Fig. 1. 3-D surface plot showing effect of factors (pH and dose) on turbidity removal. (a) Pearl millet and (b) black-eyed pea.

pea. Coagulation activity reduced at high doses which showed that overdosing could have re-stabilized the colloidal particles.

Turbidity removal efficiencies achieved, by both pearl millet and black-eyed pea, were significantly higher than many plant-based coagulants reported in literature. A graphical comparison of maximum turbidity removal achieved by pearl millet and black-eyed pea with various bio-coagulants, reported in literature, is shown in Fig. 2 [15,24].

3.2. Optimization of pH and coagulant dose

Desirability function of Design-Expert software was used to optimize the variables. The initial turbidity of raw water was 200 NTU, therefore target turbidity removal of >97.5% was set to achieve residual turbidity of 5 NTU as per WHO guidelines for drinking water quality [33]. Optimum pH of 2.3 and dose of 38 mg/L was selected for pearl millet to achieve 97.5% turbidity removal (5 NTU residual). Similarly, using the same approach, optimum pH of 5.4 and dose of 21 mg/L was selected for black-eyed pea to achieve 97.5% turbidity removal (5 NTU residual).

3.3. Statistical analysis

The data of response analyzed (i.e., turbidity removal) was fitted to various models (i.e., linear, 2FI, quadratic, cubic, quadratic) and the statistical results are given in Table 4.

The selection of best fitted model was made based on five statistical tests, that is, for a model to be best fit, its standard deviation should be lowest, PRESS should be lowest, sequential *p*-value should be less than 0.05 (alpha value), and most importantly, the difference between adjusted R^2 and predicted R^2 should be less than 0.2 (the tolerable statistical value). The prediction model fulfilling all or most of these statistical checks was designated as best fitted model. The results in Table 4 showed that turbidity removal was best represented by a quadratic model for pearl millet, whereas for black-eyed pea it is best described by a cubic model. The equations of the suggested models for each coagulant in terms of coded factors are as follows:

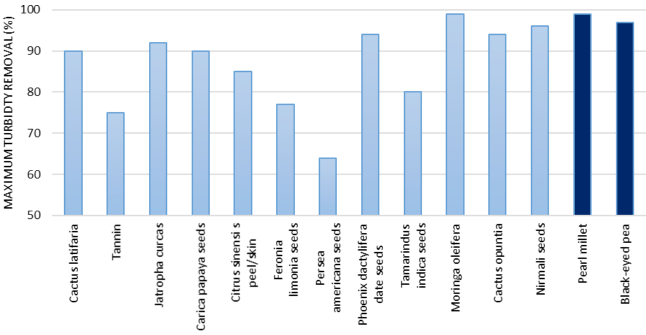

Fig. 2. Comparison of turbidity removal by pearl millet and black-eyed pea with other bio-coagulants.

Turbidity removal (for pearl millet)

$$= +54.32 - 25.89A - 0.1B + 1.04AB + 23.89A^2 - 2.78B^2$$

Turbidity removal (for black-eyed pea)

$$= +75.01 - 61.4A - 8.39B + 2.69AB - 32.04A^2 + 8.53B^2 - 2.49A^2B + 3.95AB^2 + 49.17A^3 + 1.28B^3$$

where *A* represents the pH and the coagulant dose is represented by *B*. These equations can be used to predict the response for each given level of factors. These equations are also useful for identifying the relative significance of factors by comparing the factor coefficients (higher the value of coefficient, greater is the significance of the factor). By looking at the coefficients it is evident that factor *A*, i.e., pH is more significant for both the coagulants as compared with the dose of the coagulants.

The ANOVA results, for the models and factors, are shown in Table 5.

The model *F*-value of 9.51 for pearl millet and 31.89 for black-eyed pea infer that both models are significant. There is only a 0.01% chance that *F* value this large can occur due to noise. For pearl millet, the “predicted *R*-squared” of 0.5145 is in reasonable agreement with “adjusted *R*-squared” of 0.6491, that is, the difference is less than 0.2 (the tolerable statistical value). “Adeq precision” measures the signal to noise ratio where a ratio greater than 4 is desirable. The ratio of 8.04

Table 4
Model-fitting summary statistics

Source	Standard deviation	Sequential *p*-value	Adjusted R^2	Predicted R^2	PRESS	
Pearl millet						
Linear	17.79	0.0002	0.5140	0.3881	9,162.86	
2FI	18.22	0.8884	0.4902	0.3359	9,944.28	
Quadratic	15.11	0.0134	0.6491	0.5145	7,269.60	Suggested
Cubic	15.48	0.5499	0.6320	0.2291	11,543.38	
Quadratic	12.57	0.1156	0.7572	-1.0012	29,966.26	
Black-eyed pea						
Linear	19.40	0.0064	0.3110	0.1851	10,679.93	
2FI	19.80	0.7369	0.2821	0.1104	11,659.19	
Quadratic	13.42	0.0002	0.6703	0.5489	5,912.76	
Cubic	4.59	<0.0001	0.9205	0.8133	2,446.90	Suggested
Quadratic	5.49	0.1209	0.9448	0.7218	3,646.66	

Table 5
Results of ANOVA for performance of pearl millet and black-eyed pea

Source	Pearl millet			Black-eyed pea		
	Quadratic model			Cubic model		
	F value	Coefficient	*p*-Value (Probability > *F*)	*F*-Value	Coefficient	*p*-Value (Probability > *F*)
Model	9.51	54.32 (intercept)	0.0001	31.89	75.01 (intercept)	<0.0001
A-pH	36.55	–25.89	<0.0001	104.08	–61.47	<0.0001
B-dose	5.583E-004	–0.10	0.9814	1.94	–8.39	0.1839
AB	0.029	1.04	0.8659	1.05	2.70	0.3225
A^2	10.90	23.89	0.0040	103.52	–32.05	<0.0001
B^2	0.15	–2.78	0.7052	7.35	8.54	0.0161
A^2B	–	–	–	7.35	–2.49	0.0161
AB^2	–	–	–	0.31	3.95	0.5842
A^3	–	–	–	0.79	49.17	0.3887
B^3	–	–	–	62.68	1.28	<0.0001
R-squared	0.7254			0.9503		
Adjusted *R*-squared	0.6491			0.9205		
Predicted *R*-squared	0.5145			0.8133		
Adeq precision	8.049			18.57		

Note: Significance level (α) = 0.05.

indicates an adequate signal. Similarly, for black-eyed pea, the "predicted *R*-squared" of 0.8133 is in reasonable agreement with "adjusted *R*-squared" of 0.9503, that is, the difference is less than 0.2. "Adeq precision" value of 18.57 indicates very good signal to noise ratio. All the statistical measures indicate that these models can be used to navigate the design space.

p-Value represents the implication of model terms; a value of <0.05 (significance level) indicates that model terms are significant. In this case, for pearl millet, the linear and quadratic terms of pH are significant and the terms regarding dose are not significant. Similarly, for black-eyed pea, linear and quadratic terms of pH with addition of quadratic and cubic terms of dose are significant. This implies that for black-eyed pea dose is also significant in combination with pH and in higher orders. Results of statistical tools support the inferences earlier made by Table 3 and Fig. 1.

3.4. Characterization of coagulants and flocs

3.4.1. Fourier transform infrared spectral analysis

FTIR spectra of both the bio-coagulants (Fig. 3), suspended solids present in untreated sample (Fig. 4(a)) and flocs produced from water samples treated with bio-coagulants (Figs. 4(b) and (c)) are shown. Pearl millet (Fig. 3(a)) had a broad and strong band at 3,269 cm^{-1} due to O–H stretching whereas the presence of CH_2 group (alkanes) was indicated at 2,923 cm^{-1}. Additional peaks at 1,645 and 1,542 cm^{-1}

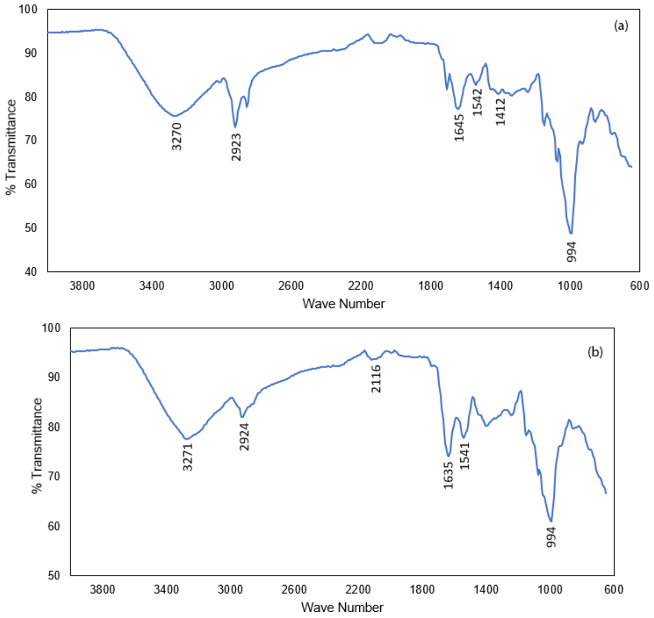

Fig. 3. FTIR spectra of bio-coagulants powder. (a) Pearl millet and (b) black-eyed pea.

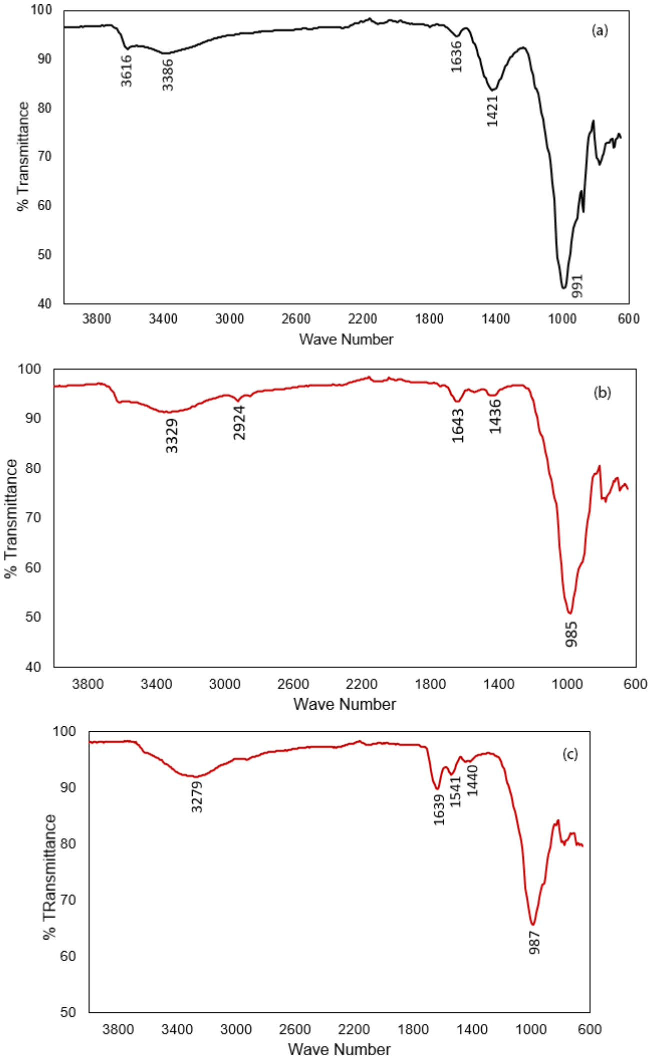

Fig. 4. FTIR of suspended solids and flocs. (a) Suspended solids, (b) flocs produced from pearl millet and (c) flocs produced by black-eyed pea.

represented the presence of NH stretching vibrations in primary and secondary amides, respectively. The weak band at 1,412 cm^{-1} was due to bending vibrations of CH_3, most likely indicating the presence of COOH (carboxyl) group [34].

Similarly, in the spectra of black-eyed pea (Fig. 3(b)), peaks at 3,271 and 2,924 cm^{-1} represented the presence of O–H stretching and CH_2 group (alkanes), respectively. Additional peaks at 1,635 and 1,541 cm^{-1} indicated the presence of NH stretching vibrations in primary and secondary amides, respectively.

It can be deduced that the flocs (Figs. 4(b) and (c)) and the suspended solids (Fig. 4(a)) showed several similarities in peaks in their spectra. The strong band at 1,421 cm^{-1} present in the spectra of suspended solids, was reduced in the flocs treated with both coagulants, indicating interaction between suspended solids and coagulants. The occurrence of an additional peak at 2,924 cm^{-1} (C–H group) was observed in flocs treated with pearl millet. This additional peak also indicated the interaction between pearl millet and suspended solids. It is supposed that carboxyl group present in pearl millet provided adsorption sites for the suspended solids during the coagulation process [15]; therefore, the coagulation mechanism seems to be adsorption. Likewise, an additional peak was observed in flocs treated with black-eyed pea at 1,541 cm^{-1} (NH group). This additional peak indicated that the N–H group formed intermolecular hydrogen bonds between the coagulant and suspended solids to aid in coagulation [35]. As adsorption may occur through hydrogen bonding [36], it is suggested that coagulation mechanism for Black-eyed pea is also adsorption.

3.4.2. Scanning electron microscopy examination

SEM examination of coagulants and flocs was performed to analyze their surface morphology and characteristics. Flocs formed after coagulation process were separated via sedimentation and drying. SEM images of pearl millet seed powder showed spherical particles (Fig. 5(a)) and a similar pattern was observed in the flocs formed by it (Fig. 5(c)). However, flocs were relatively compact and bigger in size with visible formation of aggregates (Fig. 5(c)). This relatively higher compactability and large sizes are evidence of formation of aggregates through the coagulation–flocculation process.

Conversely, SEM images of black-eyed pea showed a fibrous structure (Fig. 5(b)). Flocs formed by black-eyed pea also showed a fibrous structure similar to that of the coagulant itself but with more compactness (Fig. 5(d)). These aggregates were, presumably, formed through adsorption (as demonstrated by IR spectra) and inter-particle bridging (as indicated by the morphology of the flocs).

3.5. Cost of treatment

Cost of treatment is an important consideration, especially in the context of developing countries. The cost of bio-coagulants used in this study was estimated to treat 1,000 m^3 of water and is shown in Table 6. The cost of alum

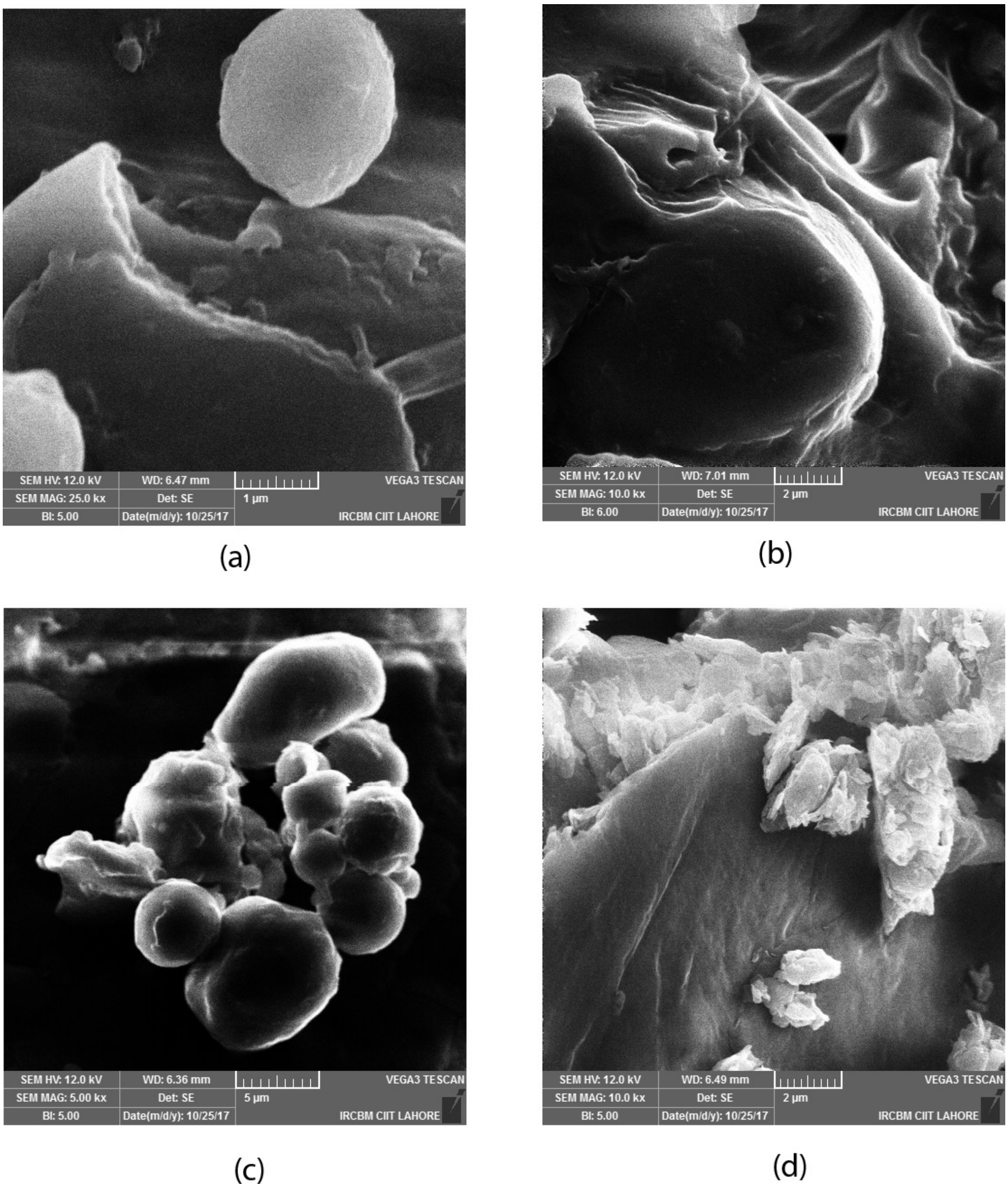

Fig. 5. SEM images of bio-coagulants and flocs produced by them. (a) Pearl millet, (b) black-eyed pea, (c) flocs produced by pearl millet and (d) flocs produced by black-eyed pea.

Table 6
Cost comparison of selected bio-coagulant with chemical coagulant

Coagulant	Optimum dose (mg/L)	Unit cost (US$/kg)	Cost of treatment (US$/1,000 m^3 of water)
Alum	12	3.47	41.6
Pearl millet	76	0.93[a]	70.6
Black-eyed pea	21	1.11[a]	23.3

[a] Cost of bio-coagulants includes cost of processing (grinding, sieving, filtration, etc.).

with its optimum dose for treating water with comparable turbidity is also provided for comparison [37].

Table 6 shows that it costs USD 41.6 to treat 1,000 m^3 of water with alum. The cost almost reduces to half with the use of black-eyed pea (USD 23.3). However, pearl millet proved to be even costlier than alum. In addition to the low costs incurred, black-eyed pea is also eco-friendly and has food grade quality. This could make it a much better replacement of alum.

4. Conclusions

This study demonstrates that pearl millet and black-eyed pea were efficient in treating turbid water. Pearl millet worked best in a pH range 2–4 with little effect of dose change on turbidity removal. Maximum turbidity removal (2 NTU from 200 NTU) was achieved for pearl millet at a pH 2 and a coagulant dose of 80 mg/L. The mechanism of coagulation activity for pearl millet is suggested to be adsorption due to the carboxyl group. Black-eyed pea, on the other hand, worked best in a pH range of 4–6 and low coagulant doses, that is, 20–40 mg/L. High doses of black-eyed pea produced lower efficiencies at selected pH, which shows the phenomena of overdosing. Maximum turbidity removal (5 NTU from 200 NTU) was achieved for black-eyed pea at pH 4 and dose of 20 mg/L. Suggested mechanism of coagulation is adsorption due to hydrogen bonding. The cost of treating 1,000 m^3 of water with alum, pearl millet and black-eyed pea is USD 41.6, 70.6 and 23.3, respectively.

References

[1] S.A.A. Jahn, Drinking water from Chinese rivers: challenges of clarification, J. Water Supply Res Technol., 50 (2001) 15–27.

[2] J. Bratby, Coagulation and Flocculation in Water and Wastewater Treatment, IWA Publishing, London, UK, 2006.

[3] M. Kang, T. Kamei, Y. Magara, Comparing polyaluminum chloride and ferric chloride for antimony removal, Water Res., 37 (2003) 4171–4179.

[4] T.P. Flaten, Aluminium as a risk factor in Alzheimer's disease, with emphasis on drinking water, Brain Res. Bull., 55 (2001) 187–196.

[5] E. Gauthier, I. Fortier, F. Courchesne, P. Pepin, J. Mortimer, D. Gauvreau, Aluminum forms in drinking water and risk of Alzheimer's disease, Environ. Res., 84 (2000) 234–246.

[6] J. Walton, Aluminum involvement in the progression of Alzheimer's disease, J. Alzheimer 's Dis., 35 (2013) 7–43.

[7] G. Newcombe, D. Dixon, Coagulation and flocculation with organic polyelectrolytes, Interface Sci. Drinking Water Treat. Theory Appl., 10 (2006) 63.

[8] C. Dorea, Use of *Moringa* spp. seeds for coagulation: a review of a sustainable option, Water Sci. Technol. Water Supply, 6 (2006) 219–227.

[9] R. Sanghi, B. Bhatttacharya, V. Singh, *Cassia angustifolia* seed gum as an effective natural coagulant for decolourisation of dye solutions, Green Chem., 4 (2002) 252–254.

[10] V. Govindan, Coagulation studies on natural seed extracts, J. Indian Water Works Assoc., 37 (2005) 145.

[11] M. Asrafuzzaman, A. Fakhruddin, M.A. Hossain, Reduction of turbidity of water using locally available natural coagulants, ISRN Microbiol., 2011 (2011) 1–6.

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

[13] S.M. Miller, E.J. Fugate, V.O. Craver, J.A. Smith, J.B. Zimmerman, Toward understanding the efficacy and mechanism of *Opuntia* spp. as a natural coagulant for potential application in water treatment, Environ. Sci. Technol., 42 (2008) 4274–4279.

[14] G. Vijayaraghavan, T. Sivakumar, A.V. Kumar, Application of plant based coagulants for waste water treatment, Int. J. Adv. Eng. Res. Stud., 1 (2011) 88–92.

[15] C.-Y. Yin, Emerging usage of plant-based coagulants for water and wastewater treatment, Process Biochem., 45 (2010) 1437–1444.

[16] M.B. Šćiban, M.G. Antov, M.T. Klašnja, Extraction and partial purification of coagulation active components from common bean seed, Acta Period. Technol., 37 (2006) 37–43.

[17] K. Gunaratna, B. Garcia, S. Andersson, G. Dalhammar, Screening and evaluation of natural coagulants for water treatment, Water Sci. Technol. Water Supply, 7 (2007) 19–25.

[18] M. Pritchard, T. Mkandawire, A. Edmondson, J. O'neill, G. Kululanga, Potential of using plant extracts for purification of shallow well water in Malawi, Phys. Chem. Earth, 34 (2009) 799–805.

[19] H.A. Devrimci, A.M. Yuksel, F.D. Sanin, Algal alginate: a potential coagulant for drinking water treatment, Desalination, 299 (2012) 16–21.

[20] S. Zhao, B. Gao, X. Li, M. Dong, Influence of using *Enteromorpha* extract as a coagulant aid on coagulation behavior and floc characteristics of traditional coagulant in Yellow River water treatment, Chem. Eng. J., 200 (2012) 569–576.

[21] N.A. Oladoja, Perspectives on the use of equilibrium isotherm equations to elucidate coagulation–flocculation mechanisms in plant-based coagulants, Ind. Crops Prod., 70 (2015) 211–212.

[22] M. Siddique, S.A. Soomro, S. Aziz, A.S. Jatoi, A. Mengal, H. Mahar, Removal of turbidity from turbid water by bio-coagulant prepared from walnut shell, J. Appl. Emerg. Sci., 6 (2016) 66–68.

[23] A. Mukhtar, W. Ali, G. Hussain, A preliminary study of *Opuntia stricta* as a coagulant for turbidity removal in surface waters, Proc. Pak. Acad. Sci., 52 (2015) 117–124.

[24] S.Y. Choy, K.M.N. Prasad, T.Y. Wu, M.E. Raghunandan, R.N. Ramanan, Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification, J. Environ. Sci., 26 (2014) 2178–2189.

[25] I. Bodlund, Coagulant Protein from Plant Materials: Potential Water Treatment Agent, KTH Royal Institute of Technology, Stockholm, Sweden, 2013.

[26] K. Manning, R. Pelling, T. Higham, J.-L. Schwenniger, D.Q. Fuller, 4500-year old domesticated pearl millet (*Pennisetum glaucum*) from the Tilemsi Valley, Mali: new insights into an alternative cereal domestication pathway, J. Archaeol. Sci., 38 (2011) 312–322.

[27] V.S. Nambiar, J. Dhaduk, N. Sareen, T. Shahu, R. Desai, Potential functional implications of pearl millet (*Pennisetum glaucum*) in health and disease, J. Appl. Pharm. Sci., 10 (2011) 62.

[28] C. Gopalan, B. Rama Sastri, S. Balasubramanian, Nutrition Value of Indian Foods, National Institute of Nutrition, Indian Council of Medical Research Hyderabad, India, 2003.

[29] F.S. Ba, R.S. Pasquet, P. Gepts, Genetic diversity in cowpea [*Vigna unguiculata* (L.) Walp.] as revealed by RAPD markers, Genet. Resour. Crop Evol., 51 (2004) 539–550.

[30] B.G. Cook, B.C. Pengelly, S. Brown, J. Donnelly, D. Eagles, M. Franco, J. Hanson, B.F. Mullen, I. Partridge, M. Peters, Tropical Forages: an Interactive Selection Tool, CIAT and ILRI, Brisbane, Australia, 2005.

[31] A. Rangel, G.B. Domont, C. Pedrosa, S.T. Ferreira, Functional properties of purified vicilins from cowpea (*Vigna unguiculata*) and pea (*Pisum sativum*) and cowpea protein isolate, J. Agric. Food Chem., 51 (2003) 5792–5797.

[32] J. Fu, Y. Zhao, X. Xue, W. Li, A. Babatunde, Multivariate-parameter optimization of acid blue-7 wastewater treatment by Ti/TiO_2 photoelectrocatalysis via the Box–Behnken design, Desalination, 243 (2009) 42–51.

[33] F. Edition, Guidelines for Drinking-water quality, WHO Chronicle, 38 (2011) 104–108.

[34] F. Ni, X. Peng, J. He, L. Yu, J. Zhao, Z. Luan, Preparation and characterization of composite bioflocculants in comparison with dual-coagulants for the treatment of kaolin suspension, Chem. Eng. J., 213 (2012) 195–202.

[35] J. Fatombi, B. Lartiges, T. Aminou, O. Barres, C. Caillet, A natural coagulant protein from copra (*Cocos nucifera*): isolation, characterization, and potential for water purification, Sep. Purif. Technol., 116 (2013) 35–40.

[36] J.C. Crittenden, R.R. Trussell, D.W. Hand, K.J. Howe, G. Tchobanoglous, MWH's Water Treatment: Principles and Design, John Wiley & Sons, New Jersey, USA, 2012.

[37] S. Haydar, H. Ahmad, J.A. Aziz, Optimization of coagulation-flocculation in the treatment of canal water, Environ. Eng. Manage. J., 9 (2010) 1563–1570.