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Turbidity removal using natural coagulants derived from the seeds of *strychnos potatorum*: statistical and experimental approach

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Abstract. Providing sufficient quantity of drinking water to the inhabitants of poor countries and rural areas is one of the difficult challenges for water treatment authorities, where it was reported, by the World Health Organisation (WHO), that nowadays there are more than 800 million people in the poor countries and rural areas do not have sufficient quantity of drinking water, which subjected those people for outbreaks of different water-related diseases. Water companies therefore seek to provide affordable and reliable methods of treatment in order to solve this life-threatening issue. In this vein, the goal of the current study is to develop and analyse the effectiveness of affordable eco-friendly coagulants that derived from the seed of *Strychnos potatorum*. The applicability of this new type of coagulants for water treatment has been validated by treating turbid water sample, which was synthesized using kaolinites. The experimental work was also focused on optimising the effects of coagulants dosage; retention period and pH of water on turbidity removal by applying the Response Surface Methodology (RSM). The final results demonstrated that the new eco-friendly coagulants are suitable for water treatment as they removed about 93% of kaolinite turbidity when the unit was operated at natural pH (7) for 70.0 min with a dosage of the new coagulants of 40.0 mg.L^{-1}.

1. Introduction

A considerable number of poor countries around the world facing a shortage in drinking water supplying [1]. The published reports and studies evidenced that 58% of the global rural areas do not have sufficient amount of safe drinking water for their daily needs, while 42% have a reliable supply of drinking water, because of the continuous discharge of wastewaters and the upsurge in water consumptions [2-6]. Moreover, the World Health Organisation (WHO) reported that the lack of drinking water, during the last 20 years, left more than 20.0% of the global population without access for drinking water [3]. The report of the WHO was based on an assumption that each person needs, at least, 20 L of safe drinking water per day [3]. Unfortunately, a considerable body of recent studies demonstrated that the several factors have contributed in increasing the lack of safe drinking water; these factors are the climate change that increased the consumption of water, the increase in the production of wastewaters (industrial, agricultural and domestic wastewaters) [7, 8]. In addition, the global population is rapidly growing that maximises the consumption of water [2, 7].

As a result of the increase in the lack of drinking water, residents of poor areas/countries nowadays depend of polluted freshwater sources to secure water for drinking, cleaning and cooking purposes. For example, people in some parts of Asia and Africa nowadays use ground water, without treatment or with poor treatment, for drinking purposes [9-12]. Poor infrastructure and economy are the main barriers for using of efficient and advanced water treatment methods, such as membrane and reverse osmosis [13-16].

Thus, scientists and engineers carried out several to develop cost-effective and efficient water treatment technologies to provide enough quantity of safe water for the inhabitants of poor areas and countries, such electrochemical and filtration methods [17-22]. Though numerous technologies, such as electrocoagulation and adsorption methods, were practiced as potential solutions to provide the poor areas/countries with enough quantity of safe water, chemical coagulation (CC) is currently considered as the most affordable and suitable method for such areas and countries [20, 21, 23, 24].

The CC method utilises the capability of certain kinds of chemicals, named coagulants, to destabilize water contaminants through the difference in surface electrical charges; followed by the formation of aggregates that removed from water as sludge [10]. Although a wide range of organic and inorganic coagulants were practiced in water treatment industry, aluminium-based coagulants (such as aluminium sulphate and aluminium chloride) and iron-based coagulants (such as ferric sulphate and ferrous sulphate) are the most commonly used kinds of chemical coagulants in water treatment [25]. The effectiveness of the aluminium-based and iron-based coagulation agents is approved in the elimination of numerous contaminants from water, for instance the literature showed that these coagulants can remove up to 99.0% of the organic matters from water. Nevertheless, the literature links the use of chemical coagulants in water purification to several health and economic problems. For instance, many studies demonstrate that the residual chemicals in the purified water, especially the residuals of aluminium-based coagulants, causes serious diseases, such as Alzheimer and different types of cancers [26]. In addition, the residuals of aluminium-based and iron-based coagulants, especially iron-based coagulants, resulted in clocking of pipes, which results in the need to replace these pipes or the need for expensive maintenance (economic losses) [26]. Furthermore, the chemical coagulants maximizes the volumes of the sludge [7, 27], that maximizes the costs of treatment as more treatment steps and management strategies are required for sludge treatment [28-30], and a proper landfills are sometimes required to dispose the sludge [31]. However, some researchers tied to minimize the economic impacts of the produced sludge by recycling it in different industries, especially in construction industry [32-35].

Hence, various researches and studies were recently aimed at the development of safe and cost-effective coagulants (from natural materials), such as *nirmali seeds and tannins*, as alternative to the artificial coagulation agents. Similarly, the present study focuses on the cost-effective production of eco-friendly coagulants from natural raw materials (seeds of *Strychnos potatorum*) that could be safely used for water treatment (kaolinite as a case study) taking into considerations the effects of coagulants dosage, pH of solution and detention periods.

2. Methodology

2.1. Artificial water sample

The artificial water samples, which used the current research, was made according the standard procedures stated by [36]:

10,000 milligrams of kaolinite were added to 1000 millilitre of deionised water and well stirred for 1 hour to create homogeneous suspension, and then it was left for one day at the room temperature to completely hydrate kaolinite particles. The hydrated solution is the stock solution that used in the presented research to create diluted samples (with lower concentrations). To carry out the coagulating experiments, 2.50 millilitre of the concentrated sample was pipetted to 0.50 L of deionised water to generate 35 NTU of turbidity (this step usually carried out immediately before any experiment to avoid any change in the turbidity level).

It should be mentioned that the artificial turbidity, in the present research, was generated using kaolinite because the latter is broadly used in industry, such as coatings, paint, ceramic and toilet detergents industries. Additionally, the removal of kaolinite from solutions is difficult because the particles of the material are very small in sizes (about 2 µm) that significantly decreases their settle-ability [37].

2.2. Coagulants preparation
Manufacturing of the eco-friendly coagulants, derived from the Seeds of *Strychnos potatorum*, was initiated by cleaning the seeds, then milling process was commenced for 2 minutes using commercial food processor (model: Magimix 5200-XL). A 0.4 mm sieve was then used to sieve the grinded seeds (to remove unwanted sizes), 250 g of the passed particles was placed in a 2 litters plastic container, and 1000 millilitres of solvent was decanted and well mixed to create homogenous solution [38].
It is noteworthy to mention that the solvent, used in the present research, was synthetized according the stated procedures by Muthuraman and Sasikala [38], where 0.5 M of NaCl was added to 1000 millilitre of deionised water and stirred until the whole amount of NaCl is completely dissolved. The solvent-seeds suspension was filtered at two stages to remove the residuals of the seeds, the initial stage of filtration was commenced using Whatman filter paper no. 42; then the filtrate was filtered again on 0.45 µm. The collected filtrate was used as eco-friendly coagulants in the present research. Problems of coagulants aging, and coagulants consumption by bacteria were avoided by preparing and using the coagulants during the same time of experiments.
Progress of turbidity removal from water was regularly measured, at the intervals mentioned in section 2.3., by taking a sample at each interval from filtration unit and filtering it through filter papers no.2 (Whatman brand) to separate unwanted particles. The filtered samples were used to calculate the remaining turbidity using a turbidity meter (TL-2350 meter).

Turbidity removal (R.E.) has been determined using the following formula [39]:

$$R.E.\ (\%) = \frac{Initial\ turbidity\ level - measured\ turbidity\ level}{Initial\ turbidity\ level} \times 100\% \quad (1)$$

2.3. Optimisation process
As it is well known that the response surface methodology (RSM) is a family of different techniques; a considerable number of the published researches used the central composite design (CCD) (a kind of RSM) to design and optimise the experimental works as this technique enjoys many merits, such the capacity to simulate the effects of several factors simultaneously, the yields accurate results and it minimises the number the experiments [40]. The CCD was applied to a pH range of 5-9, dosages of coagulants of 30-50 mg/L, and retention periods of 30-60 min. Minitab package (version 19.20) has been used in the current research perform CCD. The latter showed that the smallest required number of the experimental trails, to perform the optimisation process, are 19 experiments at a star point (α) of ± 1.680, as listed in Table 1.

Table 1. α value for the investigated factors.

D (mg. L^{-1})		**DT (min)**		**pH**	
-α	**+α**	**-α**	**+α**	**-α**	**+α**
23.180	56.820	19.770	70.230	3.640	10.360

Application of the CCD to optimise the performance of the new eco-friendly coagulants yielded a set of combinations of experiments (consists of 19 experiments), see Table 2. This set of experiments has been commenced under controlled conditions at the IC laboratories in Liverpool John Moores University. The obtained removal of turbidity, from each experiment, was recorded and analysed later to develop a regression model that could be used to simulate the performance of the new eco-friendly coagulants in the elimination of kaolinite turbidity from artificial water (see the next part of the current research).

Table 2. Design of turbidity removal experiments using the new eco-friendly coagulants.

Run	D (mg. L^{-1})					DT (min)					pH				
	-α	**30**	**40**	**50**	**+α**	**-α**	**30**	**45**	**60**	**+α**	**-α**	**5**	**7**	**9**	**+α**
1.0		×					×					×			
2.0				×			×					×			
3.0		×							×			×			
4.0				×					×			×			
5.0		×					×							×	
6.0				×			×							×	
7.0		×							×					×	
8.0				×					×					×	
9.0	×							×					×		
10.0					×			×					×		
11.0			×			×							×		
12.0			×							×			×		
13.0			×					×			×				
14.0			×					×							×
15.0			×					×					×		
16.0			×					×					×		
17.0			×					×					×		
18.0			×					×					×		
19.0			×					×					×		

3. Results and discussion

3.1. Experimental study and optimisation

As it was explained in the previous section of the present research, the CCD method was used for optimising the performance of the new eco-friendly coagulants in terms of kaolinite turbidity removal from water. The application of CCD to the studied parameters yielded the 19 experimental runs, see Table 2, which have been commenced in the laboratory under control conditions. At the end of the desired detention period, the removal of turbidity was measured and recoded to be analysed later, see Table 3. According to the recorded removals of turbidity using the new eco-friendly coagulants, the optimum removal of the turbidity was 93.30% that was attained after 70.23 min using 40 mg/L of the new coagulants. It was also noticed that the neutral pH is the optimum level for efficient performance of the new eco-friendly coagulants in terms of kaolinite turbidity removal from water.

According to the published literature, the enhancement in the removal of turbidity by the new eco-friendly coagulants at the neutral pH level is explained by the change in the surficial charges of the bio-sorbents. At low pH, the bio-sorbents acquire a positive charge that causes a decrease in the attraction force between the bio-sorbents and contaminates. Similarly, as the pH increase to basic level; anionic hydroxides are developed, which consequently decrease the removal of water contaminants [23].

In terms of effects of the detention time on the ability of the new eco-friendly coagulation agents to remediate kaolinite turbidity from solution, the obtained results in Table 3 evidence that the removal of turbidity is positively affected by the contact times, especially in the course of the initial period of treatment. The enhancement in the removal of kaolinite turbidity with detention time is due to the fact that the longer detention times increase collusions between kaolinite particles and coagulation agents, which results in better growth of flocks, and consequently in better turbidity removal. Nevertheless, after a certain period of time, a state of balance was noticed, that leads to a clear reduction in the effects of the detention period. A similar behaviour was found as the dosage of the new coagulants increased. For instance, Table 3 shows that increasing the doses of the new eco-friendly coagulants led to a clear increase in the removal of kaolinite turbidity. The explanation for the enhancement in the removal of kaolinite with the dosage of the coagulants is the increase in effective surface area of coagulants that is ready for adsorption of coagulants, leading to a better removal of turbidity [38].

The results of the present research are agree with the results of some of the published articles, such as the results of Muthuraman and Sasikala [38].

Table 3. Results of removal of kaolinite turbidity by the new eco-friendly coagulants.

Run	D ($mg.L^{-1}$)					DT (minute)					pH					Observed removal (%)	Predicted removal (%)
	-α	30	40	50	+α	-α	30	45	60	+α	-α	5.0	7.0	9.0	+α		
1		×					×					×				42.6	44.87
2				×			×					×				72.6	71.13
3		×							×			×				80	77.65
4				×					×			×				79.7	81.50
5		×					×							×		29.3	27.79
6				×			×							×		58.1	60.74
7		×							×					×		61	62.77
8				×					×					×		75.3	73.32
9	×							×					×			30.3	30.28
10					×			×					×			61.8	61.23
11			×			×							×			56.7	55.64
12			×							×			×			93.3	93.77
13			×					×			×					82.3	82.23
14			×					×							×	61.5	60.98
15			×					×					×			88.8	89.60
16			×					×					×			90	89.60
17			×					×					×			89.2	89.60
18			×					×					×			89.6	89.60
19			×					×					×			90.5	89.60

3.2. Statistical analysis and modelling

The findings gathered from the experimental work were employed to construct a regression equation to forecast the effects of the factors tested on the ability of the new eco-friendly coagulants to eliminate kaolinite turbidity from artificial water. The regression model is presented in the formula below:

$$R.E.\ (\%) = -370.4 + 14.414\times D + 4.227\times DT + 14.93\times pH - 0.155\times D^2 - 23.4\times 10^{-3}\times DT^2 - 1.59\times pH^2 - 37.33\times D\times DT + 83.7\times D\times pH + 18.3\times DT\times pH \quad (2)$$

Table 3 also indicates an agreement between theoretical and experimental removals of kaolinite turbidity from water, this table suggesting high correlation between turbidity theoretical and measured removal efficiency.

The outcomes of the experimental runs and statistical analysis indicated that the *Strychnos potatorum* seeds could be used as effective coagulants for elimination of kaolinite turbidity from water, and the CCD method could be used optimise and model the performance of the *Strychnos potatorum* seeds in terms of turbidity removal. Additionally, the recent successful developments in sensing technologies [41-45] motivate the researchers to use this technology to monitor the level of influent turbidity and manage the performance of the coagulation unit.

4. Conclusions

The current research discusses the possibility of manufacturing eco-friendly coagulants from *Strychnos potatorum* seeds to remove kaolinite turbidity from artificial water under various experimental conditions, including water pH, detention periods and dosages of the new eco-friendly coagulants. The results of the current research evidenced that the optimum removal of kaolinite turbidity could be reached by sustaining neutral pH of the artificial water, long detention time, and by using 40 mg / L of the new eco-friendly coagulants (*Strychnos potatorum* seeds). Results of the present research also showed that *Strychnos potatorum* seeds-based coagulants are not efficient in the removal of kaolinite turbidity in strong basic or acidic environments. Additionally, the research highlighted that short detention periods and/or small doses of the new eco-friendly coagulants negatively affect the removal efficiency of the kaolinite turbidity. Additionally, it was found that the CCD method belongs to RSM, is applicable to reproduce the remediation of turbidity using the coagulation agents that derived from the seeds of *Strychnos potatorum*.

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