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Application of *Moringa peregrina* seed extract as a natural coagulant for phenol removal from aqueous solutions

Edris Bazrafshan¹, Ferdos Kord Mostafapour¹, Hamed Faridi¹ and Mohammad Ali Zazouli^{2*}

¹Health Promotion Research Center, Zahedan University of Medical Sciences, Zahedan, Iran.

²Department of Environmental Health Engineering, Faculty of Health and Health Sciences Research Center, Mazandaran University of Medical Sciences, Sari, Iran.

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Phenol and its derivatives are toxic pollutants frequently found in surface and tap waters, and in aqueous effluents from various manufacturing processes such as petroleum and petrochemical, coal conversion, synthetic rubber, plastics, paper, oil refineries and phenol-producing industries. In the present study, the ability of *Moringa peregrina* seed extract to remove phenol from aqueous solutions in a batch system was investigated. The effects of major operating variables such as coagulant dose (0.05 - 4 ml/L), initial phenol concentration (5, 10, 20, 50 and 100 mg/L) and pH (3-9) were investigated. The optimum pH for phenol removal by coagulation using *M. peregrina* seed extract was 5, with coagulant dose of 0.1 and 0.3 ml/L and the highest removal was found to be 95.16, 95.84, 97.91, 98.27 and 99.9% at initial phenol concentration of 5, 10, 20, 50 and 100 mg/L, respectively, as a result of precipitation, co-precipitation and adsorption mechanisms. According to these results, it was defined that *M. peregrina* seed extract is not only an inexpensive coagulant, but also quite an effective factor in the removal of phenol from aqueous environments.

Key words: Phenol removal, *Moringa peregrina*, coagulation, natural coagulants.

INTRODUCTION

Phenol is an organic substance that is used in several manufacturing processes such as the manufacture of phenolic resin and other phenol derivative chemicals. It is also used as a solvent, as an antiseptic and as additive in disinfectant (Busca et al., 2008). Nearly all phenols are toxic and some are known to be human carcinogens. They are incorporated also in the food chain and generate important environmental problems (Gomez et al., 2006). Phenolic compounds are released in the surface water by a number of industries, mainly by pharmaceutical plants, petroleum and petrochemical, oil refineries, coke plants, pulp, plastics, ceramic, steel, resin and food-processing industries (Rivas et al., 2003;

Gernjak et al., 2003; Rengaraj et al., 2002a; Huang et al., 2007; Tepe and Dursun, 2008). These industries are distributed in wide area, which result in serious environmental problems because of their toxicity, poor biodegradability and accumulation potential in plants and tissues. Fishes living in the lake and river contaminated with phenolic compounds have a queer taste and bad dour. Therefore, the wastewater containing phenolic compounds must be treated before their discharge into water streams.

Phenol is a combustible compound that is very soluble in water, oils, carbon disulfide and numerous organic solvents (Ahmaruzzaman et al., 2008). It is characterized by a typical pungent sweet, medicinal, or tar-like odor (Amore and Hautala, 1983). In addition, their presence in water supplies is noticed by a bad taste and odour. In the presence of chlorine in drinking water, phenol forms chlorophenol, which has a medicinal taste that is quite pronounced and objectionable. Also, phenol is a potential

*Corresponding author. E-mail: zazouli49@yahoo.com or mzazouli@mazums.ac.ir. Tel: +98 1513543080. Fax: +98 1513543237.

human carcinogen and is of considerable health concern, even at low concentration. Owing to their toxic effects, including permeabilisation of cellular and cytoplasmic coagulation, phenolic contaminants can damage sensitive cells and thus cause profound health and environmental problems (Bodalo et al., 2009; Rengaraj et al., 2002b; Aksu and Yener, 2001; Viraraghavan and De Maria, 1998). Phenol has been registered as priority pollutants by the US Environmental Protection Agency (USEPA), with a permissible limit of 1.0 mg/L in wastewater (Eahart et al., 1997). In Iran, a maximum phenol level of 1.0 mg/L is permitted in wastewater for discharge to surface water resources by the Institute of Standard and Industrial Research of Iran (ISIRI).

Wastewaters containing phenols and other toxic compounds need careful treatment before their discharge into the receiving bodies of water. Various techniques have been employed for the removal of phenolic compounds from aqueous environments, including biological treatment (Li et al., 2011; Bajaj et al., 2008; Annadurai et al., 2002), ultrasonic degradation (Pandit et al., 2001), photocatalytic degradation (Agrios et al., 2003), membrane filtration (Bodalo et al., 2009; Bodalo et al., 2008), enzymatic treatments (Mao et al., 2006), solvent extraction (Juang et al., 2010), chemical oxidation and electrochemical methods (Bazrafshan et al., 2012), wet oxidation (Yang et al., 2008) and adsorption (Senturk et al., 2009; Gundogdu et al., 2012). Natural coagulants of vegetable and mineral origin were used in water treatment before the advent of chemical salts, but they have not been able to compete effectively because of the fact that a scientific understanding of their effectiveness and mechanism of action was lacking.

Thus far, the use of natural coagulants has been discouraged without any scientific evaluation. They have succumbed progressively under modernization and survived only in remote areas of some developing countries (Ndabigengesere and Narasiah, 1998). Recently however, there has been a resurgence of interest in natural coagulants for water treatment in developing countries, mainly due to the advantages such as biodegradability, low sludge production and fewer risks to health and environment (Katayon et al., 2006; Shahriari et al., 2012). The usage of natural coagulants such as *Moringa oleifera* is better than chemical coagulant (Ndabigengesere et al., 1995; Ndabigengesere and Narasiah, 1997). Moreover, sludge generated using *M. oleifera* as coagulant is less bulky compared to a chemical coagulant (McConnachie et al., 1999). Ndabigengesere et al. (1995) documented that the mechanism of coagulation with *M. oleifera* appeared to consist of adsorption and neutralization of the colloidal charges. Usage of *M. oleifera* as natural coagulant has also been reported by other researchers (Katayon et al., 2006; McConnachie et al., 1999).

Moringa peregrina is a desert species; its occurrence in Iran is restricted to the southeast of the Sistan and

Baluchestan Province. Globally, it grows in Northeast Africa and Southwest Asia. The ground where *M. peregrina* grows is usually covered with coarse rock debris, which characterizes the upstream runnels at the mountain bases and slopes (Hegazy et al., 2008). The main objective of this research was to evaluate the feasibility of using the *M. peregrina* seed extract as a natural coagulant for the removal of phenol from aqueous solutions. The effects of different parameters including solution pH, coagulant dosage and initial phenol concentration were studied.

MATERIALS AND METHODS

Preparation of *M. peregrina* seed extract

Dry *M. peregrina* seeds were collected around the vicinity of Nikshahr city (26°68' N, 59°71' E), Sistan and Baluchestan province, Iran. The preparation and extraction process was carried out as followings: the seeds coats and wings were removed and seeds were reduced into powder by a domestic mill. A 1-M NaCl solution was prepared and 5 g of powder were put into 100 ml of it. The NaCl solution with powder was stirred for 30 min at room temperature (around 20±2°C) to extract the coagulation active components. No pH modification was needed, as natural pH 7 was achieved. Subsequently, the extract was filtered twice: once through commercial filter paper on Büchner funnel and once again through a fine filtering millipore system (0.45 µm glass fiber). The result was a clear, milk-like liquid. *M. peregrina* stock solution prepared in this way was used the same day it was produced, although there are references that point the stability of the extract (Katayon et al., 2006).

Phenol solution preparation

All chemicals used for this study are of analytical grade (AR) and were obtained from Merck (Darmstadt, Germany). Stock solutions of phenol were prepared by weighing the purified grade chemicals and dissolving them in deionized water without pH adjustment. Working solutions of the desired concentrations (5, 10, 20, 50 and 100 mg/L) were obtained by successive dilutions.

Analysis

The residual phenol concentration in the solution was analyzed using ultraviolet-visible (UV-VIS) spectrophotometer (Cintra 10e, GBC, Australia), and 4-aminoantipyrene at $\lambda_{\max} = 500$ nm. The pH of solution was measured using a pH-meter (Denver Ultra basic-UB10, USA).

Coagulation experiments

Jar test is the most widely used method for evaluating and optimizing the coagulation-flocculation processes. This study consists of batch experiments involving rapid mixing, slow mixing and sedimentation. The VELP jar test apparatus (JLT6 model) was used in all the coagulation experiments. Glass beakers of 2 l filled with the model phenol solution were used. The apparatus allowed six beakers to be agitated simultaneously, and rotational speed could be varied between 0 and 140 rotations per minute (rpm), thus allowing simulation of different mixing intensities and resulting

Table 1. The ranges of experimental parameters used.

Parameter	Range
pH	3, 4, 5, 6, 7, 8, 9
Coagulant dosage (ml/L)	0.05, 0.1, 0.3, 0.5, 1, 2, 3, 4
Phenol concentration (mg/L)	5, 10, 20, 50, 100
Rapid mixing, slow mixing and sedimentation	2 min (120 rpm), 20 min (40 rpm) and 30 min
Turbidity (NTU)	10, 20, 30, 40, 50, 100, 150

Table 2. The typical water quality characteristics used for the some experiments.

Parameter	Concentration
pH	7.63
Conductivity ($\mu\text{S}/\text{cm}$)	641
Turbidity (NTU)	0.6
Iron (mg/L)	0.03
Manganese (mg/L)	0.013
Sulfate (mg/L)	37.2
Chloride (mg/L)	48.6
Sodium (mg/L)	29.3
Nitrate (mg/L)	7.3

flocculation process.

In a typical run, beakers were filled with one litre of the model phenol solution, placed on the flocculator and agitated at the preselected intensity of rapid mixing. During rapid mixing, the coagulant dosage was added into each beaker using micropipettes. The duration of rapid mixing was controlled with a stopwatch. After rapid mixing, the preselected intensity of slow mixing was quickly established and its duration was controlled again with the stopwatch. After slow mixing, the beakers were carefully removed from the flocculator and were placed in a safe place for the sedimentation phase to take place. In this study, the intensity and duration of both rapid mixing and slow mixing were fixed respectively at 120 ± 2 rpm for 2 min in the case of rapid mixing and 40 ± 2 rpm for 20 min in the case of slow mixing for flocculation. The duration of sedimentation was kept constant at 30 min. At the end of the settling period, water samples were taken from the supernatants, immediately vacuum-filtered using a $0.45\text{-}\mu\text{m}$ pore size membrane filter, and analyzed. All experiments were run at room temperature ($20 \pm 2^\circ\text{C}$) and no pH control was exercised. The ranges employed for these parameters are given in Table 1. Finally the efficiency of phenol removal (% Removal) was calculated as:

$$\% = \frac{(C_i - C_f)}{C_i} \cdot 100 \quad (1)$$

Where, C_i is the initial concentration (mg/L) and C_f is the final phenol concentration (mg/L).

Moreover, after the determination of optimum conditions (pH and coagulant dose of *M. peregrina* seed extract) for phenol removal, the effect of initial turbidity on phenol removal efficiency was investigated at an initial phenol concentration of 5, 10, 20, 50 and 100 mg/L. Synthetic turbid water for coagulation tests was prepared by adding stock kaolin suspension into distilled water. The stock kaolin suspension contained 10 g of kaolin in 1 L of distilled water.

Finally a series of jar tests were performed using one liter of tap water with 5, 10, 20, 50 and 100 mg/L of phenol concentration. The characterization of the tap water used for these experiments is listed in Table 2.

RESULTS AND DISCUSSION

Effect of initial pH

The pH of the solution is one of the most critical parameters in the coagulation process and pollutants removal from aqueous solutions (Shi et al., 2007; Chu, 2001). In order to determine the desired pH for removal of phenol by *M. peregrina* seed extract as a natural coagulant, the uptake of phenol as a function of hydrogen ion concentration was studied. Figure 1 shows the effects of initial pH values within the range of 3 - 9 on the efficiency of phenol removal by *M. peregrina* seed extract with initial dosage of 0.1 ml/L. As shown in the figure, maximum efficiency was obtained at initial pH 5 for all initial concentrations of phenol. On the other hand, the optimal phenol removal can be achieved at pH 5, and then gradually declines as the pH is further increased and decreased. The highest removal was found to be 94.132, 95.69, 98.04, 98.14 and 99.931% at initial phenol concentration 5, 10, 20, 50 and 100 mg/L, respectively, as a result of precipitation, co-precipitation and adsorption mechanisms.

Also, the results show that extract of *M. peregrina* seed has a minimal effect on the pH of the samples. This result was in agreement with that obtained by Yarahmadi et al.

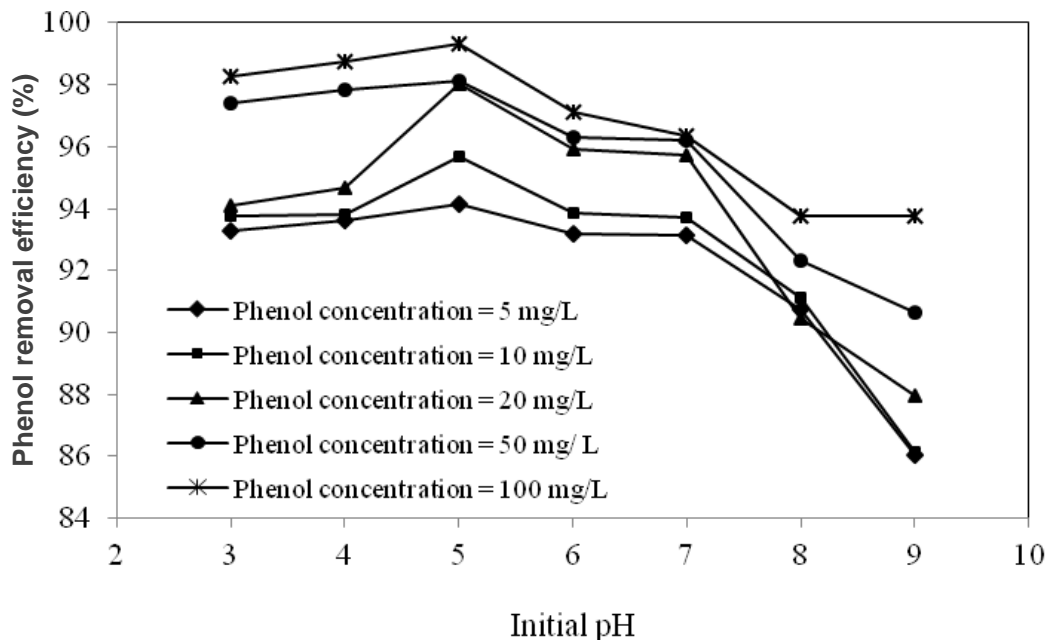


Figure 1. Effect of pH on phenol removal from synthetic solutions (*Moringa peregrina* seed extract dose = 0.1 mg/L).

(2009) on removal of turbidity by *M. oleifera* seed extract. Moreover, they reported that the efficiency of the *M. oleifera* extract as a coagulant is not affected by pH (Yarahmadi et al., 2009).

Effect of coagulant dose

Coagulation dosage is one of the most important parameters that have been considered to determine the optimum condition for the performance of coagulants in coagulation and flocculation. Essentially, insufficient dosage or overdosing would result in the poor performance in flocculation. Therefore, it is significant to determine the optimum dosage in order to minimize the dosing cost and sludge formation and also to obtain the optimum performance in treatment. The effect of coagulant dose (*M. peregrina* seed extract) on phenol uptake is depicted in Figure 2, which shows that the coagulation efficiency of phenol changed with an increase in the dosage of *M. peregrina* seed extract from 0.05 to 4 ml/L. The optimum dosage of *M. peregrina* seed extract for both low and high phenol solutions was 0.1 and 0.3 ml/L. On the other hand, the highest removal was found to be 95.16, 95.84, 97.91 and 98.266% at initial phenol concentrations 5, 10, 20 and 50 mg/L at coagulant dosage 0.1 ml/L, respectively (and 99.904% at 0.3 ml/L for initial phenol concentration 100 mg/L).

Meanwhile, more increase the dosage of coagulant did not help in improving the removal of phenol; in fact this increased the residual phenol of the coagulated sample. Therefore, in the present study, coagulant dosage of 0.1

ml/L was selected as optimum dose of coagulant. The results obtained at optimum pH values of coagulant are in good agreement with the values given in literatures (Sanghi et al., 2006). On the other hand a further increase in coagulant dose causes restabilization of the particles as the charge reversal on the colloids occurs (Duan and Gregory, 2003). Generally, it can be concluded that lower doses of investigated natural coagulants were better than higher ones. This is very important not only for process economy but also for lower organic matter load in processed water because it is known that high organic load might cause microbial growth (Sciban et al., 2009). Nevertheless, the results obtained are valid only in applied conditions.

However, higher removal (>99%) was achieved for the samples with high concentration (>99%) of phenol (100 mg/L). This was in agreement with the findings reported by other researchers (Zhang et al., 2006). They documented the increase in turbidity removal with increase in the initial turbidity of a sample. Also, it was observed that after the coagulation and flocculation experiments using *M. peregrina* seed extract, residual phenol concentrations decreased to below 1.0 mg/L which is the permissible limit of phenol in wastewater determined by the USEPA and ISIRI.

Effect of turbidity on phenol removal

The study of the effect of initial turbidity on the phenol removal at various initial concentrations of phenol (Figures 3 to 7) indicates that turbidity addition can

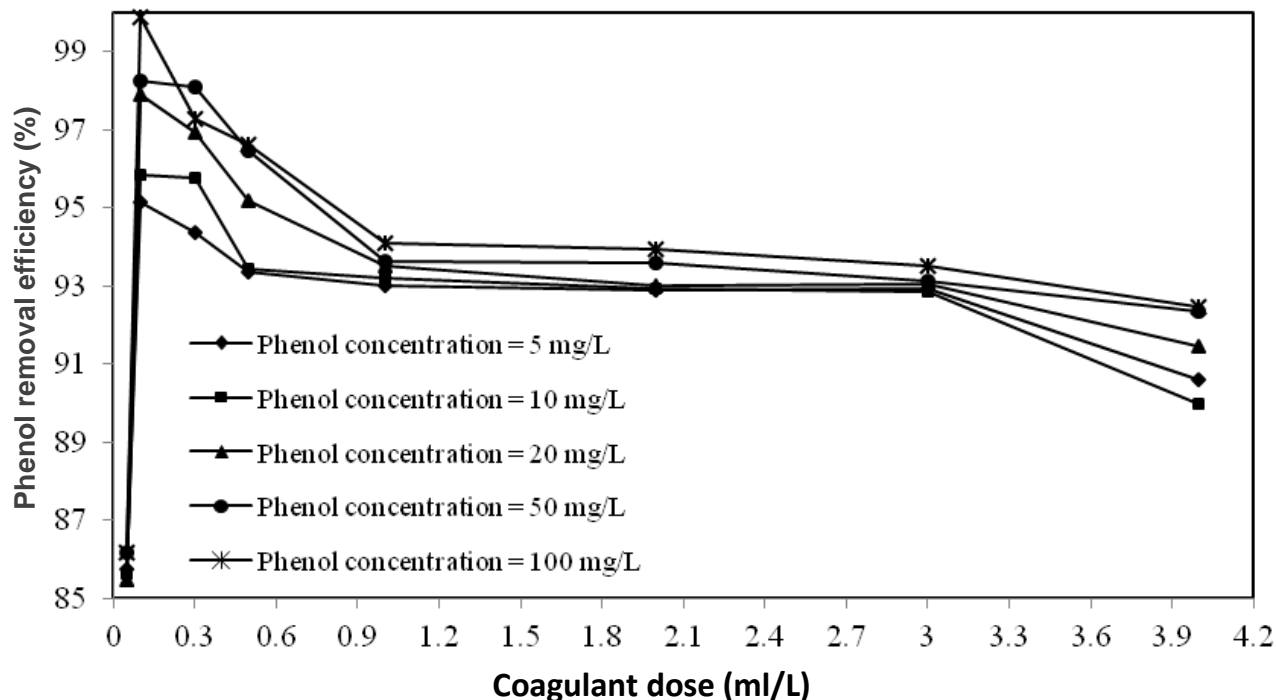


Figure 2. Effect of coagulant dose (*M. peregrina* seed extract) on phenol removal from synthetic solutions (at optimum pH = 5).

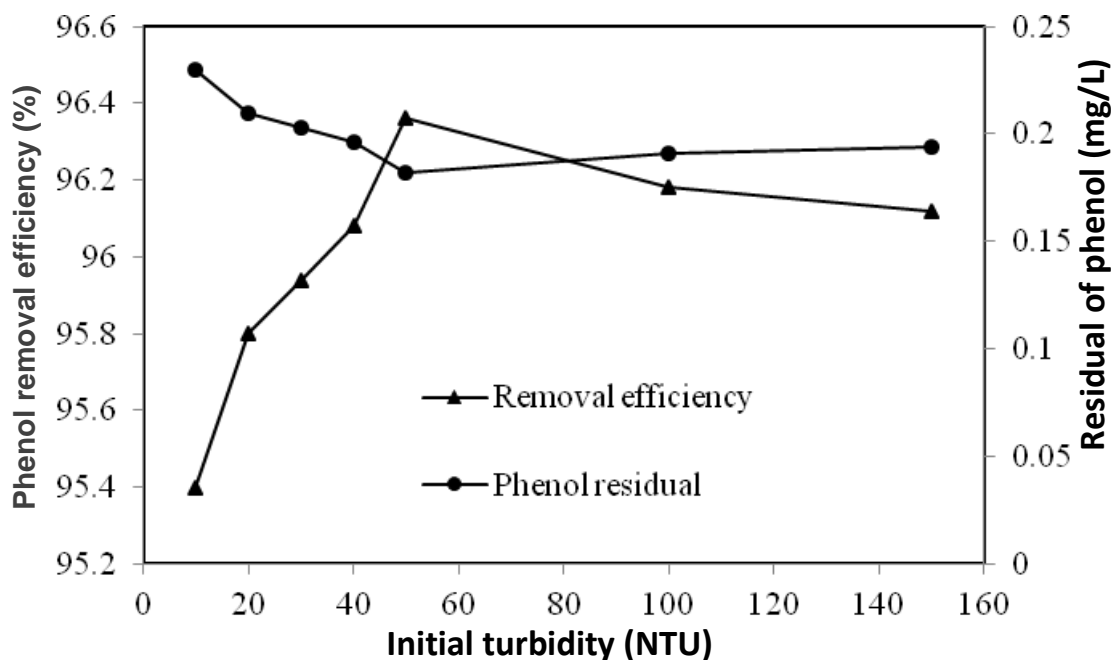


Figure 3. Effect of initial turbidity on phenol removal from synthetic solutions (initial concentration of phenol = 5 mg/L, pH = 5, coagulant dose = 0.1 ml/L).

slightly enhance the phenol removal more than when the coagulant is used alone. Nevertheless, further addition of turbidity decreased removal efficiencies of phenol. The

best efficiency for initial phenol concentrations 5, 10, 20, 50 and 100 mg/L was obtained in initial turbidity 50, 100, 150, 150 and 100 NTU and pH 5 for all experiments,

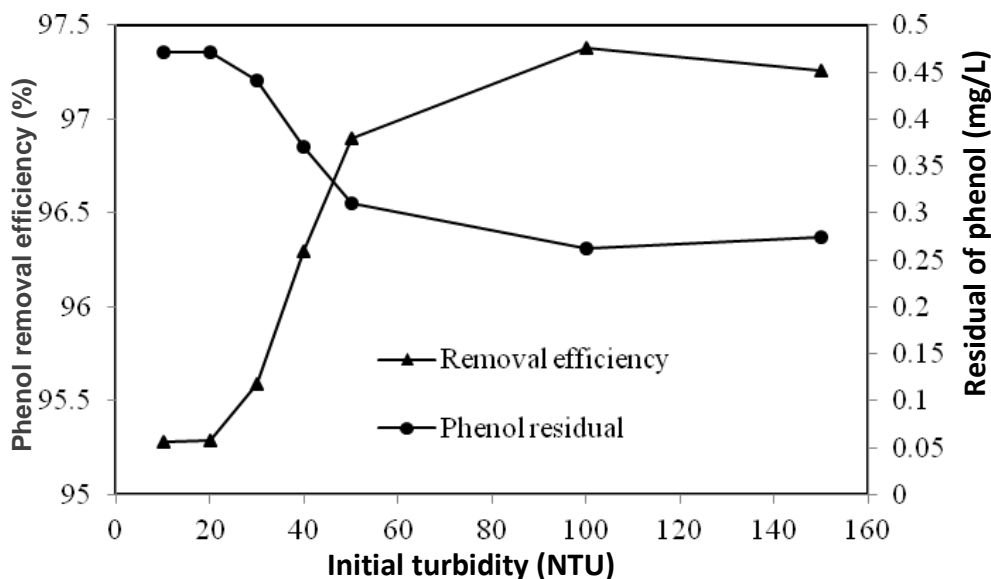


Figure 4. Effect of initial turbidity on phenol removal from synthetic solutions (initial concentration of phenol = 10 mg/L, pH = 5, coagulant dose = 0.1 ml/L).

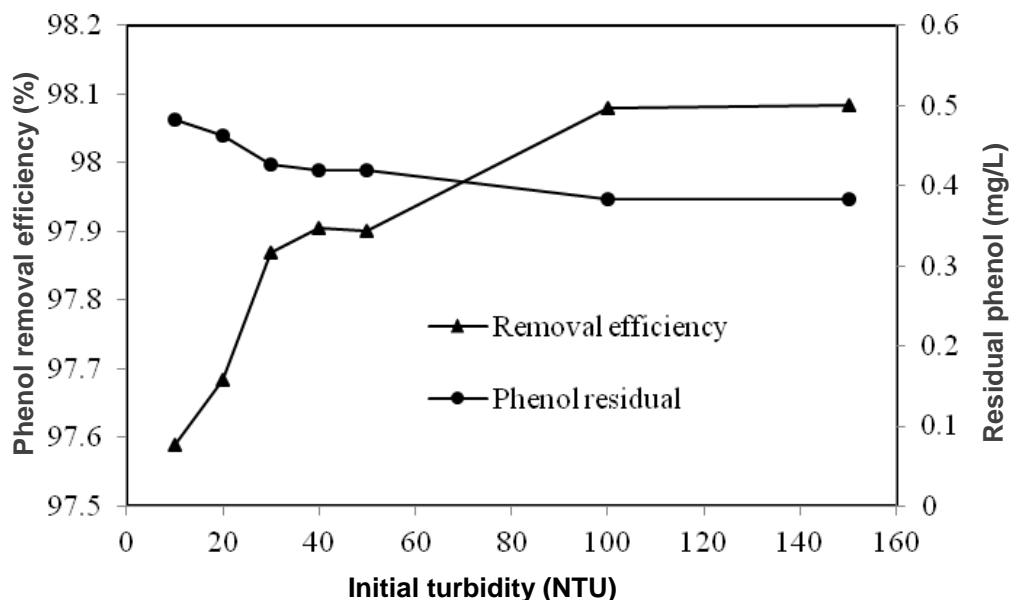


Figure 5. Effect of initial turbidity on phenol removal from synthetic solutions (initial concentration of phenol = 20 mg/L, pH = 5, coagulant dose = 0.1 ml/L).

respectively. As shown in Figures 3 to 7, the residual phenol concentration for all experiments was lower than 1.0 mg/L, except for initial phenol 50 mg/L (with initial turbidity 10 and 20 NTU).

Experiments on tap water

Some experiments were performed to study the

performance of *M. peregrina* seed extract as coagulant for phenol removal from natural (tap) water samples with different initial phenol concentrations of 5, 10, 20, 50 and 100 mg/L. The results obtained from jar test experiments with 0.1 ml/L *M. peregrina* seed extract are shown in Figure 8. The result indicates that the removal efficiency of phenol increased with its increase in initial concentration, and so maximum efficiency (98.88%) was obtained at initial concentration 100 mg/L. In addition,

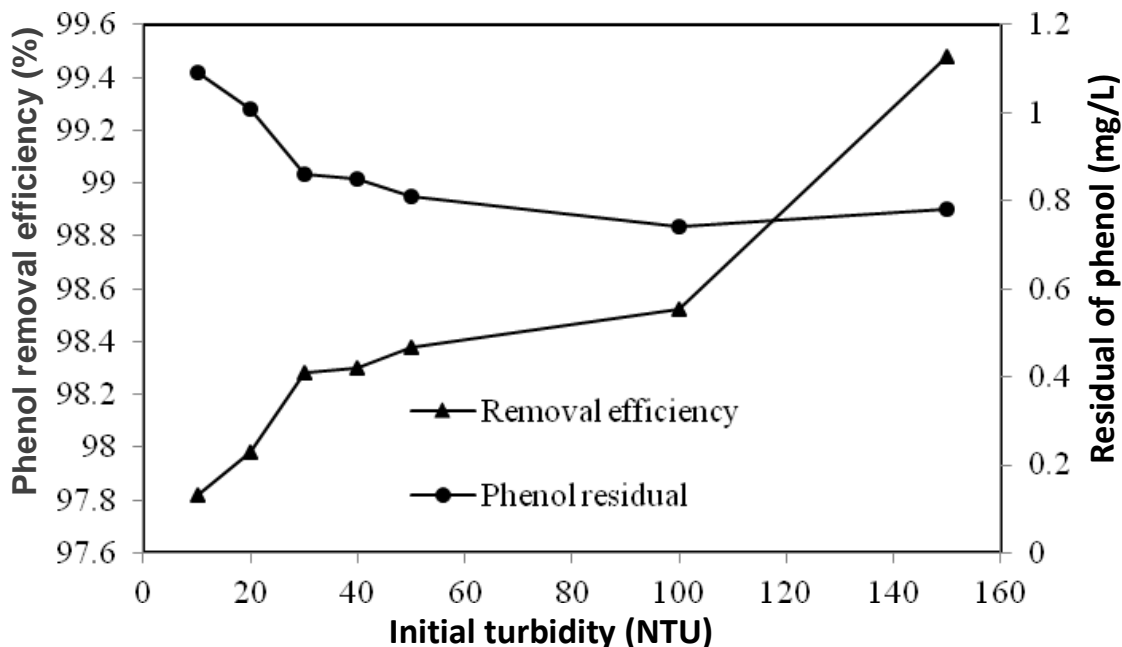


Figure 6. Effect of initial turbidity on phenol removal from synthetic solutions (initial concentration of phenol = 50 mg/L, pH=5, coagulant dose = 0.1 ml/L).

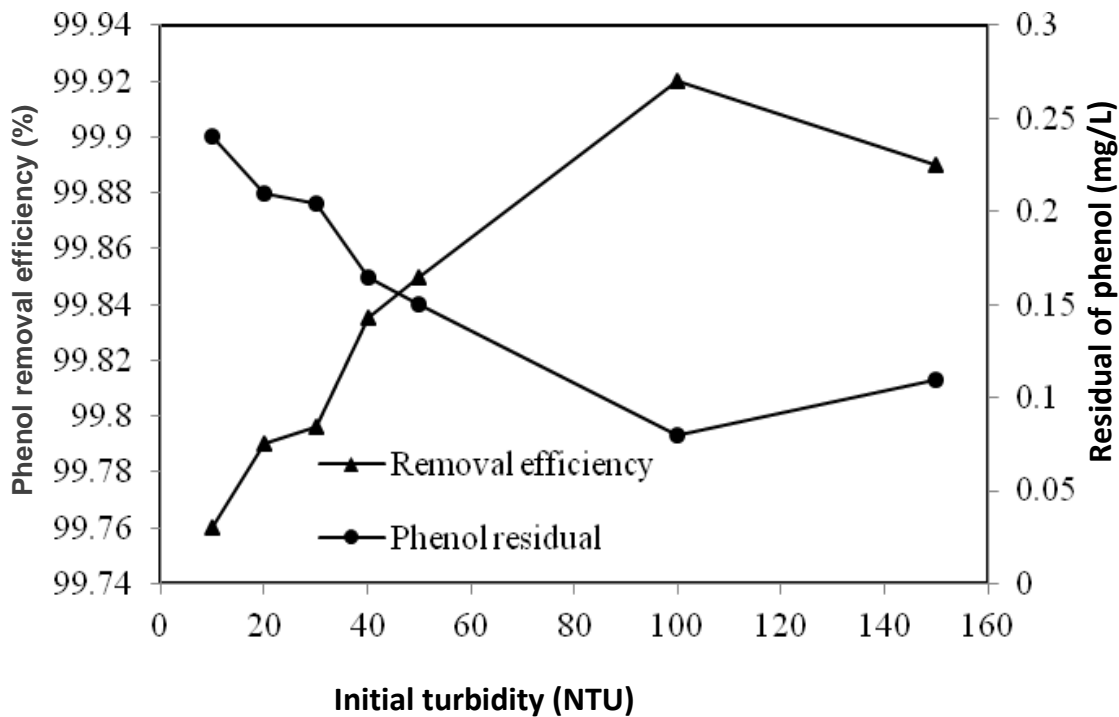


Figure 7. Effect of initial turbidity on phenol removal from synthetic solutions (initial concentration of phenol = 100 mg/L, pH = 5, coagulant dose = 0.1 ml/L).

after the coagulation and flocculation experiments using *M. peregrina* seed extract, residual phenol concentrations decreased to below 1.0 mg/L (except for initial phenol

100 mg/L), which is the permissible limit of phenol in wastewater determined by the USEPA and ISIRI. Therefore, it could be concluded that for higher

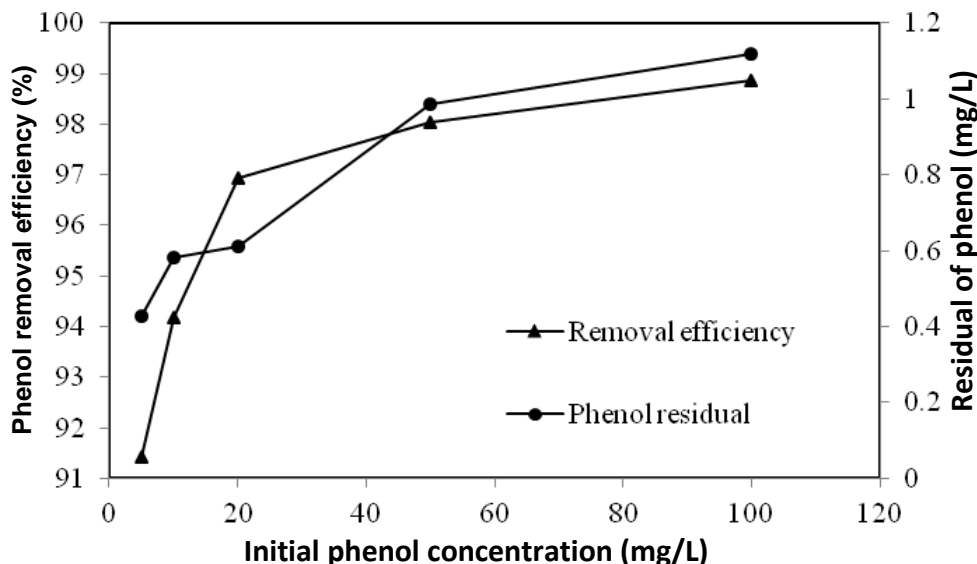


Figure 8. Removal efficiency of phenol from tap water at various initial concentrations of phenol (coagulant dose = 0.1 ml/L).

concentration of phenol, more coagulant is required to reach the current standards.

Conclusion

The present study was aimed at investigating the applicability of coagulation process using *M. peregrina* seed extract as a natural coagulant in the removal of phenol from aqueous environments. The influence of various variables such as coagulant dose, pH and initial concentration of phenol on the removal of phenol was also investigated. The results obtained with synthetic solutions reveal that the most effective removal capacities of phenol were achieved with coagulant dose 0.1 and 0.3 ml/L at pH 5, and the highest removal was found to be 95.16, 95.84, 97.91, 98.27 and 99.9% at initial phenol concentration of 5, 10, 20, 50 and 100 mg/L, respectively. Hence, it can be concluded that *M. peregrina* seed extract is an efficient coagulant for phenol removal in various concentrations from water and wastewater.

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