Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2014.928909

55 (2015) 2221–2228
August

Taylor & Francis
Taylor & Francis Group

Removal of metronidazole antibiotic from contaminated water using a coagulant extracted from *Plantago ovata*

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Received 19 October 2013; Accepted 24 May 2014

ABSTRACT

A coagulant was successfully extracted from *Plantago ovata* by a method using a $FeCl_3$-induced crude extract (FCE). FCE was investigated for its coagulation potential to remove metronidazole (MNZ) from water. Tests were done to evaluate the effects of solution pH, FCE dose, MNZ concentration, and solution temperature on removal of MNZ by FCE. A low coagulant dose, 1.75 mg/L, achieves a high MNZ removal percentage, 89.3%, at neutral pH and room temperature. MNZ removal by FCE increased from 18 to 89.3% with increased FCE quantity from 0.75 to 1.75 mg/L. The low concentration of MNZ, 10 mg/L, was completely removed by FCE. Coagulant process did not depend on water temperature. The sludge volumetric index and sludge volume were also studied. Overall, the results indicate that developed FCE was an efficient coagulant that presents an attractive coagulant method for application in treating contaminants in water.

Keywords: Coagulant; Contaminated water; Metronidazole; *Plantago ovata*

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1. Introduction

Currently, one of the most hazardous environmental pollutants is antibiotics. Antibiotics are widely used therapeutically for humans and animals, and as growth promoters in livestock and aquaculture operations. These classes of drugs are receiving considerable attention as being environmental contaminants, classified as recalcitrant bio-accumulative compounds and are, thus, regarded as hazardous chemicals [1,2]. The release of pharmaceuticals into the environment, thus, results in contamination of aquatic or terrestrial ecosystems [3]. Effluent containing antibiotics needs to be treated physically or chemically to prevent the adverse effects from contaminated water.

Many conventional treatment methods such as adsorption, oxidation, electrochemical methods, and membrane techniques are too costly for extensive deployment in the developing countries. Among these available techniques, the process involving activated carbon is the most commonly applied adsorption process. The major problem with activated carbon is that while bench-scale tests may show excellent treatment performance, full-scale operation is fraught with severe problems, such as clogging and biofouling [4,5]. Therefore, affordable technologies using low-cost materials and matching or exceeding the capability of conventional treatment technologies are required. The

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affordable technologies such as coagulation–flocculation will replace current pharmacy wastewater treatment plant based on biological techniques that has been proven inefficient for many pharmaceuticals [6]. Further, the performance of the coagulation–flocculation process is largely affected by the coagulant types that enhance the aggregation of particles to form large, rapid-setting flocs either through charge neutralization or chain-bridging mechanisms. Thus, research on water and wastewater treatment has been focused on the synthesis of new and eco-friendly coagulant to improve the coagulation–flocculation process [7,8]. These novel coagulants include inorganic, organic, and composite coagulants. However, organic coagulants have attracted more research attention due to excellent coagulation performance and have widely been used in many water and wastewater plants [9]. The most common inorganic coagulants used during the coagulation–flocculation process are hydrolyzing metal salts of aluminum and iron such as $AlCl_3$, $Al_2(SO_4)_3$, $FeCl_3$, and $Fe_2(SO_4)_3$ [6,10]. Their major drawbacks are that the coagulant species form rapidly during dilution, and the formation of hydrolysis species cannot be controlled. However, for Al-based coagulants this may result in a high residual concentration of Al in the treated water [11,12]. Recently, concerns have been raised about the potential toxicity of residual aluminum and, thus, a threat to human health and the environment [13]. In order to make the flocculation–coagulation process attractive, novel low-cost coagulants with higher coagulation capability are required. This has led to an increasing research interest in the production of natural coagulants from renewable and cheaper precursors. Natural coagulants, mainly polysaccharides, are generally nontoxic and biodegradable [14], which are essential from a sustainability point of view.

In this study, we attempt to use *Plantago ovata* as a low-cost, abundantly available, and renewable precursor for the production of coagulant for the removal of metronidazole (MNZ) from water. We consider *P. ovata* because it is fast growing plant and easily found in Iran. *P. ovata* is a small, one-year-old plant without or with a very short stem covered with soft fiber, from the *Plantaginaceae* family, and its height ranges from 7 to 30 cm [15]. *P. ovata* seeds contain mucilage, protein, fixed oil, cellulose, and starch [16]. Interestingly, as it is clear from the existing literature [17–19], the efficiency of water or wastewater treatment by a natural coagulant is significantly influenced by the characteristics and the origin of the coagulant. Therefore, the method of extraction is aimed at improving the material's coagulation capability, thereby reducing the rate of coagulant consumption to reduce the cost of water and wastewater treatment and making it more cost-effective. In this research, an innovative method is used. We use $FeCl_3$ during the purification of an extracted coagulant with the assumption that the Fe^{3+} ions are trapped in the structure of the biomaterial extracted from *P. ovata* and, therefore, according to the Schulze–Hardy rule [19], may improve the coagulation potential of the coagulant rather than K^+ and Na^+.

Therefore, the aim of this work is to assess the $FeCl_3$-induced crude extract (FCE) provided from *P. ovata* seeds, as a novel coagulant, for removal of MNZ as a model of an antibiotic, from contaminated water. MNZ, a kind of nitroimidazole antibiotic, is the drug of choice prescribed for the treatment of a number of human bacterial infections, as a growth promoter for animals and in agriculture to prevent infections [20]. Influences of different parameters, such as water pH, FCE quantity, MNZ concentration, and water temperature on the MNZ removal were evaluated. Further, the sludge volume index (SVI) and sludge volume of the sludge after the treatment were investigated.

2. Materials and methods

2.1. Materials

Dry seeds of *P. ovata* were purchased from a local herb market and stored at room temperature. Ammonium acetate, hydrochloric acid, sodium hydroxide, ferric chloride, calcium chloride, and sodium chloride were supplied by Aldrich Co. and were of analytical grade. Ethanol (98%) and *CM-Sepharose* were purchased from Merck Co. Analytical grade MNZ (chemical formula: $C_6H_9N_3O_3$, molar mass: 171.2 g/mol, and water solubility: 9.5 g/L) was purchased from Sigma Aldrich Co., and used as received. The contaminated water used in the experiments was prepared from mixing aliquots of MNZ stock solutions (1,000 mg/L) with distilled water.

2.2. Extraction and purification processes of coagulant from P. ovata

Extraction of the crude coagulant from *P. ovata* seeds were performed to obtain the proteinous coagulant as much as possible. For this purpose, *P. ovata* seeds were initially dehusked and soaked in water for 1 d to obtain the gelatinous biomaterial. The biomaterial was filtered and dried at 70˚C in an oven and then milled in a domestic blender (VARING) to obtain a powder form. The powder was defatted by mixing with ethanol 98% using a magnetic stirrer for 60 min. Then, the supernatant was centrifugally separated (3,500 rpm, 40 min) and the settled powder was dried in an oven (at 60˚C) overnight. The crude coagulant (that contained proteins) was extracted from the oil-free powder using ammonium acetate 10 mM in 5% w/w [21]. The mixture was then stirred for 50 min using a magnetic stirrer and the supernatant, namely the crude extract, was separated by centrifugation (3,500 rpm, 40 min). Purification of the coagulant protein from the crude extract was carried out as follows:

CM-Sepharose ion exchange was equilibrated using a 10-mM ammonium acetate solution [22]. Equilibrated *CM-Sepharose* ion exchange was added to the crude extract in 10% (v/v) proportion and mixed with a blade stirrer for 50 min. Further modification procedure, as a novelty, was carried out directly using two concentrations of $FeCl_3$ (0.25 and 0.5 M). For this propose, the designated concentration $FeCl_3$ was added to solution containing crude extract and magnetically stirred for 3 h. After that, the $FeCl_3$-crude extract was centrifuged. Different salt ($FeCl_3$) concentrations led to different fractionating of proteins and each achieved protein had a given coagulation properties. The pretest results showed that 0.5-M $FeCl_3$ proteins led to a higher coagulation performance and added less dissolved organic matter in the treated water than 0.25-M $FeCl_3$ proteins. Consequently, the 0.5-M $FeCl_3$ proteins produced as more active coagulant proteins. The elution stages were repeated three times in order to recover as much coagulant as possible.

2.3. Coagulation procedure

Coagulation tests were carried out in a standard jar test apparatus (Phipps and Bird, Model 300). The jar test is the most widely applied method for evaluating and optimizing the coagulation–flocculation process. The prepared water (300 mL) of different initial MNZ concentration was filled into the beakers (1 L) and the standard procedure for the jar test was performed at constant room temperature 24˚C. The standard procedure implies 1 min of rapid mixing (120 rpm) followed by 20 min of slow mixing (45 rpm) for flocculation. After that, treated water was allowed to settle for 1 h and the supernatant sample was withdrawn by a syringe from about 2 cm below the liquid level for analysis. The initial pH of the water was adjusted to a desired value with 1 M NaOH or 1 M HCl and further correction of pH was not carried out during the experiments. The same coagulation test was conducted with no coagulant as a blank. The effect of water pH, initial MNZ concentration, coagulant quantity, and water temperature as variable parameters were evaluated as different conditions in this research. Table 1 presents the details of experimental runs and conditions. All the experiments were conducted in duplicate to ensure reproducibility of the results; the mean of these two measurements are stated herein. The removal efficiency of MNZ (R_{MNZ}) was calculated from Eq. (1).

$$R_{MNZ}\ (\%) = [(MNZ_0 - MNZ_t)/MNZ_0] \times 100 \qquad (1)$$

where MNZ_0 and MNZ_t represent the initial and final concentration of MNZ (mg/L) of water, respectively.

2.4. Analysis

All analytical measures were done according to the American Public Health Association standard methods [23]. The concentration of MNZ in a solution was measured by high-performance liquid chromatography (HPLC, Shimadzu, LC10A HPLC) equipped with a UV detector (SPD-10AV) at a wavelength of 318 nm. A Diamonsil (R) C18 column (5 μm, 250 mm long × 4.6 mm ID) was used, and the mobile phase was composed of a mixture of acetonitrile and buffer phosphate with pH = 4.8 (40/60, v/v). The flow rate was set at 1 mL/min, and 20 μL injections were used.

Measures referring sludge production and SVI were carried out with a 25-mL calibrated test tube and 1-L Imhoff cone. In the first case, a 25-mL sample was taken just after coagulation and flocculation process (without sedimentation) and suspended solids were determined by Millipore fine filtration (45 μm glass fiber filter). In the second case, the Imhoff cone received a 0.5-L sample of treated water and it was allowed to settle for 1 h. Then, sludge volume was measured as Imhoff cone was calibrated.

The pH level of samples was determined using a pH meter (Sense Ion 378, Hack). A scanning electron microscopy (SEM) image of the sludge produced after the coagulation/flocculation treatment was obtained using a SEM (JSM-6390LV; Make: Jeol, Japan). An electron dispersive spectroscopy (EDAX) analysis was done to determine the elemental composition of the crude extract coagulant, fresh FCE, and FCE sludge by using the FE-SEM (QUANTA 200 FEG).

3. Results and discussion

3.1. Influences of FCE dosage

Generally, the coagulation process is a surface phenomenon; therefore, coagulation performance can significantly be affected by surface charge because of the mass of the coagulant. Thus, from an economic point

Table 1
Experimental runs and conditions for MNZ removal by FCE.

		Conditions			
Run	Purpose	Water pH	MNZ concentration, mg/L	FCE, mg/L	Water temperature, ℃
1	Effect of FCE dose	7	50	0.75–5.25	24
2	Effect of water pH	2–12	50	1.75	24
3	Effect of initial concentration of MNZ	7	10–100	1.75	24
4	Effect of water temperature	7	50	1.75	10– 40
5	SVI and sludge production	7	50	1.75	24

of view, the optimization of coagulant dose and the best-required mass of the coagulant for the scale-up and designing of large-scale equipment are necessary. Hence, assays with FCE dosage variation on MNZ removal were carried out under the conditions given in Table 1. The results are plotted in Fig. 1. The study range of the FCE dosage was 0.75–5.25 mg/L. The optimum FCE dosage was obtained as 1.75 mg/L. At the optimum coagulant dosage, the maximum MNZ removal efficiency was 89.3%. As it can be appreciated in Fig. 1, by increasing the FCE dose (up to 1.75 mg/L) the MNZ removal was increased. This could be described to a couple of reasons; the increase in the adsorptive surface due to an increase in FCE amount and the formation of flocs and acid anion complexes by electrostatic interaction. The complexes led to an enhancement of the removal of colloids. Another point made by Fig. 1 is the decrease in MNZ removal for FCE doses of more than 2.25 mg/L. With the addition of a relatively large dosage of the coagulant, the surface charge of the particles gets reversed because of the continued adsorption of the mono- and polynuclear hydrolysis species of the active group on the surface of FCE [24]. As the colloidal particles become positively charged, they cannot be removed by perikinetic flocculation. These results are quite competitive to those stated by other researchers. For example, Sansalone and Kim [25] have applied up to 150 ppm of alum and up to 100 ppm of iron chloride to achieve a turbidity reduction of 75% in a similar municipal wastewater.

3.2. Influences of water pH

To find the optimum pH for maximum removal of MNZ by FCE, the effect of MNZ solution pH level ranging from 2 to 12 was investigated under the conditions given in Table 1 (run 2). The results of pH effect on MNZ removal are depicted in Fig. 2. As seen in Fig. 2, the maximum MNZ removal was obtained at solution pH of around 7. Based on data plotted in Fig. 2, the removal of MNZ by FAC improved from 77 to over 88.9% when the solution pH was increased from 2 to 7. A further increase of pH to 12 resulted in a reduction of MNZ removal to 61%. The protonation for both MNZ and FCE functional groups at neutral pH enhances the coagulation capacity of FCE. Coagulation of MNZ onto FCE in water is strongly influenced by the solution pH so that several possible

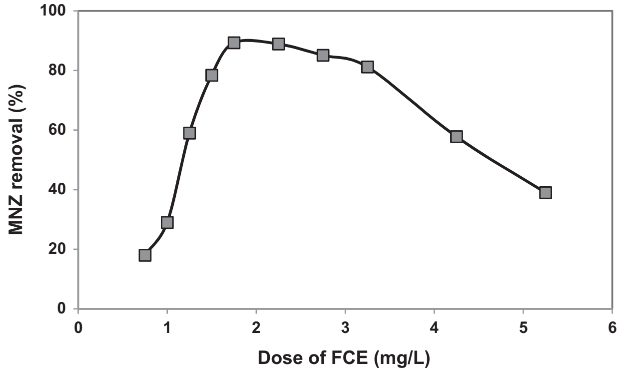

Fig. 1. Influence of FCE dosage on MNZ removal.

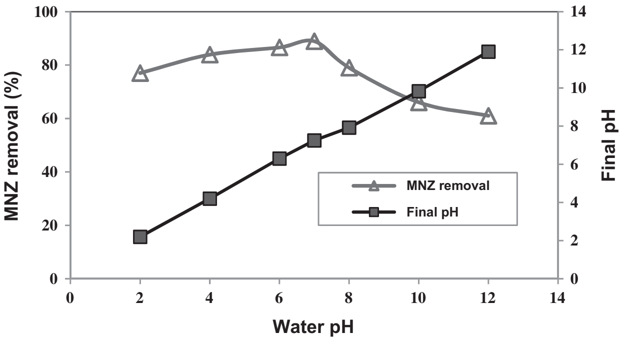

Fig. 2. Influence of water pH on MNZ removal.

coagulation mechanisms of MNZ by FCE can be proposed.

The first mechanism is cation-exchange mechanism. In principle, cations in FCE structure (see Table 2) were exchanged by protonated functional groups within MNZ which have positive charge. The positive charge of MNZ was due to the protonation of MNZ amino group by the presence of free H ions in the solution. For amino groups, the protonation take place as ($-NH_2 + H^+ \rightarrow -NH_3^+$) [26].

On the other hand, the exchange between protonated amino groups in the MNZ and cations in FCE structure such as Al^{3+}, Fe^{3+}, and Na^+ during coagulation process is indicated by the presence of these cations in solution after coagulation completes.

The second reason for increasing the removal efficiency of MNZ would be attributed to that with the increase of the water pH from 2 to 7, more hydroxyl ions reacted with the Fe and Al species in FCE to produce higher ferrite polymers, thus, improving the bridging flocculation. At the same time, the colloids particle charges were decreased by the charge neutralization mechanism, enhancing the destabilization of the colloidal particles in solution. However, at higher alkaline water, the coagulant would be susceptible to hydrolysis, inhibiting the bridging flocculation. That is, there was a distinct decrease in the MNZ reduction at pH more than 7. Therefore, the optimum pH value of 7 was attained to maximize the performance of coagulation.

The point that the maximum coagulation efficiency by FCE was attained at neutral pH is advantageous for full-scale application. This is because any necessity to changing the pH level of the water for efficient coagulation is eliminated making the process more cost-effective compared with other coagulants that require an acidic or alkaline optimum pH level to remove pollutants. Furthermore, the final pH of the solution at the end of each experiment was similar to the initial value (Fig. 2). Control of the pH at an optimal value is critical to attaining maximum performance. However, the results indicate that the adjustment of water pH during coagulation by FCE is unnecessary, which reduces the overall cost of treatment. The result is contradictory with observation that higher pH values are optimal for other natural coagulants e.g. from *M. oleifera* [18], from *P. juliflora*, and *C. latifaria* [24]. The contradictory reports can explain by differences in experimental conditions and type of coagulant.

Table 2
The EDAX analysis of the crude extract, fresh FCE, and FCE sludge

Element (wt%)	Value		
	Crude extract	Fresh FCE	FCE sludge
C	22.10	24.15	58.07
N	0.72	0.82	1.91
O	0.61	0.63	0.761
S	2.22	2.98	3.85
Al	0.16	0.170	0.132
P	0.77	0.52	0.61
Fe	0.64	4.77	4.01
Na	3.01	5.11	6.22
Cl	0.22	2.132	5.80

3.3. Influences of MNZ concentrations

The effect of various concentrations of MNZ (10–100 mg/L) on its removal by FCE was investigated under conditions specified in Table 1 (run 3). The results of the effect of initial MNZ concentration on MNZ removal by FCE are shown in Fig. 3. Generally, in all reactions, different concentrations caused changes in the driving force. Hence, increasing the MNZ initial concentration to 100 mg/L resulted in reduction in its removal percentage due to the ratio of MNZ to the constant mass of FCE. The similar result was reported by Šciban et al. [27]. Further, the percentage MNZ removal for all initial MNZ concentrations was significant (>70%). The ability of the FCE to work at different MNZ ranges may be reflected its versatility to treat various waters.

3.4. Influences of water temperature

Water temperature was evaluated as a parameter in the coagulation–flocculation process. The reason

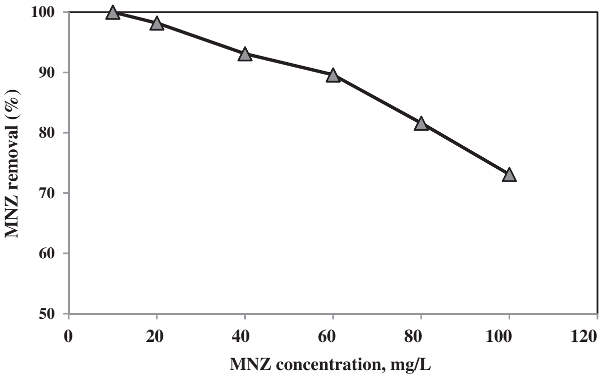

Fig. 3. Influence of initial MNZ concentration on its removal by FCE.

why it was introduced in this study has to do with seasonal variation; temperature of effluents may be rather different in summer than in winter; or affecting to lakes or ponds where wastewater temperature may be raised up. However, water temperature is also important in order to extrapolate the present results to other similar effluents, such as industrial ones, which may come into the treatment plant with very different conditions. As it can be seen in Fig. 4 as well, water temperature did not influence the effectiveness of the MNZ removal by FCE. By varying temperature from 10 to 40°C no enhancement or worsening in MNZ removal is observed. Hence, FCE may be an effective coagulant–flocculant agent even in the case of thermal-contaminated waters.

3.5. Sludge production

Sludge production is a critical task in order to evaluate efficiency in coagulation–flocculation process [28]. It may be as low as possible, and sludge volume may be reduced as well. Due to this fact, aluminum-induced coagulation usually is said to be a high-sludge production process, so it became a drawback [29]. In the case of FCE, sludge production, sludge volume, and the relationship between these two parameters, which is called SVI, were determined. SVI is defined by Eq. (2):

$$\mathrm{SVI} = V_s/W_s \tag{2}$$

where V_s denotes the volume that is occupied by the sludge (mL) and W_s denotes the sludge mass (g).

As it can be seen in Fig. 5, sludge volume increasing has a less steep slope than SVI. From 1.75 mg/L and ahead, flocculation capacity of FCE seems to be less efficient, and a sludge compression seems to appear as SVI decreases. This fact is rather normal in sedimentation process [30]. These SVI values are quite interesting because they are rather lower than others, as reported by Fernández-Leborans and Moro [31] where almost every SVI value was more than 150 mL/g. Furthermore, several disadvantages linked to alum or iron salts are avoided [32].

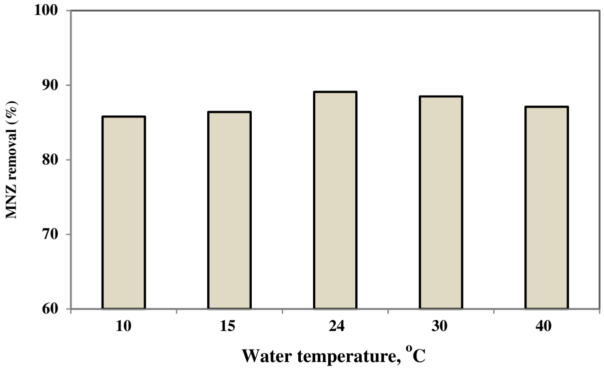

Fig. 4. Influence of water temperature on MNZ removal by FCE.

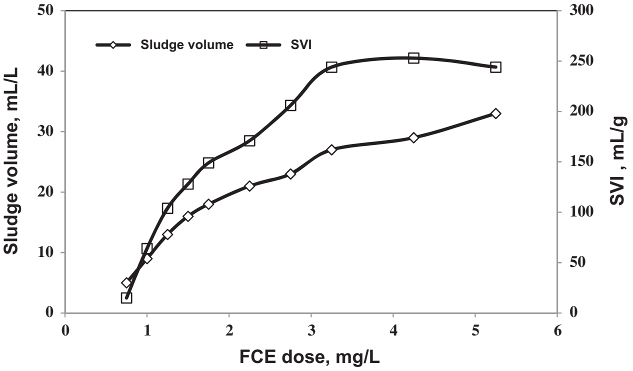

Fig. 5. SVI and sludge production during MNZ removal by FCE.

3.6. SEM micrograph and EDAX analysis

From the SEM micrograph (Fig. 6), it is clear that the sludge produced after the treatment of MNZ solution with FCE is hard and sturdy. The honeycomb-like structure, with small colloidal particles entrapped in it, might indicate the contribution of the sweep flocculation mechanism for the removal of colloidal particles in the water mixture. The chemical composition of the sludge and fresh FCE is given in Table 2. Among various elements, carbon was found to be dominant in the sludge as indicated by the MNZ removal. Regarding polysaccharide $[C_n(H_2O)_m]$, fatty acids $[CH_3–(CH_2)_n–COOH]$, and

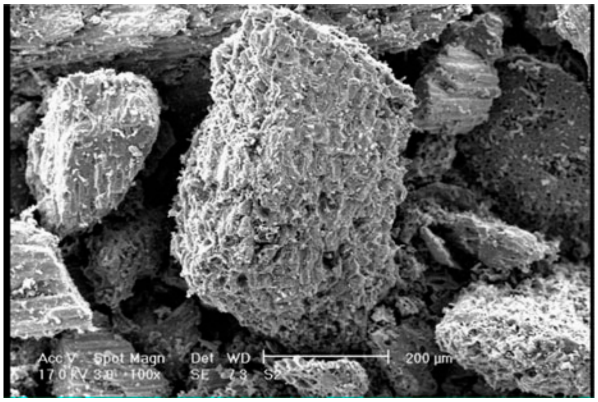

Fig. 6. SEM image of FCE floc.

protein general formula [$RCH(NH_2)COOH$], the obtained coagulant could be neither polysaccharide nor fatty acid as it contained N. Also, regarding mentioned formula, the polysaccharides contain high amount of oxygen rather than proteins and fatty acids, so the coagulant structure could not be made from polysaccharide. Further, we know that some amino acids such as *met*- and *cys*- amino acids contain S; thus, presence of S in the structure of coagulant might be another reason for proteinous of the coagulant. However, low values of P may be due to phospholipids acids in the extracted coagulant. Some proteins, namely metallo-protein, contain metals such as Fe; however, we modified the crude extract with $FeCl_3$. Na and other such metal is naturally presented in plant seeds. Therefore, we can conclude that the extracted coagulant was majority protein and minority phospholipids acids.

4. Conclusions

The removal of MNZ, an antibiotic, on the recently developed coagulant FCE, was examined under various operational variables. It was found that the removal rate of MNZ by FCE depended on solution pH level, FCE quantity, and MNZ concentration. The pH level for optimum coagulation was close to that of neutral water and this presents a significant advantage for practical application of the process. A high degree of MNZ could be removed by using a low amount of 1.75 mg/L FCE in a short contact time. Water temperature had a negligible influence on MNZ removal by FCE. Sludge production is reasonably within normal ranges, and presents no aluminum or iron salts disadvantages. Briefly, it can be concluded that the FCE has a high coagulation capacity toward the antibiotic drugs making FCE coagulation process a viable and efficient process for treatment of water containing such contaminants.

Acknowledgment

The authors acknowledge the Bushehr University of Medical Sciences for providing the technical and financial support for conducting this research.

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