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Evaluation of the Coagulation and Flocculation Process Using Plantago major L. Seed Extract as a Natural Coagulant in Treating Paper and Paperboard Industry Wastewater

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A R T I C L E I N F O

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ABSTRACT

Introduction: Paper and paperboard industry wastewater has potentially damaging effects on the ecosystem of the receiving environments due to the presence of toxic compounds. As a result, the treatment of such wastewater is necessary prior to discharge into the environment. In this study, the potential of *Plantago major* L. seed extract as a natural coagulant in treating paper and paperboard industry wastewater was investigated.

Materials and Methods: In this experimental study that was carried out in vitro, the effects of the pH (5, 6, 7, 8, 9, 10 and 11) of the solution and different doses (0.05, 0.1, 0.15, 0.2 and 0.25 g/L) of coagulant were studied. At each step of optimization turbidity, TSS and COD were measured according to the methods presented in the books on water and wastewater testing standards.

Results: Optimal pH for removal of turbidity, TSS and COD using *Plantago major* L. seed extract was determined 10, with the removal efficiency of 60%, 76% and 36%, respectively. The optimal dose of coagulant was obtained 0.1 g/L, with the maximum efficiency of 80%, 87% and 37.7% for turbidity, TSS and COD, respectively.

Conclusion: Based on the results of this study, *Plantago major* L. seed extract can be used as a natural coagulant without causing environmental risks.

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Introduction

The paper and paperboard industry is one of the industries that consume large amounts of water during the production process. For each tone of production, 30-180 m^3 of wastewater is discharged to the environment, while 20-70 m^3 of wastewater is produced per ton of paper [1]. Toxic contaminants present in the wastewater of the paper and paperboard industry include tannin, suspended solids, lignin, Chloride compounds, fatty acids, and metals [2, 3]. Although this industry helps the environment - due to use of waste papers and their collection, drainage of the wastewater without treatment and recycling

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of its useful materials, will have many adverse impacts on the ecosystem of receiving aqueous environments [4, 5]. Coagulation studies have been carried out on the treatment of paper and paperboard industry wastewater using inorganic coagulants such as alum, iron salts, poly aluminum chloride, and polyacrylamides [6]. Although inorganic coagulants produce high removal efficiency in the coagulation process, they have toxic effects on human health, generate high volumes of sludge, and are deactivated at low temperatures [7]. In recent years, there has been a particular interest in the use of natural coagulants in developing countries, due to their advantages such as biodegradability and minimum environmental risks [8, 9]. Chestnuts and acorn seed extract [10] and cactus [11], pistacia atlantica [12], tannin [13], Robinia [14] and okra seed extract [15] are a number of natural coagulants used to remove contaminants such as turbidity and color from water and wastewater. *Plantago major* L. is a perennial plant of the Plantaginaceae family. Approximately 200 Plantago species have been identified in temperate and tropical regions [16]. The species used in this study, *Plantago major* L. is a valuable and useful plant that can grow up to 70 cm. *Plantago major* L. has large and wide leaves on the ground. This plant grows all over the north, center, and south of Iran. It occurs in the vast regions of Europe, North Africa, and North America [17, 18]. The active ingredients of the plant include plantagin, succinic acid, adenine, and choline. *Plantago major* L. seeds contain gum, mucilage, and glycosidic compounds [17]. Subramonian et al. examined the function of *Cassia obtusifolia* seed extract in the treatment of paper and paperboard wastewater. They achieved efficiency of 86.9% and 36.2% for removal of TSS and COD, respectively, at pH 5 and the optimal dose 0.75 g/L, and after centrifugation at 10 rpm for 10 min [1]. Freitas et al. studied textile wastewater treatment using okra (*Abelmoschus esculentus*) mucilage. In that study, the efficiency of color, turbidity, and COD removal was obtained, respectively, 15.97%, 97.24%, and 85.69% using 3.2 mg/L okra in combination with 88 mg/L of ferric chloride at pH 5 [15]. Shamsnejati et al. examined the function of *Ocimum basilicum* seed extract in the treatment of textile wastewater, and observed that the efficiency of color and COD removal was 68.5% and 61.6%, respectively [19].

The aim of this study was to evaluate the efficiency of the coagulation and flocculation process in the treatment of paper and paperboard industry wastewater using *Plantago major* L. seed extract as a natural coagulant.

Materials and Methods

Extraction of Plantago major L. seed

Plantago major L. seeds were prepared from Mazandaran province. To obtain the extract, the seeds were at first heated in an oven at 100 °C for 2 h. After the seeds were ground, 2.5 g of the resulting dried powder was added to 100 ml of boiling water and 0.9% sodium chloride [15, 20]. The resulting solution was stirred for 2 h by a magnetic stirrer [21] to extract and activate the coagulation components [12]. The extract was then passed through a cloth filter.

Sampling

In this research, the wastewater drained from a paper and paperboard plant was sampled as combined. Temperature, pH, and EC were measured at the site of sampling, and the samples were examined in the laboratory according to the methods presented in the books on water and wastewater testing standards.

Methods and instruments for measuring parameters

In this research, pH was measured using a pH meter (151 Mi, England) and EC by an EC meter (HQ 40, HACH, USA).The turbidity was measured by a turbidity meter (A-TUR-1.16), the TSS by measuring weight (2540 D), the TDS by the (2540 C) method, and the COD by closed reflux colorimetric method [22]. A spectrophotometer (HACH, USA) at 600 nm was used to measure the COD of the samples. A Jar test (402-7790, HACH, USA) was used to perform the experiments and digital scale (mettle

AE240) to weigh materials with detection limit 0.0001 g.

Coagulation and flocculation process

In order to determine the optimal pH, samples of the same amount (1 L) of wastewater were injected into a Beaker. Then, using a solution of sodium hydroxide and normal 1 Hydrochloric Acid, their pH was adjusted to 5-11. Then, by keeping the optimal pH obtained in the previous step fixed, the coagulant at different doses (0.05, 0.1, 0.15, 0.2, and 0.25 g/L) was added to the sample. All experiments were performed in centrifugation at 170 rpm for 60 s and then a comparatively slower centrifugation at 10 rpm for 10 min [1]. The sedimentation time was examined after each step of the process for 30 min. After each step of sedimentation turbidity, TSS and COD were measured to determine optimal conditions.

Sludge Volume Index (SVI)

To determine the value of SVI at the step of determining pH and optimal dose, the liquid sample was poured in a 1 L graduated cylinder, and 30 min later, the volume deposited in the cylinder was measured. According to the deposited volume and Mixed Liquor Suspended Solid concentration of the sample, the SVI was calculated by Equation 1:

$$\mathrm{SVI}\left(\frac{\mathrm{ml}}{\mathrm{g}}\right) = \frac{\left(\text{Sluge volume settled}, \frac{\mathrm{ml}}{\mathrm{l}}\right)\left(\frac{10^3\,\mathrm{mg}}{\mathrm{g}}\right)}{\left(\text{Suspended solids}, \frac{\mathrm{mg}}{\mathrm{l}}\right)}$$

Analysis

Graphs were prepared using Excel 2010 and data were analyzed by the SPSS version 23 using one-way ANOVA. $P < 0.05$ was considered as a significance level.

Ethical issues

This article was derived from a student research project with the code of ethics IR.SSU.SPH.REC.1396.140.

Results

Wastewater characteristics

Physical-chemical characteristics of the crude wastewater of paper and paperboard industry studied are presented in Table 1.

Table 1: Physical-chemical characteristics of crude wastewater of paper and paperboard industry

Characteristic	Unit	Average ± Standard deviation
pH	-	0.42 ± 6.6
Temperature	℃	40 ± 0
Ec	mS/cm	0.48 ± 10.63
Turbidity	NTU	353.5 ± 2250
TDS	mg/L	325.2 ± 7090
TSS	mg/L	116.6 ± 1942.5
COD	mg/L	883.8 ± 15125

Determining optimal pH

Figure 1 illustrates the effect of initial pH values (7-11) on the efficiency of turbidity, TSS and COD removal by the extract of *Plantago major* L. seed. As it can be seen, with increasing pH, the efficiency of pollutants removal increases. The efficiency of turbidity, TSS and COD removal at pH higher than 10 did not significantly increase. As a result, pH 10 was determined as optimal pH, and the removal percentages of turbidity, TSS and COD were obtained 60%, 76% and 36%, respectively. Therefore, the initial pH of the solution for other tests was determined 10. There was a significant difference between pH and all studied parameters ($P < 0.05$). In addition, the effect of *Plantago major* L. seed extract on initial pH of wastewater is illustrated in Figure 2.

Determining the optimal dose of coagulant

The effect of the doses (0.05-0.25 g/L) of the coagulant is illustrated in Figure 3. The maximum efficiency of turbidity, TSS and COD removal was, respectively, 80%, 87% and 37.7% at 0.1 g/L. Therefore, the optimal dose of

Plantago major L. seed extract was 0.1 g/L. There was a significant difference between different concentrations and studied parameters ($P < 0.05$).

Investigating SVI using P. major seed extract

In Figure 4 (a), the effect of pH on the SVI is illustrated. Figure 4 (b) shows the effect of different doses of the coagulant on the SVI.

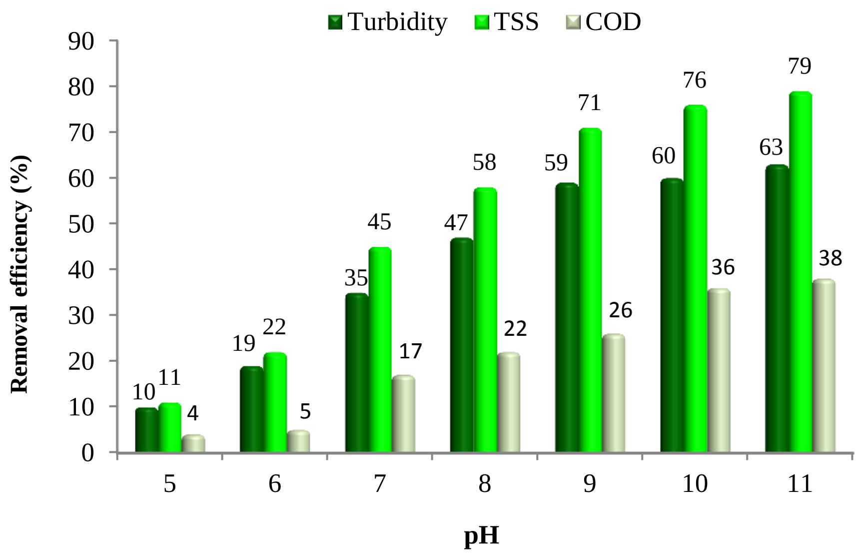

Figure 1: Initial pH effects on the removal percentage of turbidity, TSS, and COD from wastewater (Constant coagulant dose = 0.15 g/L)

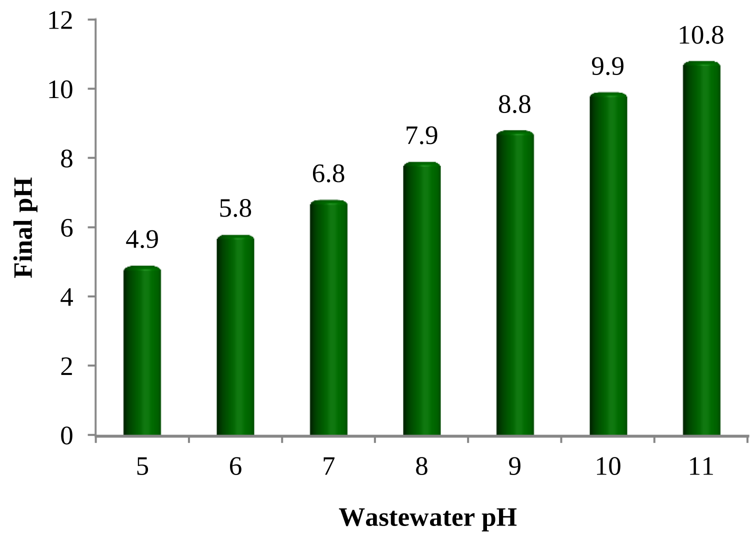

Figure 2: Effect of *Plantago major L.* seed extract on initial pH of wastewater

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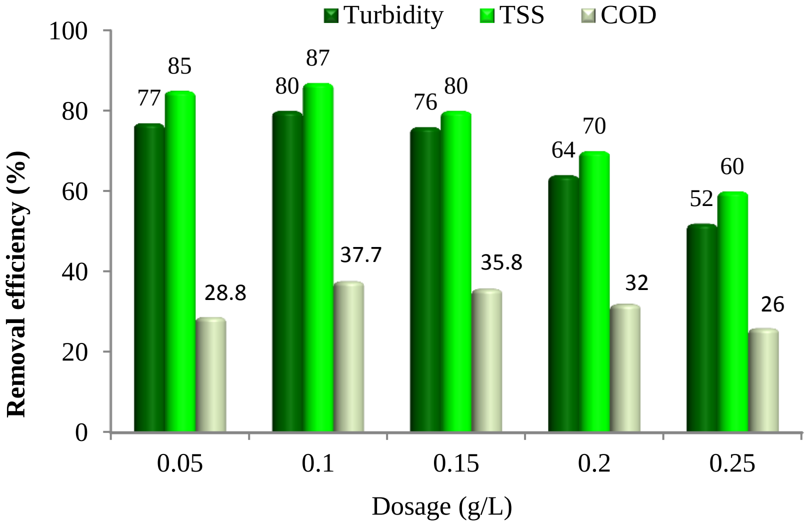

Figure 3: Effects of different doses of coagulant on the removal percentage of turbidity, TSS, and COD from wastewater (pH 10 in all samples)

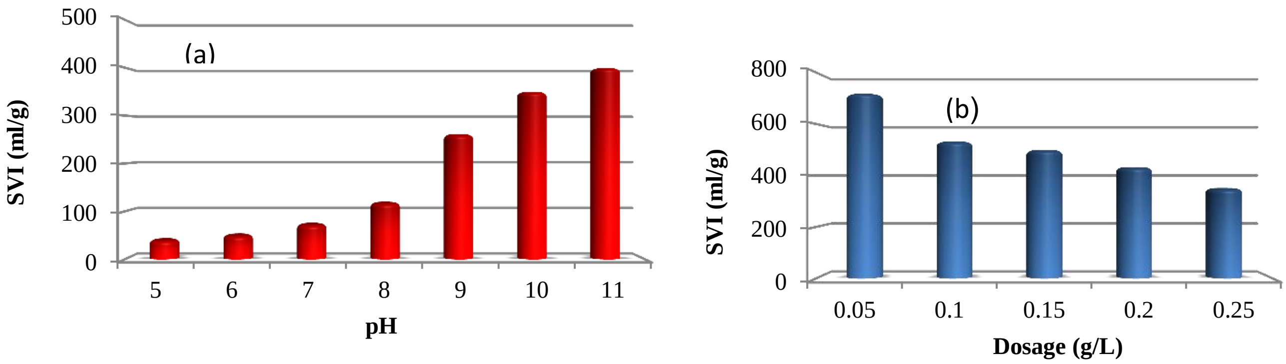

Figure 4: The effect of pH (a) and the effect of different doses of coagulant (b) on sludge volume index (SVI)

Discussion

The pH of solution is one of the important parameters for the removal of contaminants in the wastewater in the process of coagulation and flocculation [1, 23]. The pH of the solution affects the chemistry of the aqueous solution and absorbent surface bonds [12]. The results of this study showed that with increasing pH, the coagulation and removal of pollutants from the environment increased. The maximum removal of pollutants in this study at alkaline pH of 10 is probably due to the reaction of organic compounds with hydroxide ions, which forms hydroxide flocs [15]. The results of the study by Subramonian et al. showed that the acid pH of 5 was determined as the optimal pH [1], which is not consistent with our study. This result can be attributed to the coagulant's characteristics [24]. In addition, the results showed that *Plantago major* L. seed extract had no significant effect on pH, so that the difference between the initial and final values of pH was not significant (Figure 2). The study of Yarahmadi et al. on turbidity removal by Moringa olifera seed extract, showed that use of M. olifera as coagulant did not affect pH level [25]. A study was conducted by Thakur and Choubey on the removal of turbidity using natural coagulant. The results have shown that increasing pH improves the efficiency of turbidity removal, which is in line with the findings of the current study [26]. Coagulant dose is considered one of the most important parameters to determine optimal conditions for the coagulation and flocculation process. Inadequate or excessive coagulant dose may result in poor function in the flocculation [27]. Therefore, studying the effects of coagulant dose

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is important not only for evaluating economic goals, but also for preventing too much coagulant use in wastewater treatment [1]. As illustrated in Figure 3, the efficiency of turbidity, TSS, and COD removal decreased with increasing coagulant dose. The study of Ramavandi and Farjadfard on the effect of Plantago ovata at 0.5-5 mg/L doses in removing COD from textile wastewater showed that COD removal decreased at doses above 2 mg/L [28]. A further increase in the dose of the coagulant used instead of coagulation actually results in the re-stability of colloidal particles and the return of particle load [29]. In general, it can be concluded that in lower doses, natural coagulants have better efficacy in removing pollutants compared to higher doses. This point is not only economically important but also creates less concentrated organic materials. Solid sludge is formed due to the presence of organic and suspended materials that have been removed from the wastewater during the coagulation process as the coagulant was being used. In general, the amount and properties of the sludge formed during the coagulation and flocculation process depends on the type of the coagulant's material and the conditions of the experiment. The effects of polymers, osmotic pressure, and hydration affect the SVI in the coagulation and flocculation process [30]. As shown in Figure 4, with increasing wastewater pH, the SVI increases, and Separation of water and sludge becomes more difficult. By increasing the dosage of coagulant, SVI decreases, which increases the amount of sludge compression, which will result in better separation of water from sludge in the treatment process. The highest SVI value at an average dose of 720 ml/g was obtained 0.05 g/L, which disrupted the separation process, and the lowest SVI value at 0.25 mg/L was obtained 350 ml/g.

Conclusion

The results of this study showed the optimal dose of coagulant with the maximum efficiency of 80%, 87% and 37.7% for removal of, respectively, turbidity, TSS and COD was obtained 0.1 g/L. Therefore, *Plantago major* L. seed extract can be used as a natural coagulant.

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Conflict of interest

There is no conflict of interest.

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