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Paper-recycling wastewater treatment using *Ocimum basilicum L.* along with alum: Optimization by response surface methodology (RSM)

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ABSTRACT

The application of natural coagulants in coagulation-flocculation (CF) process in water and wastewater treatment includes many advantages compared with chemical agents, such as particular biodegradability, low toxicity, being economical and generating low volumes of sludge. In this research, *Ocimum basilicum L.*, as an active plant-based coagulant, in combination with alum, has been evaluated to treat paper-recycling wastewater. Response surface methodology (RSM), based on central composite rotatable design (CCRD), was employed to optimize the treatment process. The effect of three parameters including pH, alum- *O. basilicum* ratio and slow mixing contact time on the chemical oxygen demand (COD) and turbidity removal was analyzed. Results revealed that the maximum removal of COD and turbidity could be achieved in optimum conditions, i.e., pH value of 8.3, alum-*O. basilicum* ratio of 2.8 and contact time of 48.8 min, in which the reduction efficiencies of COD and turbidity were 39.7 and 81.4 %, respectively. The model predictions also agreed well with experimental data. In order to gain further insights into the properties of coagulants and produced flocs, Fourier Transform Infrared Spectroscopy (FTIR) and Thermal Gravimetric Analysis (TGA) employed as characterization methods. Results suggested presence of active functional groups on *O. basilicum* and the thermal stability of formed flocs. Furthermore, CF mechanism was most likely based on charge neutralization and adsorption with interparticle bridging. The present study showed that *O. basilicum* mucilage could be used as a coagulant aid in combination with alum in the treatment of paper-recycling wastewater.

Keywords: Ocimum basilicum; Coagulation; Paper-recycling wastewater

1. Introduction

Integrated pulp and paper industry is considered as a highly water-intensive sector and a major source of aquatic pollution [1,2]. More than 250 chemicals are produced at various stages of pulp & paper production, which are also found in the wastewater. Discharge of untreated pollutants into the environment can cause important and various environmental problems due to their aggregation in recipient water and soil [3,4].

Depending on the type of paper mills (packaging, recycling, Kraft), several treatment technologies have been applied to minimize the harmful effects of effluents on the environment [5]. However, the primary clarification, secondary treatment and tertiary processes are the main methods used in pulp and paper mills which can be simply

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divided into physicochemical and biological processes [6]. According to research conducted by Helble et al. [7] and Chamarro et al. [8], samples with biodegradability index (BOD_5/COD) smaller than 0.3 are not completely biodegradable and for complete biodegradation, the effluent should have an index of at least 0.40.

Coagulation-flocculation as a physicochemical process, has also been extensively studied to treat the pulp and paper mill wastewater. In such studies, polyaluminium chloride (PAC), chitosan, cationic and anionic polyacrylamides (PAMs) have been evaluated in the CF process, and various levels of pollutants removal efficiency have been reported [9–11].

Conventional chemical-based coagulants including alum, ferric chloride, ferric sulphate and PAC, in combination with organic polymers as coagulant aid have several drawbacks such as relatively high costs, ineffectiveness at low temperatures, adverse effects on human health, production of large volumes of sludge and a change in the pH level of treated water. Therefore to solve these problems, other environmentally acceptable and economically viable materials are desirable to be introduced and replaced with the synthetic coagulants [12,13].

Generally, natural coagulants including polysaccharides, are a family of carbohydrates and when having charge, they behave as polyelectrolytes. Commonly, the negatively charged groups are carboxylic or sulfate groups and positively charged ones are ammonium groups [14].

Therefore, since natural polymeric macromolecules extracted from plants, as plant-based coagulants, are highly biodegradable, non-toxic, non-corrosive and locally grown, they are more appropriate and cost-effective in comparison with imported chemicals [15,16].

Ocimum basilicum L., is usually called basil; that is an aromatic herb grown in the tropical regions of Asia, Africa, and Central and South America. Its leaves are applied as a spice in traditional cuisine [17].

The mucilaginous layer structure of the *O. basilicum* swollen seeds is a pectinous matrix with a large capacity for hydration. The polysaccharides obtained from *O. basilicum* seeds contain two major fractions of glucomannan and (1→4)-linked xylan and a small fraction of glucan [18,19].

The seeds of *O. basilicum* have been used as biosorbents to remove hexavalent chromium, the radionuclides and copper from aqueous solutions [19–21] .*O. basilicum* mucilage has been analyzed as an active natural coagulant in combination with alum for leachate pretreatment, and it appears to be a highly effective coagulant in combination with alum. This combination reduced the amount of required alum to achieve a high COD and color removal efficiency [22]. In addition, the *O. basilicum* has been evaluated for the removal of dye from textile wastewater and therefore, high color and COD removal efficiency was obtained using a low amount of this coagulant. It was also found that *O. basilicum* can be highly effective in treating real textile wastewater in combination with alum [23].

The recovery of alum is a very influential parameter on the operating costs of treatment plants. Optimal recovery condition of alum have been found regarding acidic pH value of 1.3 and the recovered alum was proved as desired coagulant almost the same as the fresh alum [24]. Besides, addition of sulphuric acid (at pH value of 2) to the sludge has improved the extraction of the aluminum by 70% in a 30 min contact time. Finally, savings of US \$3.78/1000 m^3 was reported by the recovery of alum and the treatment cost may be reduced by 60% if a lime stabilization process is selected [25]. Besides, *O. basilicum* is biodegradable and it is presumed to be safe for human health. In addition, natural coagulants produce readily biodegradable and less volume of sludge which was reduced by 20–30% of that the sludge treated by alum [26]. Moreover, current study was as a chemical pretreatment of MBR system for reuse of water. This pretreatment has positive effects on prevention of membrane fouling [27].

In conventional multifactor experiments, process optimization is usually carried out through varying a single factor while fixing all other factors on specific conditions. It is time-consuming and usually unable to reach the true optimum since the interactions between factors are ignored. As a solution, the response surface methodology (RSM) has been proposed to determine the influence of individual factors and also their interactive influence. In this method, an experimental design such as the central composite design (CCD) was used in order to fit a modeling by the least squares technique. Furthermore, analysis of variance (ANOVA) provides the statistical results and diagnostic checking tests that enables researchers to evaluate the adequacy of model [28–31].

The main objective of this study is to investigate the optimization of COD and turbidity removal using RSM. In addition, the chemical structure of *O. basilicum* as plant based coagulant was investigated. The current study was a pretreatment step for membrane bioreactor system for reuse of paper-recycling plant wastewater. The pH, alum-*O. basilicum* ratio and slow mixing contact time were chosen as independent parameters. Coagulant and floc characteristics were also analyzed using FTIR and TGA.

2. Materials and methods

The samples of paper-recycling wastewater were collected from Kahrizak paper mill located in approximately 10 km far from Tehran, Tehran province, Iran. All samples were collected manually in 20-l polyethylene containers. Then they were transferred to the laboratory and, stored at 4°C to reduce biodegradation owing to biological activity. Table 1 represents the range and mean values for 6 measurements from September to November 2016 (3 months).

The COD, turbidity and TSS were measured according to the Standard Methods (American public health associa-

Table 1
Paper-recycling wastewater characterization

Characteristics	Range	Mean
Color	Brown	–
Temperature (°C)	26–31	29
pH	6.5–7.4	7.1
COD (mg l^{-1})	6,000–7,000	6,830
BOD5 (mg/l)	2620–2860	2630
Total suspended solids (TSS) (mg l^{-1})	3,740–5,990	5,580
Turbidity (NTU)	1,360–4,200	3,750

tion standards) [32]. The pH of the solution was adjusted by 20% NaOH or H_2SO_4 solution during the experiment.

2.1. Preparation of O. basilicum mucilage coagulant

O. basilicum whole seeds were purchased locally. The seeds were dried in an oven at 100°C for 2 h. Then, the seeds were ground and sieved to achieve 70 mesh (210 µm) particle size. In the next step, the obtained powdered seeds were soaked in warm water and 0.9% NaCl solution and stirred for 2 h. The mixture was centrifuged at 30 rpm for 10 min, so that the *O. basilicum* mucilage was obtained [22,33].

2.2. Jar-test experiment

Coagulation process was performed in a jar test apparatus equipped with six beakers (Phipps & Bird Stirrer, Mod-7790-400). Various rates of alum and *O. basilicum* were added into the paper-recycling wastewater according to experimental design.

The alum concentration was fixed at 2000 ppm, and *O. basilicum* concentrations were changed from 200 to 2000 ppm. The solution was stirred at rapid-mixing stage at 120 rpm for 5 min. The effluent was then subjected to slow mixing at 40 rpm for various contact times and allowed to settle for 30 min [22]. Then, the supernatant was evaluated in terms of COD and turbidity reduction percentage. The coagulation efficiency of each experimental run was represented by COD and turbidity removal as shown in the following equations:

$$COD\ removal\ (\%) = \frac{COD_i - COD_f}{COD_i} \times 100 \quad (1)$$

$$Turbidity\ removal\ (\%) = \frac{Turbidity_i - Turbidity_f}{Turbidity_i} \times 100 \quad (2)$$

where COD_i and COD_f are the initial and final COD (mg l^{-1}), respectively. All experiments were performed in triplicate.

2.3. Design of experiments

The CCD and the Box-Behnken are the two most common designs being used extensively in RSM. In this work, the CCRD with three variables was used to evaluate the response pattern and it creates a design having the standard error of predictions equal at points equidistant from the center of the design. The CCRD, in comparison with Box-Behnken, is more appropriate for sequential experimentation. It also provides a reasonable amount of information to test lack of fit while not involving an unusually large number of design points, which provide high quality predictions over the entire design space [34,35].

The factors and their optimized ranges for independent variables were pH (2–12), alum- *O. basilicum* ratio (1:1–10:1) and slow mixing time (5–60 min) and each variable were at five levels, namely, –1.68, –1, 0, 1 and 1.68. COD and turbidity reduction efficiency were analyzed as the process responses. The design of experiments was shown in Table 2. A software package of design expert version 7.1.3

Table 2
Experimental design based on CCRD, and COD and turbidity reduction as responses

Run order	Independent variables						COD Reduction (%)			Turbidity Reduction (%)		
	pH (A)		contact time (min) (B)		alum- *O. basilicum* Ratio (C)		Experimental	Predicted	Residual	Experimental	Predicted	Residual
1	9.97	(1)	16.15	(–1)	8.18	(1)	22.5	20.8	1.7	68.2	67.7	0.5
2	7.00	(0)	5.00	(–1.68)	5.50	(0)	27.9	28.9	–1	69.1	69.4	–0.3
3	4.03	(–1)	16.15	(–1)	8.18	(1)	22.6	22.4	0.2	54.2	53.4	0.8
4	9.97	(1)	48.85	(1)	8.18	(1)	31.5	31.2	0.3	68.6	68.3	0.3
5	9.97	(1)	48.85	(1)	2.82	(–1)	38.4	38.2	0.2	80.7	82.5	–1.8
6	2.00	(–1.68)	32.50	(0)	5.50	(0)	20.1	19.4	0.7	50.0	49.0	1
7	7.00	(0)	32.50	(0)	5.50	(0)	37.3	35.1	2.2	77.7	76.3	1.4
8	9.97	(1)	16.15	(–1)	2.82	(–1)	34.4	31.2	3.2	79.3	79.7	–0.4
9	4.03	(–1)	48.85	(1)	8.18	(1)	30.0	29.7	0.3	60.9	61.5	–0.6
10	7.00	(0)	32.50	(0)	5.50	(0)	36.6	35.1	1.5	77.5	76.3	1.2
11	4.03	(–1)	48.85	(1)	2.82	(–1)	28.9	30.2	–1.3	63.0	64.5	–1.5
12	7.00	(0)	60.00	(1.68)	5.50	(0)	38.7	38.2	0.5	80.4	78.7	1.7
13	4.03	(–1)	16.15	(–1)	2.82	(–1)	29.4	29.3	0.1	52.8	54.1	–1.3
14	7.00	(0)	32.50	(0)	5.50	(0)	34.3	35.2	–0.9	75.8	76.3	–0.5
15	7.00	(0)	32.50	(0)	1.00	(–1.68)	40.6	39.8	0.8	73.2	70.8	2.4
16	7.00	(0)	32.50	(0)	5.50	(0)	31.8	35.1	–3.3	75.9	76.3	–0.4
17	7.00	(0)	32.50	(0)	5.50	(0)	36.1	35.1	1	78.3	76.3	2
18	7.00	(0)	32.50	(0)	5.50	(0)	34.9	35.1	–0.2	72.6	76.3	–3.7
19	7.00	(0)	32.50	(0)	10.00	(1.68)	26.7	28	–1.3	57.2	58.2	–1
20	12.00	(1.68)	32.50	(0)	5.50	(0)	23.7	24.9	–1.2	76.6	76.3	0.3

(Stat-Ease, Statistics Made Easy, Minneapolis, MN, USA) was used in this research.

3. Results and discussion

3.1. Analysis of COD reduction

The COD reduction values of the coagulation-flocculation process are listed in Table 2. The regression equation of the experimental results is as follows:

$$\begin{aligned} COD\ reduction = & +35.15 + 1.61A + 2.79B - 3.51C + 0.76AB \\ & -1.64AC + 1.61BC - 4.60A^2 - 0.57B^2 - 0.44C^2 \end{aligned} \quad (3)$$

Statistical testing of the model was carried out with the Fisher's statistical test for analysis of variance. The results of the ANOVA for COD reduction are presented in Table 3. The quadratic regression indicates that the model was significant and the value of the correlation coefficient (R^2 = 0.96) indicates that only 4% of the total variation could not be explained by the empirical model. Regarding COD reduction, the response surface plots of the quadratic model with one variable fixed at central level and the other two varying ones within the experimental ranges are shown in Figs. 1a–c.

Fig. 1(a) shows the effect of pH, contact time, and their interactions on the COD removal at center point of the experiment design. The maximum COD removal was achieved at a pH range of 6.5–8.5 and time of 60 min. At pH < 6 and pH > 9, COD reduction decreased sharply. In comparison with pH, the slow mixing time did not have significant effect on the COD reduction and it was according to usage of alum- *O. basilicum* for leachate treatment [22].

Fig. 1b depicts the effect of pH and alum- *O. basilicum* ratios on the COD removal. By increasing the amount of *O. basilicum* mucilage (low alum *O. basilicum* ratios), the COD reduction increased and the optimum pH for CF process was 6.5–8.5. *O. basilicum* as a natural polymer forms netlike structure and acts as a bridge [36]. Moreover, the pH range of 6–8 is the optimum condition of sweep floc for alum and, the negatively charged colloidal particles are entrapped in the precipitating particles of $Al(OH)_3$ [37].

Fig. 1c shows the effect of contact time and alum- *O. basilicum* ratio on the COD removal. According to the results, the effect of low mixing time on COD reduction was not very significant but as alum- *O. basilicum* ratios was reduced, the COD reduction increased. Because there

Table 3
ANOVA for COD reduction

Source	Sum of squares	DF	Mean square	F-value	P-value (P > F)
Model	661.15	9	73.46	24.82	< 0.0001
A	35.29	1	35.29	11.92	0.0062
B	106.09	1	106.09	35.84	0.0001
C	167.84	1	167.84	56.71	< 0.0001
AB	4.65	1	4.65	1.57	0.2385
AC	21.45	1	21.45	7.25	0.0226
BC	20.80	1	20.80	7.03	0.0243
A^2	304.84	1	304.84	102.99	< 0.0001
B^2	4.66	1	4.66	1.57	0.2381
C^2	2.85	1	2.85	0.96	0.3493
Residual error	29.60	10	2.96		
Lack of fit	9.96	5	1.99	0.51	0.76
Total	690.75	19			

R^2 = 0.96, Adj R^2 = 0.92, Pred R^2= 0.85

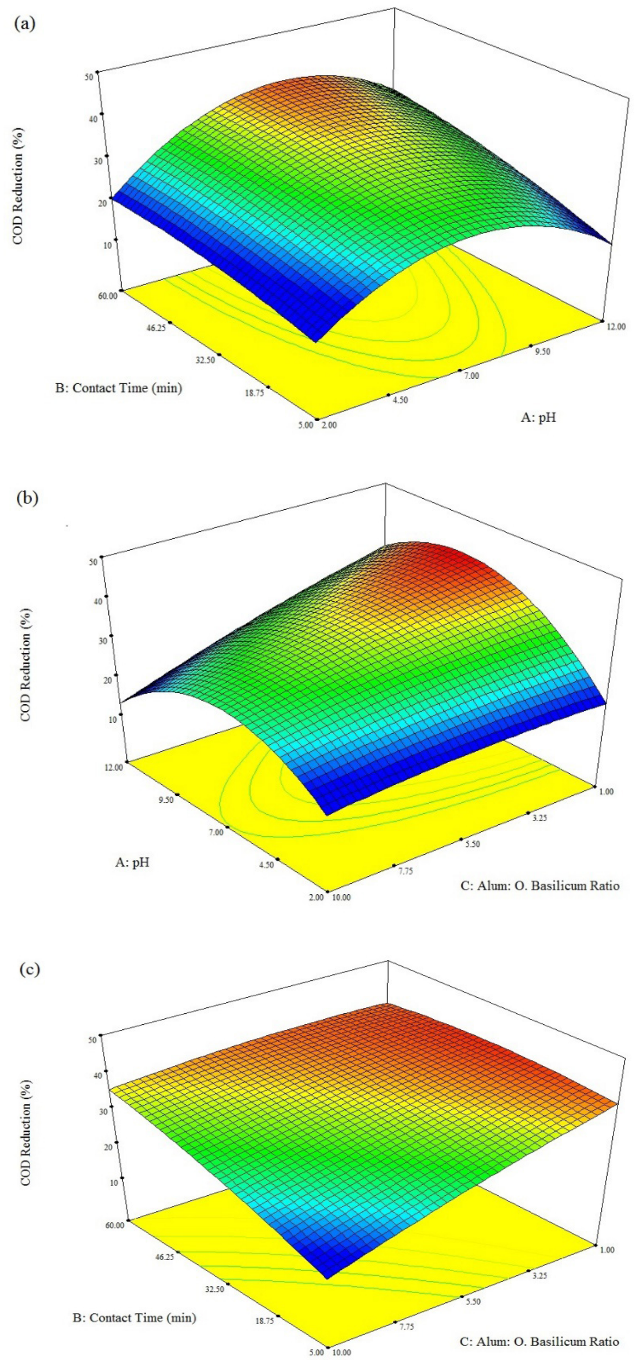

Fig. 1. Surface graphs of COD reduction (a) effect of pH and contact time (b) effect of pH and alum- *O. basilicum* ratio and (c) effect of contact time and alum- *O. basilicum* ratio.

are two main stages in an absorption reactions in which, the initial rapid absorption is a kind of surface reaction and its velocity is more quickly, in second stage velocity is very small because it is a process during which the organics transfer and diffuse into the internal pore [38].

3.2. Analysis of supernatant turbidity reduction

The experimental results of turbidity reduction are also summarized in Table 2. Similar to the COD reduction, the regression model [Eq. (4)] from results was obtained as follows:

$$\begin{aligned} \textit{Turbidity reduction} = &+76.34 + 8.10A + 2.76B - 3.72C - 1.89AB \\ &-2.81AC - 0.56BC - 4.86A^2 - 0.82B^2 - 4.19C^2 \end{aligned} \quad (4)$$

The results of the ANOVA for turbidity reduction are shown in Table 4. The quadratic regression shows that the model was significant and the value of the correlation coefficient ($R^2 = 0.98$) indicates that only 2% of the total variation could not be explained by the empirical model.

Regarding turbidity reduction, the response surface plots of the quadratic model with one variable fixed at central level and the other two varying ones within the experimental ranges are shown in Figs. 2a–c.

Fig. 2a clearly shows the effect of pH and contact time on the turbidity removal. With increasing pH and contact time to 8.5 and 45 min, respectively, turbidity reduction increased and then reduced. It can be seen that time was not, in general, a very significant parameter for the turbidity removal after 45 min. Similar to COD reduction, optimum pH for the turbidity removal of paper recycling wastewater was found to be in the range of 5.5 to 8.5. Aggregation of colloidal particles is generally due to effects of charge neutralization and sweep floc. The pH range of 4–5.5 is suitable for the charge neutralization between positively charged coagulant (such as aluminum or iron salts) and negatively charged colloids. Turbidity removal was achieved by adsorption of the amorphous $Al(OH)_3$ and pH range of 6–8 was favorable because the aluminum ions need sufficient alkalinity [29,39]. Therefore, pH range of 5.0 to 8.5 was the optimal range for charge neutralization and floc sweeping.

Fig. 2b demonstrates the effects of pH and alum- *O. basilicum* ratio on the turbidity. By increasing pH and reducing alum- *O. basilicum* ratio, the reduction of turbidity increased. Maximum turbidity removal was obtained at pH 9.5 and alum- *O. basilicum* ratio of 2. After this point, turbidity reduction reduced slightly.

Table 4
ANOVA for turbidity reduction

Source	Sum of squares	DF	Mean square	F-value	P-value (P > F)
Model	1824.90	9	202.77	48.72	< 0.0001
A	896.27	1	896.27	215.36	< 0.0001
B	104.10	1	104.10	25.01	0.0005
C	189.03	1	189.03	45.42	< 0.0001
AB	28.50	1	28.50	6.85	0.0257
AC	63.28	1	63.28	15.21	0.0030
BC	2.53	1	2.53	0.61	0.4535
A^2	340.94	1	340.94	81.92	< 0.0001
B^2	9.59	1	9.59	2.30	0.1600
C^2	253.27	1	253.27	60.86	< 0.0001
Residual error	41.62	10	4.161724		
Lack of fit	20.12	5	4.02	0.94	0.53
Total	1866.52	19			

$R^2 = 0.98$, Adj $R^2 = 0.96$, Pred $R^2 = 0.90$

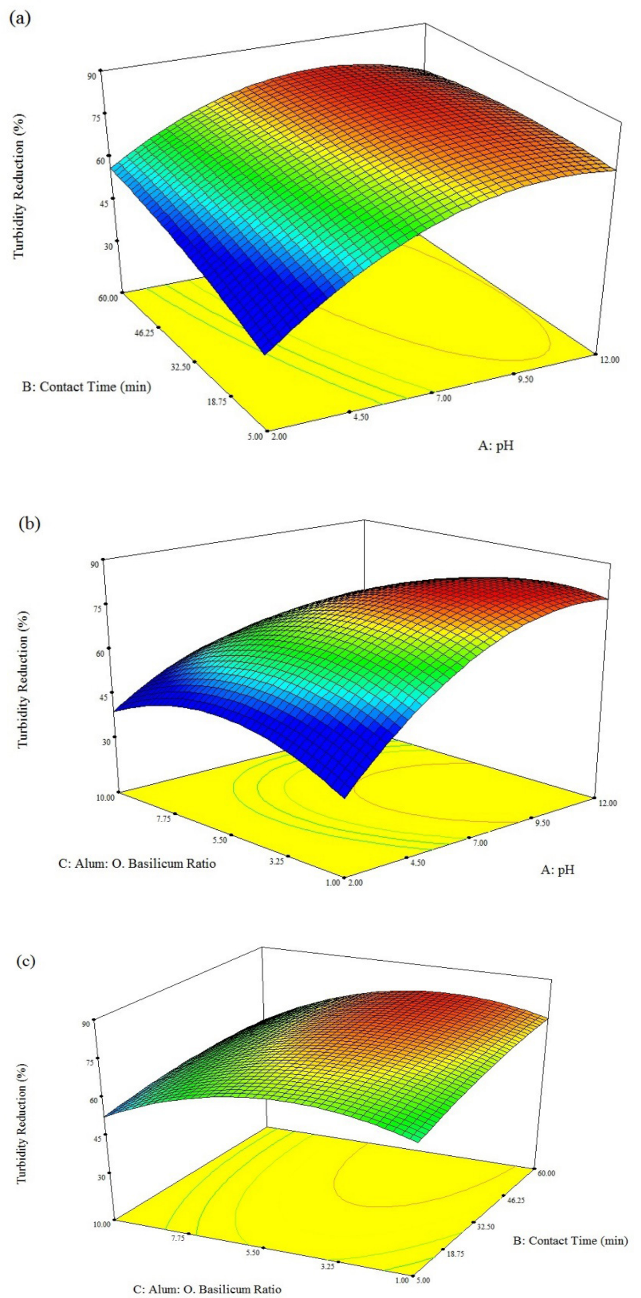

Fig. 2. Surface graphs of turbidity reduction (a) effect of pH and contact time (b) effect of pH and alum- *O. basilicum* ratio and (c) effect of contact time and alum- *O. basilicum* ratio.

At a low pH and with a low coagulant dosage, the main mechanism for colloidal destabilization is charge neutralization, and the aggregates are small in size, but by increasing of pH, the coagulation-flocculation mechanism is changed. The most important reasons for such behaviors are: 1) at low pH, the presence of monomeric aluminum species such as Al^{3+}, $Al(OH)^{2+}$, and $Al(OH)_2^+$ causes the anionic particles to neutral and therefore, deposition of pollutants settlement is best performed due to the formation positive metal complexes that help to build flocs with the negative organic pollutants, 2) with an increase in pH, the concentration of dissolved aluminum is reduced by the formation of uncharged metal hydroxide ($Al(OH)_3$) that leads to rapid precipitation and 3) with further increase in pH, the species $Al(OH)_4^-$ is dominant which it reduce coagulation effects [40,41].

Fig. 2c shows the effects of contact time and alum- *O. basilicum* ratio on the turbidity reduction. By reducing alum- *O. basilicum* ratio, the reduction of turbidity increased. However, contact time was not a very significant factor. Regarding alum- *O. basilicum* ratio, the turbidity reduction was 75–80% in the range of 2–4.

3.3. Confirmation experiments

The maximum predicted COD and turbidity removal were 39.7 and 81.4%, respectively, at pH of 8.3, alum- *O. basilicum* ratio of 2.8 and time of 48.8 min. The actual experimental values in optimum condition were 41.4% with 1.7% deviation for COD reduction and 82.3% with 0.9% deviation for turbidity reduction. Therefore, both measured COD and turbidity were close to those values predicted by RSM and the results were in good agreement with the model estimations.

In Table 5, the obtained results were compared with the data reported in literatures. It is obvious that the use of alum- *O. basilicum* led to remove most of pollutants from the paper-recycling wastewater.

3.4. Characterization of coagulant and floc

Using FTIR, the major functional groups of *O. basilicum* mucilage, alum, and also the floc of alum and *O. basilicum* mucilage are illustrated in Fig. 3. In Fig. 3a, the wave number range of 2850–2930 cm^{-1} is due to sp3 CH stretching vibrations. The broad band at 3320 cm^{-1} and also other major peaks between 1150–1250 cm^{-1} results from the presence of hydroxyl groups.

The peak patterns observed in Fig. 3b is related to the spectrum of alum; The broad O–H stretching band between 2800–3300 cm^{-1} and the peak at 1665 cm^{-1} indicated that the sample is a hydrate [47,48]. Additionally, the strong and broad band at 1116 cm^{-1} with a shoulder at 1054 cm^{-1} could be due to sulfate absorptions [48].

Regarding combined floc of alum and *O. basilicum* mucilage (Fig. 3c), the OH band was clearly indicated as the

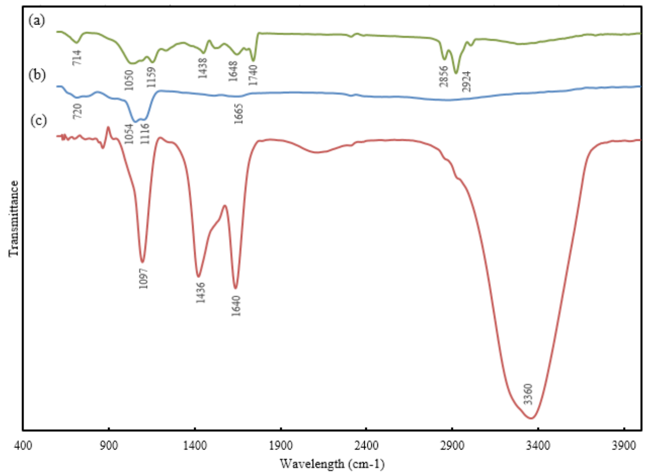

Fig. 3. FTIR spectra of (a) *O. basilicum* mucilage, (b) alum, (c) floc formed using *O. basilicum* mucilage and alum.

Table 5
Comparison of the present findings with other studies

Wastewater	Process	COD		Turbidity		References
		Initial (mg/L)	Removal (%)	Initial (NTU)	Removal (%)	
Pulp mill	Coagulation ($FeCl_3$) + Flocculation (Chitosan)	1,303	58.0	10	75.0	[42]
Paper-recycling	Coagulation (Alum) + Flocculation (PACl)	3,523	82.0	872.5	83.2	[43]
Paper-recycling	Coagulation (Alum) + Flocculation (C-PAM)	3,523	91.3	873	95.82	[44]
Pulp mill	Coagulation (aluminumchloride) + Flocculation (starch-g-PAM-g-PDMC)	1358	–	1209	95.7	[45]
Cardboard-mill	Flocculation (PACl)	500–1800	40–45	229–469	55–60	[10]
Textile wastewater	Coagulation (Alum) + Flocculation (*O. basilicum*)	2400	61.6	16.6	–	[23]
Textile wastewater	Coagulation ($Fecl_3$) + Flocculation (*A. esculentus*)	–	85.69	–	97.24	[46]
Paper- recycling	Coagulation (Alum) + Flocculation (*O. basilicum*)	6830	39.7	3750	81.4	Present study

broad and strong band at 3360 cm^{-1}. The weaker bands at 1436 cm^{-1} represents the bending vibration of CH_3 and the scissor vibration of CH_2. The peak at 1640 cm^{-1} was assigned to free moisture molecule in *O. basilicum* [49].

According to the findings of this study, the peak 1648 and 1740 cm^{-1} (Fig. 3a) may be assigned to $–COO^-$ stretching bands of carboxylate salts, probably uronic acids, which confirmed its anionic structure because partial deprotonation of the carboxyl functional group in aqueous solution provides a COO^- group, which acts as a site for cation adsorption. As suggested by Eq. (5), the quantity of available COO^- adsorption sites is affected by the OH^- concentration in the solution [26,50].

$$R-COO^- + H_2O \leftrightarrow R-COOH + OH^- \quad (5)$$

The second peak (1438 cm^{-1}) may be related to C-N stretching and primary amide. The peak at 1050 cm^{-1}, represents various vibrations of C-O-C glycosidic and C-O-H bonds [50].

3.5. Thermal stability analysis of floc

Thermo gravimetric analysis was applied to investigate the floc thermal stabilities and thermal decomposition of carbonaceous materials while the temperature rises [51]. According to Fig. 4a, a weight loss of 8% at a temperature below 100°C was the result of the release of light volatile compounds and moisture loss during drying [52].

As the temperature elevated from 100 to 900°C, significant amount of weight loss (55%) was observed due to gradual decomposition of the floc. Differential thermal gravimetric (DTG) curve represented decomposition stages of the floc of *O. basilicum* mucilage and alum. (Fig. 4b). At the temperature less than 100°C, the molecular moisture was evaporated from the floc. The loss of mass observed above 100°C was attributed to the loss of absorbed species and therefore several decomposition of organic components occurred. The present result was in accordance with other reports regarding that most components in the floc was decomposed at high temperatures [51,53].

4. Conclusions

This work demonstrated the effectiveness of *O. basilicum* mucilage to be used as a coagulant aid in CF process of paper-recycling wastewater in order to enhance both COD and turbidity removal. Up to 39.7% COD removal and 81.4% turbidity reduction was achieved at 48.8 min, pH 8.3 and alum- *O. basilicum* ratio of 2.8.

The characteristics of floc, alum and *O. basilicum L* mucilage revealed considerable results in the presence of functional groups and thermal stability of the floc. According to the combined floc morphology, possible coagulation mechanisms included adsorption with interparticle bridging due to presence carboxyl functional group in the *O. basilicum* and charge neutralization. The present study successfully indicates that *O. basilicum* mucilage, as an emerging natural coagulant, could be potentially used as a coagulant aid combined with alum for paper-recycling wastewater treatment.

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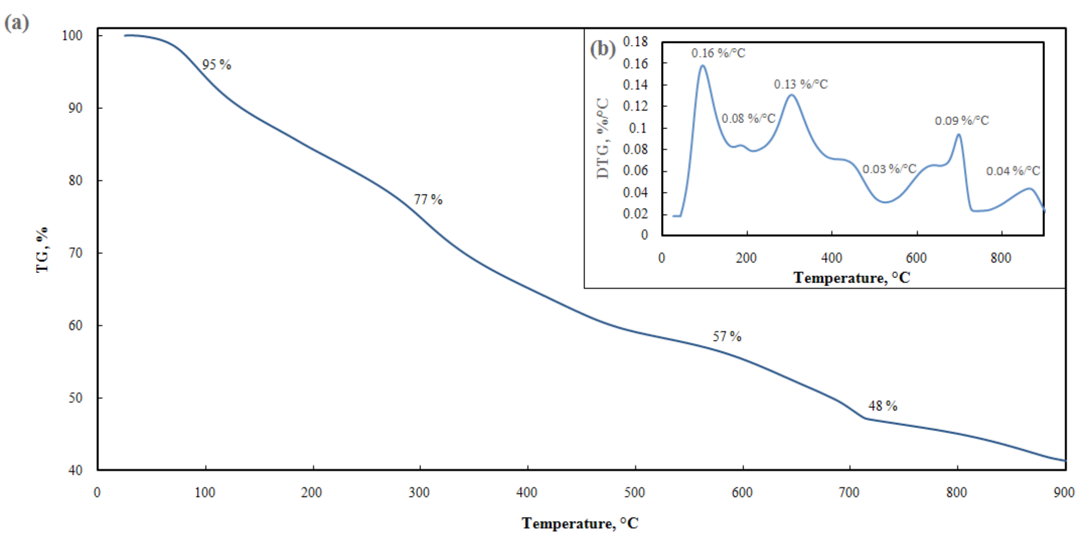

Fig. 4. (a) TG plot, (b) DTG plot of formed floc using *O. basilicum* mucilage and alum.

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