



# Enhanced turbidity removal in water treatment by using emerging vegetal biopolymer composite: a characterization and optimization study

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#### ABSTRACT

There are many factors which affect flocculation process in water treatment process. In this study, it is hypothesized that physico-chemical properties such as molecular weight, functional groups, charge density, mixing speed, mixing time, sedimentation time, concentration of cation (Fe), and polymers (polyacrylamide (PAM) and pectin) added, and pH of water influence treatment capability. The treatment is carried out using conventional coagulation–flocculation (two stage mixing mode). Secondly, coag–flocculation (one stage mixing mode) is carried out where a composite of Fe–PAM or Fe–pectin is added. The result suggests that coag–flocculation process is favorable for treating turbid wastewater. Furthermore, pH and concentration of Fe–PAM or Fe–pectin affect treatment process. Unlike Fe–pectin, mixing speed is a significant factor when using Fe–PAM. Moreover, Fe–PAM requires a higher mixing speed, higher pH and lower concentration added than that of Fe–pectin. Considering the importance for energy conservation worldwide, composite Fe–pectin is preferable as it uses less energy.

*Keywords:* Biodegradation; Coag–flocculation; PAM; Pectin; Photodegradation; Thermal degradation

#### 1. Introduction

Coagulation-flocculation process is an essential treatment process for solid-liquid separation. It can be used as a pre-treatment, post-treatment, or even a main treatment process [1]. It is commonly used to remove turbidity, color, and natural organic matter [2]. Hydrolyzing metal salts are commonly used in water treatment to induce coagulation by charge destabilization or neutralization. In addition, a widely used polyelectrolyte, polyacrylamide (PAM) has deg-

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radation products that are neurotoxic and carcinogenic [3]. Furthermore, countries like Japan and Switzerland are the forerunners of legislating policy to prevent the use of polyelectrolyte in water treatment. Whilst, Germany and France prohibited sludge treated with PAM to dispose on areas under cultivation by the end of 2013 [4]. Therefore, there is a need to replace the use of PAM in treatment plants and/or to reduce the amount required during water treatment. As such, natural flocculant becomes a viable alternative for liquid–solid separation in water and wastewater treatments [5]. Pectin, as an emerging natural polysaccharide flocculant exhibits good flocculating activity and

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contains polygalacturonic acids [6]. Pectin can be extracted from fruits such as apple pomace [7], beet, citrus [8], cocoa husk [9], pomelo [10], a few to name.

Generally, polymer addition induces simple charge neutralization, charge patch neutralization, polymer bridging, and polymer depletion [11,12]. There could be a combination of a few mechanisms depending on the nature of the colloids and polymer conformation at the solid–liquid interface. Polymer conformations at the solid–liquid interface depends on the concentration of polymer in solution and at the interface, nature of the solvent, temperature, pH, concentration of salt, polymer molecular weight, and surface charge density of the substrate [13]. Therefore, these factors should be taken into account in order to understand flocculation mechanism in single-mixing mode or conventional mixing mode.

In this study, pH, concentration of salts, concentration of polymer, functional group, molecular weight, and surface charge density of polymer are investigated. The objectives include, (1) investigate the influence of physical parameters during flocculation, (2) determine confirmation mechanisms of polymer on surface of particles, and (3) evaluate the efficiency when using composite of Fe–PAM and Fe–pectin.

### 2. Materials and methods

# 2.1. Molecular weight (M<sub>w</sub>) determination

 $M_w$  was determined using gel permeable chromatography (GPC) supplied by GPCmax, from Viscotek with TDA305 triple detector array. A column of A6000 M, general mixed Ag 300 × 0.8 mm was installed. 0.1 M sodium nitrate as mobile phase was prepared and samples were filtered using 0.20 µm nylon filter. The injection volume was 20 µL at 1 mL/min. The result was analyzed using OmniSEC software. The standard used was PolyCAL<sup>TM</sup> PEO 19k while Dextran was used for verification test.

# 2.2. Functional groups determination

Mid-infrared region (4,000–400 cm<sup>-1</sup>) was used to determine functional groups in PAM and pectin. The sample pallet was analyzed using Perkim Elmer, System 2000 FTIR, and used potassium bromide (KBr) pallet [14].

## 2.3. Charge density

The samples were prepared using 0.0001 M NaCl at a sample concentration of 1 mg/mL. They were analyzed using Malvern Zetasizer (Nano Z) at 25 °C.

This analysis was carried out at triplicate. The results were interpreted using Zetasizer software.

#### 2.4. Water treatment

Ferric chloride was used as a coagulant (from here onwards, it is noted as Fe). It is chosen based on the potential contribution of Al salts to Alzheimer's disease and produces large volumes of sludge [1]. Furthermore, Fe is effective for wider pH range of 4–11 and the formation of floc from ferric hydroxide is denser than alum. Subsequently, it improves sedimentation characteristics [15]. Polymer used in water treatment is PAM and pectin was supplied by Chemical Systems and extracted based on Ho et al. [6], respectively.

For conventional coagulation–flocculation process (two-stage mixing), Fe was added followed by rapid mixing at 150 rpm for 3 min and subsequently, addition of polymer with slow mixing at 30 rpm for 20 min.

For coag-flocculation process (one-stage mixing), composites of Fe-PAM or Fe-pectin are added. Subsequently, it underwent mixing at the same speed. pH, concentration of Fe-PAM or Fe-pectin, mixing speed, mixing time, and settling time are variables in the coag-flocculation process.

# 2.4.1. Assay of turbidity reduction

Turbidity reduction was carried out according to the Standard Methods, Method 2130 B [16]. Value of turbidity was determined by HACH Turbidimeter, 2100Q. Percent reduction is calculated as in Eq. (1):

Percent reduction, 
$$\% = \frac{A_{\rm o} - A}{A_{\rm o}} \times 100$$
 (1)

where,  $A_{o}$  is the control sample before treatment, NTU, A is the sample after treatment, NTU.

# 2.4.2. Design of experiment

Screening and optimization analyses were carried out in order to evaluate the significant factors and optimum treatment settings, respectively. Design Expert was used for screening and optimization study.

In this study, screening of the significant factors were designed using two-level factorial design and involved three and six factors for conventional twostage coagulation–flocculation process and one-stage coag–flocculation process, respectively. A, B, C, D, E, and F were denoted for factor of pH, concentration of cation, concentration of pectin or PAM, mixing speed, mixing time, and settling time, respectively. Tables 1–4 show the design boundaries based on preliminary study. Lastly, response surface methodology (RSM) was employed to optimize turbidity reduction in water treatment. RSM is commonly in environmental field to analyze and optimize the influence of independent variables on the responses [17–20].

#### 3. Results and discussion

# 3.1. Effect of molecular weight

The molecular weight of PAM and pectin determined by GPC was found to be  $2.356 \times 10^4$  kDa and  $6.039 \times 10^2$  kDa, respectively. In comparison, PAM has higher molecular weight. This indicates that PAM may have better flocculating ability as there are many repeating functional groups that may influence the flocculating performance. The optimization results obtained show that the concentration of PAM added is lesser than pectin, whether in coagulation-flocculation process or coag-flocculation process. Polymer with high molecular weight usually results in a bridging mechanism due to its long dangling tails. The tails and loops are extended far beyond the surface and interact with other particles. Furthermore, the flocs formed are stronger as the strength of polymer surface bond is high and the adsorbed polymer chain has attachments at multiple sites on the surface.

#### 3.2. Effect of functional group

The FTIR spectra for PAM and pectin are given in Figs. 1 and 2, respectively. Whereas, Tables 5 and 6 show the FTIR wavelength for PAM and pectin, respectively. Due to electrostatic repulsion, carboxyl and hydroxyl groups stretch out and bind and bridge the particles together. Bridging between fine particles with linearly extended polymer chain leads to the formation of flocs [21]. In addition, PAM adsorbed onto the surface of particles via hydrogen bonding with

Table 1

Design boundary for PAM using conventional coagulation-flocculation treatment method

| Factor                     | Low level | High level |
|----------------------------|-----------|------------|
| pH                         | 4         | 7          |
| Concentration of Fe, mM    | 0.01      | 0.10       |
| Concentration of PAM, mg/L | 10        | 35         |

#### Table 2

Design boundary for pectin using conventional coagulation-flocculation treatment method

| Factor                        | Low level | High level |
|-------------------------------|-----------|------------|
| pH                            | 2         | 4          |
| Concentration of Fe, mM       | 0.1       | 0.8        |
| Concentration of pectin, mg/L | 10        | 60         |

#### Table 3

Design boundary for composite of Fe-PAM in coag-flocculation treatment method

| Factor                     | Low level | High level |
|----------------------------|-----------|------------|
| рН                         | 3         | 9          |
| Concentration of Fe, mM    | 0.01      | 0.10       |
| Concentration of PAM, mg/L | 0.02      | 1.00       |
| Mixing speed, rpm          | 30        | 150        |
| Mixing time, min           | 5         | 15         |
| Sedimentation time, min    | 5         | 10         |
|                            |           |            |

Table 4

Design boundary for composite of Fe-pectin in coag-flocculation treatment method

| Factor                        | Low level | High level |
|-------------------------------|-----------|------------|
| pH                            | 3         | 9          |
| Concentration of Fe, mM       | 0.1       | 2          |
| Concentration of pectin, mg/L | 0.5       | 5          |
| Mixing speed, rpm             | 30        | 150        |
| Mixing time, min              | 5         | 15         |
| Sedimentation time, min       | 5         | 10         |

their primary amide functional groups [22]. Thus, the free carboxyl, hydroxyl, and primary amide groups in polymers may reduce electrical potential between particles and enhance electrostatic attraction on positive segment of surface of particles that leads to the formation of floc.

Apart from having higher molecular weight, PAM has additional primary amide groups that may induce better flocculation process as lower concentration of PAM is required to add to both systems.

Nonetheless, the magnitude of electrical charge in both polymers should be known in order to understand its role in reducing electrical potential gradient on the particle surface. High-charge polymer is aimed to greatly reduce the potential difference between particles and therefore, bind and bridge particles.



Fig. 1. FTIR spectrum for PAM.



Fig. 2. FTIR spectrum for pectin.

# 3.3. Effects of electrostatic potential

Zeta potential distribution of PAM and pectin analyzed using Zetasizer software indicated that the zeta potential for PAM and pectin is -46.1 and -22.0 mV, respectively. Evidently, both polymers are anionic polymers. According to the magnitude of charge density, PAM has higher strength than pectin. The electrostatic attraction in solution was affected by the addition of Fe and the surface charges of particles. Therefore, the zeta potential values of Fe and kaolin were calculated. The results show that the zeta potential values in Fe and kaolin are +47.4 and -44.0 mV,

| Table | 5          |    |     |
|-------|------------|----|-----|
| FTIR  | wavelength | of | PAM |

| Wavelength, cm <sup>-1</sup> | Description                 | Reference |
|------------------------------|-----------------------------|-----------|
| 3,443                        | OH-stretching               | [23]      |
| 2,923                        | C–H stretching              | [24]      |
| 2,360, 2,340                 | CO <sub>2</sub>             | [25]      |
| 1,624                        | NH bending from amide group | [26]      |
| 1,455                        | CH <sub>2</sub> scissoring  | [26]      |
| 1,409                        | C–N stretching              | [26]      |
| 1,033                        | C–O–C stretching            | [24]      |
| 866                          | C–C stretching              | [27]      |
| 686 to 653                   | C-H bending                 | [27]      |

Table 6 FTIR wavelength of pectin

| Wavelength, cm <sup>-1</sup> | Description                              | Reference |
|------------------------------|--|-----------|
| 3,420                        | OH stretching                            | [23]      |
| 2,933                        | C–H stretching from the aliphatic groups | [24]      |
| 2,360, 2,340                 | CO <sub>2</sub>                          | [25]      |
| 1,748                        | C = O esterified                         | [28]      |
| 1,637                        | Stretching of COO-                       | [29]      |
| 1,380                        | C–H bending                              | [30]      |
| 1,234–1,019                  | C=O stretching                           | [31]      |
| 831                          | C–O bonding                              | [32]      |
| 669                          | C–H bonding                              | [27]      |

respectively. Kaolin used in this study is similar to colloid in water which is usually negative in charge.

During the coagulation–flocculation process, the Fe added may destabilize the negative surface charge of the particle and depends on the amount of Fe added, it may be, (1) having excess of positive charges from Fe or (2) the particles may have remnants of negative segments on the surface. When the polymer is added, the anionic polymer would (1) get attracted to the positive edge of surface of particle destabilized by Fe or, (2) on the surface of particle itself to bind and bridge the particles and form floc.

On the other hand, during coag-flocculation process, composite of Fe–PAM and Fe–pectin are added into the solution. Polymers do not only serve bridging mechanism, it is also possible to neutralize the surface charges and bridge particles together when collision occurs during mixing. Owing to the fact that surface charges on the particle can be destabilized by adding both cationic Fe and anionic polymers like PAM and pectin. It effectively destabilized at opposite charge segment on surface of particles and bind in one go. The flocculation mechanism is suggested to be in Fig. 3. The result suggests that coag-flocculation has a more successful binding process. Moreover, PAM has higher charge than pectin, therefore, concentration of PAM added is lower than pectin. It portrays the relationship whereby the concentration added is proportionally inversed with the charge density of polymer. This result is in accordance to optimization setting of water treatment.



 $\sim$  Attraction to opposite charge

Fig. 3. Destabilization on surface of particle and bridge particle during coag-flocculation process.

#### 3.4. Effects of factors in water treatment

Factorial design shows the influential factors in water treatment study. Figs. 4 and 5 show the half-normal probability plot of conventional coagulation–flocculation process and coag–flocculation process, respectively.

Pectin shows significant to factors such as pH, concentration of cation, and concentration of pectin for both flocculation systems. It suggests the use of pectin to be able to operate under any condition (individually or composite Fe–pectin). On the other hand, coag– flocculation process using Fe–PAM requires a specific mixing speed. Higher mixing speed in Fe-PAM increases the chances for particles to collide and therefore, increases the treatment capability. To examine the capability in water treatment, optimization experiments were carried out to determine the optimum setting of each of the factors.

The second-order regression models obtained for the optimization were satisfied since the value of coefficient of determination ( $R^2$ ) was high and close to 1 for all treatment settings. The analysis of variance (ANOVA) using Fe and PAM in coagulation–flocculation process is given in Table 7.

It showed the quadratic effects of pH  $(x_1^2)$  and concentration of PAM  $(x_3^2)$  were insignificant, the rest were showed to have significant effects (p < 0.05). Second-order polynomial model in coded form that correlated turbidity reduction with all the significant variable terms is given in Eq. (2). Furthermore, the Three-dimensional response surface is shown in Fig. 6 for turbidity reduction.

Turbidity reduction = 
$$93.04 - 5.30 x_1 + 9.42 x_2$$

$$-7.23 x_3 + 1.31 x_1^2 - 5.76 x_2^2 -3.01 x_3^2 + 5.61 x_1 x_2 -4.82 x_1 x_3 + 7.05 x_2 x_3$$
(2)

As for coagulation–flocculation process using Fe and pectin, the linear effect and quadratic effect for turbidity reduction showed significant effect as given in the ANOVA (Table 8).

Obviously, the linear effect of pH ( $x_1$ ), concentration of Fe ( $x_2$ ), and concentration of pectin ( $x_3$ ) were significant (p < 0.05). Additionly, the quadratic effect of pH ( $x_3^2$ ), concentration of Fe ( $x_2^2$ ), and concentration of pectin ( $x_3^2$ ) were significant. Second-order polynomial model in coded form that correlated turbidity reduction with all the significant variable terms is given in Eq. (3). The 3-D response surface is shown in Fig. 7.

Turbidity reduction = 
$$85.69 - 5.80 x_1 + 3.96 x_2$$
  
- 3.17  $x_3 - 6.47 x_1^2 - 2.92 x_2^2$   
+ 5.34  $x_3^2 + 1.46 x_1 x_2 - 1.86 x_1 x_3$   
+ 1.41  $x_2 x_3$  (3)

Table 9 shows the ANOVA for turbidity reduction for Fe-PAM composite in coag–flocculation process. It shows that linear effects for pH ( $x_1$ ), concentration of Fe ( $x_2$ ), and concentration of PAM ( $x_3$ ) were significant



Fig. 4. Half-normal probability plot of (a) PAM and (b) pectin in conventional coagulation-flocculation process.



Fig. 5. Half-normal probability plot for composite of (a) Fe-PAM (b) Fe-pectin in coag-flocculation process.

| Table 7   |   |
|---|---|
| The results of ANOVA for turbidity reduction by using Fe and PAM in | conventional coagulation-flocculation process |

| Source                                      | Sum of squares | DF | Mean square | <i>F</i> -value | <i>p</i> -value |
|---|----------------|----|-------------|-----------------|-----------------|
| Model                                       | 1959.11        | 9  | 217.68      | 21.65           | < 0.0003        |
| pН  | 224.51         | 1  | 224.51      | 22.33           | < 0.0021        |
| Concentration of Fe                         | 709.89         | 1  | 709.89      | 70.62           | < 0.0001        |
| Concentration of PAM                        | 418.47         | 1  | 418.47      | 41.63           | < 0.0003        |
| pH×pH                                       | 7.26           | 1  | 7.26        | 0.72            | < 0.4236        |
| Concentration of Fe × concentration of Fe   | 139.55         | 1  | 139.55      | 13.88           | < 0.0074        |
| Concentration of PAM × concentration of PAM | 38.07          | 1  | 38.07       | 3.79            | < 0.0927        |
| pH × concentration of Fe                    | 125.89         | 1  | 125.89      | 12.52           | < 0.0095        |
| $pH \times concentration of PAM$            | 93.12          | 1  | 93.12       | 9.26            | < 0.0188        |
| Concentration of Fe × concentration of PAM  | 198.53         | 1  | 198.53      | 19.75           | < 0.0030        |
| Residual                                    | 70.37          | 7  | 10.05       |                 |                 |
| Total                                       | 2029.48        | 16 |             |                 |                 |

Note:  $R^2$  for turbidity reduction is 0.9653.

(p < 0.05). Moreover, the quadratic effect of concentration of Fe ( $x_2^2$ ) was significant (p < 0.05). The interaction between pH and concentration of Fe ( $x_1x_2$ ), and concentration of PAM and mixing speed ( $x_3x_4$ ) showed to be significant (p < 0.05).

Second-order polynomial model in coded form that correlated turbidity reduction with all the significant variable terms is given in Eq. (4). Fig. 8 shows threedimensional response surface plot for PAM.

Turbidity reduction = 
$$97.57 - 20.87 x_1 + 20.07 x_2$$
  
+ 5.60  $x_3 - 3.44 x_4 - 7.69 x_1^2$   
- 20.24  $x_2^2 - 2.99 x_3^2 + 0.81 x_4^2$   
+ 19.48  $x_1x_2 - 1.57 x_1x_3$   
+ 1.86  $x_1x_4 + 1.63 x_2x_3$   
+ 1.21  $x_2x_4 + 3.96 x_3x_4$  (4)

On the other hand, the ANOVA for Fe-pectin composite in coag–flocculation process is given in Table 10. It shows that linear effects for pH, concentration of Fe, and concentration of pectin were significant (p < 0.05). The quadratic effect of concentration of Fe and concentration of pectin showed to be significant (p < 0.05). The interaction between pH and concentration of Fe, and interaction between pH and concentration of pectin showed to be significant (p < 0.05) as well.

Second-order polynomial model in coded form that correlated turbidity reduction with all the significant variable terms is given in Eq. (5). Three-dimensional surface response plot (Fig. 9) is built to illustrate the optimum setting of turbidity reduction. Obviously, turbidity reduction exhibited a clear surface and illustrated optimum condition for maximum turbidity reduction which is well defined inside the design boundaries. 1786 Table 8

| Source  | Sum of squares | DF | Mean square | F-value | <i>p</i> -value |
|---|----------------|----|-------------|---------|-----------------|
| Model   | 825.09         | 9  | 91.68       | 15.49   | < 0.0008        |
| pH  | 269.58         | 1  | 269.58      | 45.54   | < 0.0003        |
| Concentration of Fe                               | 125.22         | 1  | 125.22      | 21.15   | < 0.0025        |
| Concentration of pectin                           | 80.45          | 1  | 80.45       | 13.59   | < 0.0078        |
| pH×pH   | 176.41         | 1  | 176.41      | 29.80   | < 0.0009        |
| Concentration of Fe × concentration of Fe         | 35.78          | 1  | 35.78       | 6.04    | < 0.0436        |
| Concentration of pectin × concentration of pectin | 120.05         | 1  | 120.05      | 20.28   | < 0.0028        |
| pH× concentration of Fe                           | 8.58           | 1  | 8.58        | 1.45    | < 0.2676        |
| $pH \times concentration of pectin$               | 13.84          | 1  | 13.84       | 2.34    | < 0.1701        |
| Concentration of Fe× concentration of pectin      | 7.92           | 1  | 7.92        | 1.34    | < 0.2852        |
| Residual  | 41.44          | 7  | 5.92        |         |                 |
| Total   | 866.53         | 16 |             |         |                 |

The results of ANOVA for turbidity reduction of Fe and pectin in conventional coagulation and flocculation process

Note:  $R^2$  for turbidity reduction model was 0.9522.

Turbidity reduction = 
$$66.99 - 26.40 x_1 - 5.90 x_2$$
  
+ 10.63  $x_3 - 0.52 x_1^2 + 12.78 x_2^2$   
- 19.67  $x_3^2 + 5.54 x_1 x_2$   
+ 6.76  $x_1 x_3 + 3.41 x_2 x_3$  (5)

It can be seen that turbidity reduction exhibited a clear surface. It suggests that the optimum condition for maximum turbidity reduction is well defined inside the design boundaries. The optimum condition for turbidity reduction is shown in Table 11.

Polymers added suggest neutralizing the surface charges, likewise the addition of salts; this explains why coag-flocculation process is a better treatment option. With this mechanism, the concentration of Fe added can be reduced. The result shows that when using Fe-pectin composite in coag-flocculation process, the concentration of Fe reduced 0.1 mM. The concentration of polymer added is a highlight here as it reduced from 13–0.05 to 10–3 mg/L for Fe-PAM and Fe-pectin composites, respectively. It shows a great reduction when using coag-flocculation process with small amount of Fe required at optimum condition.





Fig. 6. Three-dimensional response surfaces for turbidity reduction using Fe and PAM in conventional coagulation and flocculation processes.

Fig. 7. Three-dimensional response surfaces for turbidity reduction using Fe and pectin in conventional coagulation–flocculation processes.

Table 9

| Source                                      | Sum of squares | DF | Mean square | F-value | <i>p</i> -value |
|---|----------------|----|-------------|---------|-----------------|
| Model                                       | 2,7687.96      | 14 | 1,977.71    | 37.52   | < 0.0001        |
| pH  | 7,837.52       | 1  | 7,837.52    | 148.71  | < 0.0001        |
| Concentration of Fe                         | 7,252.09       | 1  | 7,252.09    | 137.60  | < 0.0001        |
| Concentration of PAM                        | 564.48         | 1  | 564.48      | 10.71   | < 0.0056        |
| Mixing speed                                | 213.56         | 1  | 213.56      | 4.05    | < 0.0638        |
| $pH \times pH$                              | 153.10         | 1  | 153.10      | 2.90    | < 0.1104        |
| Concentration of Fe × concentration of Fe   | 1,059.79       | 1  | 1,059.79    | 20.11   | < 0.0005        |
| Concentration of PAM × concentration of PAM | 23.19          | 1  | 23.19       | 0.44    | < 0.5179        |
| Mixing speed × mixing speed                 | 1.68           | 1  | 1.68        | 0.032   | < 0.8610        |
| pH × concentration of Fe                    | 6,068.41       | 1  | 6,068.41    | 115.14  | < 0.0001        |
| pH × concentration of PAM                   | 39.69          | 1  | 39.69       | 0.75    | < 0.4001        |
| pH × mixing speed                           | 55.50          | 1  | 55.50       | 1.05    | < 0.3222        |
| Concentration of Fe × concentration of PAM  | 42.25          | 1  | 42.25       | 0.80    | < 0.3857        |
| Concentration of Fe × mixing speed          | 23.52          | 1  | 23.52       | 0.45    | < 0.5150        |
| Concentration of PAM × mixing speed         | 251.22         | 1  | 251.22      | 4.77    | < 0.0465        |
| Residual                                    | 737.86         | 14 | 52.70       |         |                 |
| Total                                       | 28,425.82      | 28 |             |         |                 |

Note:  $R^2$  for turbidity reduction model was 0.9740.

Table 10 The results of ANOVA for turbidity reduction of Fe-pectin composite in coag–flocculation process

| Source  | Sum of squares | DF | Mean square | F-value | <i>p</i> -value |
|---|----------------|----|-------------|---------|-----------------|
| Model   | 10,417.44      | 9  | 1,157.49    | 67.69   | < 0.0001        |
| pH  | 6,969.60       | 1  | 6,969.60    | 407.56  | < 0.0001        |
| Concentration of Fe                               | 348.10         | 1  | 348.10      | 20.36   | < 0.0011        |
| Concentration of pectin                           | 1,129.97       | 1  | 1,129.97    | 66.08   | < 0.0001        |
| pH×pH   | 0.74           | 1  | 0.74        | 0.043   | < 0.8396        |
| Concentration of Fe × concentration of Fe         | 449.28         | 1  | 449.28      | 26.27   | < 0.0004        |
| Concentration of pectin × concentration of pectin | 1,063.80       | 1  | 1,063.80    | 62.21   | < 0.0001        |
| pH × concentration of Fe                          | 245.31         | 1  | 245.31      | 14.34   | < 0.0036        |
| $pH \times concentration of pectin$               | 365.85         | 1  | 365.85      | 21.39   | < 0.0009        |
| Concentration of Fe × concentration of pectin     | 93.16          | 1  | 93.16       | 5.45    | < 0.0418        |
| Residual  | 171.01         | 10 | 17.10       |         |                 |
| Total   | 10,588.45      | 19 |             |         |                 |

Note:  $R^2$  for turbidity reduction model was 0.9838.

Furthermore, both Fe-PAM and Fe-pectin composites are preferable to operate in pH region approaching to neutral. Moreover, the mixing speed required for Fepectin is lower than Fe-PAM during coag-flocculation process. As coag-flocculation process requires lesser treatment time and energy during mixing, it gains advantages over coagulation-flocculation process where it reduces operating cost in a treatment plant in terms of energy consumed, quantity of materials added and time used. The findings strongly suggests that coag-flocculation of using Fe-pectin composite is favorable in water treatment process.

The findings in this study shows that at optimum setting in coag-flocculation process may improve the flocculation rate and turbidity reduction by overcoming the electrical repulsion and is enhanced by bridging. Furthermore, coag-flocculation process may eliminate the possible saturation of adsorption site on surface of particle as it finetunes the coagulation and flocculation processes.



Fig. 8. Three-dimensional optimization plot for Fe-PAM composite (coag–flocculation process) for turbidity reduction.



Fig. 9. Three-dimensional optimization plot for Fe-pectin composite (coag–flocculation process) for turbidity reduction.

| Table 11 |           |     |       |           |
|----------|-----------|-----|-------|-----------|
| Optimum  | condition | for | water | treatment |

| Solid–liquid separation<br>process | Polymer | pН   | Fe, mM | Polymer,<br>mg/L | Mixing speed,<br>rpm | Mixing time,<br>min | Turbidity<br>reduction, % |
|------------------------------------|---------|------|--------|------------------|----------------------|---------------------|---------------------------|
| Coagulation-flocculation           | PAM     | 4.00 | 0.05   | 13.48            | _                    | 23                  | 99.85                     |
| 0                                  | Pectin  | 2.73 | 0.61   | 10.00            | -                    | 23                  | 95.16                     |
| Coag-flocculation                  | PAM     | 6.30 | 0.07   | 0.05             | 56.91                | 5                   | 99.40                     |
|                                    | Pectin  | 3.38 | 0.52   | 3.48             | 30                   | 5                   | 98.56                     |

# 4. Conclusion

Based on the findings, molecular weight, functional group in a polymer, and its charge density influence the flocculation process. PAM reduces the concentration of Fe and PAM added to the system as it contains high molecular weight, more functional groups, and high charge density. However, due to the differences in mixing condition, coag-flocculation process gains advantages where, (1) pH of water is nearer to neutral, (2) greatly reduces chemicals added, and requires lower energy. Though the results from water treatment show that PAM has an advantage over pectin, pectin shows to be able to treat water in any mixing condition while PAM has a rigid requirement at higher mixing speed. As energy is a worldwide priority nowadays, thus, pectin works as an emerging plant-based flocculant especially in Fe-pectin composite

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