Removal of Phosphate from Aqueous Solution by using Natural Coagulant: Optimization Response Surface Methodology (RSM)

N K Faraj\(^1\) and Z N Abudi\(^2\)

\(^1\) Department Environmental Engineering, Mustansiriyah University, Baghdad, Iraq; 
\(^2\) Assist prof, Mustansiriyah University, Baghdad, Iraq.

Abstract. This study aims to remove phosphate from synthetic solution using the coagulation-flocculation process by the natural coagulants (Rice Husk (RH), Moringa Oliveira Seed pods (MOSP)) and optimize coagulant conditions using response surface methodology (RSM). The effects of operating conditions like initial concentration (40-80 ppm), coagulant dosing (2-3 g/L), and pH of (2-6) were implemented. The coagulation-flocculation was improved utilizing Box–Behnken Design (BBD) under RSM. Quadratic model produced by BBD, with a value of R\(^2\) equal to 0.9894 and 0.9808 for RH and MOSP, respectively. The results of RSM show that the optimum operating conditions for the RH coagulant were an initial phosphate concentration of 60 ppm, dosing 2 g/l, and an initial pH of 4. Which are the same conditions for another coagulant (MOSP) with a difference of dosing 2.5 g/l. The main results showed that the coagulants used in this work have a high ability to treat phosphates efficiently instead of using chemicals at a natural pH. The current study shows that it is possible to use RH and MOSP as new natural coagulants that are cost-effective in treating industrial wastewater.

1. Introduction

The water resources in Iraq are affected by many sources of pollution. Wastewater on surfaces and rivers is formed from other uses (human, agricultural and industrial) as a source of deteriorating water quality, and rivers water suffers from pollution and environmental problems as a result of industrial abuses practiced by factories, by throwing their industrial waste\(^[1]\). Industrial wastewater is directly discharged to the river without treatment, where this wastewater carried a large amount of phosphate and other elements of industrial wastes that are discarded by discharged them in residential neighborhoods sewerage system\(^[2]\). The discharge of phosphate from both non-point and point sources into runoff may impose a menace on Water resources\(^[3]\). As a growth-limiting alimentary, high concentration phosphate can boost organisms in produce phosphates in large quantities during the process of photosynthesis in natural water and the end becomes a prime factor in the eutrophication of lots of freshwater ecosystems\(^[4]\).

The presence of phosphates in large quantities in water causes (1) abnormal growth of toxic bacteria, algae, and lichens because it is one of the nutrients for water organisms. (2) It affects the aesthetic aspect of the water source. (3) Causes the reduction of dissolved oxygen in the water.
It is consequently important to develop active technologies to remove phosphate from polluted water before discharge to natural water bodies [4]. Lots of phosphate removal technologies such as physical, biological, and chemical treatment methods have been sophisticated for various applications, especially for the removal of phosphate from industrial and municipal effluents [5].

Biological and chemical treatments have been certified and proven to be active in remove phosphate from industrial wastewater. Addendum of chemicals, such as iron salts, calcium, and aluminum into the industrial wastewater is it deems a simple phosphate removal technique, which split up the phosphate from the aqueous system through coagulant [6,7] then disposal of the sludge [8,9]. Chemical compounds have been linked to diseases like cancer and Alzheimer's [10]. In the visibility of these, the use of the by-products of crops in water treatment is one of the most efficient methods of according to the statistics of the World Food Organization [8]; the present study had been carried out on the evolution of natural coagulants and eco-friendly such as RH and MOSC.

[9] Rice production is about (582) tons per year and the percentage of rice husks is about 22-25% of the total amount of rice produced, that is, (45) million tons per year is the percentage of rice husks [9]. The cultivated areas according to the statistics of the Ministry of Agriculture are about 125 hectares per year. Iraq's production of the rice husk crop is (91) tons annually. There are economic and environmental problems caused by this waste due to the accumulation of increasing amounts of it around the world. Therefore, many scientists and researchers directed studies and research to remove pollutants using natural and non-traditional sources [11–13]. Rice husks are insoluble in water; their outer surface is often composed of sharp-angled silica and has capillaries. In addition to being resistant to fungi, and depending on geographical conditions, the chemical content varies from sample to sample. These conditions include different climatic conditions, type of fertilizer used in agriculture, rice quality, and soil quality.

Moringa Olifeira Seed Pods (MOSP) include the soluble protein rateable stabilizes the particles and clarifying the water. In recent studies [14] utilized Moringa oleifera seed in the treatment of coffee fermentation wastewater [15] unfold Moringa oleifera seed to the treatment of s.003urface water [16] combined aluminum sulfate with moringa seed powder in the treatment of water. Yet, none of these researchers using optimized the process variable factor in the optimization of the coagulation-flocculation process of water.

Modeling and optimization are very significant in any operation to improve output [17]. The use of a single variable method for optimization and modeling is obsolete and it no displays reaction between other variables in a process [18]. Response Surface Methodology (RSM) is a method of optimization that included experimental design, modeling, and analysis during the partial fall back fitting of the experimental factors [19]. It has the potency to merge many variables at a time and display reciprocal interaction on the output of operations, it also decreases the number of empirical runs needed to Supply sufficient information for statistically suitable results [20]. RSM has been used in many processes especially in chemical and biological processes but none has been reported on the natural coagulants (like RH and MOSP) for the phosphate removal by the coagulation-flocculation process.

This work aims to optimize the process operation variables affecting the coagulation-flocculation process to remove phosphate from polluted water by using natural coagulants. To minimize the phosphate in the synthetic wastewater, RSM was used to determine the effects of three operation variables (initial phosphate concentration, coagulants dosing, and pH).
2. Materials and method

2.1. Chemical reagents

Ammonium molybdate, Tincture of phenolphthalein, Sulfuric acid (H\textsubscript{2}SO\textsubscript{4}), HCl (0.1 N), NaOH (0.1 N), distilled water, Mono-tin chloride (SnCl\textsubscript{2}.H\textsubscript{2}O), Di-Potassium Phosphate (K\textsubscript{2}HPO\textsubscript{4}).

2.2. RH powder Preparation

Rice husk (RH) was agricultural residues obtained from Najaf Governorate, central of Iraq. The rice husks were washed well using tap water to remove dust and dirt and then washed with distilled water to remove the salts from it. To dry it, it was placed under sunlight for 48 hours. After making sure that it was completely dry, it was crushed using an electric grinder and sifted with a sieve with a diameter of (200) micrometers, and then the powder was stored in a dry place (plastic boxes) until it was used in the experiments. Table (1) displays the material composition makes it ideal for use as a coagulant.

Table 1. Show the Composition analysis of RH [21]

| Chemical constituents | Weight (%) |
|-----------------------|------------|
| Fe\textsubscript{2}O\textsubscript{3} | 0.51       |
| AL\textsubscript{2}O\textsubscript{3} | 0.63       |
| CAO | 1.96       |
| MgO | 0.77       |
| SIO2 | 93.26      |
| K2O | 2.85       |

2.3. MOSC powder Preparation

The other natural coagulant used in this study, Moringa Olifeira Seed Pods (MOSP), was obtained from the Moringa Oliveira tree in College of engineering/Mustansiriya University, Baghdad, Iraq. To prepare MOSC powder, seeds were removed from inside Moringa Olifeira's cap; the seed coats were washed well at first with tap water to remove dust and dirt then washed with distilled water to remove the salts from it. In sunlight, MOSP was lifted out for 72 hours to dry. After making sure that it is well dry, it crushed using an electric grinder for 40 seconds, as well as sifting with a sieve diameter of 200 micrometers. The MOSP is stored in a dry place (plastic boxes) until it is used in the experiments. Table (2) shows that the MOSP composition makes it ideal for use as a coagulant.

Table 2. Show the Composition analysis of MOSP [22].

| Parameter | Weight (%) |
|-----------|------------|
| Moisture  | 5.169      |
| Ash       | 2.989      |
Protein \hspace{1cm} 38.281 \\
Crude fire \hspace{1cm} 17.2 \\
Fat \hspace{1cm} 11.29 \\
Carbohydrate \hspace{1cm} 25.071 \\

2.4. Synthetic phosphate water preparation

It is called Potassium phosphate or MKP (also known as potassium dihydrogen phosphate, or KDP, or monopotassium phosphate), and \( \text{KH}_2\text{PO}_4 \) is a soluble salt consisting of potassium phosphate and a dihydrogen ion. The compound is used in fertilizers, food additives, and fungicides. The prepared phosphate solution is from dissolving the \( \text{KH}_2\text{PO}_4 \) powder (2.634 g) in 1 liter of distilled water to synthesis an aqueous solution at a concentration of 600 ppm.

2.5. Method

A six-paddle tester, Phipps and Bird, was used with six 1-liter glass beakers and filled with polluted water. Rapid mixing was performed at 200 rpm for 1 minute, then, mixed slowly for 20 seconds at 40 rpm, after which 30 minutes left to precipitate. The arrangement of the operation parameters checked was the natural coagulant dose, the solution pH, and initial phosphate concentration, to find the optimal dose of the natural coagulant (RH and MOSP) doses ranged from (0.5 - 3) g/liter. To find the optimum pH it was varied in the range of (2-12). Finally, the initial phosphate concentration range was 40-80 ppm. Phosphates were measured using a spectrophotometer at the wavelength of 669 nm. Then the phosphate removal rate was found through the use of equation (1).

\[
\text{Removal efficiency (\%)} = \frac{\text{initial concentration} - \text{final concentration}}{\text{initial concentration}} \times 100\% \quad (1)
\]

2.6. Design of experiments

Response surface methodology (RSM) software was used to optimize the variables affecting three varied coagulation factors; the initial concentration, coagulant doses, and the solution pH on synthetics wastewater treatment were studied. 17 playback experiences were created with Box Behnken Design (BBD). The suitability of the model was estimated using analysis of variance (ANOVA) and test of significance; the variables identified represent initial concentration and doses (RH and MOSP) and pH X1, X2, and X3, respectively, as shown in Table 3. The coding of the selected variants was reported by [21]. Design expert regressions were used to fit the parameter of the polynomial response model shown in equation (2).

\[
Y = \beta_0 + \sum_{i=1}^{k} \beta_i K_i + \sum_{i=1}^{k} \beta_{ii} K_i^2 + \sum_{i<j}^{k} \beta_{ij} X_i X_j + e \quad (2)
\]

Where \( Y \) = dependent variable (Phosphate removal rate), \( X_j = k \) (number of variables studied), \( X_i \) = independent variable, \( \beta_i \) = regression coefficients for linear \( \beta_0 \) = regression coefficients for intercept, \( \beta_{ij} \) = interaction terms, \( \beta_{ii} \) = regression coefficients for quadratic and is the random wrong. The design expert (11) program software was utilized to analyze and design the experience data. Design – expert (11) is a computer software bundle that implements the design of experiences, optimization of operation variables, and comparative tests. It also contains graphic tools that help define the effect of each parameter on the outcome of the process.
Table 3. The Independent variables and their levels for BBD.

| Independent variables | Code units | Coded level | -1 | 0 | 1 |
|-----------------------|------------|-------------|----|---|---|
| Concentration(PPM)    | A          | 40          | 60 | 80|
| Dosing (g/L)          | B          | 2           | 2.5| 3 |
| PH                    | C          | 2           | 4  | 6 |

Table 4A. BBD design matrix for three variables with response values of Phosphate removal efficiency using the RH.

| Run | Factor 1 A: Concentration ppm | Factor 2 B: dosing g/l | Factor 3 C: PH | Response Percentage removal |
|-----|--------------------------------|-------------------------|----------------|-----------------------------|
| 1   | 60                             | 1.5                     | 6              | 92.73                       |
| 2   | 40                             | 2                       | 6              | 97.33                       |
| 3   | 60                             | 2                       | 4              | 96.63                       |
| 4   | 80                             | 2                       | 6              | 89.2                        |
| 5   | 40                             | 2                       | 2              | 94.48                       |
| 6   | 80                             | 1.5                     | 4              | 86.25                       |
| 7   | 60                             | 2                       | 4              | 96.63                       |
| 8   | 60                             | 2.5                     | 6              | 94.5                        |
| 9   | 60                             | 1.5                     | 2              | 91.39                       |
| 10  | 80                             | 2.5                     | 4              | 89.16                       |
| 11  | 40                             | 2.5                     | 4              | 98.57                       |
| 12  | 40                             | 1.5                     | 4              | 94.69                       |
| 13  | 80                             | 2                       | 2              | 87.41                       |
| 14  | 60                             | 2                       | 4              | 96.63                       |
| 15  | 60                             | 2.5                     | 2              | 92.55                       |

Table 4 B. BBD design matrix for three variables with response values of Phosphate Removal efficiency using the MOSP.

| Run | Factor 1 A: Concentration ppm | Factor 2 B: dosing g/l | Factor 3 C: PH | Response Percentage removal |
|-----|--------------------------------|-------------------------|----------------|-----------------------------|
| 1   | 40                             | 3                       | 4              | 97.53                       |
| 2   | 80                             | 2                       | 4              | 90.52                       |
| 3   | 60                             | 2                       | 2              | 93.47                       |
3. Results and discussion

3.1. Model fitting and regression analysis

Coagulant processes RH and MOSC were improved based on BBD for the high removal of phosphate from synthesis wastewater. Phosphate removals from the synthetic polluted solution, the programs of designed experiments (11) by using RH, and MASC (Table 4) were modeled as a function of initial phosphate concentration (A), dosing (B), and initial pH (C). To find the adequacy of the model, the experimental data were applied to different models such as the quadratic, cubic models, linear and interactive (2FI), and cubic models. It was found in this study that quadratic polynomial models to be appropriate, as it displays maximum $R^2$, minimum p-values, and adjusted $R^2$. The experimental relationship expresses by a quadratic equation with react terms was fitted between the experimental results obtained based on Box-Behnken design (BBD) in experimental design and entered variables. The definitive equation obtained in conditions of coded factors was produced as display in Table 4. The positive signal of the stipulation inspired the presence of mutually exclusive effects between individual effects while mutual interactions negative signal show inverse effects [23]. It was used ANOVA (Table 6) to observe the effect of concentration of phosphate, coagulant dosage, and initial pH on the removal of phosphate. In the present study, indication analysis by F-test discovered all the models for phosphate removals were significant, which are 95% and the confidence level with values-p of less than 0.005. Likelihood values less than 0.05 the model expression are important while values more than 0.100 the model terms are not important, whole the model terms are shown in Table 7. In the case of phosphate removal using RH, Note that A, B, C, A2, B2, and C2 their importance. High elevated $R^2$ models indicate general differences in phosphate removal interpreted according to the models [24].

When the value $R^2$ is greater than 0.75 this evidence good accuracy that it gives models. Another statistical tool to notice is adequate precision (AP) which represents the ratio indicative to noise [23]. Indicate AP top from 4 desired ratios and the consequences, the last equation was obtained through the coding factors as shown in Table 5.
Table 5. The final quadratic model for the removal of phosphate from the synthetic solution was obtained from ANOVA at a 95% confidence level (p < 0.05).

| Treatment | Final quadratic model (coded factors) |
|-----------|--------------------------------------|
| RH        | $37.725+0.6409X_1+33.375X_2-4.488X_3-0.0242X_1^2X_2^2\cdot 0.00662X_1^2X_2^2-0.48750X_3^2$ |
| MOSP      | $28.812+0.914X_1+29.272X_2+4.0725X_3+0.117X_1^2X_2^2-0.0053X_1^2X_3^2+0.94X_2^2X_3^2-0.7298X_3^2$ |

Table 6. Summary of the estimated regression model for all responses.

| Factor  | Phosphate removal (%) | SS  | DF | MS  | F-value | P-value |
|---------|-----------------------|-----|----|-----|---------|---------|
| RH      | Model                | $R^2=0.9855$ | Adj$R^2=0.9594$ | 201.84 | 9 | 22.43 | 37.78 | 0.0005 |
| Lack of fit | 2.97 | 3 | 0.9894 | - | - |
| Pure error  | 0.0000 | 2 | 0.0000 | - | - |
| MOSP    | Model                | $R^2=0.9273$ | Adj$R^2=0.8545$ | 125.19 | 9 | 13.91 | 10.55 | 0.0092 |
| Lack of fit | 9.59 | 5 | 1.92 | - | - |
| Pure error  | 0.0000 | 2 | 0.0000 | - | - |

Table 7. Summary of the estimated regression model for all responses.

| Factor  | Phosphate removal (%) | SS  | D.F | MS  | f-value | p-value |
|---------|-----------------------|-----|-----|-----|---------|---------|
| RH      | Model                | 122.20 | 7 | 17.46 | 12.75 | 0.0017 |
| A-Concentration | 59.68 | 1 | 59.68 | 43.58 | 0.0003 |
| AB      | 5.50 | 1 | 5.50 | 4.02 | 0.0851 |
| AC      | 0.1849 | 1 | 0.1849 | 0.1350 | 0.7241 |
| BC      | 3.53 | 1 | 3.53 | 2.58 | 0.1522 |
| A²      | 22.28 | 1 | 22.28 | 16.27 | 0.0050 |
| B²      | 6.54 | 1 | 6.54 | 4.78 | 0.0651 |
| C²      | 31.46 | 1 | 31.46 | 22.97 | 0.0020 |
| MOSC    | Model                | 125.19 | 9 | 13.91 | 10.55 | 0.0092 |
| A-Concentration | 59.68 | 1 | 59.68 | 45.26 | 0.0011 |
| B-dosing | 0.7875 | 1 | 0.7875 | 0.5973 | 0.4745 |
| C-PH    | 2.21 | 1 | 2.21 | 1.67 | 0.2525 |
3.2. Effect of concentration and dosage

Figures 1(a) and 2(a) show the effects of phosphate initial concentration and natural coagulants (RH and MOSP) dosage and on the phosphate removal percentage at a constant pH of 4. The results showed that the effect of initial concentration exceeds the dosage effect for both coagulants used (RH and MOSP).

When using RH the F-value for concentration (230.01) was higher than the dosage (19.89), indicating that initial concentration is the variable affected by the coagulation process for getting a high removal percentage. Also, when used MOSP, the F value for the concentration (42.26) was higher than the F-value the dose value was (0.5973). The high initial concentration had a dominant effect on the removal of phosphate with RH and MOSP. An increase of phosphate removal was observed with increment dosage and decreases the initial concentration to 80 ppm using dosages of between 2.5-3 g for RH and MOSP. Finally, the removal rate of phosphate was more than 95% when using an initial phosphate concentration of 60ppm and coagulants dosages of 2 and 2.5g for RH and MOSP, respectively. The removal efficiency decreases with the increase in phosphate concentration. This is because the increase in the initial concentration leads to an increase in the number of polluted ions. Several ions are linked to the active amine groups on the surface of the coagulants used (RH, MOSP) thus leading to saturation of the active sites of the coagulants [25]. In other words, the phosphate may be absorbed through the limited active sites and there remain several phosphate molecules that are not removed, causing the decreased removal rate [21].

However, the removal capacity can be increased by changing the dosages. In the expression of dosage, it can be clear that an insufficient amount of dosage has a small effect on removal efficiency, whilst excess dosing led to the reflection effect on phosphate removal from synthetic wastewater. In general, a sufficient dosage of coagulant leads to the formation of weighty flocks, which makes it precipitate faster than scattered particles because of the effect of gravity in the precipitation process [26].

3.3. The Effects of change concentration and initial pH

The effects of the factors, initial concentration (A) and pH (C), on the removal rate of phosphate at constant conditions of setting time 30 min for both coagulants RH and MOSC were shown in Figures 1(b) and 2(b), respectively. The effect of the initial concentration factor on the removal efficiencies was more significant than the effect of pH (another factor) when RH was used as a coagulant. From Table 6, the F-value for RH (concentration - 230.01 and pH - 13.24) this value was higher than the F-values when MOSC was used (concentration - 42.26 and pH-1.67). The results displayed that 97% of phosphate can be removed at a low pH when used RH as shown in Figure 1(b). While removal efficiencies reached, when used MOSC, 93% (Figure 2(b)), the acidic condition of phosphate was convenient for treatment using MOSC and RH. The activity of the suspended solids that capable of coagulate is increasing when reducing the pH of the wastewater [27].
This may happen due to the charge neutralization when a precipitator where cationic surroundings have strong rapprochement toward colloidal particles. When the pH value increases above 4, removal efficiencies begin to decrease. This could be because of the nature of colloidal particles while it tends to be negatively charged when the pH increment and continue its cationic form when the pH low. Moreover, the positive charge on the RH and MOSP surface gets down significantly low as pH of the solution increment thus produce not enough charge neutralization of natural coagulant and destabilizes the particles.

To best comprehend the mechanisms concerned [28] expound that at low pH the zone surface of the coagulant is hugely protonated at the positive ions (H+) give an electrostatic strength between the polluted of molecules and surface, leading to high adsorption. In the meanwhile, led increment pH low in degree the protonation of the surface, causing tine in a lower diffusion and adsorption reason to electrostatic repulsion. The results offer that at pH 4, that 99.15% and 60.73% when using RH and MOSP, the removal rate of phosphate while removal efficiencies become low when it is adjusted the pH alkaline and neutral.

**Figure 1** Response surface curves of phosphate removal using RH in the coagulation-flocculation process under different conditions:
(a) initial concentration and RH dosage
(b) initial concentration and pH
(c) RH dosage and pH

**Figure 2** Response surface curves of phosphate removal using MOSP in the coagulation-flocculation process under different conditions:
(a) initial concentration and MOSP dosage,
(b) initial concentration and pH
(c) MOSP dosage and pH
3.4. Effects of coagulant dosage and initial pH

Curves of responses surface are a diagrammatic representation of providing polynomial equation utilized to envisage the relationship between empirical levels of all factors and the responses also deduce the best condition for the process. Figures 1(c) and 2(c) display the effect of different coagulant dosages (B) and solution pH (C), and the reciprocal react on phosphate removal, respectively at a stationary condition at 30 min settling time. The F-value from ANOVA in Table 7 for RH coagulant dosage (19.89) versus pH (13.24) displayed that the impact of dosage was more important than pH, the dosage of RH possesses more effect at phosphate removal. At the same time, when used MOSC for phosphate removal, the F-value of the pH (F-value = 1.67) was more important compared to the impact of dosage (F-value = 0.5973). The elliptical lineaments obtained for phosphate removal denote a strong interplay between dosage and pH, as display in Figure1(c), removal of phosphate as high as 98.57% could be obtained at RH dosage between 2.5 and 3 g and a pH of 4. And at the same conditions at use MOSP, also high albeit with the removing 97.53% of phosphate could be obtained at MOSP dosage between 3 and 3.5 g and a pH of 4, the coagulation-flocculation process for removal of phosphate by using RH and MOSP happening due to the particle’s span mechanisms and charge neutralization. Also, RH and MOSP have a profusion of cationic charge, which helped to equalize anionic conditions in an aqueous solution. Increasing the dosage of natural coagulant at its optimum zone leads to more pollutant particles were unstable via electrostatic attraction between anionic particles in aqueous solution and natural coagulant cationic species [27]. However, the lower dosage leads to a lower than optimum value inhomogeneous removal of phosphate [28]. When a coagulant dosage surpasses its optimal point, can be the reason to restore the stability that is pH alteration.

3.5. FTIR analysis

In this study, FTIR spectroscopy was used to analyze a set of current functionalities of the blocks formed by the phosphate coagulation process by natural coagulants (RH, MOSP) and in spectra within (400-4000). From Figure (3), we note phosphate produces suspended solids materials after treatment using (RH, MOSP). Also, the presence of hydroxyl ions in phosphates and the region has a wide range of 3448 and 3446 [29]. The highest value for each sample 2587-2941 was assigned to the C=H bonds to indicate the lipids and fatty acids present in suspended solids and due to the formation of the solid due to the presence of phosphates [30]. The peak was observed in 1733, 1731 and this peak represented the expansion of the C=O group. In contrast, the recent study revealed that the peak at the values 1800-1500 represents the expansion of C=N and this group indicates the presence of hydrogen compounds in phosphates [31]. According to [32] the peaks are at the boundary 1750-900 with a biowaste and are composed of carboxylic acids, aldehydes, cellulose, ketones, and a chain of C, but they are not long. Finally, the results showed that the used agricultural waste (RH, MOSP) contains multiple fibbers and naturally occurring organic materials, and through the spectra, we note that can detect silica and aluminum and within the limits of 460, 613, 748, and 1633 through the use of RH, but when using MOSP, it was found that silica and aluminum were higher than RH, reaching its maximum limits in MOSP 1081. These characteristics indicate that the coagulants used are affected by the cationic properties of suspended pollutants, which makes the neutralization of the plankton charge happen easily. Through Figures 3(a, and c), we note that the FTIR results for the coagulants showed that they mainly contain, amines, hydroxyl, carboxyl, and alkane. These properties reflect peak properties, either iron formation or permanently disappearing.
Figure. 3 FTIR spectra for coagulants and flocks produced by coagulation treatment of phosphate using RHA and MOSP (a) RH (b) MOSP (c) RH after phosphate removal (d) MOSP after phosphate removal

4. Conclusion
   The current study focused on scrutinizing the effect of three operating parameters which as polluted concentration, coagulant dosing, and pH on the phosphate removal in the treatment of synthetic wastewater using natural coagulant in the coagulation-flocculation process and dependence Box-Behnken design (BBD) as a way to optimization. The experience data were installed to a second-order equation which was used for the improvement of operation factors. To amelioration, the water quality, water purity technology by utilized membrane filtration can be purity utilized to produce potable water. There are however combined troubles in the membrane filtration process as membrane pollute can be beaten by the utilization of coagulant process as it assists in elimination from the colloidal particles restrained the flow of liquid during the membranes. Treatment of phosphate using a natural coagulant (RH and MOSC) gives good results for the percentage removal as 98.57% and 97.53%, respectively. The RSM results show that the optimum conditions when using RH as a coagulant were dose 2g, pH 4, and initial concentration of 60ppm, while for MOSC were dose 2.5g, pH 4, and initial concentration of 60ppm. The results showed the high ability to used natural coagulants RH and MOSC as they can process phosphate at a natural pH with a safe and environmentally friendly concentration.

References
[1] Blackall L L, Crocetti G R, Saunders A M and Bond P L A review and update of the microbiology of enhanced biological phosphorus removal in wastewater treatment plants 11
[2] Sonune A and Ghate R 2004 Developments in wastewater treatment methods Desalination 167 55–63

[3] Bhargava D S and Sheldarkar S B 1993 Use of TNSAC in phosphate adsorption studies and relationships. Literature, experimental methodology, justification and effects of process variables Water Research 27 303–12

[4] Karaca S, Gürses A, Ejder M and Açikyildiz M 2004 Kinetic modeling of liquid-phase adsorption of phosphate on dolomite Journal of Colloid and Interface Science 277 257–63

[5] Huang C P 1977 Removal of Phosphate by Powdered Aluminum Oxide Adsorption Journal (Water Pollution Control Federation) 49 1811–7

[6] Zeng L, Li X and Liu J 2004 Adsorptive removal of phosphate from aqueous solutions using iron oxide tailings Water Research 9

[7] Tanada S, Kabayama M, Kawasaki N, Sakiyama T, Nakamura T, Araki M and Tamura T 2003 Removal of phosphate by aluminum oxide hydroxide Journal of Colloid and Interface Science 257 135–40

[8] Oguz E 2004 Removal of phosphate from aqueous solution with blast furnace slag Journal of Hazardous Materials 114 131–7

[9] A.N S Nathen and Ravi.S R 2017 Indian Rice Husk Ash – Improving the Mechanical Properties of Concrete: A Review International Journal of Engineering Research and Applications 07 76–9

[10] Baum L, Chan I H S, Cheung S K-K, Goggins W B, Mok V, Lam L, Leung V, Hui E, Ng C, Woo J, Chiu H F K, Zee B C-Y, Cheng W, Chan M-H, Szeto S, Lui V, Tsoh J, Bush A I, Lam C W K and Kwok T 2010 Serum zinc is decreased in Alzheimer’s disease and serum arsenic correlates positively with cognitive ability Biometals 23 173–9

[11] Ash H 2013 SELF COMPACTING CONCRETE INCORPORATING RICE Maximum resisted Load 6

[12] Abbas M N, Al-hermizy S M M, Abudi Z N and Ibrahim T A 2019 Phenol Biosorption from Polluted Aqueous Solutions by Ulva lactuca Alga Using Batch Mode Unit 20 225–35

[13] Abudi Z N 2018 Using sawdust to treat synthetic municipal wastewater and its consequent transformation into biogas Journal of Ecological Engineering 19 10–8

[14] Garde W K, Buchberger S G, Wendell D and Kupferle M J 2017 Application of Moringa Oleifera seed extract to treat coffee fermentation wastewater Journal of Hazardous Materials 329 102–9

[15] Paula H M De, Sangoi M, Ilha D O and Andrade L S 2014 Concrete plant wastewater treatment process by coagulation combining aluminum sulfate and Moringa oleifera powder Journal of Cleaner Production 1–6

[16] Adedayo O, Abdulkareem F, Yusuf A S and Lala M 2019 South African Journal of Chemical Engineering Response surface methodology approach to optimization of process parameter for coagulation process of surface water using Moringa oleifera seed 28 46–51
[17] Wang M, Wang J, Tan J X, Sun J F and Mou J L 2011 Optimization of ethanol fermentation from sweet sorghum juice using response surface methodology Energy Sources, Part A: Recovery, Utilization and Environmental Effects 33 1139–46

[18] Hasnain M, Henry E and Ahmed Z 2013 ScienceDirect Boron removal by electrocoagulation and recovery Water Research 51 113–23

[19] Nariyan E, Sillanpää M and Wolkersdorfer C 2017 Uranium removal from Pyhäsjärvi/Finland mine water by batch electrocoagulation and optimization with the response surface methodology Separation and Purification Technology

[20] Kakoi B, Kaluli J W, Ndiba P and Thiong G 2017 Optimization of Maerua Decumbent bio-coagulant in paint industry wastewater treatment with response surface methodology. Journal of Cleaner Production

[21] Azadi F and Saadat S 2018 Experimental Investigation and Modeling of Nickel Removal from Wastewater Using Modified Rice Husk in Continuous Reactor by Response Surface Methodology Iranian Journal of Science and Technology, Transactions of Civil Engineering 0123456789

[22] Hou D, Chen D, Wang X, Wu D, Ma H, Hu X, Zhang Y, Wang P and Yu R 2020 RSM-based modelling and optimization of magnesium phosphate cement-based rapid-repair materials 263

[23] Hou D, Chen D, Wang X, Wu D, Ma H, Hu X, Zhang Y, Wang P and Yu R 2020 RSM-based modelling and optimization of magnesium phosphate cement-based rapid-repair materials 263

[24] Hashim K S, Jasim N, Shaw A, Phipps D, Kot P, Alattabi A W, Abdulredha M and Alawsh R 2019 Separation and Puriﬁcation Technology Electrocoagulation as a green technology for phosphate removal from river water 210 135–44

[25] Singh V and Kumar P 2011 Cassia Seed Gums: A Renewable Reservoir for Synthesizing High Performance Materials for Water Remediation Biopolymers: Biomedical and Environmental Applications 267–90

[26] Pui K, Shak Y and Wu T Y 2014 Coagulation – flocculation treatment of high-strength agro-industrial wastewater using natural Cassia obtusifolia seed gum : Treatment efficiencies and flocs characterization CHEMICAL ENGINEERING JOURNAL 256 293–305

[27] Xia M, Ye C, Pi K, Liu D and Gerson A R 2018 Cr(III) removal from simulated solution using hydrous magnesium oxide coated fly ash: Optimization by response surface methodology (RSM) Chinese Journal of Chemical Engineering 26 1192–9

[28] Freitas T K F S, Oliveira V M, Souza M T F De, Geraldino H C L, Almeida V C, Fávaro S L and Garcia J C 2015 Optimization of coagulation-flocculation process for treatment of industrial textile wastewater using okra (A. esculentus) mucilage as natural coagulant Industrial Crops & Products 76 538–44

[29] Choy S Y, Murthy K, Prasad N and Wu T Y 2016 Isolation, characterization and the potential use of starch from jackfruit seed wastes as a coagulant aid for treatment of turbid water Environmental Science and Pollution Research

[30] Liu Q and Cui B 2011 Impacts of climate change / variability on the streamflow in the Yellow River Basin, China Ecological Modelling 222 268–74
[31] Hoong K, Wang Y, Sheng Z and Cheng C K 2018 ScienceDirect A study into syngas production from catalytic steam reforming of palm oil mill effluent (POME): A new treatment approach *International Journal of Hydrogen Energy* 1–14

[32] Smidt E, Eckhardt K, Lechner P, Schulten H and Leinweber P 2005 Characterization of different decomposition stages of biowaste using FT-IR spectroscopy and pyrolysis-field ionization mass spectrometry 67–79