Using of Coagulation Process of Pomegranate Peels for the Tertiary Treatment of Wastewater

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Abstract
The aim of this study is to evaluate the performance of alum and local material like pomegranate peels in a tertiary treatment of wastewater. Experimental work was carried out through the design of a pilot plant scale, where consisting of three units: coagulation, flocculation, and sedimentation units.

From the experimental work, it can be noted that when using alum only, the best removal efficiencies of biochemical oxygen demand (BOD), chemical oxygen demand (COD), and nitrate (NO₃), were 35.9%, 38.24%, and 48.68%, respectively, at a dosage of 200 mg/l of alum with pH value of 6. The removal of phosphate (PO₄) was 76% at a dosage of 250 mg/l of alum with pH value of 6. The using of pomegranate peels (as a powdered activated carbon) leads to obtain best removal efficiencies of BOD, COD and NO₃ were 37.5%, 40.28%, and 62.58%, respectively, at a dosage of 20 g/l of powdered activated carbon. The best removal of PO₄ was 73% which has been satisfied with a dosage of 15 g/l. The best pH value was 5 to satisfy optimum removal efficiencies of BOD, COD and PO₄, while for NO₃ removal, the value of pH was 6.

Finally, the best dose of alum was added with each doses of the pomegranate peels to increase the removal efficiency of sewage effluent pollutants. The removal efficiencies of BOD, COD and NO₃ were 68.24%, 70.59%, and 78.01%, respectively, these values have been obtained at the best a dosage of alum of (200 mg/l) with 20 g/l dosage of pomegranate peels.

Keywords: Coagulation, Tertiary Treatment, Powdered Activated Carbon

INTRODUCTION
In the last years, water drought happened due to the shortage in rainfall. This is a problem in arid and semi-arid countries such as Iraq. During the period of drought, water in streams, rivers, and wells can be severely diminished. On the other hand, rapidly growing population with associated change in lifestyle and consumption patterns; competition between sectors, such as industry, agriculture, and energy for precious land and water demand for different for water use figures [1].

Continued population growth, contamination of both surface water and groundwater, uneven distributions of water resources, and periodic droughts have forced water agencies to search for new sources of water supply. Use of highly treated wastewater effluent, now discharged to the environment from municipal wastewater treatment plants, is receiving more attention as a reliable water resource [2].

The primary goal of municipal wastewater treatment is to protect the health, safety, and welfare of the public. A secondary but essential goal is to protect and provide for the propagation of fish, and wildlife. A concomitant goal is to provide for recreation in and on the receiving water. To achieve sustainable development, maximum beneficial reuse of the wastewater [3].

Reuse of effluents from the municipal wastewater treatment plant is becoming a matter of great importance as available and economical resources for different of applications. The major pathways of water reuse include irrigation, industrial use, surface water replenishment, and groundwater recharge [2].

Tertiary wastewater treatment (or advanced wastewater treatment) is defined as the methods and processes that remove more contaminants (suspended and dissolved substances) from wastewater than are taken out by conventional biological treatment. Put another way, advanced wastewater treatment is the application of a process or system that follows secondary treatment or that includes phosphorus removal or nitrification in conventional secondary treatment [4].

The purpose of advanced wastewater treatment is to remove one or more of the following constituents: organic matter and total suspended solids remaining after the conventional secondary treatment process to meet more stringent discharge and reuse requirements, residual total suspended solids to condition the treated wastewater for more effective disinfection, nutrients beyond what can be accomplished by conventional secondary treatment processes to limit eutrophication of sensitive water bodies, and specific inorganic (e.g., heavy metals) and organic constituents to meet more stringent discharge and reuse requirements for surface water, groundwater recharge, and industrial reuse [2].

Many physico-chemical processes can be applied in the advanced treatment of wastewater, such as ozonation, Fenton reagent oxidation, active carbon adsorption, ion exchange, chemical precipitation, membrane separation, coagulation and zero-valent iron (ZVI), etc. In these processes, coagulation and ZVI are relatively cheaper, easier to operate. As a result, the two processes are widely used in wastewater treatment [5 and 6]. The selection of these processes depended mainly on the required quality of treated effluent or type of removed constituent [7].
Processes studied in the bibliography for tertiary treatment include coagulation-flocculation, sedimentation, filtration, disinfection and ultrafiltration [8]. Coagulation, flocculation and sedimentation systems are aimed at reducing SS, biodegradable organics and nutrients (nitrogen and phosphorus compounds) in the feed [9].

[10] studied the decolorization and COD removal of secondary yeast wastewater effluents by coagulation using alum. They concluded that the best dosage of alum was 4.5 g/l with pH value 8 to obtain removal efficiency of COD and color equal to 67% and 89% were respectively. [11] studied the advanced wastewater treatment by coagulation and zero-valent iron (ZVI) processes for treatment of effluent discharged from coking wastewater. [12] studied the tertiary treatment which was including (coagulation, flocculation, sedimentation, and disinfection) for secondary effluent from the urban wastewater treatment plant, to reuse effluent in irrigation purpose. [13] indicated to use coagulation and flocculation together with microsieve filtration in advanced treatment for phosphorus removal from effluent wastewater treatment plant.

Powdered activated carbon (PAC) is used for removal of low molecular weight organics, taste, and odor causing materials in raw water. It can be used in conjunction with coagulation, enhanced coagulation to improve the removal of natural organic matter from raw water [14], [15] used powdered activated carbon as a coagulant aid for removing of total organic carbon. In their study, they used two types of primary coagulants which are a ferric chloride and poly aluminum chloride with powdered activated carbon.

The first main aim of this study is to find local material and evaluating the performance of such material for tertiary wastewater treatment by using coagulation process. The second main aim is to design and construct a pilot-scale plant for the purpose of obtaining the required data that prove the success of the local materials as reliable coagulants.

**MATERIALS AND METHODS**

**Characteristics of The Secondary Effluent**

In this study, the effluent sewage was taken from secondary clarifiers of Al-Barakia wastewater treatment plant in Al-Najaf city/ Iraq. Al-Barakia wastewater treatment plant is of activated sludge type. The secondary effluent characteristic are given in table (1).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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</thead>
<tbody>
<tr>
<td>BOD₅ (mg/l)</td>
<td>35-43</td>
</tr>
<tr>
<td>COD (mg/l)</td>
<td>64-81</td>
</tr>
<tr>
<td>NO₃ (mg/l)</td>
<td>45-93</td>
</tr>
<tr>
<td>PO₄ (mg/l)</td>
<td>7-8</td>
</tr>
</tbody>
</table>

**Coagulation Experiments**

A pilot plant which has been constructed to consist a coagulation unit (rapid mixing tank), flocculation unit (slow mixing tank), and settling unit (sedimentation tank). The schematic diagram of the pilot plant used is shown in figure (1). The figure shows units, pipes, and pumps that are used in the present study. The pilot plant was manufactured from glass with (10 mm) thickness and glued with silicon.

Each experiments begins by passing wastewater in the rapid mixing basin at impeller speed of 125 rpm for 3.8 min detention time, and then the wastewater is passing through a slow mixing tank and settling tank for a detention time of 1.54 hr.

![Figure 1: Schematic Diagram of the Pilot Plant Used.](image-url)
Materials
The aluminum sulfate or alum used was of chemical formula of \((\text{Al}_2\text{(SO}_4\text{)}_3\cdot18\text{H}_2\text{O})\). The pH of the solutions was adjusted by means of NaOH / HCl solutions. Also, some materials used in preparation of pomegranate peels.

Pomegranate peels preparation
Pomegranate peels were as wastes that produced in large quantities from different sources such as juice mill. In the present study it was produced an activated carbon from pomegranate peels. The pomegranate peels were cut into small pieces, dried in an electrical oven at a 105 °C for two hours, and crushed. The pomegranate peels were washed with hot water and dried in an electrical oven at a 105 °C. The dried peels were placed in glass containers. A phosphoric acid \((\text{H}_3\text{PO}_4)\) was poured carefully into the containers containing the peels with an impregnation ratio of \((1 \text{ g of peels impregnated in } 2 \text{ ml of } 3\text{M of } \text{H}_3\text{PO}_4)\) at 25 °C and an impregnation time of twenty for hours [16]. At the end of the soaking time, the soaked peels were left in the air for partial dryness and then dried in an oven at a 120 °C for one hour. After then, the peels were placed on a metallic plate and subjected to an average temperature of 500 °C for one hour [17]. After one hour of carbonization time, product was taken to a flask and distilled hot water was added. Then, peels were washed with distilled boiling water until the desired pH was achieved (6.5), and the product was dried in an electric oven at 120 °C in order to remove any undesired moisture within the particles. And then, the peels were grinded and sieved through 75µm stainless steel sieve (sieve No. 200) to get powdered activated carbon of pomegranate peels to be used in the present study. Sample of pomegranate peels is shown in figure (2).

RESULTS AND DISCUSSION
Evaluation the Performance Efficiency of the Alum for the Treatment of Sewage Effluent
Effect of pH
Figure (3) shows the effects of pH on the reduction efficiency of pollutants of effluent sewage such as (COD, nitrate, and phosphate concentrations) using alum only as a coagulant. Different values of pH used in this study and these values were 4, 5, 6, 7, 8, and 9.

![Figure 2: (a) Pomegranate Peels After Drying, and (b) Powdered Activated Carbon of Pomegranate Peels.](image)

![Figure 3: Variation of Removal Efficiency with pH of Alum for Reduction concentrations of COD, Nitrate, and Phosphate, and the Other Conditions were Kept Constant (such as a Dosage of Alum, Speeds Mixing (Rapid and Slow Mixing), Times Mixing, and Settling Time).](image)
The results of this figure refer that the best values of removal efficiencies are 38.24% for COD, 48.68% for nitrate, and 76% for phosphate and these values were obtained by using alum only as a coagulant with pH value of 6.

From the analysis of the results of above figure it can be indicated that the using of alum as a coagulant gives better efficiency for removal of wastewater pollutants at acid value of pH.

Figure (4) explains the variation of efficiency of alum in removing the biochemical oxygen demand (BOD$_5$), chemical oxygen demand (COD), nitrate (NO$_3^-$), and phosphate (PO$_4^{3-}$) of wastewater. The experiments were done by adding different dosages of alum of values of 50, 100, 150, 200, and 250 mg/l.

The examination of the results of this figure indicated that the best removal efficiencies are 35.9% for BOD$_5$, 38.24% for COD, and 48.68% for NO$_3^-$. These values of efficiencies are obtained with alum dosage of 200 mg/l, and with the best removal efficiency of phosphate ions of 76% at a dosage of 250 mg/l of alum only.

Based on the analysis of results of this figure it can be concluded that the use of alum only in advanced wastewater treatment leads to obtain good ability for removing of pollutants of effluent sewage but within specific range as shown in figure (4). Destabilization is referred to as charge neutralization or coagulation of colloidal particles of wastewater (colloidal particles are normally charged negatively) in the presence of Al(OH)$_3$ is termed as enmeshment or sweep floc. Dissolved organic can be removed by adsorption on aluminum precipitation. This conclusion is in well agreement with the previous study [18].

Evaluation the Performance Efficiency of the Pomegranate Peels for the Treatment of Sewage Effluent

In this study the pomegranate peels were used after carbonization as a powdered of activated carbon because the using of these peels in organic phase leads to increase the organic content in wastewater.

Effect of pH

Figure (5) shows the effects of pH on the reduction efficiency of pollutants of effluent sewage (COD, nitrate, and phosphate ions) using powdered activated carbon of pomegranate peels as a coagulant. Different values of pH used in this study and these values were 4, 5, 6, 7, 8, and 9.

The results of this figure indicate that the best values of removal efficiency of 40.28% of COD and 73% of phosphate ions are obtained by adding powdered activated carbon of pomegranate peels at pH value of 5. While, the optimum removal efficiency of 62.58% of nitrate ions occurs at pH value of 6. The other conditions were kept constant (such as a dosage of pomegranate peels, speeds mixing (rapid and slow mixing), times mixing, and settling time).

The analysis of the results of this figure, reveals that the using of powdered activated carbon of pomegranate peels in coagulation process can give better efficiency for the removal of wastewater pollutants under acidic value of pH, and this conclusion is in good agreement with previous study [19] which have indicated that the increase of pH favored the ionization of organic matter and decreasing the pH could redound to neutralize the negative charge on the surface of powdered activated carbon using hydrogen ions.

The reason for the better adsorption capacity observed at acidic value of pH may be attributed to the larger number of H$^+$ ions present, which in turn neutralize the negatively
charged adsorbent surface, thereby reducing, hindrance to the diffusion of organics and negative ions at higher pH. At higher pH, the capacity of the adsorbent decreased. The reduction in adsorption may be possible due to the abundance of OH ions, causing increased hindrance to diffusion of organic matters contributing to (COD) and negative ions (such as nitrate and phosphate). Lower removal efficiency at pH lowering is due to the competition of Cl⁻ ions (from HCl added externally to adjust the pH) with negative ions to the adsorbent sites, at higher pH the highly negatively charged adsorbent surface sites do not favor the adsorption of oxanion due to the electrostatic repulsion, this conclusion is in well agreement with previous studies [20 and 21].

Figure (6) shows the variations of efficiency of pomegranate peels in treatment of pollutants of wastewater such as BOD₅, COD, NO₃⁻, and PO₄⁻ with the variations of dosages of pomegranate peels. The experiments were done by adding different dosages of pomegranate peels. And these dosages are of values of (1, 5, 10, 15, and 20) g/l.

**Figure 5:** Variation of Removal Efficiency with pH of Pomegranate Peels for Reduction concentrations of COD, Nitrate, and Phosphate, and the Other Conditions were Kept Constant (such as a Dosage of Alum, Speeds Mixing (Rapid and Slow Mixing), Times Mixing, and Settling Time).

**Figure 6:** Variation of Efficiency of Removal BOD₅, COD, NO₃⁻, and PO₄⁻ with Dosages of Pomegranate Peels.
The examination of the results of figure (6) leads to obtain a good conclusion which that the best value of removal efficiency of BOD$_5$ was 37.5%, 40.28% of COD, and 62.58% of NO$_3$. These values are achieved by using pomegranate peels with a dosage of 20 g/l. The pH values for removal BOD$_5$ and COD was 5, and was of a value of 6 for nitrate removal. The optimum removal of phosphate was 73% at a dosage of 15 g/l of pomegranate peels under pH value of 5.

From the analysis of the results of this figure, it can be concluded that the using of activated carbon of pomegranate peels as a coagulant for removal efficiencies of BOD$_5$, COD, and nitrate concentrations were better than using of alum only. The reason for this could be due to the fact of the very active surface of powdered carbon and its high capability in adsorbing organic matters, this conclusion is in well agreement with the previous study [22].

**Effect of Alum with Pomegranate Peels on the Pollutants of Wastewater**

Figure (7) shows the efficiency of removal of the pollutants of wastewater such as BOD$_5$, COD, and NO$_3$ versus different dosages of pomegranate peels (g/l) with optimum dosage of alum (200 mg/l) for the removal of BOD$_5$, COD, and NO$_3$.

It can be noted from the analysis of the results that the removal efficiencies were 68.42%, 70.59%, and 78.01% for BOD$_5$, COD, and of NO$_3$ respectively. The results are obtained by using pomegranate peels with optimum dosage of alum (optimum dosage of alum was 200 mg/l for removal efficiencies of BOD$_5$, COD, and NO$_3$) together with optimum pH value of pomegranate peels. The pH value for the removal of BOD$_5$ and COD was 5, and the pH value of nitrate ions removal was 6. The optimum removal of phosphate was 73% which has been achieved with a dosage of 15 g/l of pomegranate peels and with a pH value of 5.

From the analysis of results of this figure it can be obtained that the adding of pomegranate peels as a coagulant aid with alum leads to increase the values of removal efficiencies of pollutants of wastewater which is considered a better than using of alum only or pomegranate peels only. Since coagulation is effective in removing high molecular weight organic matter and ineffective in low molecular weight organic compounds, the possibility of using powdered activated carbon as a coagulant aid in removing organic matter in a vast molecular weight range, this conclusion is in well agreement with the previous study [19].

**CONCLUSION**

1) It is possible to use coagulation-flocculation-sedimentation processes with local materials in advanced wastewater treatment. These above processes have good ability to remove pollutants of sewage effluent.

2) The best removal efficiencies obtained with using of alum only were 35.9% for BOD$_5$, 38.24% for COD, and 48.68% for nitrate ions with 200 mg/l of alum, and 76% for phosphate ions with a dosage of 250 mg/l of alum.

3) The best removal efficiencies when using of pomegranate peels only were 37.5% of BOD$_5$, 40.28% of COD, and 62.58% of NO$_3$ with a dosage of 20 g/l with pH of 5 for removing of BOD$_5$ and COD, while the pH value of 6 was good to remove nitrate. Also, the optimum removal efficiency of phosphate was 73% at a dosage of 15 g/l of pomegranate peels under pH value of 5.

4) The removal efficiencies of pollutants of sewage effluent increased when using local materials as a coagulant aid with alum. The maximum removal efficiency of BOD$_5$, COD, and NO$_3$ when using pomegranate peels as a coagulant aid with alum were 68.42%, 70.59%, and 78.01% respectively.
REFERENCES


