

# Evaluating the Efficiency of Clarifier Returned Sludge with Poly-aluminum Chloride Coagulant (PAC) for Improving the Removal of Turbidity, COD and PVC

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Received: 3 July 2021

Revised: 6 August 2021

Accepted: 16 September 2021

#### Abstract

**Background:** Nowadays, the discharge of effluents containing chemicals through manufacturers or consumer industries causes major environmental problems. In this study, we assessed the effect of sludge reversal of the clarifier unit on enhancing the coagulant performance of Poly-Aluminum chloride in the removal of turbidity/COD/PVC from PVC effluent of Bandar Imam Khomeini petrochemical plant and reduction of the amount of coagulant and costs.

**Methods:** The experiments were carried out by Jar test with poly aluminum chloride (PAC); the returned sludge, as the assisting coagulant, and the amplitude of the differentiates including (5.5-11) pH of coagulant concentration (10-140mg/L) and concentration of returned sludge (2-20 ml) were investigated. To ensure the accuracy and precision, we repeated each stage of the experiment three times.

**Results:** The results showed that the most optimal conditions for coagulant were at pH=8 and the optimum dose of poly aluminum chloride was 25 mg/L plus 5ml of the clear sludge volume in the clarifier. The removal efficiency of turbidity, COD, and PVC under optimum conditions of Poly-Aluminum chloride were reported 99.71%, 97.19%, and 99.55%, respectively.

**Conclusion:** The results indicated that the sludge of the clarifier, as the main assisting coagulant, in addition to increasing the efficiency of the removal turbidity of COD and PVC reduced the dose of the main coagulant.

Please cite this article as: Kahvaei S, Takdastan A, Jalilzadeh Yengejeh R. Evaluating the Efficiency of Clarifier Returned Sludge with Poly-aluminum Chloride Coagulant (PAC) for Improving the Removal of Turbidity, COD and PVC. *J Health Sci Surveillance Sys*. 2021;9(4):298-304.

**Keywords:** Turbidity, Coagulant, Waste water, Clarifier, Environment

## Introduction

The oil refining industry produces a large amount of wastewater due to the amount of water needed for specific processes, including cooling systems. As the discharge of untreated wastewater leads to major environmental problems, refining the wastewater is considered the main concern for these industrial units.<sup>1</sup>

In Bandar Imam Khomeini Petrochemical

Refinery PVC unit, the flocculation process of particulate matter in the effluent is carried out by applying alum coagulant. The flocculation process is acquired in the refinery unit of this company to improve the effluent treatment process in the PVC unit, alleviate environmental problems, and separate the PVC powder particles from the effluent.<sup>2</sup>

Polyvinyl chloride (PVC) is considered one of the most extensive petrochemical products and one of the

mostly used synthetic polymers. PVC is provided in two types, namely hard and soft.<sup>3</sup> Due to the harmful effects of these particles, which are based on chlorine, before taking any actions toward their discharge into the environment, they should be removed.<sup>4</sup>

Poly aluminum chloride is one of the most important types of IPFs that, in comparison to other types, is widely applicable. From the combinational respect, the Polyaluminum chloride or hydrated aluminum chloride is a mineral macromolecule in which Monomer is a dual-core complex of aluminum. In low concentrations, in the abiotic environment, this compound creates a multicore complex formation and this property makes this coagulant unique in the coagulation process.<sup>5</sup>

The returned sludge of the secondary settling tank consists of high amounts of coagulant residues that will be discarded in case of not being retrieved.<sup>6</sup> Since the feedback efficiency of Bandar Imam Khomeini Petrochemical Refinery should be based on the environmental standards, by selecting a suitable coagulant with appropriate pH injection concentration to the effluent, it will be able to remove significant amounts of turbidity, PVC, and COD; also, by the sludge returning of the clarifying unit to the beginning of coagulation and flocculation stage, it will be effective in improving the flocculation act and the possibility of reusing the produced sludge.

A large number of studies have been conducted on this case so far. In the research conducted by Mostafizur et al., Tamarindus indica and Litchi chinesis seeds were applied as coagulants in the treatment of river water.<sup>7</sup> Adams et al. used rice peel ash to remove water turbidity.<sup>8</sup>

Abdolmoteleb et al. used chitosan as a coagulant to remove turbidity; they indicated that in comparison with the electrocoagulation process without the help of coagulants, applying electrocoagulation with aluminum electrode along with chitosan, as a coagulant, increased the removal of turbidity.<sup>9</sup> Hashemi Nejad et al. in their research used tannic acid as a coagulant for reducing water turbidity and found that tannic acid had the ability to coagulate and, therefore, can be replaced with the conventional coagulants in preliminary procedures of water treatment.<sup>10</sup> Takdastan et al. compared the coagulant efficiency of poly ferric sulfate (PF) and poly aluminum chloride (PAC) and stated that there was a partial but statistically non-significant difference between the two groups of PFS and PAC in terms of removal efficiency of the turbidity.<sup>11</sup>

In their research, Moradian Fard et al. found that the maximum turbidity and coliform removal efficiency of water at a concentration of 10 ppm of poly aluminum chloride was above 90%.<sup>12</sup>

## Methods

This descriptive-analytical study was conducted on a laboratory scale in the PVC unit laboratory of Bandar Imam Khomeini Petrochemical Company. The samples were taken from the effluent of the petrochemical PVC unit. The procedures, including collecting, storing, and transferring the samples, were carried out according to the predefined guidelines. The average annual turbidity entrance of raw water to the sewage-treatment plant in non-flood conditions was calculated to be 120 Nephelometry units (NTU).

In operating all the stages of this procedure, in 2019, devices of high quality and standards, namely the Jar Test Device (the AQUALYTIC Model made in Germany), Turbidity Meter (with the brand name HACH, made in the USA), and pH meter (Model 3510, made in the United Kingdom) were applied. Effluent samples taken from the PVC unit of Bandar Imam Petrochemical Company were used in performing the experiments. The fresh sludge of the Clarifier tank was also taken from the same place. PAC and the returned sludge were applied as coagulants and coagulant aids in this study. Major variables under study were assessing the turbidity, COD, and PVC.

1 g/L amount of PAC solution (1000 mg/L) was collected. The first stage i.e. rapid mixing in the Jar Test, was done applying a mixture with a round of 130 rpm in 2 minutes, and the slow mixing stage, with the speed of 40 rpm, took 30 minutes. And the final sediment stage also occurred in 30 minutes. In this stage, to achieve an optimum mixture, we determined the poly aluminum chloride coagulant with an optimum dose of 40 mg/L, an optimum pH equal to 8, and the optimum sludge volume of the clarifier as 5 ml. Determining the consuming coagulant concentration (PAC) in this experiment was done along with the sludge of the clarifier. At first, different doses of consuming coagulant within a range of 10-40 mg/L along with the sludge of the clarifier with an optimum volume equal to 5 ml were injected into the samples, and the optimum dose of 25 mg/L was obtained according to the stage.<sup>13-17</sup> The effect of each parameter separately on turbidity/COD/PVC removal in the volume of the sediment sludge was determined at the end of each stage, by applying the covariance analysis, and the corresponding diagrams were drawn using Excel software.

## Results

### *Determining the Efficiency of Turbidity Removal Under Optimum Conditions of PAC Performance Along with Returned Sludge*

By maintaining the optimal conditions of pH on a

constant level of 8, the optimum volume of the sludge of the clarifier unit equal to 5 ml with the incoming turbidity of 1055 NTU, in this stage, poly-aluminum chloride was injected in different doses with the following amounts:10/15/20/25/30/35/40; the outlet turbidity at the optimum dose of 25 mg/L was equal to 3 NTU and the removal efficiency of turbidity was 99.71%, as shown in Figure 1.

The obtained results of correlation analysis ( $P < 0.05$ ) revealed a meaningful relationship between the acquired dose of poly-aluminum chloride along with the sludge of clarifier and the turbidity removal.

Figure 1 illustrates the effect of poly-aluminum chloride along with the sludge clarifier on the efficiency of turbidity removal. As the Figure indicates, the maximum efficiency of turbidity removal of 99.71% was observed in the optimum dose of 25 mg/L poly aluminum chloride plus 5 mg/l of the optimum volume of injectable sludge. In these conditions, the mechanisms of turbidity removal depend on the concentration of the coagulant, which is considered as the dominant mechanism of adsorption or vacuum coagulation or a combination of both mechanisms. As shown in the Figure, by increasing the dose of the coagulant matter up to 25 mg/L, the flocs become more adhesive and heavier, leading to the increase of the sediment rate and the removal efficiency. However, at higher doses, the removal efficiency decreases, due to re-stability.

Similar results have been reported that confirm the increase of the turbidity removal efficiency, due to particle aggregation in fluke formation in higher coagulant values.<sup>18</sup> Furthermore, other results have shown that the volume of optimum returned sludge along with poly aluminum chloride was observed in turbidity removal with maximum efficiency of 98.31% at 10 ppm dose in the volume of injectable sludge equal to 10 ml. Also, the maximum efficiency of 98.92% at a dose of 30 ppm in injectable sludge was reported as 4 ml. Another fact indicated in this study was that by increasing the sludge to the rapid mixing stage, the turbidity of the sample decreased.<sup>19</sup>

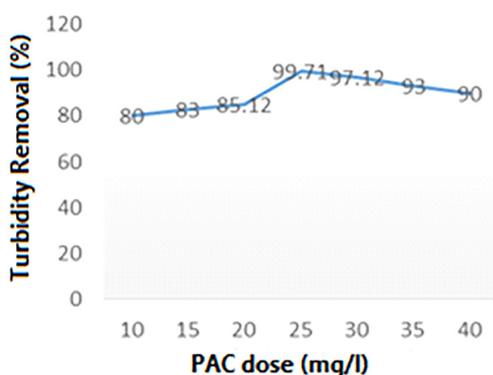


Figure 1: Effect of coagulant with clarifying sludge on Turbidity Removal

#### Determination of COD Removal Efficiency in the Determining the Stage of Optimum Coagulant Dose with Clarifier Sludge

At this stage, the mean of the input COD was 712 and the output COD in the optimum coagulant dose was 25 mg/L equal to 20 mg/L, and the maximum COD removal efficiency, in this period was 99.19%. The percentage of the COD removal efficiency is shown in Figure 2.

Under optimum conditions of poly-aluminum chloride coagulant yield and the optimum volume of return sludge, COD removal efficiency increased up to 25 mg/L by increasing the dose of coagulant, and removal efficiency reached 97.19%.

A study was conducted, in which the removal of COD and dye from the adhesion industry was investigated by applying coagulation. Aluminum chloride and ferric chloride were used as coagulants. The results showed that the optimum coagulant dose obtained, applying jar test experiment, was between 100-1000 mg/L and ferric chloride 1000-6000 mg/L. At pH=4 and concentration of 1000 mg/L poly-aluminum chloride, dye removal was 75% and COD removal was 65%.<sup>20, 21</sup>

#### Pvc Removal Efficiency in the Stage of Determining the Optimum Coagulant Dose Along with the Clarifier Returned Sludge

At this stage, the mean PVC input was 1700 mg/L and the output PVC at the optimum dose of coagulant was 25 mg/L, 7.5 mg/L, and the maximum removal efficiency value of PVC in this period was 99.55%. The graph below illustrates the percentage of PVC removal (Figure 3).

A study was conducted on the enhancement of the wastewater coagulation and flocculation process in a PVC company, showing that aluminum sulfate as a coagulant along with polyelectrolyte had better efficiency in comparison with ferric chloride and calcium chloride. Furthermore, the two factors of pH and mixing rate played an important role in improving

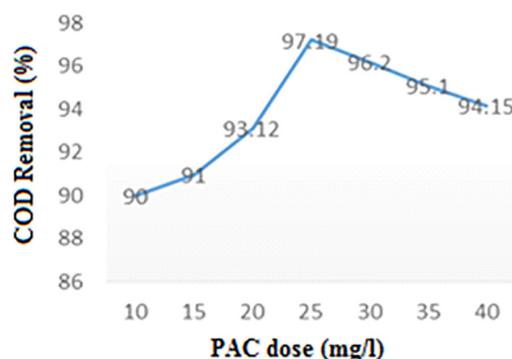
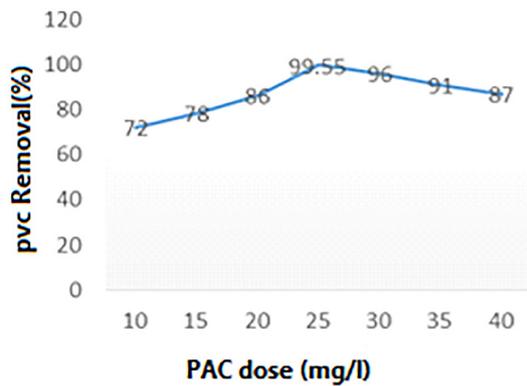


Figure 2: Effect of coagulant along with clarifier sludge on COD Removal



**Figure 3:** The effect of coagulant along with the clarifier sludge on PVC removal (%)

the coagulation and flocculation process.<sup>19</sup> It has been shown that in practical applications, the coagulation effect of poly-aluminum chloride is far greater, about 2 to 3 times that of common aluminum salts. One of the advantages of PAC is that it creates fluke information; thus, it takes a short time to react and sediment. By using the same dose of coagulants, the pH reduction caused by poly-aluminum chloride consumption is lower than another mineral coagulant.<sup>22</sup>

*Determining the volume of sediment sludge in the determination stage of the coagulant dose with the clarifier sludge. At this stage, the volume of the sediment sludge was 7 ml/L.*

According to the results of correlation analysis ( $P < 0.05$ ), a significant relationship was noticed between the dose of poly-aluminum chloride along with the clarifier sludge and the percentage of turbidity removal /COD/PVC/ from the volume of sediment sludge.

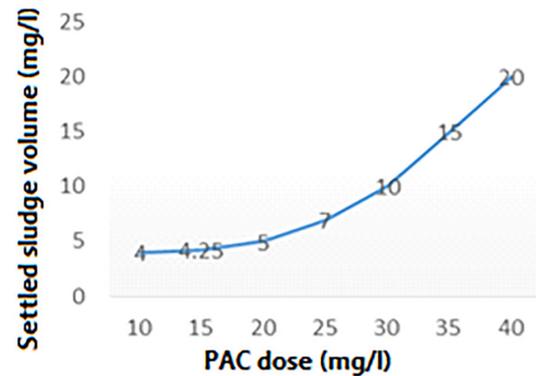
According to Figure 4, the volume of the sediment sludge increased with the increase in the dose of poly-aluminum chloride coagulant.

In other similar studies, it has been reported that under optimum conditions, removing the chromium of the PASIC coagulant with the concentration of 150 mg/L, pH equal to 5, and sedimentation time of 45 minutes can reduce the chromium initial concentration of 10 mg/L to  $0.29 \pm 118$  mg/kg, which is beyond the standards of effluent discharge into receiving waters.<sup>11</sup>

The results showed that 30 minutes was the time of optimum sedimentation for cadmium to be removed by poly aluminum chloride.<sup>25</sup>

## Discussion

In this study, the removal of zinc and nickel by poly aluminum chloride along with chitosan as an aid-coagulant was assessed; the results indicated that chitosan, as coagulant aid along with poly aluminum chloride, led to a significant increase in metal removal



**Figure 4:** The effect of the coagulant with the clarifier sludge on settled sludge volume

efficiency to 99%.

The entry of organic compounds, due to the degradation of plant residues and the discharge of urban, industrial, and agricultural wastewaters into water resources, is considered one of the major concerns and issues regarding water resources. Many studies have been conducted in the field of comparing the effect of various coagulants, in turbidity removal of different water resources, including poly aluminum chloride, ferric chloride, aluminum sulfate, and other mineral and organic coagulants. In this research, the experiments were carried out by Jar test with poly aluminum chloride (PAC).

The results showed that by increasing the amount of PASIC coagulant, the removal rate of turbidity also increased; moreover, an increase in the amount of silicate, due to the bridge construction mechanism, led to an increase in the turbidity removal.<sup>20</sup> Previous research has shown that in petrochemical wastewater refinery, applying poly aluminum chloride, as a substitute to ferric chloride, has led to the formation of larger flocs; therefore, poly aluminum chloride was selected as the effective coagulant and also pH=7.5 was considered as the optimal pH.<sup>20</sup> The results indicate that, with the increase of dose, the formulated flocs become smaller and lighter. As a result, their sedimentation rate is lower. Accordingly, the best flocs have been observed at a dose of 25 mg/L poly-aluminum chlorides. A meaningful relationship was noticed between the increase of poly-aluminum chloride coagulant and the increase in lead removal efficiency.<sup>24</sup>

Similar results have been reported that indicate in case of using an optimal dose of the returned sludge in Minab refinery plant, an average amount of 30% of poly aluminum chloride reagents is reduced; therefore, we will save an amount of 138 kg of this substance per day, that is 37.50 tones in a year; from an economic point of view, this is a considerable amount.<sup>26</sup>

In 2007, Avid Banihashemi conducted a study to investigate the result of comparing PAC1 as a

coagulant with ferric chloride, and optimizing the application conditions, and investigated its combined use with bentonite for opacity removal from Tehran raw water. With poly aluminum chloride, less pH loss was observed than ferric chloride. Adding bentonite, as a coagulant, has dramatically improved turbidity removal operations in a way that in addition to the significant reduction of the optimum concentrations, turbidity percentage also improved with ferric chloride, especially with poly aluminum chloride. Tokdestan and his colleagues (2006) conducted a study on the use of starch as a natural coagulant aid in the Ahvaz water treatment plant; the results of this study showed that starch had a high efficiency in eliminating water turbidity and microorganisms.

Fazeli et al. in 2014 in a study to investigate the efficiency of five coagulants including poly ferric sulfate, chlorofric, alum, poly aluminum chloride, and poly aluminum ferrous chloride in the removal of turbidity and organic matter using the AHP method concluded that in high input turbidity up to NTU 300 the performance of the coagulants tested was relatively similar; the highest turbidity removal efficiency did not show much difference, but in low turbidity, NTU 10 coagulants Polymeric compounds had better performance than mineral coagulants. Of all the coagulants tested, poly frame sulfate showed better performance in the removal of turbidity and organic matter.

In another study, it was found that the effects of optimum dose of returned sludge led to a 20% reduction in the consumption of poly aluminum chloride (PAC), which is equivalent to saving 191 kg of this coagulant in a day. Also, there was a reduction of 4.2% of the final disposal sludge of the refinery plant, equivalent to 51 cubic meters per day, which had a significant positive role in protecting soil resources and its environment.<sup>27</sup>

The findings of a study indicated that under optimum conditions, chitosan application as an aid-coagulant in turbidity removal along with poly-aluminum chloride reduced the concentration of the coagulant poly-aluminum chloride to about 50%. Furthermore, molecules formed by chitosan were larger and had a high sedimentation rate.<sup>28</sup>

The results obtained in the research conducted by Moradianfard and his colleagues showed that as much as the volume of the returned sludge added to the Bandar Abbas water refinery plant increased, the efficiency of turbidity removal by poly aluminum chloride also increased. However, the efficiency of coliforms removal decreased at doses higher than 5 ml, which is consistent with the results of the present study.<sup>12</sup>

Research conducted by Jamali and his colleagues showed that the optimal PAC value was 2.5% and its

efficiency in COD and turbidity removal was 48.5% and 73.5%. Besides, this method is considered very suitable for leachate purification produced in Qazvin Burial land, due to the efficiency of poly aluminum chloride in the removal of these pollutants. On the other hand, no advanced technology is required and the low expenses add to the quality of this method.<sup>29</sup> The obtained results are consistent with those of this study.

## Conclusion

In this study, the sludge of the clarifier tank was used as the coagulant aid. By adding the sludge of the clarifier as an aid-coagulant, turbidity removal efficiency/COD/PVC increased; also, the volume of the sediment sludge decreased. Given that the aid-coagulant can reduce the dose of the consumed coagulant, the experiment went on at the last stage with doses less than 40 mg/L. At the end, we found that in the optimum dose of poly aluminum chloride of 25 mg/L, the turbidity removal efficiency increased to 99.71%; the removal of COD increased to 97.19% and PVC removal to 99.55%. Also, the volume of the sediment sludge reached 7 ml. The use of clarifier sludge as an aid coagulant, in addition to increasing the efficiency of turbidity and COD/PVC removal, contributed to saving up to 40% of the main coagulant (poly aluminum chloride).

In general, the results of this study indicated that the maximum efficiency of the turbidity/COD/PVC removal at the optimum dose of 25 mg/L polyaluminum chloride plus 5 ml of the clarifier unit sludge at an optimum pH was 8. Besides, we can save consumption of the main coagulant up to 40%, by adding the sludge of the clarifier as an aid-coagulant.

In general, evaluation of the factors affecting turbidity removal including coagulant type, concentration, ph, and primary turbidity using covariance analysis and regression for each coagulant indicated that all the mentioned factors had a significant effect on the removal of turbidity, COD, and PVC in effluents. By reviewing other studies conducted in our country and abroad, it was found that increasing coagulant dose resulted in more removal of turbidity, COD, and PVC in effluents, so that an increase of 5 to 10 units in coagulant dose increased the removal efficiency. On the other hand, changing the ph value also affected the turbidity of COD and PVC. In this regard, the relationship was inverse, meaning that the reduction of pH led to an increase in opacity removal.

## Acknowledgment

Hereby, I announce my gratitude and appreciation to the Research Center and PVC Unit of Bandar Imam Khomeini Petrochemical Company for their cooperation in conducting this research (master's thesis).

**Conflicts of interest:** None declared.

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