

Using of powdered activated carbon as coagulant aid in Total organic carbon removal in Koot Amir Water treatment plant

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Abstract

Introduction: The removal of organic precursors of disinfection by-products (DBPs), i.e. natural organic matter (NOM), prior to disinfection is considered as the most effective approach to minimise the formation of DBPs. Trihalomethanes (THMs) as the main group of DBPs are categorized and considered to have the potentiality of increasing the rate of liver, kidney and CNS adverse effects.

Methods and Materials: This study which presents experimental results, aimed at the reduction of organic matters using the powdered activated carbon (PAC) in the process of coagulation in Koot Amir water treatment plant in Ahvaz city. The affect of two coagulant such as ferric chloride and poly aluminum chloride (PACl) with PAC in removing Total Organic Carbon (TOC) considering different parameters such as concentration and the kind of coagulants, concentration of PAC, pH changes and contact time has been surveyed.

Results: The results of this study clearly indicated that maximum removal in optimum pH of ferric chloride was 40% while 44% TOC removal was achieved at pH 6.5 for Poly Aluminum Chloride coagulant. In use of powdered activated carbon with optimum pH and concentration of ferric chloride, TOC reduction will increase with increasing PAC concentration and up to 90%. Also by addition of powdered activated carbon at similar conditions together with poly aluminum chloride 87% reduction of TOC was occurred.

Conclusions: Poly aluminum chloride was more effective in TOC removal than ferric chloride and PAC adsorption as coagulant aid improved TOC removal efficiency.

Keywords: Disinfection By-Products, Powdered Activated Carbon, Coagulant aid, organic precursors, Total Organic Carbon.

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Introduction

Natural organic matter (NOM) in rivers and lakes is a complex mixture of molecules with varying molecular weight and chemical nature and originates from a variety of sources (degradation of terrestrial and aquatic organisms, biological activity in the water body, human inputs...). NOM is by far the largest source of organic material in raw water, and it comprises both particulate and dissolved components [1,2,3,4,5].

NOM is primarily composed of humic substances, such as humic acids (HA) and fulvic acids (FA) that result from decomposition of terrestrial and aquatic biomass, but it can contain a range of organic species and microorganisms and their discharges [2] NOM has been associated with unpleasant taste and odor, growth substrates for bacteria, potential transport of adsorbed hydrophobic organic and inorganic contaminants, increased coagulant and disinfectant demands, and most important for the drinking water treatment industry, disinfection by-products (DBPs) that include such as halogenated substances as trihalomethanes (THMs)[2,4,5,6]. Through epidemiological studies, some DBPs have been associated with a number of adverse human health effects e.g. cancers of the urinary and digestive tracts, bladder and colon cancers, low birth weight, intrauterine growth retardation, and spontaneous abortion, although some of these effects are still somewhat controversial and require further studies. [3] So, attention to the removal of NOM from surface water has increased in recent years. The formation of DBPs can be controlled and minimised using one, or a combination of, the following approach: removal of DBP precursors prior to disinfection, change of disinfectant, and

removal of DBPs following disinfection [5,6,7,8]. The removal of organic precursors of DBPs i.e. natural organic matter (NOM) prior to disinfection and distribution is considered to be the most effective approach to minimise the formation of DBPs. Due to the increasingly stringent DBP regulations, advanced precursor removal technologies are being used to maximise NOM removal. These include membrane filtration (microfiltration, nanofiltration, reverse osmosis), activated carbon (granular activated carbon, powdered activated carbon (PAC)), and ion exchange (MIEX resin)[5,7].

The US Environmental Protection Agency (EPA) has recognized either enhanced coagulation (EC) or granular activated carbon (GAC) as the best available technology (BAT) for controlling DBP precursors [4]. Enhanced coagulation corresponds to the use of coagulant dosages effective for TOC removal. In USA, enhanced coagulation was selected as the Stage 1 treatment of choice because it was effective for the TOC removal and could be implemented at most water plants treating surface waters using existing treatment processes [7]. The removal of organic precursors of DBPs using enhanced coagulation is related to pH, alkalinity, type and coagulant type and doses and also the nature and concentration of the organic matter. The D/DBP Rule has two steps [6]; the first two steps TOC removal requirements based upon raw water TOC and alkalinity, obviously the higher the raw water TOC, the greater the percentage TOC removal achievable by enhanced coagulation. Coagulation and softening tests conducted on natural waters, showed that TOC (and therefore NOM) removal is strongly pH dependent. Therefore, the higher the alkalinity

of the water, the lower the NOM removal achievable using enhanced coagulation.[8]

Generally, coagulation removes humic and high molecular weight organic matter better than it removes non-humic and low molecular weight organic matter. Thus, non humic fraction of NOM can be removed by using adsorption process. Powdered activated carbon (PAC) is used for removal of low molecular weight organics and taste-odor causing materials in raw water. Besides, it can also be an effective adsorbent for organic precursors of DBP. PAC can thus be used in combination with enhanced coagulation (EC) to remove NOM from raw water supplies. There are several advantages to adding PAC during EC. Thus, EC-PAC combination may have greater DBP precursors removal capacities. [7]

PAC treatment has been used in conjunction with coagulation, enhanced coagulation, or ultrafiltration to improve the removal of NOM [7,9,10]. In a pilot-scale study, Jacangelo et al. reported 12–80% removal of NOM from US river water, depending on PAC dose, when PAC is added as a pre-treatment to ultrafiltration [9]. Based on the results from a series of laboratory scale jar test experiments, Najm et al. claimed that the combination of enhanced coagulation and PAC provides a more cost-effective treatment process than enhanced coagulation only, in order to produce drinking water that meets US water quality regulations [10]. In another

laboratory-scale study, Uyak et al. demonstrated that supplementing enhanced coagulation with PAC in the treatment of Turkish lake water resulted in an increased DOC removal from 45% to 76% at an optimum PAC dose of 40 mg L⁻¹[7]. Recently, in a laboratory-scale study on the effect of PAC addition on the removal of NOM, Alvarez-Uriarte et al. reported that the addition of small amounts (up to 50 mg L⁻¹) of PAC during coagulation increased the removal of THM precursors from 40% to 70% [11]. However, Carrière et al. found that the application of PAC (11 mg L⁻¹) combined with enhanced coagulation at a WTP in Canada only resulted in a small increase (7%) in the reduction of DOC and did not improve the removal of THM precursors [12].

The purpose of this study was to evaluate enhanced coagulation with ferric chloride and poly aluminum chloride as coagulant combined powdered activated carbon (PAC) as coagulant aid to achieve the required TOC removal in Koot Amir Water treatment plant.

Methods and Materials

Collecting samples

The raw water used for this research was obtained from the Intake basin of Koot Amir Water treatment plant, situated nine kilometers south west of Ahvaz. The raw water samples (Table 1) were carried to the laboratory of this water treatment plant in 20-liter plastic containers.

Table 1: Water quality characteristics of the collected water samples

Alkalinity (mg/l)CaCO ₃	TOC (mg/l)	Turbidity (NTU)	EC (µm/c)	pH	Temperature (°C)
160	7.2	68	1830	8.08	24

The used chemical materials

The chemical materials used in this research consist of poly aluminum chloride and ferric chloride were of analytical grade purity (AR grade P99% pure) or better and laboratory powdered activated carbon, with a trade name of 'Merck', which is made in Germany. As well as sulphuric acid, NaOH has been used to adjust the pH.

Method of experiments performance

The executed experiments in this research were consisting of jar test and measuring the Turbidity, Alkalinity, pH and TOC. The total alkalinity analysis involves titration with 0.02 N H₂SO₄ and pH analysis was carried out using digital pH meter of Jenway company number 3520. Temperature and electrical conductivity was measured using digital analyser of Hatch company. To be more acute, all samples experienced three times. Totally, at least 270 jar testing and TOC analysis were carried out for measuring TOC, turbidity and pH.

Method of jar test

The jar test was performed using six Analytic model 1-liter jars. The concentration of the used coagulant was fluctuating between 5 to 80 mg/l and the powdered activated carbon between 10 to 100 mg/l. At the first stage, ferric chloride and poly aluminum chloride were added to water separately as coagulant and the enhanced coagulation was performed in the jar test set. The standard jar test procedure consisted of a rapid mix at 120 rpm for one minute, flocculation at 30 rpm for 20 min. Then the floc was allowed to settle for 30 min based on the recommendation of reputable bodies especially the US environmental protection agency have been adhered to. [5] During the next stage, the powdered activated carbon was

added to each of the coagulant, poly aluminum chloride and ferric chloride.

Method of measuring TOC

After execution of jar test, samples were taken from depths of water of deeper than 5 centimeters inside the jar using a pipette in order to measure the total organic carbon. Due to lack of TOC measuring facilities at the laboratory of Koot Amir water treatment plant, 250 milliliter opaque glass with sandpaper textured caps were used for storing the samples in compliance with standard method for water and wastewater analysis procedure. All the samples were fixed with Acid for pH reduction down to below 2 and kept in the refrigerator at 4° Celsius. Measuring of TOC was done at Tehran Research Institute of Petroleum Industry using TOC analyzer according to 5310 B (combustion- infrared) method published in the standard method book [13].

Statistical Analysis

In order to investigate the effects of different agents on TOC changes, statistical variance method was used. Then Duncan method was applied to analyze data obtained by Anova and Pierson's regression analysis was employed to study the correlation between TOC concentration and other agents such as pH, coagulant dosage and powdered activated carbon dosage. Descriptive statistic method like Duncan analysis was used to examine variable dependence of TOC. Graphs were drawn by MS Excel and the results were statistically significant ($P < 0.05$).

Results

Ferric chloride

The reduction levels of TOC for tested values of pH have been shown in Fig. 1. As reported

in Fig.1 maximum TOC removal was occurred at pH 5.5.

After determining the optimum value for the pH, different concentrations of Ferric chloride were investigated. It can be seen from Fig. 2 that the most reduction in the TOC concentration occurred at 80mg/l and as the dosage of coagulant increased the TOC values were decreased.

In Fig. 3 the increase of powdered activated carbon as coagulant aid is shown at optimum pH value of 5.5 and Ferric chloride concentration of 10 mg/l. As it has been shown in the fig.3 at 60 mg/l of powdered activated carbon, the value of TOC has reached below 2 mg/l, which is the recommended value by EPA [4]. After that, the rate of TOC decreases even more with increasing the concentration of powdered activated carbon.

The studies on effects of flocculation retention time on reduction of total organic carbon have been shown in Fig. 4. The results of statistic tests for flocculating durations of 15, 30, 60, 120 and 180 minutes show that the average values of TOC in durations of 15 to 180 minutes for a remarkable 5% value, do not differ considerably. It is needless to say that the least rate of TOC during flocculation has occurred during the 60 minutes.

Ferric chloride and powdered activated carbon at their optimum concentration and at different pH values of raw water were tested in order to determine the optimum pH value of powdered activated carbon in TOC reduction. The results (Fig. 5) show that the

lowest value of total organic carbon is achieved at pH 5.5.

Poly aluminum chloride

Fig. 6 shows the jar test results for determining the optimum pH value of poly aluminum chloride in TOC reduction. According to the obtained results the most TOC reduction is achieved at pH 6.5.

In order to achieve the optimum dose of poly aluminum chloride, different doses of PACl were tested at the optimum pH of 6.5. According to Fig. 7, the most rate of TOC reduction was achieved at 80 mg/l and to a value of 44% and the least value of TOC reduction was 3.3mg/l and this coagulant did not achieve the value of TOC to the value recommended by EPA (less than 2mg/l) [4], however, poly aluminum chloride was most effective in TOC removal than ferric chloride. As it is shown in Fig 7, TOC values were decreased with increase in coagulant dose.

In Fig.8 the effects of adding powdered activated carbon as a coagulant aid coupled with poly aluminum chloride on optimum concentration and pH have been demonstrated. The jar test results showed that with adding 60mg/l of powdered activated carbon, TOC concentration reached to around 2 mg/l which is the recommended value by EPA. By increasing the amount of powdered activated carbon, TOC was decreased and the least rate of TOC achieved at 100mg/l powdered activated carbon.

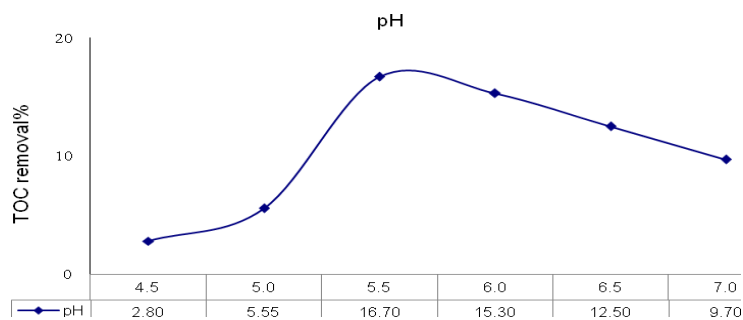


Fig 1: Determining the optimum value of pH for ferric chloride coagulant in elimination of organic carbon ($\text{FeCl}_3=10\text{mg/l}$)

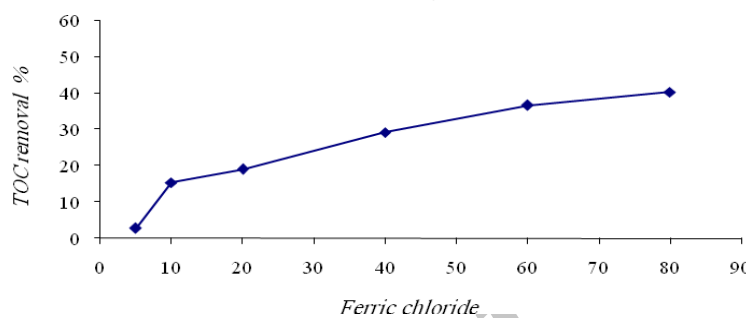


Fig 2: The average percentage reduction of TOC for different concentrations of Ferric chloride (pH=5.5)

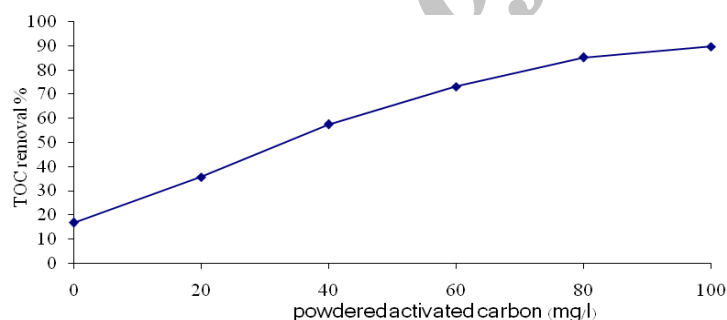


Fig 3: The average percentage reduction in total organic carbon for different concentrations of powdered activated carbon and optimum dose and pH for Ferric chloride ($\text{FeCl}_3=10\text{mg/l}$, pH=5.5)

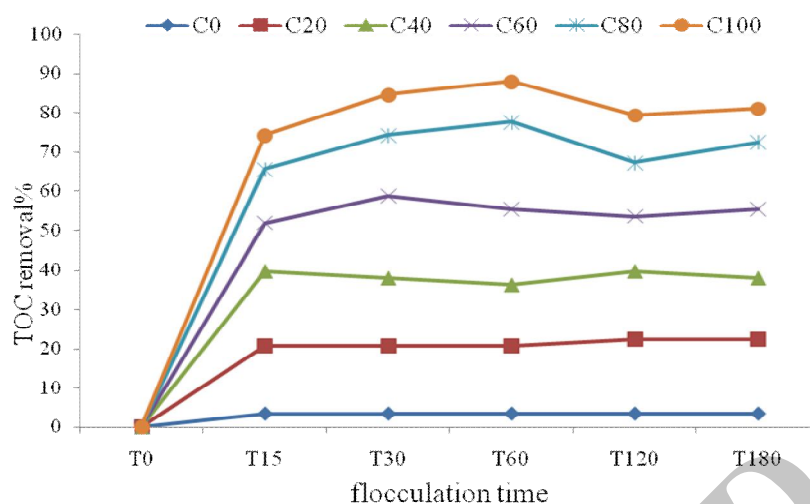


Fig 4: Average percentage reduction of total organic carbon for 15 to 180 min flocculation retention time ($\text{FeCl}_3=10\text{mg/l}$, $\text{pH}=5.5$, various PAC concentrations)

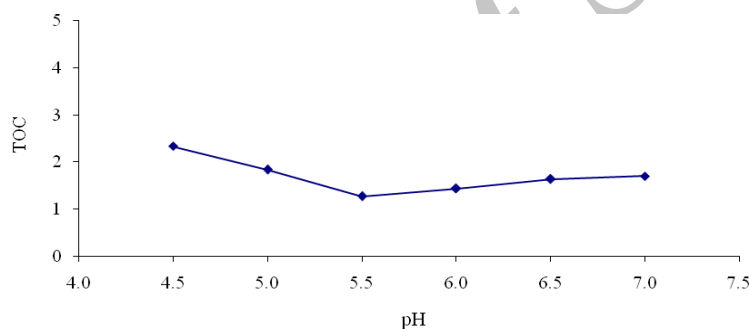


Fig 5: Determining the optimum pH of powdered activated carbon for reduction of total organic carbon

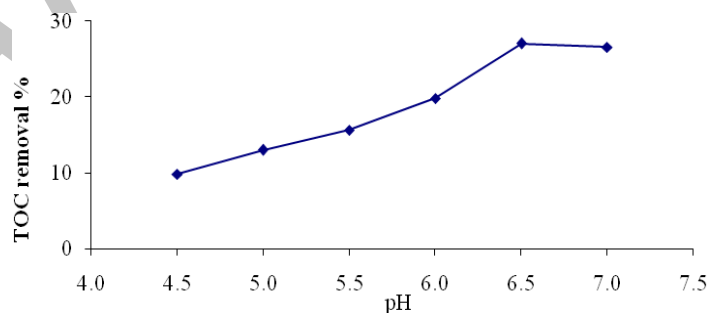


Fig 6: Determining the optimum pH value of poly aluminum chloride in reduction of total organic carbon ($\text{PACl}=20\text{mg/l}$)

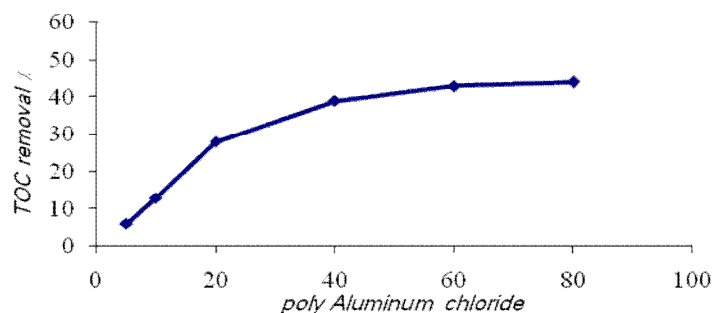


Fig 7: The average percentages of total organic carbon at different concentrations of poly aluminum chloride (pH=6.5)

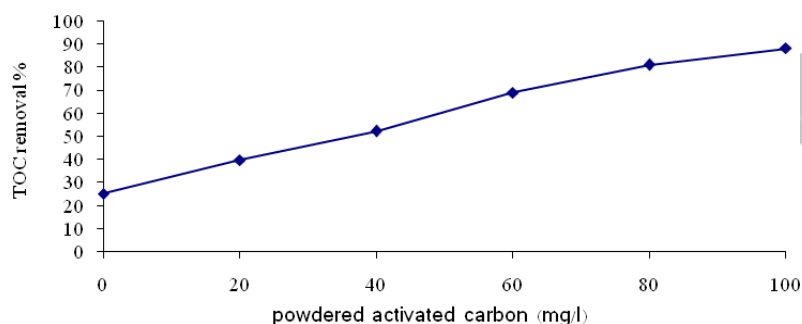


Fig 8: The average percentages of total organic carbon at different concentrations of powdered activated carbon and optimum concentration and pH values of poly aluminum chloride (pH=6.5 , PACI=20mg/l)

Discussion

Measuring the concentration of effluent TOC in Koot Amir water treatment plant demonstrated the poor efficiency of the present treatment processes in elimination of organic matter indicator and this could be due to the fact that this plant was not designed to eliminate organic matters in the first place. Study of determining the most suitable value of pH in application of ferric chloride, the optimum obtained pH was shown to be in the region of 5.5 to 6. Literature findings reported that the pH of coagulation was dependent on the coagulant type and treated water sample [14,15]. It appears with increasing pH from 4.5 to 5.5, contribution of the charge neutralization to NOM removal increased, whereas, after pH 5.5 contribution of the charge neutralization decreased and contribution of the adsorption and entrapment

predominated for NOM removal. Numerous researchers have identified coagulation pH as the parameter having a great effect on achieving optimal organic precursors removal by coagulation process [16,17,18]. Vaezi et al., reported that without any changing in ferric chloride coagulant dose and only with pH reduction from 0.5 to 2 degree, COD reduction was occurred in Karaj river samples up to 90% [19].

Study of best effects of pH or efficiency in elimination of TOC by use of poly aluminum chloride showed an optimum pH of nearly 6.5 to 7. The results mean that polymerized metal coagulants can widen their optimum coagulation pH range when used in water treatment. At very low pH, very few NOM molecules can be accommodated around each metal species because organic matter contains

few anionic sites and NOM is tightly wound. As the pH is raised, more anionic sites are generated by deprotonation of acidic carboxyl groups, and at the same time, the molecules unwind and become more linear, which is the result of repulsion of the many negative charges. At this time, in one hand, cationic metal species interacts electrostatically with anionic NOM to form insoluble charge-neutral product because of charge neutralization. In this way, organic molecules are linked together to form flocs and then can be easily removed from water. As the pH is raised further, say higher than optimum pH value, the increase of pH favored the hydrolysis of metal-ions, and decreased the formation of positively charged ions. Thus, the ability for coagulants to neutralize the negative charge on NOM became small, and NOM could not be removed effectively.[8]

For the application of various quantities of ferric chloride at optimum pH, the most reduction in the TOC concentration occurred at 80mg/l and as the dosage of coagulant increased, the TOC values were decreased. This is due to the fact that in enhanced coagulation, increasing in coagulant concentration causes formation of the settleable flocs that it is effective in organic matter reduction.[4] While the lowest measured TOC value with increasing 80 mg/l ferric chloride has been 4.1 mg/l. Uyak et.al found maximum DOC removal efficiency by 45% with injecting ferric chloride.[7] Also statistic results showed that the average values of TOC in 60 and 80 mg/l and also for 10 and 20 mg/l of ferric chloride at a meaningful level of 5% have not made a meaningful difference. In addition to this meaningful difference, a remarkable level of TOC average values for 5 and 10 mg/l have shown an optimum value of 10 mg/l for ferric

chloride. As mentioned in literature, the lower pH, by increasing the protonation of the NOM and increasing the positive charge of the coagulating species, reduces the coagulant demand and favors the adsorption of humics onto metal hydroxides [5,7,8].

The optimal coagulation of poly aluminum chloride is believed to correspond to prehydrolyzed metal-ion coagulants with the higher cationic charge and larger polymeric species. Thus, coagulating species with high cationic charge and large polymeric species should be performing better than those with low cationic charge and species. Because PACl has higher cationic charge and larger polymer size than ferric chloride at the same pH value, PACl gave higher TOC removal efficiency than ferric chloride at the same conditions[20,21,22] Comparing two coagulants of poly aluminum chloride and ferric chloride in enhanced coagulation resulted in better efficiency of poly aluminum chloride over ferric chloride. Making matters worse using ferric chloride as a coagulant added a great amount of iron to the water.

Study the effects of pH on the elimination efficiency of TOC using powdered activated carbon, has shown the optimum pH range is between 5.5 and 6. As a result at pH of around 7, the rate of TOC reduction was 76%. Taking into consideration the closeness to the natural pH of water, this showed the high efficiency of powdered activated carbon in the natural pH of water. It has also shown that a considerable reduction in total organic carbon concentration is obtained in pH ranges of 5.5 to 7. This is due to the fact that TOC solubility decreased and organic matter adsorption increased in this ambient pH. 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.[5]

Study the effects of flocculation time on TOC reduction using powdered activated carbon and ferric chloride, shown a maximum reduction of TOC in the first hour, but beyond that time even with increasing the flocculation duration to three hours, there is not a noticeable reduction of TOC. The reason is probably related to the fact of quicker adsorption of organic matter of a lighter molecular weight at shorter periods of time [7]. Increasing PAC retention time can significantly decrease the required dose. These findings are in line with the results of Uyak et al., studies in 2006 in disinfection by-products precursors removal by enhanced coagulation and PAC adsorption [7].

Optimizing the coagulation process using powdered activated carbon along with poly aluminum chloride and ferric chloride, increased the rate of TOC elimination. The maximum elimination of total organic carbon by powdered activated carbon as a coagulant aid in application of ferric chloride occurs at 100 mg/l and at a rate of 90%. Uyak et al., studies in removing DBPs precursors by enhanced coagulation and PAC adsorption proved that supplementing enhanced coagulation with PAC adsorption increased the removal of DOC to 76%, and PAC adsorption removed mostly low molecular weight and uncharged NOM substances.[7] Although adding powdered activated carbon enhanced organic matter removal, it caused decreasing coagulant consumption. For the application of poly aluminum chloride, by

increasing the amount of powdered activated carbon, TOC was decreased and the least rate of TOC achieved at 100mg/l powdered activated carbon. Results showed that powdered activated carbon addition resulted in dramatically reduction of coagulant consumption. The reason for this could be due to the fact of the very active surface of powdered carbon and its high capability in absorbing the organic matters [10]. Carriere et al., surveyed using powdered activated carbon in conjunction with poly aluminum chloride in TOC removal and proved that with adding only 11mg/l of powdered activated carbon, TOC removal efficiency was improved and Taste and odor agents was decreased [12]. There are researches indicating negative effects of large-molecule organic matter spread velocity on powdered activated carbon adsorption [1,5,7,12,23]. It was also concluded that PAC effect on removing organic matter may reduce in high molecular weight despite of its capability to remove vast variety of organic matter [5,7]. Since coagulation is effective in removing high molecular weight organic matter and ineffective in low molecular weight organic compounds, the possibility of using PAC as a coagulant aid in removing organic matter in a vast molecular weight range has been proposed [5,7,12,20].

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