Filter backwash water treatment by coagulation: A comparison study by polyaluminium ferric chloride and ferric chloride

Afshin Ebrahimi^a, Mohammad Mehdi Amin^a, Yaghoub Hajizadeh^a, Hamidreza Pourzamani^a, Mohsen Memarzadeh^b, Amir Hossein Mahvi^c, Mokhtar Mahdavi^{d,*}

^aEnvironment Research Center, Research Institute for Primordial Prevention of Non-communicable disease, Isfahan University of Medical Sciences, Isfahan, Iran, & Department of Environmental Health Engineering, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran

^bManager of Isfahan's Water Treatment Plant, Isfahan, Iran

^cCenter for Solid Waste Research, Institute for Environmental Research, Tehran University of Medical Science, Tehran, Iran ^dEnvironment Research Center, Isfahan University of Medical Sciences, Isfahan, Iran, & Student Research Committee and Department of Environmental Health Engineering, School of Health, Isfahan University of Medical Sciences, Isfahan, Iran, Tel. +989394766913, email: ShamaLL6@yahoo.com

Received 9 February 2016; Accepted 27 June 2016

ABSTRACT

Now a day population increase and water shortage in many countries compels them to use reclaimed water. One of the options to combat this matter is to spent filter backwash water (SFBW) which is notably generated in most water treatment plants. There are many techniques for SFBW treatment, and coagulation is the most common process. The aim of this study was to investigate the effective-ness of polyaluminium ferric chloride (PAFCI) and ferric chloride (FeCl₃) for treatment of SFBW at water treatment plant in Isfahan, Iran. The results showed that the optimum pH ranges for the coagulation of both SFBW with PAFCI and FeCl₃ were 7 and 15 mg/L, respectively. The initial turbidity, colour, dissolved organic carbon, UV254 absorbance, aluminum and iron in settled SFBW were 38 NTU, 16 Pt. Co. units, 2.8 mg/L, 0.06 cm⁻¹, 0.2 and 0.15 mg/L, respectively. The removal efficiency by PAFCI for above parameters were 98.68, 100, 39.29, 40.68, 76.5 and 90%, respectively. While, the removal efficiency by FeCl₃ were dose. Also, the quality of treated water by PAFCI was better than raw water entered to the WTP. Therefore, SFBW can be returned to the water treatment plant and solution of a steries of the coagulation of steries of the coagulation of steries of the steries of

Keywords: Water treatment plant; Spent filter backwash water; Water treatment; Coagulation; PAFCl; FeCl₃

1. Introduction

Today, many countries are facing water shortage [1]. Water and wastewater reuse is one of the main options that can be considered as a new water source in regions with water stress [2]. Spent filter backwash water (SFBW) is generated during most water treatment processes. Reclamation

*Corresponding author.

of filter backwash water is a potential source of water that can be useful in communities with water supply shortage. Nonetheless, there are some concern regarding the reuse of SFBW water as a matter of heavy metals, natural organic matters (NOM) and microorganisms.

Surface water conventional treatment processes known as coagulation, flocculation, sedimentation and filtration [3]. During filtration process, the filters become loaded with particles, organic matters and bacteria. Thus, to maintain better performance and treatment efficacy of the filters, backwashing with clean water is conducted periodically to remove

1944-3994 / 1944-3986 © 2017 Desalination Publications. All rights reserved.

contaminants and material that captured in the filter media [4]. The quantity of backwash water typically averages from 2 to 10% of the total water plant production [5]. On average, it accounts to 2.5% of total plant production [6]. The quality of the SFBW depends on the volume and the quality of water used for filter backwashing. This is characterized by concentration of microbial pollution, suspended solids, natural organic matter (NOM) and inorganic materials such as aluminium (Al), iron (Fe) or other heavy metals [7-10]. The concentration of organic matters and other contaminants in SFBW streams has been shown to be significantly higher than raw water [11,12]. So, reclamation and recycling of SFBW could jeopardize the quality of finished water and threaten the health of communities [13]. NOM is responsible for colour and odor in water body [14] and results in the formation of disinfection by-products such as trihalomethanes (THMs), which are harmful to humans [15,16]. On the other hand, Al- or Fe-based coagulants may result in elevated concentrations of residual Al or Fe in finished water [17,18]. Al could increase the risk of Alzheimer's disease. Therefore, it is important to minimize the amount of residual Al in drinking water [19]. To reduce these risks, the SFBW must be treated to ensure human health and environmental protection. The most common process in conventional water treatment plants is coagulation and it is capable for removal of NOM, colloidal particles and even some ionic contaminants [20-22]. Coagulation process can also eliminate dissolved organic carbon by 30-60% [23].

Today, traditional monomeric inorganic coagulants (FeCl₃ and Al₂(SO₄)₃) have changed to inorganic pre-hydrolyzed coagulants such as poly-ferric sulphate (PFS), polyaluminium ferric chloride (PAFCl) and polyaluminium chloride (PACl) [24,25]. These coagulants are less sensitive to low temperatures, require lower dosage, react at wider pH range and are more efficient than traditional coagulants in NOM removal [26–28]. PAFCl is a composite coagulant, which has been developed by the pre-hydrolyzation of AlCl₃ and FeCl₃, or prepared from hydrochloric pickle liquor or blast furnace dust [29].

The main goals of this research were to identify the levels of contaminants in the spent filter backwash water, to investigate the feasibility of reclamation of backwash water after treating by PAFCl and ferric chloride, to evaluate the performance of PAFCl and FeCl₃ on spent filter backwash water and the effect of dosage and pH on the coagulation process.

2. Materials and methods

2.1. Sampling and filter backwash water descriptions

Isfahan water treatment plant treats 12 m³/s of water by coagulation, flocculation, sedimentation and rapid sand filtration processes using PACl as a coagulant. There are 48 filter units in the plant, where their backwash water is considered as a waste. Backwash water was sampled from the plant during annual operation. In winter, autumn and in the rainfall periods the backwash water had a high turbidity amounting about 600 NTU, while in other seasons, it had a turbidity of about 200–250 NTU. Thus, the data of this article focuses on spring and summer, as the quality of water is almost constant during these seasons owing to the lack of rainfall. During backwashing of each filter, some 500 m³ of wastewater was generated. Considering 48 filter with 24 h cleaning interval it accounts for about 2.25% of the raw water entering to the plant. Therefore, during the water treatment process approximately 24000 m³/d of SFBW is generated. In general, 2–10% of the finished water is used for backwashing in each WTP [8,30]. Table 1 presents a summary of quality analysis (the average values of experiments during two sampling period, in spring and summer) for raw, finished, and filter backwash water in the WTP.

2.2. Experimental methods

To investigate the coagulation effects on SFBW, Jar test experiments were conducted using a Phipps and Bird standard jar-test unit consisting of six 2-L square beakers. One liter of the SFBW water was transferred into each beaker. After coagulant addition, the water samples underwent rapid mixing (120 rpm for 2 min), flocculation (40 rpm for 10 min) and settling for 20 min [27]. All experiments were performed at room temperature ($23 \pm 20^{\circ}$ C). For optimum pH, a constant dose of PAFCl and FeCl₃ (7 mg/L) was used separately at different pH (5-9.5). The pH value which led to gain the best results for turbidity and colour removal was adopted as an optimal pH. Optimum dose selection was conducted by applying the identical optimal pH, but, with different doses of PAFC and FeCl₃. Then, the coagulation procedures were carried out for the SFBW using the optimal doses of PAFCl and FeCl₂ under the optimal pH during spring and summer sampling, and the results were recorded.

2.3. Analytical methods

Samples were analyzed for total organic carbon (TOC), dissolved organic carbon (DOC), UV absorbance at 254 nm (UV₂₅₄), residual turbidity, true colour, residual Al and Fe and some other heavy metals such as arsenic, lead and cadmium. All the experiments were conducted according to the standard methods for the examination of water and wastewater. DOC, UV₂₅₄ and true colour were analyzed after filtering the samples by a 0.45 μ m membrane filter.

TOC was analyzed using a Phoenix 8000 system. Turbidity, UV₂₅₄, colour, TDS, EC and pH of the samples were measured by TN-100 (EUTECH) Turbidimeter, DR 5000-HACH LANGE, EC meter SENSION5 (HACH LANGE) and pH-meter model CG 824, respectively.

2.4. Coagulants and reagents

Both FeCl₃·6H₂O and PAFCl were used as coagulants in this study. The commercial products were provided by chemical suppliers from Merck and China Company. PAFCl is a commercial grade product with a composition of 28% Al₂O₃, 1.84% Fe2O3 and basicity of about 81.22%. For preparation of PAFCl stock solution, 1 gr of the PAFCl was dissolved in 1 liter distilled water to achieve a concentration of 1 g/L. For 1 g/L FeCl₃ stock solution, 1.662 gr of FeCl₃·6H₂O was dissolved in 1 liter distilled water. pH adjustment of the samples were conducted with 0.1 M HCl or 0.1 M NaOH solutions. 322

3. Results and discussion

3.1. Raw water characteristics

Typical characteristics of the SFBW samples are summarized in Table 1. Results showed that pH of the SFBW was weakly alkaline. Total amount of its DOC varied between 8 and 12 mg/L during the study period. Differences in the amount of turbidity, DOC and other parameters among the sampling period were nearly constant. The quality of raw water was almost constant as well due to the lack of rainfall and good quality of water resources. Furthermore, raw water is introduced to pre-chlorination and pre-ozonation processes. In this regard, the majority of organic matter content is oxidized resulting in low UV₂₅₄ absorbance, and DOC and colour are reduced in treated water and backwash effluent. As shown in Table 1, the average values of DOC $_{54}$ absorbance in raw water were 2 (±0.28) mg/L and UV and 0.052^{-1} (±0.03) cm⁻¹, respectively, while in the finished water reached to 1.1 (± 0.14) mg/L and 0.034 (± 0.001) cm⁻¹, respectively.

3.2. Effect of coagulation on turbidity and colour removal

3.2.1. Optimum pH selection

In coagulation process the optimum pH is an important parameter for proper coagulation as it affects the surface charge of colloids, organic matters and the charge of NOM functional group [31]. Also, the pH plays an important role on stabilization and destabilization mechanism especially on surface charge neutralization. Most particles in water have electrically charged surfaces that are usually negative [32]. At very low pH there are more concentration of H⁺ that affects the surface charge of particles and organic matter. By addition of coagulant, positive cation like Fe³⁺ or Al³⁺ react with negative charge particles in water body. At very acidic condition these two positive charges repel each other, so the results of coagulation are not very well. This drawback occurs at high pH condition as well. When pH increases, the surface charge becomes increasingly negative owing to the presence of OH-, thus this situation causes destabilization of particles and subsequently deteriorates coagulation. In fact, optimum pH is a value at which these negative and positive charges are shifted to a situation that can be attracted by positively charged oxides and hydroxides of Al or Fe. For this reasons, water with high alkalinity may require high coagulant consumption to depress the pH value favorable for coagulation [33]. As pH increases, carboxyl groups of humic substances will loss protons, so, they become more ionized and the positive charges on metal coagulants will decrease. Consequently, higher coagulant dosages will be required at higher pH values.

The degree of turbidity removal by PAFCl and FeCl₃ was investigated separately at different pH from 5 to 9.5 (with an interval value of 0.5) and constant dose of 7 mg/L. The results indicated that high removal efficiencies (to obtain turbidities ≤ 0.8 NTU) for PAFCl was at pH rage of 5–9 and that for FeCl₃ was at pH rage of 6.5–8.5 (Fig. 1). Performance of the PAFCl and FeCl₃ at pH values upper than 9 and 8.5, respectively, was dramatically alleviated. The results achieved by PAFCl are in accordance with the previous study [27]. At pH values greater than 9 the turbidity removal efficiency was decreased due to the hydrolysis of coagulants into negatively charged precipitates. In this study, the SFBW had a mean pH of about 8.35. On the other hand, both coagulants showed good efficiencies at pH

Table 1

Average quality of raw water, SFBW and sand filter effluent in summer and spring

Parameter	Units	Raw water	Filter backwash	Filter backwash water after primary sedimentation	Filters effluent
Turbidity	NTU	7 (±0.7)	275.5 (±2.1)	38 (±1.4)	0.25 (±0.01)
Colour	Pt. Co. units	11 (±1.4)	35 (±2.8)	16 (±1.4)	0
EC	µs/cm	333 (±2.8)	335 (±1.4)	344 (±2.8)	334 (±1.4)
TDS	mg/L	165 (±2.8)	167 (±1.4)	166 (±1.41)	166 (±1.41)
pН		8.22 (±0.02)	8.4 (±0.1)	8.35 (±0.07)	8.2 (±0.02)
Alkalinity	mg/L	132 (±1.4)	150 (±2.8)	144 (±1.41)	126 (±1.4)
Fe	mg/L	0.1 (±0.01)	0.7 (±0.002)	0.15 (±0.02)	0.02 (±0.002)
Al	mg/L	0.035 (±0.01)	0.31 (±0.002)	0.2 (±0.02)	0.049 (±0.002)
Arsenic	µg/L	0	0	0	0
Lead	µg/L	2 (±0.28)	16 (±0.002)	13 (±1.4)	0.5 (±0.002)
Cadmium	µg/L	0.43 (±0.028)	0.7 (±0.002)	0.52 (±0.02)	0.38 (±0.002)
Temperature	°C	23 ± 2	23 ± 2	23 ± 2	23 ± 2
Sludge volume	ml/L	negligible	12 (±1.4)	negligible	0
UVA-254 nm	cm ⁻¹	0.052 (±0.03)	0.18 (±0.01)	0.06 (±0.014)	0.034 (±0.001)
DOC	mg/L	2 (±0.28)	10 (±2.8)	2.8 (±0.14)	1.1 (±0.14)
TOC	mg/L	2.2 (±0.14)	11 (±2.8)	3 (±0.14)	1.2 (±0.14)



Fig. 1. Optimum pH selection for turbidity removal by FeCl_3 and PAFCl at 7 mg/L dosage.

range of 6 to 8.5. The difference between efficiency removal at optimum pH (8) and at neutral pH of SFBW was about 0.03 NTU which is negligible. Therefore, to avoid any chemical addition, pH value of 8.35 was selected as an optimum pH.

By comparison between the two coagulants, it was concluded that at constant dose and pH, PAFCl showed better turbidity removal efficiency than FeCl_3 at pH 8.5 (98.3% vs 97%).

Optimum colour removal was observed at pH range of 7 to 8.5. At this pH the colour removal efficiency for PAFCl and FeCl₃ was 100% and 93.8%, respectively. Fig. 2 shows when the pH is above 8.5 the colour removal efficiency is decreased, and at pH range of 7 to 8.5 they have constant effect in colour removal. Traditional coagulants like ferric salts undergo rapid and uncontrolled hydrolysis reactions upon their addition to water, so they form a series of products such as monomers, oligomers and polymeric hydroxyl complexes.

Alkalinity has a very useful effect on the pre-hydrolyzed coagulants to produce Fe- and Al-species that are quite stable and very efficient in NOM and particle removal. Therefore, they are hardly affected by alkalinity and pH in water [34]. For these reasons, by increasing the pH value from 7 to 9, PAFCl shows a better efficiency than FeCl_a.

3.2.2. Optimum dose selection

Coagulation was examined with a various doses of PAFCl and $FeCl_{37}$ from 1 to 40 mg/L in 11 steps. Experi-



Fig. 2. Optimum pH selection for colour removal by FeCl₃ and PAFCl at constant dose (7 mg/L) and initial colour of 16 Pt. Co. units.

ments were conducted two times in summer and spring for both coagulants, and the results are presented in Figs. 3 and 4. For PAFCl, by increasing the dose up to 13 mg/L, the removal efficiency for turbidity and colour were increased. When the dose increased above 13 mg/L, it worsened the removal efficiency due to re-stabilizing the colloidal suspension. As shown in Figs. 3 and 4, PAFCl has constant performance for turbidity and colour removal at 7 to 13 mg/L dose. At these doses, the residual turbidity and colour are reduced to 0.5 NTU and 0 Pt. Co. units. In comparison, by increasing the dose of FeCl₂ up to15 or 20 mg/L the removal efficiencies for turbidity and colour were increased and then decreased in the upper tested dosage, whereas at 40 mg/L the residual turbidity and colour reached to 0.9NTU and 5 Pt. Co. units. Thus, from Figs. 3 and 4 it was concluded that this coagulant shows a good efficiency at 15 to 20 mg/L. To prevent additional chemical consumption and sludge production, 15 mg/L was selected as an optimum dose. At this dose, residual turbidity was reduced down to 0.51 NTU.

In comparison, PAFCl removes more turbidity and colour at lower dosage than FeCl₃ and it is affected quickly by over dosing. Applying 5 mg/L of each coagulant, a residual turbidity of 0.7 NTU for PAFCl and 1 NTU for FeCl₃ were achieved. Also, residual colour was reduced to 1 Pt. Co. units and 2 Pt. Co. units, respectively. Removal efficiencies of turbidity and colour worsened at PAFCl doses over 13 mg/L and at FeCl₃ doses over 25 to 30 mg/L.

At low doses, the coagulant was not able to compress the double layer of the colloid particles or to bind the colloid particles to form bridging. As coagulant dosages were exceeded the charge neutralization requirement, the for-



Fig. 3. Optimal dose selection for turbidity removal by ${\rm FeCl}_3$ and PAFCl on pH 8.35.



Fig. 4. Optimal dose selection for colour removal by FeCl_3 and PAFCl, with initial colour of 16 Pt. Co. units.

mation of precipitates led to the relatively rapid formation of visible flocs. This process is usually called sweep floc or enmeshment in precipitate. Also, at a higher dosage of the coagulant, there are no enough negative charges on the surfaces of particles or colloidal matter to stabilize the positively charged particles. Subsequently, the precipitate suspension becomes unstable and it affects the quality of treated water. Apparently, 5 mg/L of PAFCl is adequate to overbear double layer compression, but for FeCl₂ it requires 15 mg/L. One reason for this matter is related to the PAFCl positively charge property. In fact, the higher charge density of PAFCl species results in a decrease in the coagulant dose. Previous study showed that pre-hydrolyzed coagulant like PACl contains stable preformed aluminium species that are more effective at charge neutralization than conventional coagulants (like FeCl₂) due to a higher charge density [35]. Increasing the coagulants doses resulted in a decrease in turbidity removal which can be attributed to charge reversal and destabilization of colloidal particles due to coagulant overdosing [36]. So, the superior performance of PAFCl compared to FeCl, is attributed to its wider working pH range and lower dose requirement to achieve equivalent treatment efficiency [37].

In this study, we found that the required dosage of PAFCl was 53.3% less than that of FeCl₃. Halvadiya et al. reported same results for PACl and alum [38]. In fact, SFBW contains destabilized solids that play two important roles in coagulation efficiency. First of all, it increases the number of collision sites that enhances physical removal of colloids and solids, and secondly, it contains neutralized sites that by applying a little amount of coagulants they get charged and act as new flocs for coagulation and adsorption of turbidity.

3.3. Effect of coagulation condition on organic matter removal

3.3.1. Optimum pH selection:

The effect of pH (5 to 9.5) on the removal of organic matter was studied by reduction of UV₂₅₄ absorbance with PAFCl and FeCl₃ with dosage of 7 mg/L (Fig 5). According to Table 1, DOC and UV₂₄₅ absorbance of the SFBW after primary sedimentation were 2.8 mg/L and 0.06 cm⁻¹, respectively. Fig. 5 show that pH has a significant effect on the coagulation efficiency as UV₂₅₄ absorbance reduction. For both coagulants, it was observed that in low pH they had more efficiency. At pH 5, reduction efficiency of PAFCl and



Fig. 5. Optimum pH selection for UV254 absorbance reduction by FeCl₃ and PAFCl at 7 mg/L dosage.

FeCl₃ for UV₂₅₄ absorbance was 50 and 41.6%, respectively. However, by increasing the pH, two coagulants showed different behaviors which were mainly related to their various chemical properties and chemical interactions. It is interesting that for PAFCl, the reduction of UV₂₅₄ absorbance was constant at wider range than for FeCl₃. For FeCl₃ by a unit change in the pH, it affects the level of UV₂₅₄ absorbance. For example, at pH 5 to 7, PAFCl had constant reduction efficiency of about 50% and at pH 7.5 to 8.5 reductions was about 45%. In contrast, for FeCl₃, reductions were decreased from 41.6% at pH 5 to 23.3% at pH 8.5.

Previous studies showed that the traditional coagulants like FeCl, provide optimal organic matter removal at low pH values (pH < 6.0) [39]. Electrostatic interactions occur since molecules still bear some charged functional groups. The suppression of the negative charge ions at low pH values with H⁺ or Al³⁺ and Fe³⁺ leads to formation of aggregates [40] and subsequence negative organic matter removal take place due to charge neutralization by the monomeric Fe or Al species which exists under that pH. The results showed that optimum pH for the removal of organic matter with PAFCI was 5 to 7 but at pH from 7.5 to 8.5, no significant difference was observed. Thus, to avoid chemical addition for pH adjustment, pH 8-8.5 (pH of sample) was selected as an optimum pH. FeCl₃ removed more organic matter at low pH, but, at this pH, turbidity removal was poor. To obtain better removal for both turbidity and organic matter, pH 8–8.5 was selected as an optimum for FeCl₂ as well. Because of both Al(OH)₃ and Fe(OH)₃ species which exist at higher pH (7.5-8.5), both charge neutralization and adsorption could be responsible for organic matter removal [41].

It is obvious that alkalinity and pH affect the interactions of coagulant and organic matter removal. For traditional coagulants, organic matter is removed better at low pH. In high alkaline water, removal is generally obtained by acidifying the raw water or by applying increased dose of coagulants [42]. By applying pre-hydrolyzed coagulants such as PAFCI, this problem can be handled.

3.3.2. Optimum dose selection

To study the effect of the coagulants dose on the removal of organic matter, coagulation was performed with different doses of the coagulants from 1 to 40 mg/L. Fig. 6 shows the changes in UV₂₅₄ absorbance during the coagulation process by PAFCl and FeCl₃. The efficiency of NOM removal (UV₂₅₄ absorbance reduction) increases with an increase in the coagulants dose. For both coagulants low doses up to 4



Fig. 6. Optimal dose selection for organic matter removal (UV254 absorbance reduction) by FeCl₃ and PAFCl on pH 8.35.

mg/L did not properly remove organic matter. The coagulation does not take place at low dosage, because the electrostatic repulsion between negatively charged functional groups within the organic molecules does not allow them to approach together. In this study, the two coagulants showed different behavior for organic matter removal at various doses. Both coagulants had maximum removal efficiencies of organic matter at 40 mg/L, but this dose is not optimum for turbidity removal. At a dosage of 7 to 13 mg/L PAFCl a removal efficiency of 40% was attained, but for FeCl₃ with the same dose was 30%. To achieve 40% removal efficiency by FeCl₂, it is necessary to apply 20 mg/L of it. Therefore, the suitable dose for PAFCl and FeCl, was selected 7 and 15 mg/L, respectively (Fig. 6). The average values obtained for TOC and DOC at optimum doses and pH for PAFCl were 1.43 (± 0.05) and 1.3 (± 0.02) mg/L, and for FeCl₂ were 1.6 (± 0.14) and 1.45 (± 0.01) mg/L, respectively.

At lower pH, hydrogen ions could dominate the metal hydrolysis products for organic ligands. Thus, the amount of unsatisfied organic ligands decreases and the organic matter can be removed more efficiently [34]. In this study, pH of samples was 8.35, so, the amount of H⁺ is lower than OH⁻. It can be concluded that at high pH condition, another mechanisms may control the organic matter removal. For PAFCl, increases in dose didn't reduce the pH very much, but it is obvious that pre-hydrolyzed coagulants had more positive charge in comparison to the traditional coagulants. So, positively charged PAFCl flocs can adsorb negatively charged organic matter. In contrast, for FeCl₂, it is necessary to add more doses to get enough positive charges. The aggregation mechanisms which takes place and NOM are removed include charge neutralization, entrapment, adsorption and complexation with coagulant ions into insoluble masses [43-45].

Another mechanism that improves PAFCl efficiency is adsorption of negatively charged organic matter on flocs. PAFCl has both Al and Fe ions, so, it uses the both properties of Fe(OH)₃ and Al(OH)₃ complex. Pre-hydrolyzed coagulant has been found to be superior compared to the traditional coagulants for particulate or organic matter removal under some conditions in which significant amounts of high-charged polynuclear aluminium and iron hydrolysis products are present [46–47]. In case of FeCl₃, the removal of DOC under relatively higher coagulant concentration (15 mg/L) could result by the sweep flocculation process.

Another important parameter for study of coagulation performance and controlling the quality of treated water is specific UV absorbance (SUVA) which is defined as the UV absorbance of a sample at 254 nm divided by the DOC concentration of the sample. This ratio describes the nature of natural organic matter (NOM) in the water sample in terms of hydrophobicity and hydrophilicity. Also, it has been reported that there is a good correlation between the SUVA value and THMs formation potential [48]. Water with SUVA≥4 indicates that the NOM composition is dominated by humic substances. Water with SUVA between 2 and 4 indicates that it contains a mixture of humic and non-humic substances and water with SUVA ≤2 indicates that the NOM composition is dominated by non-humic substance [33]. The amounts of SUVA in raw water entering to the Isfahan WTP, filters output and SFBW before and after coagulation are shown in Table 2.

The SUVA value in the raw water was 2.6 and after coagulation with PACl it reached to 3. It was concluded that water contains a mixture of humic and non-humic substances. In addition, SUVA of SFBW before and after coagulation by PAFCl and FeCl₃ were 2.1, 2.7 and 2.6, respectively. The results indicate that the raw water samples were comprised of a varied mixture of hydrophobic and hydrophilic organic compounds [4].

The results presented in Table 2 show that by applying PAFCl and FeCl₃, the DOC content was reduced by 53 and 48%, respectively. Based on the SUVA levels of the water in Table 2, it is interesting to say that FeCl₃ has to some extent higher affinities to hydrophobic humic acids in comparison to PAFCl. SUVA value for PAFCl reached from 2.1 to 2.7 after coagulation and for FeCl₃ it reached from 2.1 to 2.6, which are in accordance with Yan et al results [49].

3.4. Effects of coagulant type and dose on pH and alkalinity

The pH and alkalinity affect the performance of coagulants significantly. Fig. 7 shows the changes of pH in treated water after coagulation with various doses of PAFCl and



Fig. 7. pH changes after coagulation with various doses of PAF-Cl and FeCl_3 (initial pH was 8.35).

Table 2

The average amounts of SUVA in Isfahan raw water, filters output and SFBW before and after coagulation

0			1	0	
Parameters	Raw water	Filter output*	SFBW after settling	Coagulation by PAFCl	Coagulation by FeCl ₃
UV ₂₅₄ (cm ⁻¹)	0.052 (±0.03)	0.034 (±0.001)	0.06 (±0.014)	0.036 (±0.001)	0.038 (±0.001)
DOC (mg/L)	2 (±0.28)	1.1 (±0.14)	2.8 (±0.14)	1.3 (±0.02)	1.45 (±0.01)
SUVA	2.6	3	2.1	2.7	2.6
TOC (mg/L)	2.2 (±0.14)	1.2 (±0.14)	3 (±0.14)	1.43 (±0.05)	1.6 (±0.14)

*In Isfahan WTP Coagulation is carried out using PACl

FeCl₃ during two experiments in summer and spring. By increasing the dose of both coagulants, pH was decreased. Also, it can be concluded that pH variation range for PAFCl is lower than FeCl₂. The change of the solution pH may be related to the metal coagulants property that are commonly acidic and can consume water alkalinity, depending on their type. Coagulation in low alkaline water may consume all of the available alkalinity and depress the pH values too low for effective treatment [34]. In this study, the SFBW had high alkalinity, so, the reduction of pH or alkalinity by coagulants didn't notably affect them and there was no need for pH or alkalinity adjustment. High alkaline water may require high coagulant addition to depress the pH values favorable for coagulation. However, for pre-hydrolyzed coagulant, it is not very important, because PAFCl performance is steady at wide range of pH. From the results, it can be seen that application of PAFCI has a lower impact on pH and alkalinity reduction compared to FeCl₂.

3.5. Effect of coagulation on metals removal from the SFBW

One of the major concerns regarding SFBW reuse or treatment is heavy metals. During the water treatment processes, metals are removed by various mechanisms such as adsorption, precipitation and enmeshment. Most of these metals are concentrated in the clarifiers or top surface of filters. So, most of them enters to SFBW during filter's backwash. High concentrations of aluminium in drinking water are also of concern because of its potential adverse health effects such as causing Alzheimer disease [50,51]. Determination of Al concentration is very important from the standpoint of human health. In this study, the concentration of iron, aluminium, arsenic, lead and cadmium in raw water were 0.1 mg/L, 0.035 mg/L, zero µg/L, 2 µg/L and $0.43 \,\mu\text{g/L}$, respectively. In the filters output they amounted 0.02 mg/L, 0.049 mg/L, zero µg/L, 0.5 µg/L and 0.38 µg/L, respectively (Table 1). It can be concluded that concentration of all the above metals in raw and treated water were low and met WHO guideline. Only the concentration of aluminium in treated water was increased due to the use of PACl as coagulant in the process.

Table 3 shows the metals concentration in the SFBW before and after coagulation with both PAFCl and FeCl₃ at optimum dose. The concentrations of all tested metals in treated water with PAFCl were lower than those in treated water with FeCl₃. The use of coagulant for water treatment often leads to higher concentrations of aluminium or iron in

the treated water than in the raw water itself. In this study, the concentration of aluminium and iron in treated water by PAFCl reduced to 0.047 mg/L and 0.015 mg/L from 0.2 mg/L and 0.15 mg/L, respectively. Also, for FeCl, the concentration of aluminium reduced from 0.2 mg/L to 0.051 mg/L and the iron level increased from 0.15 mg/L to 0.2 mg/L. PAFCl produces flocs that have good adsorption properties, so they adsorbed heavy metals especially Al and Fe better than FeCl₃. On the other hand, large amounts of Al and Fe in the SFBW were enmeshed in tiny flocs that escape from primary sedimentation and PAFCl removes this particulate better than FeCl₃. Another parameter that affects Al and Fe concentration is release properties of the coagulant. From Table 3 it can be concluded that FeCl₂ increases Fe concentration, but PAFCl reduces both Al and Fe concentrations. This is an interesting property of per-polymerized coagulants in comparison to traditional coagulant.

The residual aluminium or iron is low in neutral pH due to the formation of amorphous precipitates like Al $(OH)_3$ or Fe $(OH)_3$ which is then removed by sedimentation. Therefore, residual aluminium or iron drops significantly around neutral pH [52]. In this study the pH and alkalinity of water were 8.35 and 144 mg/L as CaCO₃. Thus, the formation of amorphous precipitates keeps them insoluble. On the other hand, co-precipitation of organic contaminants with metal hydroxides by precipitation and adsorption property of aluminium- or iron-based flocs can reduce metals in treated water [53]. The maximum recommended range of aluminium in drinking water by WHO is 0.05–0.2 mg/L and by Iranian drinking water standard is 0.1–0.2 mg/L.

3. 6. Sludge production

During coagulation process, coagulants produce sludge in the form of metal hydroxide which removes colour and colloidal matter from raw water. Of course, not all inorganic coagulants behave in the same way, but pre-hydrolyzed coagulants such as PACl and PAFCl produce less sludge than alum or ferric chloride with equivalent doses [54]. Fig. 8 shows the sludge production by PAFCl and FeCl₃ at different dose. The sludge volumes were measured using Imhoff cones. Coagulation with 7, 10, 15 and 40 mg/L of PAFCl produced 2.4, 3, 4.1 and 8.9 ml/L of sludge, respectively.

To investigate the effect of pH on sludge production, coagulation was conducted with constant dose of PAFCl and FeCl₃ (7mg/L) at various pH (Fig. 9). It is clear that in low pH; the solubility of Al and Fe is high. So, in this

Table 3

The average amounts of metals and heavy metals concentration in SFBW before and after coagulation by PAFCl and FeCl_3 during two seasons

Metals and heavy metals	SFBW before coagulation	SFBW after coagulation with PAFCl	SFBW after coagulation with FeCl3	WHO guideline	Iran standard
Fe (mg/L)	0.15 (±0.02)	0.015 (±0.001)	0.2 (±0.028)	0.3	0.3
Al (mg/L)	0.2 (±0.02)	0.047 (±0.002)	0.051 (±0.0141)	0.2	0.1-0.2
Arsenic (µ/L)	0	0	0	10	10
Lead (μ/L)	13 (±1.4)	10 (±1.41)	12 (±1.4)	10	10
Cadmium (µ/L)	0.52 (±0.02)	0.35 (±0.028)	0.38 (±0.028)	3	3



Fig. 8. Sludge production by PAFCl and FeCl3 at various doses.



Fig. 9. Sludge production with constant dose of PAFCl and ${\rm FeCl}_3$ (7 mg/L) at various pH.

condition the chance for aluminum and iron hydroxide precipitate is very low. As the pH increases and hydrolysis proceeds, micro crystals and the amorphous metal hydroxide precipitates are formed. These products can adsorb many types of organic matter or trap colloidal matter. Eventually, they form a precipitate with higher volume and weight. Visual observation revealed that flocs forming were larger at higher pH values than at optimum or low pH values.

3.7. Effect of blending

Previous studies have shown that blending 10% of untreated SFBW with raw water prior to coagulation did not affect treated water quality and in some cases it improved removal efficiency [4,55,56]. In this study, by coagulation of the SFBW we attained good quality which allows us to use more portion of SFBW. All tested parameters like turbidity, colour, organic matter and heavy metals in treated SFBW by coagulation were lower than raw water, so all the treated SFBW can be recycled to the WTP entrance. Also, the measured metals in the treated water had low concentrations that meet WHO guideline. Only for FeCl₃ the concentration of Fe in the treated water was more than raw water. By Eq. (1) we can determine the amount of treated water that can be blended with raw water without notable effect on quality of treated water.

$$C_{mix} = \frac{(Q_{raw} \times C_{raw}) + (Q_{bw} \times C_{bw})}{(Q_{raw} + Q_{bw})}$$
(1)

where C_{mix} = concentration of heavy metals, DOC or other parameters in combined water; Q_{naw} = flow rate of raw water; C_{naw} = concentration of heavy metals, DOC or other parameters in raw water; Q_{bw} = flow rate of backwash water; C_{bw} = concentration of heavy metals, DOC or other parameters in backwash water

4. Conclusions

A. Ebrahimi et al. / Desalination and Water Treatment 66 (2017) 320-329

- (1) Compared to FeCl₃, PAFCl showed superior performance in most cases during coagulation & flocculation on SFBW treatment. At a dosage of 7 mg/L, PACl resulted in an optimum removal efficiency of turbidity, colour, DOC and UV254 of 98.68%, 100%, 39.29%, and 40.68%, respectively. Also optimum removal efficiencies of FeCl₃ for turbidity, colour, DOC and UV₂₅₄ absorbance were 98.66, 100, 35.71 and 35.59, respectively. The optimum pH for turbidity and colour removal by both the coagulants was 6 to 8.5. Thus, natural pH of SFBW (8.35) was selected as an optimum pH.
- (2) Both the coagulants remove the turbidity and colour almost with similar efficacy, but PAFC needs very low dose in comparison to FeCl₃. For the two coagulants, organic matter removal was very good at acidic pH. However, in comparison to FeCl₃, PAFCl showed a better efficiency for organic matter removal especially in pH above 7. So, its removal efficiency was not significantly affected by pH variation. As SFBW contains tiny hydroxide precipitates, it assists the coagulation process by increasing the volume of flocs and impaction between flocs and contaminants in water body. Subsequently it improves coagulation through sweep coagulation mechanisms.
- (3) The results of this research suggest that PAFCl removes heavy metals better than FeCl₃. PAFCl does not add any Fe or Al to treated water, but FeCl₃ releases Fe into the treated water. One reason for very good efficiency of heavy metals removal by both the coagulant is that the majority of heavy metals removed by main water treatment processes were trapped by flocs that remain on filter surface. So, by application of coagulation, flocculation, sedimentation and subsequent removal of tiny flocs that contains heavy metals, large portion of the metals is removed during SFBW treatment.
- (4) It can be concluded that regardless of the raw water quality we can treat SFBW by coagulation as an important pretreatment method for reuse or reclamation of water especially communities facing water shortage or scarcity. Coagulation with PAFCl is an efficient process for SFBW, compared to FeCl_a.

Acknowledgements

The authors acknowledge the financial support of Isfahan University of Medical Science for this research as a Ph.D thesis (grant No: 394281).The authors also gratefully acknowledge the very kind editorial board and reviewers of Desalination and Water Treatment journal, for their technical comments and assistances.

References

- [1] AWWA, Water Reuse Rates and Charges Survey Results, Denver, 2008.
- [2] F.M. Moghadam, M. Mahdavi, A. Ebrahimi, H.R. Tashauoei, A.H. Mahvi, Feasibility study of wastewater reuse for irrigation in Isfahan, Iran, Middle East. J. Sci. Res., 23 (2015) 2366–2373.
- [3] EPA, Implementation Guidance for the Filter Backwash Recycling Rule, Office of Water, 2004.
- [4] N.J. McCormick, M. Porter, M.E. Walsh, Disinfection by-products in filter backwash water: Implications to water quality in recycle designs, Water. Res., 44 (2010) 4581–4589.
- [5] P.W. Frederick, Regulating filter backwash water, J. AWWA., 89 (1997) 14.
- [6] A. Nasser, Z. Huberman, L. Dean, F. Bonner, A. Adin, Coagulation as a pretreatment of SFBW for membrane filtration, Water. Sci. Technol: Wat. Supp., 2 (2002) 301–306.
- [7] D.A. Cornwell, Treatment options for Giardia, Cryptosporidium and other contaminants in recycled backwash water, Amr. Water. Works. Assos., 2001.
- [8] J.C. Bourgeois, M.E. Walsh, G.A. Gagnon, Comparison of process options for treatment of water treatment residual streams, J. Environ. Eng. Sci., 3 (2004) 408–416.
- [9] M.E. Walsh, G.A. Gagnon, Blending membrane treated WTP waste residuals with finished water: impacts to water quality and biofilm formation, J. Wat. Supp. Res & Techno – AQUA., 55 (2006) 321–334.
- [10] M.E. Walsh, G.A. Gagnon, Z. Alam, R.C. Andrews, Biostability and disinfectant by-product formation in drinking water blended with UF treated filter backwash water, Water. Res., 42 (2008) 2135–2145.
- [11] A. Gottfried, A.D. Shepard, K. Hardiman, M.E. Walsh, Impact of recycling filter backwash water on organic removal in coagulation sedimentation processes, Water. Res., 42 (2008) 4683– 4691.
- [12] J.K. Edzwald, J.E. Tobiason, M.B. Kelley, H.J. Dunn, P.B. Galant, G.S. Kaminski, Impacts of Filter Backwash Recycle on Clarification and Filtration, Amr. Wat. Work. Assoc. Res. Found, Denver, USA, 2001.
- [13] US-EPA, Filter Backwash Recycling Rule Technical Guidance Manual, 2002.
- [14] G.A. Edwards, A. Amirtharajah, Removing colour caused by humic acids, J. AWWA, 77 (1985) 50–57.
- [15] M.C. Kavanaugh, Modified coagulation for improved removal of trihalomethane precursors, J. AWWA, 70 (1978) 613–620.
- [16] H. Gallard, U.V. Gunten, Chlorination of natural organic matter: kinetics of chlorination and of THM formation, Water. Res., 36 (2002) 65–74.
- [17] J.E.V. Benschoten, J.K. Edzwald, Measuring aluminium during water treatment: methodology and application, J. AWWA, (1990) 71.
- [18] P.T. Srinivasan, T. Viraraghavan, K.S. Surbramanian, Alumnium in drinking water: an overview, Wat. SA., 25 (1999) 47–56.
- [19] J. Tomperi, M. Pelo, K. Leiviska, Predicting the residual aluminium level in water treatment process, Drink. Wat. Eng. Sci., 6 (2013) 39–46.
- [20] K.J. Choi, S.G. Kim, S.H. Kim, Removal of antibiotics by coagulation and granular activated carbon filtration, J. Hazard. Mater., 151 (2008) 38–43.
- [21] A. Ebrahimi, E. Taheri, A. Pashaee, M. Mahdavi, Effectiveness of poly aluminium ferric chloride for turbidity and colour removal from low to medium turbid water, Arch. Hygi. Sci., 3 (2014) 175–183.
- [22] X. Wu, X. Tan, S. Yang, T. Wen, H. Guo, X. Wang, A. Xu, Coexistence of adsorption and coagulation processes of both arsenate and NOM from contaminated groundwater by nanocrystallined Mg/Al layered double hydroxides, Water. Res., 47 (2013) 4159–4168.
- [23] D. Ghernaout, The hydrophilic/hydrophobic ratio vs. dissolved organics removal by coagulation – A review, J. King. Saud. Univ. Sci., 26 (2014) 169–180.
- [24] B. Gao, Q. Yue, Effect of ratio and OH⁻/Al³⁺ value on the characterization of coagulant poly-aluminium-chloride-sulfate

(PACS) and its coagulation performance in water treatment, J. Chemos., 61 (2005) 579–584.

- [25] A. Zouboulis, P. Moussas, Polyferric silicate sulphate (PFSiS): preparation, characterisation and coagulation behaviour, Desalination, 224 (2008) 307–316.
 [26] A. Zouboulis, N. Tzoupanos, Polyaluminium silicate chlophanetication.
- [26] A. Zouboulis, N. Tzoupanos, Polyaluminium silicate chloride—a systematic study for the preparation and application of an efficient coagulant for water or wastewater treatment, J. Hazard. Mater., 162 (2009) 1379–1389.
- [27] A. Ebrahimi, E. Taheri, A. Pashaee, M. Mahdavi, The effectiveness of polyaluminium ferric chloride (PAFCI) for turbidity and colour removal from Isfahan raw water, Desal. Wat. Treat., 55 (2014) 1966–1972.
- [28] G. Zhu, H. Zheng, Z. Zhang, T. Tshukudu, P. Zhang, X. Xiang, Characterization and coagulation–flocculation behavior of polymeric aluminium ferric sulfate (PAFS), Chem. Eng. J., 178 (2011) 50–59.
- [29] W. Lan, H. Qiu, J. Zhang, Y. Yu, K. Yang, Z. Liu, G. Ding, Characteristic of a novel composite inorganic polymer coagulant–PFAC prepared by hydrochloric pickle liquor, J. Hazard. Mater., 162 (2009) 174–179.
- [30] A. Adin, L. Dean, F. Bonner, A. Nasser, Z. Huberman, Characterization and destabilization of spent filter backwash water particles, Wat. Sci. Technol: Wat. Supply., 2 (2002) 115–122.
- [31] Z.L. Yang, B.Y. Gao, Q.Y. Yue, Y. Wang, Effect of pH on the coagulation performance of Al-based coagulants and residual aluminium speciation during the treatment of humic acid-kaolin synthetic water J. Hazard. Mater., 178 (2010) 596–603.
- [32] W. Stumm, Chemistry of the solid-water interface: processes at the mineral-water and particle-water interface in natural systems, John Wiley & Son Inc., 1992.
- [33] J.K. Edzwald, J.E. Benschoten, Aluminium coagulation of natural organic matter, Chem. Wat. Wastewater Treat., (1990) 341–359.
- [34] M. Yana, D. Wang, J. Yu, J. Ni, M. Edwards, J. Qu, Enhanced coagulation with polyaluminium chlorides: Role of pH/Alkalinity and speciation, Chemos., 71 (2008) 1665–1673.
- [35] K. McCurdy, K. Carlson, D. Gregory, Floc morphology and cyclic shearing recovery: comparison of alum and polyaluminium chloride coagulants, Wat. Res., 38 (2004) 486–494.
- [36] Y. Shen, Treatment of low turbidity water by sweep coagulation using bentonite, J. Chem. Technol. Biotechnol., 80 (2005) 581–586.
- [37] J.Q. Jiang, Development of coagulation theory and pre-polymerised coagulants for water treatment, Sep. Purif. Methods., 30 (2001) 127–141.
- [38] H.B. Halvadiya, D. Gangadharan, K.M. Popat, P.S. Anand, Deionization of coagulated, clarified, turbid gaurishankar lake waters by using ion-exchange technology, Sep. Sci. Technol., 43 (2008) 2183–2195.
- [39] T. Tseng, M. Edwards, Predicting full-scale TOC removal. J. AWWA, 91 (1999) 159–170.
- [40] M. Pivokonsky, J. Naceradska, T. Brabenec, K. Novotna, M. Baresova, V. Janda, The impact of interactions between algal organic matter and humic substances on coagulation, Wat. Res., 84 (2015) 278–285
- [41] H. Zhao, C. Hu, H. Liu, X. Zhao, J. Qu, Role of aluminium speciation in the removal of disinfection byproduct precursors by a coagulation process, Environ. Sci. Technol., 42 (2008) 5752– 5758.
- [42] US-EPA, Enhanced Coagulation and Enhanced Precipitative Softening Guidance Manual, EPA, Office of Water and Drinking Ground Water, Washington, DC, 1998, pp. 20–50.
- [43] J. Duan, J. Gregory, Coagulation by hydrolysing metal salts, Adv. Colloid. Interface. Sci., (2003) 100–102.
- [44] A. Matilainen, M. Vepsäläinen, M. Sillanpää, Natural organic matter removal by coagulation during drinking water treatment: a review, Adv. Colloid. Interface. Sci., 159 (2010) 189–197.
- [45] A.K. Verma, R.R. Dash, P. Bhunia, A review on chemical coagulation/flocculation technologies for removal of colour from textile wastewaters, J. Environ. Manage., 93 (2012) 154–168.

328

- [46] J.K. Edzwald, Coagulation in drinking water treatment: particles, organics and coagulants, Wat. Sci. Technol., 27 (1993) 21–35.
- [47] M. Yoshihiko, Y. Akira, F. Yuji, K. Tasuku, Dynamic analysis of coagulation with alum and PACl, J. AWWA, 90 (1998) 96–106.
 [48] Q.S. Wei, C.H. Feng, D.S. Wang, B.Y. Shi, L.T. Zhang, Q. Wei, M.Y. Shi, L.T. Zhang, C. Wei, M.Y. Shi, L.T. Zhang, C. Wang, B.Y. Shi, L.T. Shi, L.T. Shi, L.T. Shi, L.T. Shi, L.T. Zhang, C. Wang, B.Y. Shi, L.T. Shi, L.T.
- [48] Q.S. Wei, C.H. Feng, D.S. Wang, B.Y. Shi, L.T. Zhang, Q. Wei, H.X. Tang, Seasonal variations of chemical and physical characteristics of dissolved organic matter and trihalomethane precursors in a reservoir: a case study, J. Hazard. Mater., 150 (2008) 257–264.
- [49] M. Yan, D. Wang, S. You, J. Qu, H. Tang, Enhanced coagulation in a typical North-China water treatment plant, Wat. Res., 40 (2006) 3621–3627.
- [50] G. Berthon, Aluminium speciation in relation to aluminium bioavailability, metabolism and toxicity, Coord. Chem. Rev., 228 (2002) 319–341.
- [51] E. Gauthier, I. Fortier, F. Courchesne, P. Pepin, J. Mortimer, D. Gauvreau, Aluminium forms in drinking water and risk of Alzheimer's disease, Environ. Res., 84 (2000) 234–246.

- [52] G. Gyawali, A. Rajbhandari, Investigation on coagulation efficiency of polyaluminium silicate chloride (PASiC) coagulant, Sci. World., 10 (2012) 33–37.
- [53] S.J. Randtke, Organic contaminant removal by coagulation and related process combinations, J. AWWA, 80 (1988) 40–56.
- [54] P. Gebbie, An operator's guide to water treatement coagulants, 31st Annual QLD Water Industry Workshop-Operations Skills, 2006.
- [55] H. Arora, G. Di Giovanni, M. LeChevallier, Spent filter backwash water contaminants and treatment strategies, J. AWWA, 93 (2001) 100–111.
- [56] P. Assavasilavasukul, B.L.T. Lau, G.W. Harrington, R.M. Hoffman, M.A. Borchardt, Effect of pathogen concentrations on removal of Cryptosporidium and Giardia by conventional drinking water treatment, Wat. Res., 42 (2008) 2678–2690.