# Comparison of the Ability of Three Coagulants to Enhance Filter Performance

Po-Hsun Lin, M.ASCE<sup>1</sup>; Leonard W. Lion<sup>2</sup>; and Monroe L. Weber-Shirk<sup>3</sup>

**Abstract:** In the operation of porous media filtration systems, poor initial performance is often observed, in which particle removal is less than desired. Alum  $(Al_2(SO_4)_3 \cdot 14H_2O)$ , ferric chloride (FeCl<sub>3</sub>), and polyaluminum chloride (PACl), are often used in drinking-water treatment to modify the surface properties of the particles being filtered and enhance their removal by filters; however, modification of the filter medium is not a common practice. In this research, these three coagulants were used to artificially modify a sand medium in situ by adding metal hydroxide precipitates onto the top of the filter before challenging the filter with an otherwise untreated kaolin suspension. After modification of the filter medium, high particle-removal efficiencies were achieved using a short (7.5 cm) sand column with  $\cong 1$  mm diameter sand. The best observed particle removal (96%) using alum and PACl occurred at the surface loading of 550 mmol of Al/m<sup>2</sup>. The filter performance deteriorated at higher surface loadings. Fractional colloid removal increased with increased ferric chloride surface loading over the entire range of dosages tested (up to 97.5% at 2,200 mmol Fe/m<sup>2</sup>). The experimental results suggest that pretreatment of filter media by application of Fe or Al hydroxides can eliminate the initial poor performance. **DOI: 10.1061/(ASCE)EE.1943-7870.0000339.** © *2011 American Society of Civil Engineers*.

CE Database subject headings: Chloride; Filtration; Coagulation; Performance characteristics.

Author keywords: Alum; Ferric chloride; Polyaluminum chloride; Enhanced filtration; Coagulants.

# Introduction

Filtration through porous media is a commonly used method to remove particles present in surface waters, precipitated hardness from lime-softened water, and precipitated iron and manganese present in many well-water supplies (Weber 1972). Filtration has been widely used following treatment of raw waters by chemical coagulation, flocculation, and sedimentation processes. The removal of suspended particles within a filter is considered to involve at least two sequential steps: transport and attachment. In the first step, the particles are transported from the bulk fluid to the immediate vicinity of solid-liquid interface presented by the filter (i.e., to a grain of the media or to another particle previously retained in the filter bed). The transport of particles to the filter medium may occur through three mechanisms: Brownian diffusion (molecular effects), interception (contact as a result of fluid flow near the surface of the porous media), and sedimentation (gravity effects). Particle attachment to the media surface is thought to be dominated by electrical and chemical interactions such as electrostatic attraction or repulsion within the electrical double layer and van der Waals attractive forces that act between particles and surfaces at short distances (Yao et al. 1971; O'Melia 1980; Elimelech and O'Melia 1990).

<sup>1</sup>Research Fellow, School of Civil and Environmental Engineering, Cornell Univ., Ithaca, NY 14853-3501. E-mail: pl254@cornell.edu

<sup>2</sup>Professor, School of Civil and Environmental Engineering, Cornell Univ., Ithaca, NY 14853-3501. E-mail: LWL3@cornell.edu

<sup>3</sup>Senior Lecturer, School of Civil and Environmental Engineering, Cornell Univ., Ithaca, NY 14853-3501 (corresponding author). E-mail: mw24@cornell.edu

Note. This manuscript was submitted on March 25, 2010; approved on November 18, 2010; published online on November 20, 2010. Discussion period open until October 1, 2011; separate discussions must be submitted for individual papers. This paper is part of the *Journal of Environmental Engineering*, Vol. 137, No. 5, May 1, 2011. ©ASCE, ISSN 0733-9372/2011/5-371–376/\$25.00.

Sand media is a key, if not the sole, component of most rapid sand filters. However, sand media may not be efficient in removing fine or submicron particles including colloids, bacteria, and viruses because of electrostatic repulsion arising from the fact that both the particles and the sand media are negatively charged at pH 6-8, which is typical of surface waters. Small particles close to 1  $\mu$ m also have poor transport, impairing their removal (Yao et al. 1971). As a result, Fe and Al are commonly added as coagulants to destabilize particles (Weber 1972; Letterman 1999; Tchobanoglous et al. 2003; MWH 2005) and improve particle removal. Destabilization involving adsorption of Al or Fe monomers is called charge neutralization; coagulation of colloidal particles in the presence of  $Al(OH)_{3(am)}$  and  $Fe(OH)_{3(am)}$  is termed enmeshment or sweep floc (Van Benschoten and Edzwald 1990). Destabilization causes colloidal particles to aggregate, and larger aggregates are more readily transported to the filter medium by interception or gravity.

In some cases, iron and aluminum hydroxides have been used to modify filtration media to improve particle-removal efficiency (Edwards and Benjamin 1989; Lukasik et al. 1999; Ahammed and Meera 2006). In these studies, the surface modification of Fe and Al was achieved by soaking the sand medium in aluminum chloride or ferric chloride solutions followed by treatment with an ammonium hydroxide solution. The pretreatment improved particle capture, but the treatment method required removing the sand from the filter and thus would not be easily applied to operating filtration systems.

In the research described in this paper, three common coagulants, alum, ferric chloride, and polyaluminum chloride (PACl), were chosen to modify a sand-filter medium before challenging the filter with a colloidal suspension of kaolin. The resulting modification of the filter could include coating the sand surface with precipitated aluminum or iron hydroxides; deposition of Al or Fe precipitates in the pore space of the filter medium; layering of Al or Fe precipitates onto the top of the sand filter, forming

JOURNAL OF ENVIRONMENTAL ENGINEERING © ASCE / MAY 2011 / 371

a dual-filter medium; or some combination of these three processes. The point of zero charge (PZC) of aluminum hydroxide and ferric hydroxide are 9.6 (Scholtz et al. 1985) and 8.5 (Benjamin 2002), respectively. Thus, at neutral pH, the modified sand media is expected to be positively charged and to have an enhanced ability to capture negative colloidal particles. In this study, a process was used for in situ modification of the sand before challenging the filter. The resulting particle-removal efficiencies are compared as a function of Al and Fe surface loading and are shown in the following to be significantly improved during the initial filtration run. Head loss was also monitored during pretreatment and during the challenge to the filter.

# Materials and Methods

#### Apparatus

An automated filtration test apparatus described by Weber-Shirk (2008) was used to conduct parametric tests of the use of alum, ferric chloride (Fisher Scientific, Pittsburgh, PA), and polyaluminum chloride (PCH-180, Al 5.5%, Cl 12.1%,  $SO_4$  1.8%, basicity 69%; Holland Company, Inc., Adams, MA) to artificially modify the medium in a sand filter. A schematic of the apparatus is shown in Fig. 1.

In all tests, 22°C Cornell University tap water (pH  $\approx$  7.6 ± 0.2; total organic carbon  $\approx$ 2.0 mg/L; hardness  $\approx$ 150 mg/L as CaCO<sub>3</sub>; alkalinity  $\approx$ 110 mg/L as CaCO<sub>3</sub>; chlorine residual  $\approx$ 0.7 mg/L) was used as the raw-water source. Kaolin clay (R.T. Vanderbilt Co., Inc., Norwalk, CT) was added to distilled water as a concentrated stock (350 mg/L, pH = 4.3) and then diluted with tap water to achieve an influent turbidity of 55–60 NTU (nephelometric turbidity units) and pH of 7.5 ± 0.2. At this pH, the zeta potential of the clay was determined to be  $-18 \pm 2$  mV by laser Doppler electrophoresis (Zetasizer Nano-ZS; Malvern Instruments, Ltd., Malvern, UK). The zeta potential of most particles in surface waters at neutral pH is in the range of -15 to -30 mV (Ongerth and Pecoraro 1996). The clay particle size was approximately 1  $\mu$ m with a mean polydispersity index of 0.6 as measured by dynamic light scattering (Zetasizer Nano-ZS).

Many sand filters are operated with an influent turbidity of less than 5 NTU. Results from experiments at two different clay concentrations ( $\sim$ 5 and  $\sim$ 60 NTU) show that a  $\sim$ 10 fold variation in turbidity did not cause significant difference in the performance of an untreated filter but resulted in a better performance for an

alum-pretreated filter when using the ~5-NTU clay suspension (Fig. S1 in the supplemental data). A higher influent turbidity of ~60 NTU was chosen in the experiments reported here to provide the worst-case scenario for particle removal and to ensure that significant differences in particle concentration could be accurately measured (the initial effluent turbidity for the pretreated filter was close to the detection limit of the turbidimeter). A higher influent turbidity also allowed the use of a shorter run time while still providing significant particle loading in the filter. The concentration of the stock solutions, clay suspension, and the coagulant doses used are given in Table 1.

The alum, PACl, and ferric chloride concentrations at the filter influent were 74 mg/L as alum, 6.7 mg/L PACl as Al, and 67 mg/L as FeCl<sub>3</sub>, respectively. The filter column was 2.5 cm in diameter and contained 7.5 cm of 0.8-1 mm in diameter filter sand (U.S. Filter, East Orange, NJ). In effect, the shallow filter column and test conditions provide an experimental model of the top 7.5 cm of a rapid sand filter.

Coagulants were mixed with base (sodium bicarbonate) and tap water to maintain the pH at 6.8 for alum, 7.1 for PACl, and 6.6 for FeCl<sub>3</sub> and were then applied to the top of the filter column to treat the filter medium before challenging the filter with the clay suspension. The pH of the filtered effluent using three coagulants was monitored and close to the tap water pH (7.5  $\pm$  0.2) in all experiments. We report the amount of coagulant applied to the filter as a surface loading in  $mM/m^2$  for the cross-sectional area of the filter. The cross-sectional area of the filter is used to express dosage rather than filter volume because the  $Al(OH)_{3(am)}$  or  $Fe(OH)_{3(am)}$  accumulates in the top few centimeters of the filter bed. Presenting the dose as a surface loading facilitates generalization of the experimental results to other filter sizes. The total surface loading of the coagulant,  $\Gamma$ , is a function of the coagulant flow rate, Q, filter surface area, A, influent concentration, C, and application time, t, as described in Eq. (1):

$$\Gamma = \frac{QCt}{A} \tag{1}$$

Experiments were performed over a wide range of surface loadings (0 as a control, 70, 140, 280, 550, 1,100, and 2,200 mmol/m<sup>2</sup>) to explore the dose-dependence of particle-capture efficiency. The filtration rate was held constant at 1.4 mm/s in all experiments. Although the experimental results may be affected to some extent by scale [the column diameter/sand size ratio was  $\approx$  25, and thus bed porosity may have been increased



Fig. 1. Schematic of filter test apparatus showing valve and pump configuration for pretreatment with aluminum sulfate

372 / JOURNAL OF ENVIRONMENTAL ENGINEERING © ASCE / MAY 2011

Table 1. Stock and Feed Concentrations of Reagents Used

Chemical name	Concentration of the stock solution	Dilution factor	Concentration at filter influent
Kaolin clay	350 (mg/L)	7	50 (mg/L)
Aluminum sulfate	3.4 (mmol/L as Al)	13.6	250 ( $\mu$ mol/L as Al)
Polyaluminum chloride	3.4 (mmol/L as Al)	13.6	250 ( $\mu$ mol/L as Al)
Ferric chloride	3.4 (mmol/L as Fe)	13.6	250 ( $\mu$ mol/L as Fe)
Sodium bicarbonate	12 (mmol/L as $HCO_3^{-1}$ )	40.8	300 ( $\mu$ mol/L as HCO <sub>3</sub> <sup>-1</sup> )
Hydrochloric acid	$0.1 \text{ (mol/L as H}^+\text{)}$	1	0.1 (mol/L as H <sup>+</sup> )

by 1.25% (Zou and Yu 1995)], these variables were systematically retained for all experiments; therefore, trends in our results are expected to be valid for full-scale filters. Backwash and an acid wash (0.1 M HCl) were used between each experiment to remove residual clay and metal hydroxide precipitates. Duplicate control experiments with no coagulant addition were conducted at the start of each series of experiments and after the experiment with the highest surface loading to confirm the ability of the filter-washing steps to restore the sand to its original (untreated) condition.

A process control software (Weber-Shirk 2008) for the automated filtration apparatus cycled through states shown in Table 2 for each test. The hydraulic retention time from the clay stock pump to the effluent turbidity meter was 3.8 min, based on the filtration rate of 1.4 mm/s. In the filter column, the volume of water above the sand was 35 cm<sup>3</sup>, and the hydraulic retention time from the top to the bottom of filter was 1.2 min (porosity = 0.4, reported by the manufacturer).

The concentration of the applied coagulants was approximately the same during the pretreatment state, but the pretreatment state duration was varied to achieve different surface loadings.

During the particle challenge state, the filter was operated at a constant flow rate of 1.4 mm/s (0.68 mL/s) for 20 min, and the filter performance was measured by inline turbidimeters (Fig. 1). The turbidimeters were chosen because they have small-volume sample cells (30 mL) that make it possible to achieve reasonable response times at the flow rates used in this research. The 20-min challenge with an influent turbidity of 60 NTU made it possible to obtain significant particle loadings for each pretreatment condition and to cycle over a wide range of coagulant dosages in a reasonable length of time. The hydraulic residence time from the inlet of the influent turbidimeter to the outlet of the effluent turbidimeter at a flow rate that corresponds to a filtration velocity of 1.4 mm/s was 4 min. We do not report the data from the first 1.5 hydraulic residence times (i.e., approximately 6.5 min) at the beginning of each particle challenge because that data show an artificially high particle removal caused by the clean pore water that is still exiting the

Table 2. Process Controller States

State name	Purpose	Duration (min)	
Backwash filter-1	Clean the filter	4	
Acid wash	Wash out the coagulant	2	
Backwash filter-2	Clean the filter	4	
Wash turbidity meters	Wash the turbidity meters	4	
Pretreatment	Modify the filter media	Variable	
Down flow	Challenge the filter	20	

filter into the effluent turbidimeter and being displaced from the tubing and turbidimeter sample vial.

# Measurement of Effluent AI and Fe Concentrations

Retention of applied Al or Fe precipitates in the filter medium was assessed by measuring aluminum and iron concentrations in the effluent during pretreatment at the highest  $(2, 200 \text{ mmol/m}^2)$  and lowest  $(70 \text{ mmol/m}^2)$  surface loading used in experiments. In addition, the Al concentration in the effluent was measured during challenge at the alum surface loadings of 70, 550, and 2,200 mmol of Al/m<sup>2</sup>. All effluent samples for metal analysis were acidified, and the concentrations reported are for total metal (i.e., they include any metals that were originally present in particulate form).

A colorimetric assay, the modified eriochrome cyanine R method, was selected for Al analysis, in which the volume of reagents and samples were scaled down to fit into 4.5-mL cuvettes (*Standard Methods for the Examination of Water and Wastewater* 1998). The assay can detect aluminum at concentrations above 20  $\mu$ g/L. All glassware used was first acid washed for 24 h with 10% nitric acid (reagent grade) made with distilled deionized (DDI) water followed by a 24-h wash in 10% nitric acid (trace metal grade). The glassware was rinsed with DDI water six times in between the 24-h wash steps. Aluminum chloride (AlCl<sub>3</sub> • 6H<sub>2</sub>O, Fisher Scientific) was used to prepare Al standard curves.

Atomic absorption spectrometry was used to determine iron in the effluent water during pretreatment (*Standard Methods for the Examination of Water and Wastewater* 1998). An air-acetylene flame was used. The assay can detect iron at concentrations above 20  $\mu$ g/L. All glassware was acid washed, as previously described. Iron standards were made by dilution of iron stock (1,000 mg/L, Fisher Scientific). The measurements were carried out in triplicate, and the coefficient of variation in analysis of duplicate samples was  $\leq 5\%$ .

# **Results and Discussion**

#### Aluminum and Iron Concentration in Effluent

All effluent during pretreatment was collected and measured for Al or Fe concentration. Mass balance showed that  $\geq 78\%$  of applied Al or Fe was retained in the filter at a surface loading of 70 mmol/m<sup>2</sup>, and  $\geq 82\%$  of Al or Fe remained in the filter at a surface loading of 2, 200 mmol/m<sup>2</sup> (Table 3). The two applied dosages are used as maximum and minimum surface loadings in the following discussion and comparison of results, so that the retained Al or Fe concentrations for the other dosages between these extremes can be estimated.

Effluent Al concentrations for alum application at the surface loadings of 70, 550, and 2,200  $\rm mmol/m^2$  during challenge are

**Table 3.** Fraction of Applied Al and Fe Retained in Sand Medium During

 Pretreatment

Sample name	Surface loading (mmol/m <sup>2</sup> Al or Fe)	Al or Fe mean concentration in effluent (mg/L)	Al or Fe fraction retained in filter (%)
Low alum	70	0.66	78
High alum	2,200	1.07	82
Low PAC1	70	0.24	92
High PACl	2,200	0.83	87
Low FeCl <sub>3</sub>	70	0.90	93
High FeCl <sub>3</sub>	2,200	0.74	97

#### JOURNAL OF ENVIRONMENTAL ENGINEERING © ASCE / MAY 2011 / 373



**Fig. 2.** Effluent Al concentrations after pretreatment with alum at three surface loadings during 20-min challenge

presented in Fig. 2. At loadings of 70 and 550 mmol/ $m^2$ , the Al concentrations were below the maximum secondary drinking water standards of the Environmental Protection Agency (EPA) (200  $\mu$ g/L). For the surface loading of 2,200 mmol/m<sup>2</sup>, the Al concentration at 0 min exceeded the predicted solubility of  $Al(OH)_{3(am)}$  (i.e., 170  $\mu$ g/L at pH 7.5). The high Al concentrations were likely caused by an initial washout of precipitated Al from the column. Subsequent Al concentrations were slightly higher than the  $Al(OH)_{3(am)}$  solubility limit and may also have been caused by the release of small amounts of precipitated Al. The effluent Al concentration increased with increasing surface loadings. We did not measure Al and Fe concentrations for PACl and ferric chloride during challenge because the solubility at pH 7.5 for both metal hydroxide precipitates were much less than that of alum [i.e., 85  $\mu$ g/L as Al, determined by experiments for PACl (Van Benschoten and Edzwald 1990) and 0.002  $\mu$ g/L as Fe, based on the solubility of  $Fe(OH)_{3(am)}$  (Snoeyink and Jenkins 1980)].

## Improvement of Filter Performance

Three coagulants, alum, ferric chloride, and PACl, were applied to the sand filter. Particle-capture efficiency is expressed here as  $pC^*$ , where  $C^*$  = turbidity of the effluent water normalized by the turbidity of the influent water; and p = -logarithm function (base 10) [i.e.,  $pC^* = -\log(C_{\text{effluent}}/C_{\text{influent}})$ ]. Fig. 3 shows representative results from the experiments with alum. The correspondence of duplicate before and after control experiments (i.e., without application of alum) shows that the cleaning procedure was effective. The untreated sand achieved a baseline particle removal of  $\simeq 60\%$ . This removal efficiency may be higher than is commonly observed under no chemical pretreatment. However, the synthetic raw water contained only a low level of organic matter (total organic carbon  $\approx 2.0 \text{ mg/L}$ ) that had passed through a water treatment plant where surface active organics would be expected to be removed. Dissolved organic matter in raw water can act to stabilize the clay suspension and reduce the filter capture efficiency. Fig. 3 also shows clay removal (pC\*) as a function of different aluminum surface loadings. The agreement of filter performance at duplicate dosages was excellent. Clay removal increased as a function of Al surface loading up to a dose of 550 mmol/ $m^2$  and then declined. Similar results were obtained with PACl, with the best particle removal obtained at loadings of 550 mmol/m<sup>2</sup> of aluminum (Fig. S2 in the supplemental data). Polyaluminum chloride has recently been reported to be superior to alum in removing particles, with advantages of reduced alkalinity consumption, less sludge production, decreased temperature and pH dependence, and reduction of



**Fig. 3.** Alum pretreatment; clay removal (pC\*) for different pretreatment surface loadings  $(mmol/m^2)$  of Al; data are shown at 30-s intervals

cost (Hu et al. 2006). For all three coagulants, the performance after pretreatment declined during the filtration run, which could result from a combination of loss of coagulant through dissolution, aging of precipitated metal hydroxides (with concomitant loss of reactivity), and site blocking or charge neutralization by captured clay. Accumulation of captured clay in controls likely caused a slight change in pore geometry that is responsible for improved particle capture.

Readers are cautioned that the value of the maximum effective alum and PACI surface loading applies to the conditions used in these experiments and is expected to be a function of the application method used to pretreat the sand medium, the size distribution of the sand, solution conditions (e.g., pH and natural organic matter concentration), and type of colloidal particle. Particle removal using iron increased with increasing surface loading over the entire range of ferric chloride dosages (Fig. S3 in the supplemental data).

The performance, pC\*, of the filter for each coagulant is shown in Fig. 4 as a function of surface loading. Performance values shown are for a 5-min average for the effluent data centered at the fifteenth minute (i.e., average the data from 12.5 to 17.5 min) and then averaged with the results of the duplicate experiment. As previously noted, maximum clay removal occurred at 550 mmol/m<sup>2</sup> of aluminum, and the performance deteriorated at higher alum and PACl doses.



**Fig. 4.** Clay removal (pC\*) by filter as function of different surface loadings of three coagulants

The performance using ferric chloride is slightly better than alum and PACl when all treatments are compared at a surface loading of 550 mmol/m<sup>2</sup>. There is a larger difference for pC\* between iron and Al at surface loadings of 1,100 and 2,200 mmol/m<sup>2</sup> because of the deterioration of performance using alum and PACl at higher surface loadings.

## **Head-Loss Profiles**

Addition of the three coagulants caused increased head loss during the pretreatment stage. Fig. 5 shows head loss as a function of surface loading of alum, ferric chloride, and PACI. In each case, the head loss increase was directly proportional to the surface loading. Ferric chloride had the largest head loss per millimole of square meter applied, followed by PACl and alum. The difference in head loss between alum and PACl suggests that the precipitates formed from these two different sources of Al are distinct. Some researchers have shown that the polymeric structure within the PACl precipitate remains intact, and the PACl precipitate often appears as small (< 25  $\mu$ m) spheres, clusters of small spheres, and even chainlike structures; wheres alum flocs are reported to consist of an amorphous solid that usually appears as fluffy, porous structures, ranging in size from 25 to 100  $\mu$ m (Van Benschoten and Edzwald 1990). Therefore, PACl flocs may create smaller pores between flocs, with a corresponding lower hydraulic conductivity than do the alum flocs; thus, for the same surface loading, the PACI flocs create higher head loss than alum flocs.

Lines were fit to the data in Fig. 5 by least squares regression, and the slopes were used to predict head loss for pretreatment at the surface-loading values shown in Fig. 4. These calculated head-loss values are compared in Fig. 6 to the observed clay removal in the subsequent challenge (pC\* data from Fig. 4). At a head loss of  $\leq$  25 cm, the relationship between the clay removals and the head losses for the three coagulants were almost identical. As noted in the following (see Fig. 7 and discussion), negligible additional head-loss accumulation occurred during the clay challenge to the pretreated filter media. Given that the metal hydroxide precipitate and not the captured clay caused the head loss, then the correlation between pC\* and head loss (for head loss of  $\leq 25$  cm) suggests that the Al and Fe precipitates may have had similar porosities and pore size distributions. Both head loss and particle-capture efficiency are strongly influenced by porosity and pore size distribution.

Precipitation of aluminum and iron hydroxides would both act to decrease void space in the porous medium. It is possible that the



**Fig. 6.** Clay removal (pC\*) by filter as function of predicted head loss of three coagulants tested

molar volumes of the Al and Fe precipitates are different. The higher head losses experienced with Fe versus Al suggest that, if volume differences exist, Fe occupies a greater pore volume on a molar basis (see Fig. 5). Analysis of the distribution of head loss caused by Al hydroxide precipitate in the filter (Fig. S6 in the supplemental data) indicate that, at high Al loadings (1,100 and 2,200 mmol Al/m<sup>2</sup>), the head loss per unit depth was focused near the top of the filter bed. Because this head loss reflects a reduction in porosity and concomitant increase in shear stress, it is possible that the resulting shear stress inhibited clay attachment to the media surfaces. As a result, performance deteriorated. For ferric chloride, the best clay removal was at the highest surface loading of iron. The better colloid removal by Fe despite higher head loss (see Fig. 6) suggests that the strength of clay–ferric hydroxide interactions was strong enough to resist the higher shear stress.

Fig. 7 presents the head loss as a function of time during the kaolin suspension challenge to the filter when the applied surface loading of Al or Fe was 550 mmol/m<sup>2</sup>. The control head loss was approximately 7 cm during the filter run. The small head-loss change during filter operation (Fig. 7) relative to the head-loss change during filter pretreatment (Fig. 5) suggests that the space the metal hydroxide precipitates occupied in the pores was much larger than the volume of clay particles that accumulated during the challenge. Alternately, head loss caused by flow through



**Fig. 5.** Head loss during pretreatment as function of surface loading of three different coagulants applied to filter



Fig. 7. Head loss during kaolin challenge to filter as function of run time after pretreatment with three different coagulants; Al or Fe concentrations were 550 mmol/ $m^2$ 

JOURNAL OF ENVIRONMENTAL ENGINEERING © ASCE / MAY 2011 / 375

porous Al or Fe precipitates dominated the observed energy loss relative to that caused by captured clay particles.

# Conclusions

The addition of alum, ferric chloride, or PACl as a pretreatment to a sand filter resulted in greatly enhanced removal of clay relative to the untreated sand medium. Surface loadings of 140 mmol/m<sup>2</sup> improved  $pC * by \sim 1$  while increasing head loss to 3 cm. The maximum observed particle removal using alum and PACl occurred at the surface loading of 550 mmol  $A1/m^2$ , and at this loading for alum application, the Al concentrations in the filtered effluent were below the EPA secondary maximum contaminant level (MCL). Fractional removal of clay increased with increased ferric chloride over the entire range of dosages tested (maximum = 2,200 mmol  $Fe/m^2$ ). However, high surface Fe loadings may not be practical in application because of the associated high head loss. For alum and PACl, the filter performance deteriorated when the surface loading was higher than 550 mmol  $Al/m^2$ . At the same surface loading on a molar basis, pretreating with ferric chloride achieved better particle-removal efficiency than the other two coagulants. However, all coagulants achieved the same clay removal when compared on the basis of the head loss associated with their application at a head loss of  $\leq 25$  cm.

In this study, a pC\* of 1.6 (97% removal) was achieved using a 7.5-cm-deep sand column with 1-m-diameter sand. All three pretreatment agents tested improved performance during the initial period of operation of a rapid sand filter. The results show good potential that media pretreatment can be incorporated with the operation of conventional treatment to improve particle removal during the ripening stage of filter operation. Real natural waters, however, contain different sizes and types of particulates and natural organic matter (NOM) such as humic and fulvic acids. For the pH conditions of most surface water, pH 6-8, NOM is negatively charged and can adsorb onto the surface of clay and other suspended particles, further increasing their negative charge (Pernitsky and Edzwald 2006). The presence of NOM in the influent of the coagulant-pretreated filter is expected to result in a lower particleremoval efficiency. Further research is required to account for the effect of NOM and the diverse types of particulate matter in the raw water, and different methods of applying coagulants that may reduce head loss (e.g., apply coagulants during backwash) should be considered.

## **Supplemental Data**

The supplemental data contains clay removal (pC\*) for an alumpretreated and untreated filter using two different clay concentrations, clay removal as a function of different PACl and FeCl<sub>3</sub> surface loadings, and analysis of the distribution of head loss caused by Al hydroxide precipitate in the filter. These materials, including Figs. S1–S6, are available online in the ASCE Library (www.ascelibrary.org).

#### References

- Ahammed, M. M., and Meera, V. (2006). "Iron hydroxide-coated sand filter for household drinking water from roof-harvested rainwater." J. Water Supply: Res. Technol.-AQUA, 55(7–8), 493–498.
- Benjamin, M. M. (2002). *Water chemistry*, McGraw-Hill Science, New York.
- Edwards, M., and Benjamin, M. M. (1989). "Adsorptive filtration using coated sand: A new approach for treatment of metal-bearing wastes." *Res. J. Water Pollut. Control Fed.*, 61(9), 1523–1533.
- Elimelech, M., and O'Melia, C. R. (1990). "Kinetics of deposition of colloidal particles in porous media." *Environ. Sci. Technol.*, 24(10), 1528–1536.
- Hu, C., Liu, H., Qu, J., Wang, D., and Ru, J. (2006). "Coagulation behavior of aluminum salts in eutrophic water: Significance of Al<sub>13</sub> species and pH control." *Environ. Sci. Technol.*, 40(1), 325–331.
- Letterman, R. D. (1999). *Water quality and treatment*, 5th Ed., American Water Works Association, Denver.
- Lukasik, J., Cheng, Y.-F., Lu, F., Tamplin, M., and Farrah, S. R. (1999). "Removal of microorganisms from water by columns containing sand coated with ferric and aluminum hydroxides." *Water Res.*, 33(3), 769–777.
- MWH. (2005). Water treatment—Principles and design, 2nd Ed., Wiley, New York.
- O'Melia, C. (1980). "ES&T features: Aquasols: The behavior of small particles in aquatic systems." *Environ. Sci. Technol.*, 14(9), 1052–1060.
- Ongerth, J. E., and Pecoraro, J. P. (1996). "Electrophoretic mobility of cryptosporidium oocysts and giardia cysts." *J. Environ. Eng.*, 122(3), 228–231.
- Pernitsky, D. J., and Edzwald, J. K. (2006). "Selection of alum and polyaluminum coagulants: Principles and applications." J. Water Supply: Res. Technol.-AQUA, 55(2), 121–141.
- Scholtz, E. C., Feldkamp, J. R., White, J. L., and Hem, S. L. (1985). "Point of zero charge of amorphous aluminum hydroxide as a function of adsorbed carbonate." *J. Pharm. Sci.*, 74(4), 478–481.
- Snoeyink, V. L., and Jenkins, D. (1980). *Water chemistry*, Wiley, New York.
- Standard methods for the examination of water and wastewater. (1998). 20th Ed., American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC.
- Tchobanoglous, G., Burton, F. L., and Stensel, H. D. (2003). *Wastewater* engineering treatment and reuse, 4th Ed., McGraw-Hill Science, New York.
- Van Benschoten, J. E., and Edzwald, J. K. (1990). "Chemical aspects of coagulation using aluminum salts—I. Hydrolytic reactions of alum and polyaluminum chloride." *Water Res.*, 24(12), 1519–1526.
- Weber, W. J. (1972). *Physicochemical processes for water quality control*, Wiley, New York.
- Weber-Shirk, M. (2008). "An automated method for testing process parameters." (https://confluence.cornell.edu/display/AGUACLARA/Process +Controller) (Sept. 25, 2008).
- Yao, K.-M., Habibian, M. T., and O'Melia, C. R. (1971). "Water and waste water filtration. Concepts and applications." *Environ. Sci. Technol.*, 5(11), 1105–1112.
- Zou, R. P., and Yu, A. B. (1995). "The packing of spheres in a cylindrical container: The thickness effect." *Chem. Eng. Sci.*, 50(9), 1504–1507.