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Influence of Particle Characteristics on Filter Ripening

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Abstract: To investigate the dependence of filter ripening on particle size and surface charge, multiple experiments were conducted under different particle destabilization conditions including pH control, alum, and polymer destabilization. Laboratory-scale filtration experiments were performed at a filtration velocity of 5 m/h using spherical glass beads with mean diameter of 0.55 mm as collectors. Particle suspensions with a broad size distribution and a 1.7 μm mean particle size were filtered through a 10 cm depth filter column. Better initial solids removal was confirmed under favorable particle and collector conditions (i.e., under smaller surface charge), but better initial particle removal does not necessarily mean better overall particle removal efficiency. It was shown that changes of the particle size distribution (PSD) in the effluent can significantly influence overall particle removal efficiency. Chemical parameters such as zeta potential can be important during the initial stage of filtration, but their importance can decrease over time depending on the specific chemical conditions. The influent PSD and the removal of certain size particles during the initial stage of filtration can significantly influence ripening, which in turn, can influence the overall particle removal efficiency.

Keywords: Particle, filtration, ripening, particle size distribution (PSD), surface charge

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INTRODUCTION

Physicochemical characteristics of particles such as particle size and surface charge have significant influence on the particle removal efficiency in granular media filtration. Much research on filtration has focused on clean bed removal, because solid removal at that stage is influenced mainly by interactions between particles and collectors (media grains). But clean bed removal is relatively short in an overall filter run, and ripening can significantly influence the overall filtration efficiency in later parts of the run. To improve filter effluent water quality, the period and efficiency of ripening is important. Ripening was investigated in this research under three different chemical conditions. The main objective of this research was to investigate the influence of particle characteristics on filter ripening from the viewpoint of surface charge and particle size.

When a filter run starts, the filter bed is clean, and this stage of filtration is called clean bed filtration. From this point onwards, the filter bed accumulates particles. The particles that accumulate on the filter medium work as additional collectors, and the effluent quality usually improves. This period is known as ripening. That is, ripening can be defined as the improvement in effluent quality early in a filter run.

In operating water treatment plants (WTPs), there is an initial degradation in effluent quality right after backwashing and before the clean bed removal stage. Amirtharajah and Wetstein (1) noted two peaks during this initial degradation. The first one resulted from the residual backwash water remaining within the filter media at the end of the backwash, and the second peak was caused by mixing of a new influent with the remnant of backwash water above the media at the end of backwashing. They recommended several alternatives to reduce the initial degradation or shorten the ripening time, even though it may not be possible to entirely eliminate it: air scour or surface wash, lower filtration rates, or starting the filtration run at a very low rate and gradually increasing flow rate. Huck et al. (2) also recommended using filter-to-waste, recycling filter effluent during ripening, storing filter effluent produced during ripening for backwash water if facilities are available, and adding coagulant to backwash water or filter influent during ripening.

Moran et al. (3) investigated the extent of ripening and breakthrough for different particle sizes and the effects of design and operational characteristics (e.g., media depth, media size, and filtration velocity) on ripening and breakthrough using a natural heterodisperse suspension obtained from a lime-softening WTP after sedimentation. They found that clean bed removal efficiency increased with increasing particle size, and ripening and breakthrough were strongly dependent on particle size. They also noted that the smaller particles ripened for the longest duration, the intermediate size ripened early but decreased significantly in removal as the deposit in the bed increased, and the largest particles exhibited the most consistent removal, but as bed deposit reached high values, removal of these particles eventually degraded.

Tobiason et al. (4) used 0.27, 1.32, and 10 μm particles to investigate particle removal efficiency and found 10 μm particles had the best removal efficiency and 1.32 μm particles had the lowest removal efficiency during the ripening period. This finding is in good agreement with the model of Yao et al. (5) which predicted poor removal for the particles around 1 μm.

The duration and pattern of ripening can be influenced by many parameters including filter influent water characteristics, backwash procedure, coagulation efficiency, and other factors. These factors could either increase or decrease the effect of the different mechanisms involved and significantly affect the duration and severity of filter ripening. The filter ripening process is still not completely understood, and the initial particle breakthrough into the filtered water is not always well managed at WTPs (6). In addition, not much research has been done on the influence of particle size and surface charge of particles on filter ripening. Therefore, to ensure the safety of tap water, the understanding of ripening process in filtration is essential, and that is what this paper addresses.

EXPERIMENTAL METHODS

Min-U-Sil 5 (U.S. Silica, WV) was used for particle material during this research. These particles are white, natural crystalline silica powders in the size range of 0.6 to 5.0 μm, and the manufacturer claims that Min-U-Sil 5 is at least 99.2% SiO_2. Solid glass spheres (Potters Industries, Inc., NJ) in a size range of 0.5 to 0.6 mm were chosen for filter media. Glass beads were thoroughly cleaned before every experiment with acid and base to remove any surface impurities. The sizes of particles and glass beads used in this research represent those of full scale WTPs (7, 8).

Particle and collector surface chemistry can be modified with many techniques. pH control, alum, and polymer destabilization were used during this research. Min-U-Sil 5 was dispersed in distilled/deionized (Milli-Q water purification system, Millipore Corp., Bedford, MA) water to make the particle suspension. A schematic diagram of the experimental filtration system utilized in this research appears in Fig. 1. The clear acrylic filter column has an inner diameter of 3.8 cm with media depth of 10 cm. With this configuration, complete particle removal was avoided, and yet the filter bed was deep enough to capture the majority of particles that would be captured in a much deeper column (e.g., a full scale column). Each experiment was implemented for 4 hours with the filtration velocity of 5 m/h, yielding a final cumulative hydraulic loading (CHL, volume applied per cross-sectional area) of 20 m^3/m^2. To establish equilibrium between the solution and media in terms of surface charge, a particle-free solution with identical chemistry as in the subsequent experiment was pumped through the filter for at least 60 minutes before time zero.

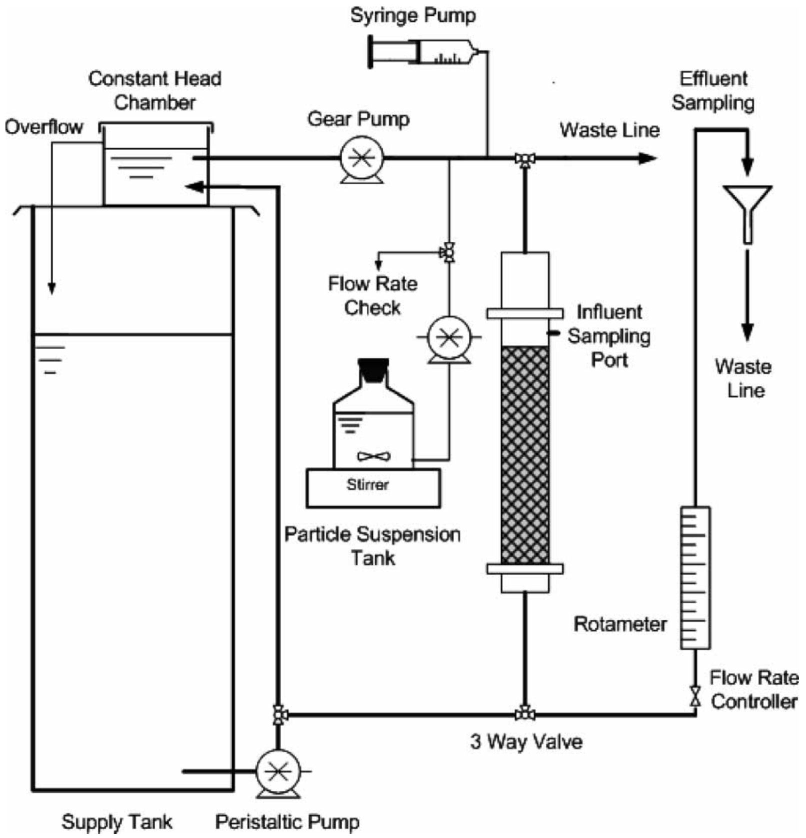

Figure 1. Schematic of experimental filtration system (not to scale).

The target influent concentration was 20 mg/L of Min-U-Sil 5; this value was decided based on the applicability of measurement techniques, the expected effluent concentration, and the applicability of the influent particle volume concentration range for a WTP. The influent concentration was always constant, but the effluent conditions varied depending on the destabilization methods.

The zeta potential distribution (ZPD) of particles was measured by a Zetaphoremeter IV (CAD, France), and particle size distribution (PSD) was measured by using Coulter Counter (Coulter Multisizer, Coulter Electronics Inc., Hialeah, FL). Solids concentrations were measured (estimated) using a Perkin-Elmer UV/VIS spectrophotometer (lambda 3), based on a calibration curve with known concentrations. The majority of particles (88.4%) have diameters in the range of 0.8 to 2.5 μm ($-0.1 < \log d_p < 0.4$). A more detailed description of the filter system and experimental methods can be found elsewhere (9).

EXPERIMENTAL RESULTS AND DISCUSSION

Ripening under pH Control

As particle transport efficiency for non-Brownian particles is proportional to the square of particle size (5), bigger particles can be more easily attached to the collectors than can smaller particles. In this research, larger particles were generally better attached than smaller particles under the same chemical conditions (e.g., same pH), especially during the initial stage of filtration. However, a more detailed view of the results reveals that the patterns of particle attachment as a function of particle size and CHL were different depending on the specific chemical conditions.

Based on the zeta potential measurements at different pH values, three different pH conditions ($\text{pH} = 3.0$, 4.0, and 5.0) were chosen at a constant ionic strength (10^{-2} M), all above the point of zero charge which is approximately pH 2.0. The pH was controlled by adding HCl, and the ionic strength was bolstered as necessary by KCl. At these low pH values, no other buffering was provided. The mean zeta potentials ($\pm$ one standard deviation) at pH 3.0, 4.0, and 5.0 were -26.4 ± 7.1 mV, -38.7 ± 9.2 mV, and -54.7 ± 9.3 mV, respectively.

Better solids removal was confirmed under more destabilized chemical conditions (i.e., smaller absolute value of zeta potential of particles) especially at the initial stage of filtration, as many other researchers have noticed. During the pH control experiments, all of the particles had negative surface charges, so less negative conditions resulted in better solids removal, at least during the clean bed removal period (i.e., the first 20 minutes of filtration). During clean bed removal, the filtration result at pH 3.0 shows slightly better solids removal, i.e., lower solids concentration remaining in the effluent, than results at pH values 4.0 and 5.0, because pH 3.0 makes less negative surface charges. On the other hand, better clean bed removal does not necessarily mean better overall particle removal. Although better clean bed removal was achieved at pH 3.0 as expected, the filters operated at pH 4.0 and pH 5.0 achieved better solids removal after a certain amount of CHL.

The particle number remaining (N/N_o) at pH 3.0 for different particle size classes was analyzed. At the beginning of the run, bigger particles were well-attached to the collectors while smaller particles were not, as larger particles have better transport efficiency. Particle removal efficiency was improved after 6 minutes ($0.5\,\text{m}^3/\text{m}^2$ CHL) for smaller particles, but for larger particles (4.0–5.0 μm) the removal efficiency did not improve. The removal of 4.0–5.0 μm size range particles exhibited some instability (some improvement, some degradation between samples) until $1.67\,\text{m}^3/\text{m}^2$ CHL, but then continuously degraded thereafter. After $10\,\text{m}^3/\text{m}^2$ CHL, better particle removal of smaller rather than larger particles was noticed. As larger particles were not relatively well removed at pH 3.0, this poor removal can be one of the main reasons for the slow ripening.

Two reasons can possibly explain the poorer particle removal of larger particles after the first few minutes. First, the surface charge conditions of bigger particles were not as favorable for attachment as they were for smaller particles. Particle number removal efficiency of smaller particles in the size range 1.0–2.0 μm kept improving, while that of larger particles such as 4.0–5.0 μm kept worsening. Second, there was a possibility of the break-off of flocs, which could increase the number of larger particles; that is, some of the 4.0–5.0 μm particles in the effluent at the later stages were flocs of smaller particles that attached individually earlier but broke off together as a floc. The latter explanation (floc break-off) seems more likely, as there is not much reason to believe that the chemical conditions would be different for different particle sizes, nor that they would change differently over time.

The particle number remaining at pH 4.0 was also evaluated. When these results were compared to those of pH 3, the 1.0–2.0 μm particles improved continuously; the 2.0–3.0 μm particles improved for a while and then were nearly flat in both cases. However, unlike the PSD at pH 3.0, at the beginning of the filtration, better particle removal efficiency was noticed for the larger size particles (e.g., 4.0–5.0 μm), and the particle removal efficiency improved from 0.33 to 5.0 m^3/m^2 CHL for all particle size ranges. Therefore, it can be assumed that, in this case, ripening was faster than it was at pH 3.0, because accumulation of larger particles can accelerate the ripening process. Solids concentration removal efficiency was better at pH 4.0 than pH 3.0 after 3.33 m^3/m^2 CHL, though the chemical conditions at pH 4.0 were less favorable than at pH 3.0. At a value of CHL $>5.0\ m^3/m^2$ at pH 4.0, only the smallest size range (1.0–2.0 μm) showed continued improvement; all three large sizes (2.0–5.0 μm) showed a slight deterioration in removal. The results for the different pH values were partially presented elsewhere (10).

Table 1 shows particle number remaining (N/N_o) at pHs 3 and 4 as a function of particle size and two different CHL values. The lower CHL value was chosen as the value where N/N_o was highest, which varied slightly between experiments. The higher CHL value was chosen as 15 m^3/m^2

Table 1. Comparison of particle number remaining in the effluent (N/N_o) at pHs 3 and 4 as a function of particle size and cumulative hydraulic loading

Particle size range (μm)	Highest value of N/N_o (CHL $\approx 0.5\ m^3/m^2$)		Value of N/N_o (CHL $= 15\ m^3/m^2$)	
	pH 3	pH 4	pH 3	pH 4
1.0–2.0	0.59	0.77	0.34	0.28
2.0–3.0	0.52	0.57	0.32	0.19
3.0–4.0	0.45	0.37	0.38	0.12
4.0–5.0	0.39	0.34	0.42	0.13

for both pH values, and is representative of the results in the latter part of the experiments. Lower values of the particle number remaining were found for all particle size ranges at pH 4.0 at the end of run (15 m^3/m^2 CHL) in comparison to the early values. These results are consistent with solids concentration remaining (C/C_o) results. In the early part of the two runs, better removal for the two larger particle size ranges (3.0–5.0 μm) was found at pH 4.0 in comparison to pH 3.0.

Figure 2 presents the mean zeta potential values and corresponding solids concentration remaining (C/C_o) with error bars at pH 4.0. Error bars indicate one standard deviation. The solids concentration remaining increased during the early stage of filtration and then kept decreasing due to the ripening effect. The mean zeta potential values moved from less negative to slightly more negative values during the clean bed removal stage (CHL < 0.5 m^3/m^2). However, during the later stage, they moved slightly from more negative to less negative values. The solids concentration remaining increased during the early stage of filtration and then decreased later. The changes of mean zeta potential values in the effluent during the initial stage of filtration can be attributable to surface charge interactions between particles and collectors. On the other hand, those of the later stage of filtration can be influenced by surface charge interactions between particles and collectors as well as by other factors (e.g., interactions between particles and previously attached particles).

Ripening under Alum Destabilization

Three alum doses (i.e., 0.06, 0.2, and 0.8 mg/L) were chosen to make negative, near zero, and positive surface charge conditions, and the

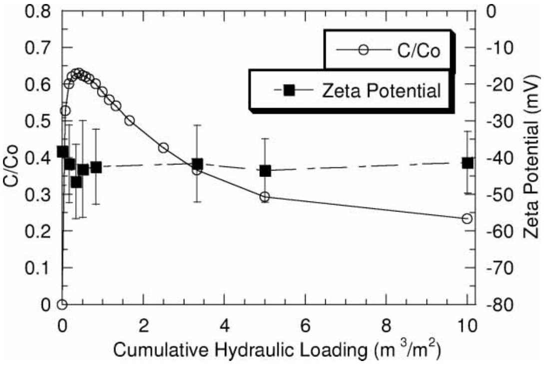

Figure 2. Effluent characteristics: solids concentration remaining (C/C_o) and mean zeta potential values of Min-U-Sil 5 as a function of cumulative hydraulic loading at pH 4.0 (error bars show one standard deviation).

corresponding mean zeta potentials (± one standard deviation) were -38.9 ± 12.1 mV, 7.9 ± 12.2 mV, and 54.3 ± 9.9 mV, respectively. To supply alkalinity that was reasonably consistent with conditions in water treatment, Min-U-Sil 5 particles were dispersed in 2×10^{-3} M $NaHCO_3$ throughout the alum experiments while pH was set to 5.2.

Figure 3 presents the solids removal at the three different alum doses as a function of CHL during the initial stage (Part A) and the 4 hour filtration experiment (Part B). Unlike the pH control experiment, no significant difference among the three different alum doses was noticed in the solids concentration remaining during the initial stage. The alum dose of 0.2 mg/L produced a favorable condition in terms of surface charge, but better particle removal was noticed for a only short period, i.e., less than $0.33\ m^3/m^2$ CHL, and after that its particle removal efficiency was worse than those of other doses.

It can be assumed that only during the clean bed removal stage is the particle removal pattern influenced mainly by interaction between collectors and particles. After that, the deposited particles can also influence the filtration efficiency, i.e., the ripening can be influenced by the PSD and morphology of the deposited particles. From Fig. 3, it is seen that a slightly better particle removal occurred at the optimum dose at least during the clean bed removal period, but that optimum dose does not always necessarily make better ripening.

The pattern of particle ripening can be assumed to be closely related to the particle removal pattern of each specific size range, i.e., a different effluent PSD (or more precisely, a different pattern of capture with respect to particle size) can cause differences in ripening even though the influent PSD is the same. As noticed during the pH control experiments, a certain size of particle removal can accelerate the ripening effect. During these experiments, as the majority of particles were non-Brownian particles, deposition of bigger particles was assumed to speed up the ripening effect when the same number of particles was removed regardless of the particle size.

The particle number remaining at the alum dose of 0.2 mg/L is shown for several size classes in Fig. 4(a). This dose was optimal from the point of view

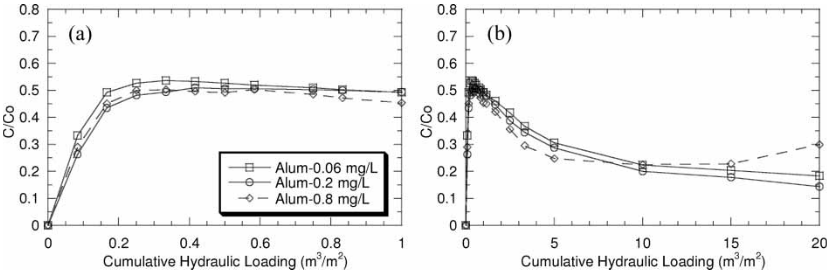

Figure 3. Solids concentration remaining (C/C_o) during the initial (a) and overall stage (b) under alum destabilization.

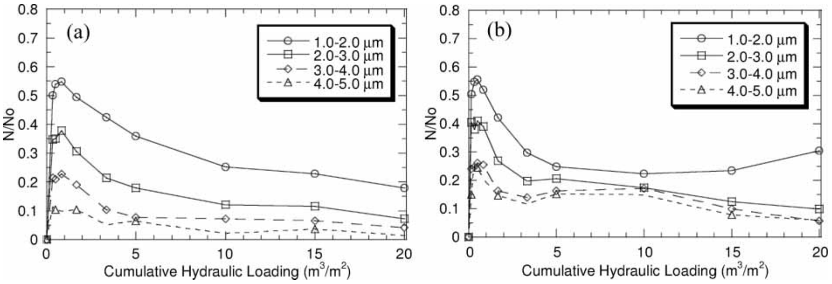

Figure 4. Particle number remaining in the effluent (N/N_o) at alum doses 0.2 mg/L (a) and 0.8 mg/L (b).

of surface charge. A relatively good particle number removal was noticed regardless of particle size, and no significant particle breakthrough occurred. On the other hand, results for particle number remaining at alum dose of 0.8 mg/L are shown in Fig. 4(b). During the initial stage of filtration, ripening happened for all particles sizes, and this may be the main reason for the good particle concentration removal shown in Fig. 3(b) between 1.7 and 6.7 m^3/m^2 CHL. Particle breakthrough was noticed at smaller particle sizes, i.e., 1.0–2.0 μm after 10 m^3/m^2 CHL. A similar trend (i.e., particle breakthrough) was noticed in Fig. 3(b) using the absorbance (i.e., solids concentration) measurement. The reasons for breakthrough of smaller particles in Fig. 4(b) can be approached from two angles.

First, the surface charge of smaller particles was unfavorable for attachment during that time. Before 5 m^3/m^2 CHL, removal of smaller particles showed good ripening, like the other size ranges of particles. After this point, though, an effect such as charge adsorption/desorption in the filter column might have made conditions unfavorable for accumulation of small particles (9).

Second, the increase of particle accumulation can lead to the increase of head loss, which in turn, leads to break-off of attached particles. Usually bigger particles are liable to be detached under increased head loss or hydraulic shock load (11) assuming surface charge is independent of particle size; thus, larger particles can be easily detached. During the detachment, there is a possibility that flocs were broken into smaller individual particles, because floc strength was so small (12).

Ripening under Polymer Destabilization

Based on the jar-test results, three different polymer (Superfloc C-572, Cytec Industries Inc., IN, dimethyl amine polymerized with epichlorohydrin) doses were chosen to make negative, near zero, and positive particle surface charges. Those doses were 0.001 ppm_v with a mean zeta potential (± one standard deviation) of −53.3 ± 13.1 mV, 0.01 ppm_v (1.0 ± 18.1 mV), and 0.1 ppm_v

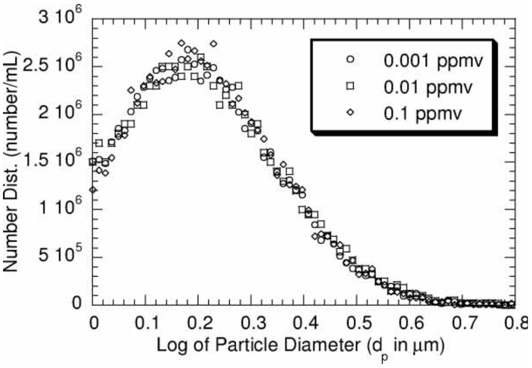

Figure 5. Particle size distribution of influents at different polymer doses (pH-5.2).

(50.9 ± 7.9 mV). Water chemistry during the polymer destabilization experiment was the same as with alum destabilization: the pH was 5.2, and the supply water was buffered with 2×10^{-3} M $NaHCO_3$.

The PSD of influent samples was analyzed for the three different polymer doses, as shown in Fig. 5. No striking differences were noticed among the different polymer doses. Mixing was sufficient to create homogeneous coagulant dispersion, but there was not enough time for coagulation and flocculation after the polymer was injected.

The values of the solids concentration remaining at different polymer doses during the early stages of filtration are presented in Fig. 6(a). The three different polymer doses led to three different particle removal efficiencies. The optimum dose (0.01 ppm_v) had the best particle removal efficiency, as expected, while the above-optimum dose (0.1 ppm_v) showed the worst particle removal efficiency. In terms of ripening, quick ripening was noticed at the optimum dose, while no significant ripening was noticed at the below-optimum dose (0.001 ppm_v) during the initial stage of filtration. The effluent concentration kept deteriorating slightly with time at the

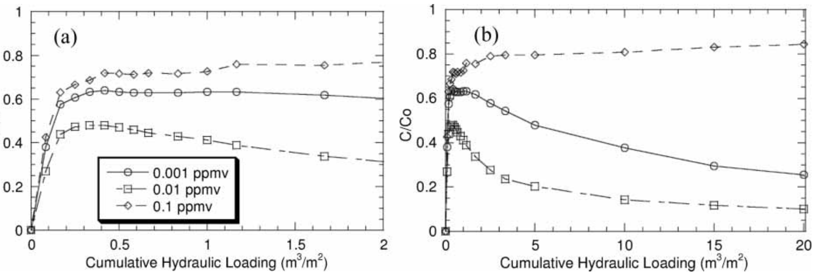

Figure 6. Solids concentration remaining (C/C_o) in the effluent at different polymer doses during the initial stage (a) and overall (b).

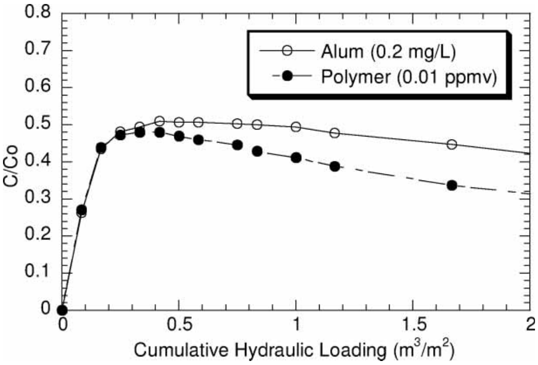

Figure 7. Solids concentration remaining in the effluent (C/C_o) at optimum doses of alum and polymer.

above-optimum dose (0.1 ppm_v), which means ripening did not happen at this polymer dose in the first few minutes of filtration.

The solids concentration remaining during $20\,m^3/m^2$ CHL is shown in Fig. 6(b). Though not much ripening was noticed during the early stage of filtration at the polymer dose of 0.001 ppm_v, substantial ripening was noticed after a certain amount of CHL (e.g., 2 m^3/m^2). However, ripening did not happen for the above-optimum polymer dose (0.1 ppm_v). Unlike alum coagulation, where ripening was noticed at all of the alum doses, ripening was not noticed at the higher polymer dose (i.e., when the surface charges of particles were positive).

Figure 7 shows the solids concentration remaining at the optimum doses (i.e., similar surface charge) of both alum and polymer; more rapid ripening was noticed with polymer than with alum coagulation. From this graph, it can be deduced that the polymer is a more efficient coagulant for accelerating the ripening process. Alum is capable of coagulating both turbidity and organics to a good extent, but it produces fragile and relatively weak flocs. On the other hand, the polymer can make larger and stronger flocs, but it has no effect on organics removal and is effective only over a smaller dose range. Therefore, to maximize the benefits of both coagulants, they can be used simultaneously at WTPs (13).

SUMMARY AND CONCLUSIONS

The major observations based on these filtration experiments are stated below.

1. Conditions with a smaller surface charge (i.e., smaller absolute value of the influent zeta potential) lead to better particle solids removal, at least during the initial particle removal stage. In this case, the initial particle removal efficiency was mainly determined by chemical interactions between particles and collectors, i.e., the zeta potentials of particles and collectors.

2. Better initial particle removal does not necessarily mean better overall particle removal. It can be assumed that during the initial stage, chemical parameters such as zeta potential are important, but after a certain amount of time their importance is decreased, depending on the specific chemical conditions.
3. The influent PSD and the removal of certain size particles during the initial stage can significantly influence ripening, which in turn, can influence the overall particle removal efficiency. When the same numbers of particles are removed during filtration, removal of larger particles leads to fast ripening because of their large volume.
4. When the time needed for the ripening process at the optimum doses of alum and polymer was compared, less time was needed for polymer destabilization. Therefore, to reduce the time needed for ripening, the polymer can be used.

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