Particle count and size alteration for membrane fouling reduction in non-conventional water filtration

A. Adin
Environmental Sciences Division, The Hebrew University of Jerusalem, Jerusalem, Israel 91904

Abstract If coagulation is not completely successful and produces aggregates which are too small, fouling may increase. In some cases, a deep-bed filter could perhaps provide a solution. The paper examines these effects using experimental results for different waters. Activated sludge effluents, stormy seawater containing microalgae and spent filter backwash water (SFBW) were coagulated by alum or ferric chloride. Sand filtration tests were carried out. Tests were performed in a membrane filtration stirred cell, filtration pilot plant equipped with SDI analyzer (seawater) and pilot UF plant (SFBW). For activated sludge effluent, alum residual ratio curves of turbidity and total particle count (TPC) followed one another. With ferric chloride, low coagulant dosage showed negative turbidity removal. Contact granular filtration reduced membrane fouling intensity. Increasing the dose resulted in higher improvement in membrane flux. For seawater, a filter run period under storm conditions reached 35 hours with satisfactory filtrate quality. An iron chloride dose of 0.3 mg/l during normal conditions and 0.5 mg/l for stormy condition should be injected, mixed well before the filters, while maintaining 10 m/hr filtration rate and pH 6.8 value. For SFBW, alum flocculation pretreatment of SFBW was effective in reducing turbidity, TPC, viruses and protozoa. SFBW settling prior to flocculation did not enhance turbidity and TPC removal. The largest remaining particle fraction after alum flocculation was 3–10 µm in size, both Cryptosporidium and Giardia are found in this size range. Coagulation enhanced the removal of small size particles, a positive impact on reducing membrane fouling potential.

Keywords Membrane filtration; membrane fouling; membrane pretreatment; particle separation; tertiary treatment

Introduction Various types of water which are potentially to be treated by membranes, e.g. municipal effluents, sea water and spent filter backwash water, contain dissolved matter, colloidal suspensions and large particles, a considerable part of them organic in nature. Particles and dissolved organic matter (DOM) separation can protect water transport systems and membranes from clogging and, as a result, make the process more economical. Coagulation has a significant effect on the particle size distribution (PSD) in comparison to the original PSD of the water (Adin et al., 1998). The efficiency of the coagulation affects directly the efficiency of the back transport mechanism of particles from the membrane surface and as a result the fouling intensity. In general, efficient flocculation produces larger flocs which can be transported away from the membrane by lateral migration or shear forces. At the same time, there is a probability to produce small flocs (colloid–colloid interaction) or small colloids which are created by complexion of DOM-iron or aluminium species. The above mentioned possibilities can affect the membrane fouling negatively, due to the fact that these new flocs are too large for brownian motion (another back transport mechanism) than with their primary size before the physical chemical treatment (Soffer et al., 1999).

Treatment of surface waters by granular filtration has long been recognized as a major barrier to the spread of disease causing agents by drinking water. Granular filtration has been incorporated as a major treatment phase in surface water treatment especially for the removal of Giardia and Cryptosporidium. These protozoan parasites have been found insensitive to the traditional disinfection methods used in the water industry. Material retained within granular filters during filtration (i.e. organic and inorganic particles,
bacteria, viruses and parasites) is removed by backwashing the filter with clean water (Nasser et al., 2002). The volume of Spent Filter Backwash Water (SFBW) generated is typically between 3–7% of the finished water produced and it is often discharged to the sewage system or recycled within the treatment plant.

Seasonal weather changes promote southern high-speed winds in the northern Red Sea coast, mixing the water column and releasing nutrients from the bottom of the sea. In that case, turbidity level instantly goes up while micro-algae of the *Synechococcus* type, starts blooming, causing fouling in desalination RO membranes and eventually shut-off of the plants for long periods (Tenzer and Adin, 1999). Neither filtration mathematical models nor field experience under normal filtration conditions have been able to provide appropriate pretreatment solutions to this problem.

This paper examines pre-treatment by coagulation and possibly filtration of these different waters in order to reduce membrane fouling, with the notion that a common denominator exists which is the fouling mechanisms themselves.

**Materials and methods**

**Secondary effluents**

This part of the work examines the possibility of improving quality of activated sludge effluents treated with iron coagulation before using membrane filtration as an advanced treatment process. It is done by right selection of physical–chemical treatment combinations and determination of pretreatment capability with iron salt as a substitute for aluminum salt, increasing membrane run time and providing better understanding of granular and membrane filtration mechanisms. Activated sludge effluents from the Dan Region Sewage Reclamation Project served in the investigation. Two flocculants were tested separately: alum Al₂(SO₄) which is long standing in water and wastewater treatment and ferric chloride FeCl₃. The flocculants were tested by the conventional jar test procedure to define optimal conditions (pH, dose). Filtration tests were carried out in bench-scale columns, 20 cm and finally 100 cm deep, 1.79 mm sand grain size. Membrane filtration tests were performed in a stirred cell using different MWCOs (molecular weight cut off). The membrane inlet was fed with wastewater with or without coagulant addition (in-line flocculation).

**Backwash water**

Spent filter backwash water (SFBW) was collected from four different water treatment plants and analyzed for microbial and physical parameters. The drinking water plants all treat surface water, either from the Jordan River or from Lake Kinnaret (the Sea of Galilee). SFBW samples were collected in sterile bottles and transported to the laboratory on ice in a cooler. Fecal coliform, Cox A9 virus, naturally occurring bacteriophages and parasite cysts and oocysts were enumerated and identified applying known methods. SFBW samples were also collected in 20 L containers and transported to the laboratory and refrigerated at 4°C. Turbidity and pH measurements were taken every minute over the course of the backwash run. SFBW samples were analyzed for turbidity, pH, TSS, DOC, UV-254 absorbance and alkalinity. All analyses were conducted as specified in *Standard Methods* (Standard Methods, 1999). Standard jar tests were conducted with alum. Samples were analyzed for turbidity, UV 254 nm, DOC, total particle count (TPC) and size distribution (PSD), alkalinity and pH. Jar tests were also conducted on SFBW samples spiked with *Cryptosporidium parvum* and Cox A9 Virus. UF tests were performed in the lab and in one of the plants, applying the ZW-10 model.

**Sea water**

Filtration rate, filter bed depth, sand grain size (fine vs coarse), iron chloride dose and polymer aid have been the main parameters of the study. Membrane feed water quality criteria
were less than 0.2 NTU for turbidity and SDI less than 3. The research consisted of two parts. First, flocculation and simulated contact and direct filtration of artificial and natural sea water with various clay-algae-pH combinations were studied in the laboratory. Secondly, a dual-column pilot filtration system was installed at the “Sabha” RO plant which treats Red Sea water and is operated during winter, spring and summer, varying the above design parameters. When the sea water did not provide a turbid situation, the specific algae (grown in situ) and clay were injected before the filters to simulate storm conditions of up to 10⁶ cells/ml and 5 NTU respectively. Iron residual, SDI, particle size distribution (PSD), pH, temperature, microbial count and head loss across the filter columns were monitored as well.

Results and discussion

Wastewater

The flocculation tests showed that the efficient range of pH for alum is pH 6–8. In this range there is a direct relationship between the dose and the turbidity and TPC (Total Particle Count) residual ratio. The optimal point is pH 6 and alum dose of 30 mg/l. At this point the turbidity residual ratio was 0.11 and the residual ratio of TPC was 0.07. For ferric chloride pH 4.5 was the best for removal efficiency. At pH 5 the residual ratio of turbidity was 0.16 and the residual ratio of TPC was 0.01. For ferric chloride, in contrast to alum, it was found that the efficient pH range was larger, pH 3–10. Nonetheless, it has been found that the charge neutralization mechanism that prevails in the acidic pH range is more effective for higher removal. While with alum, the residual ratio curves of turbidity and TPC followed one another, for ferric chloride it was different. At low coagulant dosage the turbidity residual ratio exceeded 1 (i.e. negative removal), leading to a second minimum in the turbidity curve. This phenomenon was probably a result of the formation of tiny colloids of iron hydroxide Fe(OH)₃(s).

In light of the jar test results, ferric chloride was used in the contact filtration process. At first, filtration tests were done in small 20 cm deep filters, at a filtration rate of 10 m/h, which is in the middle of the economic domain. Initial tests led to filter media grain size selection of 1.79 mm. While the optimum pH and particle interaction mechanisms for contact filtration corresponded well with the above mentioned jar test results, the optimum doses that were observed in the filtration were half or less than half than the optimal doses produced by the jar tests. It was also found that overdose for filtration was noticed in lower dosages than for the jar test. From this it can be understood that the filtration process is more sensitive to deviations from effective conditions. This sensitivity becomes higher for the sweep coagulation (basic pH) domain as a destabilization mechanism. Head loss increased moderately with increasing coagulant dose and with pH decrease from 10 to 7. Under such conditions the filter experienced short filtration runs and deteriorated filtrate quality. Energy loss increased considerably below pH 7 (charge neutralization zone).

Jar test results, at pH 7.5 (natural pH) and pH 5.5 show that the optimum coagulant dosage which was required for DOM (expressed in terms of UV-254 nm or DOC-dissolved organic) removal was higher than for particle removal, and the removal percentage was lower. Under such conditions, the positive influence of decreasing the pH level was even more significant than the case of particle separation as the above mentioned. It was observed that there is no guarantee that applying UF (ultrafiltration), even in conditions of coagulant addition, can succeed in improving DOM removal (e.g., membrane with MWCO of 50 kDa gave DOM removal close to jar test results, around 30–50%). This MWCO membrane also was penetrated by small colloids of Fe(OH)₃(s) and as a result supplied negative turbidity removal. From here, it was necessary to use LMWCO (low MWCO) UF. Significant improvement of DOM removal was achieved just with the decreasing of
membrane MWCO to 4 kDa. The DOM removal increased with increasing coagulant dose and decreasing pH. Usually, the contribution of membrane filtration on DOM removal efficiency relative to the jar test was higher for the natural pH than acidic pH.

In order to reduce the sensitivity of membrane filtration to PSD varieties, in-line filtration of wastewater coagulant suspension was performed before the membrane filtration stage. The granular filtration system included 100 cm deep bed filters containing coarse sand of 1.79 mm average grain size. In most of the cases, this pre-treatment can reduce membrane fouling intensity. The fouling improvement was dependent on the coagulant dose. In general, increasing the dose gave better improvement in membrane flux. Nanofiltration tests provided better removal efficiency, but the price was increasing sensitivity to internal fouling.

**Sea water**

Jar test results showed less dependency of removal efficiency on pH at 5 and 10 NTU (better kinetics) while lower turbidity was better removed at acidic pH (better charge neutralization). Micro-algae/clay mineral mixtures at pH 6 were better removed than the mineral colloids alone. Modified jar tests resulted in 99% removal at pH 6 for clay and somewhat less for algae alone; contact filtration outperformed direct filtration. Clay/algae mixtures were 95%–99% purified by both processes at natural pH at a wide range of flocculant doses (0.5–20 mg/l).

Filter column pilot results recommend, considering storm conditions, deeper (1.2 m) and coarser (1.0 effective grain size) media than commonly applied. An iron chloride dose of 0.3 mg/l during normal conditions and 0.5 mg/l for storm conditions should be injected, well mixed before the filters while maintaining 10 m/hr filtration rate and pH 6.8 value. The coarse media reduces pressure loss, while the extra depth compensates for the reduction in grain surface area available for particle contact. Thus, filter run length under storm conditions reached 35 hours while achieving water quality goals. Experiments with existing filter media (0.9 m deep, 0.9 mm g.s.), resulted in shorter runs. However, the existing filters can still perform well if the flocculant dose is raised to 0.5 mg/l and polymer is added. The way the filters reacted to the design parameters and water quality variations confirms with the preliminary hypothesis, that deep-bed filtration theory, mechanisms and process optimization for surface water may be applied to sea water. In addition, column and field observations indicate that the controlled formation of micro-flocs ahead of the filter (i.e. direct filtration) bed may further improve the filtration process, a matter which deserves future investigation. The ripening stage of the fine filter regarding the residual total iron, was 3 hours until reaching values below 0.020 ppm vs. 5 hours in the coarse filter (Figure 1).

**Backwash water**

Fecal coliform was detected in all SFBW samples from two tested drinking water plants at a concentration range of 2–4 MPN/100 ml in one plant and 500–1,600 MPN/100 ml in the other. Fecal coliform was not detected in the SFBW from two swimming pool treatment plants. SFBW from the two drinking water plants was also found to be positive for F+ bacteriophages and cultureable enteric viruses, whereas swimming pool samples were found to be negative for both groups of microorganisms. Cryptosporidium oocysts were detected in 5 out of 6 (83.3%) SFBW samples from drinking water plants, at a concentration range of 0–3 oocysts/l. Giardia cysts were detected only in one sample of SFBW from each location (33.3%) at a concentration range of 0–0.5 cysts/l.

There was considerable variability recorded in the physico-chemical quality of SFBW samples from the different drinking water treatment plants due to raw surface water quality and operational practices. The highest variability was recorded for TSS and turbidity,
46–384 mg/l and 28–370 NTU, respectively. In comparison, lower variability was observed in the concentration of organic material in the SFBW (composite DOC ranged between 3–13 mg/l).

Jar-tests were performed to determine the reduction of turbidity, Cryptosporidium oocysts and Coxsackie A9 Virus by alum flocculation. Coxsackie A9 virus and Cryptosporidium were seeded into composite SFBW samples (turbidity 145 NTU) and coagulation was conducted with 40 ppm alum. Reductions of 93% were observed for turbidity and Cryptosporidium and of 73% for Cox A9 (Figure 2).

To evaluate the retardation of microbes by ultrafiltration (UF), a hollow-fiber immersed membrane (Zeeweed-10) unit (production 20 l/hr) with a molecular weight cutoff of...
approximately 200,000 Da was applied for the removal of MS2 bacteriophage (25 nm). A reduction of 99.999% was observed for MS2 from tapwater. Moreover, MS2 removal was not influenced by kaolin turbidity.

The results of this study indicate that the microbiological quality of SFBW depends on the water source and on the treatment practices. Furthermore, coagulation/flocculation reduces the microbial and particle loading of SFBW which may result in improved quality and reduced fouling potential of the UF membrane. Figure 3 shows a greater than 80% removal of both turbidity and particle counts for SFBW treated with alum.

Conclusions
1. **Activated sludge effluent.** The charge neutralization mechanism was more effective for higher removal. While with alum residual ratio curves of turbidity and total particle count (TPC) followed one another, for ferric chloride it was different. At low coagulant dosage, turbidity showed negative removal, leading to a second minimum in the turbidity curve. That phenomenon was probably a result of formation of tiny colloids of iron hydroxide Fe(OH)₃(s). Contact granular filtration reduced membrane fouling intensity. Increasing the dose resulted in higher improvement in membrane flux. Nanofiltration provided better removal efficiency, but sensitivity to internal fouling increased greatly.

2. **Seawater.** Column pilot experiments showed good results with deeper (1.2 m) and coarser (1.0 mm effective grain size) than usually applied. A filter run period under storm conditions reached 35 hours with satisfactory filtrate quality. An iron chloride dose of 0.3 mg/l during normal conditions and 0.5 mg/l for stormy condition should be injected, mixed well before the filters, while maintaining 10 m/hr filtration rate and pH 6.8.

3. **SFBW.** Alum flocculation pretreatment of SFBW was effective in reducing turbidity, TPC, viruses and protozoa. SFBW settling prior to flocculation did not enhance turbidity and TPC removal. This finding might imply that settling would not be required prior to UF. The largest remaining particle fraction after alum flocculation was 3–10 µm in size, both Cryptosporidium and Giardia are found in this size range. Coagulation enhanced the removal of small size particles, a positive impact on reducing membrane fouling potential.

4. **General.** Particle size variation by coagulation directly affects particle back transport mechanism efficiency from the membrane surface and, as a result, fouling intensity. In general, efficient flocculation produces large, light flocs that can be transported away from the membrane by lateral migration or shear forces. At the same time, formation of small flocs, or small colloids which are created by complexion of DOM-iron species, is also possible. The latter may enhance membrane fouling, since they are too small to be affected by the above mentioned mechanism but also too large for effective brownian motion (another back transport mechanism).

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