Contact filtration of humic waters: performance of an expanded clay aggregate filter (Filtralite) compared to a dual anthracite/sand filter

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Abstract Contact filtration performance of a coarse expanded clay aggregates filter (Filtralite) is compared to a conventional dual media anthracite/sand filter. In a contact filtration pilot plant, humic water was coagulated using aluminium sulphate (AS), iron chloride sulphate (ICS), poly-aluminium chloride (PAC) or the bio-polymer chitosan. Filtration experiments were performed at optimum pH and dose conditions for each coagulant found from previous optimisation experiments in the pilot filter plant. The results reveal two main problems in coagulation/filtration of humic waters: short filtration run times for the most commonly used metal salts, and metal residuals in treated water. The best ways to overcome these problems in raw waters high in organic matter, was found to be by the use of a coarse filter bed in combination with chitosan, or a metal salt with the addition of polymer as filter aid. An increased turbidity level in the raw water (as bentonite clay) was found to increase the removal of humics in most cases, as well as increase the filterability of the flocs formed.

Keywords Contact filtration; expanded clay aggregates; humic substances

Introduction

The removal of humic substances is a major challenge in water treatment in Norway as well as in many other parts of the world. Humic substances may not be harmful as such, but they are causing colour, taste and odour, adsorbing micro-pollutants and causing formation of disinfection by-products (DBPs) upon chlorination. Humic lake water with high colour, low turbidity and low temperature is often used as a water source in Norway.

Coagulation with metal-salts, i.e. aluminium sulphate, in a conventional dual media anthracite/sand filter optimised for humic substances removal, results in short filter runs (Eikebrokk, 1999). This is a result of early breakthrough of weak voluminous hydroxide-humic flocs (Rebhun *et al*., 1984). A typical problem is too high residual metal concentration in the produced water (Eikebrokk, 1999), when a metal salt is used. There is, therefore, a need for improvement of the traditional coagulation–filtration process in which both the coagulation and the filtration step is optimised with regards to the removal of humic substances.

Use of expanded clay aggregates as a filter media

Expanded clay aggregates (Filtralite) are made from burning of clay at about 1,200°C, followed by crushing and sieving to get the desired size fractions. One has the possibility of making clay aggregates with different density, and this is utilised in a deep bed down flow filter. The challenge of this work was to utilise the storage capacity of the more coarse grained Filtralite filter by choosing the correct pre-treatment.

In an anthracite/sand filter, the specific grain density difference of anthracite and sand ensures that stratification of the two layers are obtained. The Filtralite filter beds used in

this study consisted of two different density materials. To obtain the desired stratification of the filter bed after backwashing, the top layer of large grains should have a low settling velocity, i.e. low density, compared to the bottom layer of smaller grains. The top layer consisted of Filtralite NC 1.6–2.5 mm, and the bottom layer of Filtralite HC 0.8–1.6 mm, where NC and HC are normal and high density crushed material, respectively. The majority of the experiments have compared a Filtralite filter with an anthracite/sand filter at equal filter bed depths. In addition, some experiments were performed with a deeper Filtralite filter. The filter beds are schematically illustrated in Figure 1.

Experimental

A pilot filter plant consisting of two parallel filter columns made it possible to compare two filter beds receiving equally treated raw water. The plant consisted of a raw water tank, where humic concentrate and/or bentonite clay suspension were mixed with tap water (turbidity <0.3 NTU). Four different raw water types were tested: RW15, RW50, RW15/3 and RW50/3, where 15 and 50 is mg Pt/l colour, and /3 denotes the addition of bentonite equalling 3 NTU in turbidity (about 9 mg/l). The raw waters was pH adjusted with HCl to optimum pH for the particular coagulant, before coagulant addition in an in-line mixer. The coagulated water was then directed to the two filter columns. Filtrate pH and turbidity were measured on-line, as well as head loss distribution in the filter bed, and data were stored on a computer. Flow through the filter columns was also automatically recorded and connected to a regulation circuit ensuring constant flow conditions.

Samples of raw water and filtrate were collected and analysed for pH, turbidity, true colour and TOC in the lab, while samples of coagulated water were analysed for suspended solids and zeta-potential.

Results

Effects of grain size and bed depth

The filter media grain size is an important treatment plant property which influence both filtrate quality and plant operation. In the experiments the effect of different grain sizes on
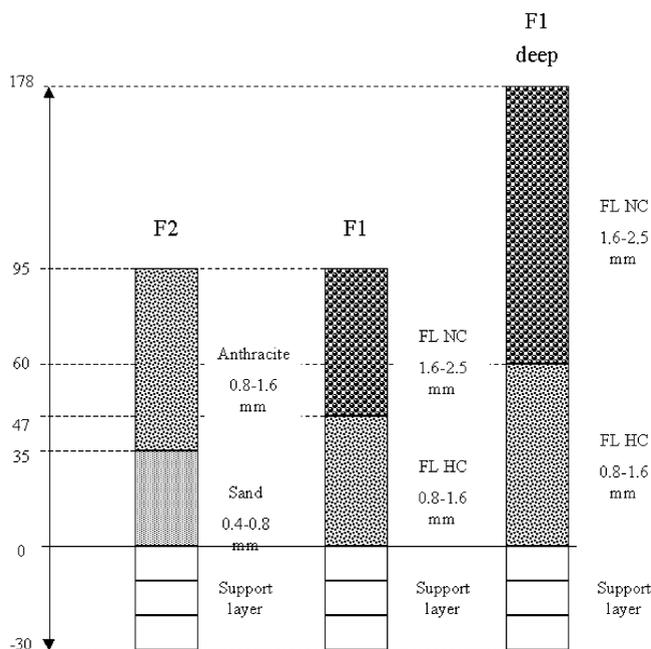

Figure 1 Schematic illustration of the filter beds used in this study (measures in cm)

filter performance could be studied for several different coagulants. The effect of filter bed depth was studied by creating a deeper Filtralite filter. Initially the depth of the Filtralite filter (F1) was chosen to be the same as for the anthracite/sand filter (F2) in order to compare results at equal depths. In the deeper Filtralite filter (F1+), depths were selected from the relationship of L/d_e (depth/effective grain diameter) (Ives and Sholji, 1965). To see if an increasing bed depth could compensate for coarser grain sizes the layer depths of the coarse Filtralite filter was increased to equal L/d_e as in each layer of the anthracite/sand filter.

The results in Figure 2 show the differences in on-line effluent turbidity, effluent metal residuals and removed fractions of TOC for the three filters treating RW50 and RW50/3. Some of the results are especially high in residual metal (total metal concentration), and this is probably a result of post-precipitation of hydroxide. The coagulation pH was lower than in the effluent water pH due to alkaline filter media (both Filtralite and anthracite gave a pH increase) resulting in high non-removable soluble metal fractions before and in the filter. From Figure 2 it can be seen that using aluminium sulphate as coagulant with RW50 result in a high effluent turbidity from the coarse F1 filter.

This high effluent turbidity is mainly particulate aluminium hydroxide penetrating the filter, giving high metal residuals. The effluent turbidity leakage does not have a significant effect on TOC removal, implying that precipitated aluminium with little humics associated with it is responsible for the effluent turbidity. The effluent turbidity using AS is about 0.25 NTU for the deeper F1+ filter, and even lower for the F2 filter. This shows that when aluminium sulphate is used when coagulating humic substances it is not sufficient to expand the filter depth according to the relationship of L/d_e in order to achieve equal effluent turbidity. The reason for this is probably weak voluminous hydroxide-humic flocs that break up in the filter, and create a rapid head loss increase because the deposits occupy relatively large volumes.
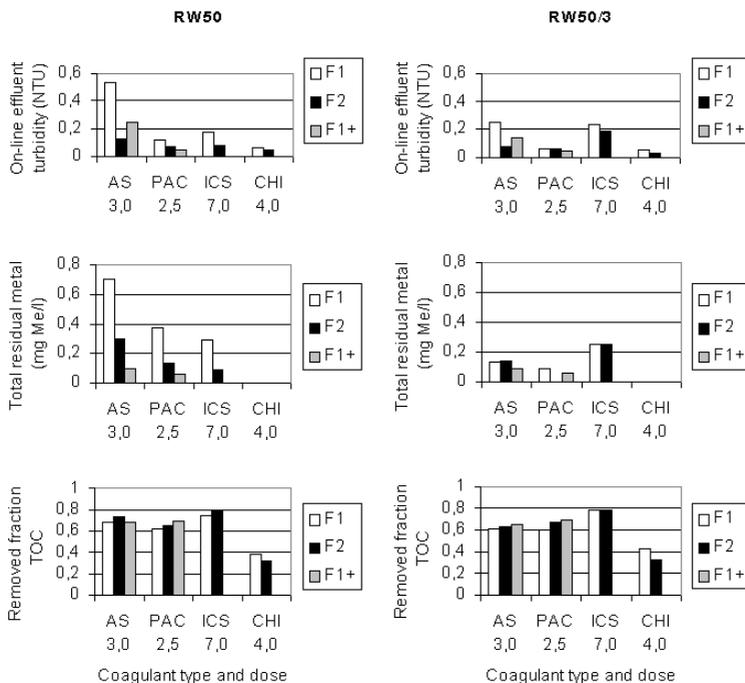

Figure 2 Effluent water quality from filter F1, F2 and F1+, treating RW50 (left) and RW50/3 (right) at a filtration rate of 7.5 m/h

Using PAC for coagulation gives a much lower effluent turbidity from all three filters, especially from F1. The aluminium residual is still high from F1, despite the low turbidity, probably a result of some post-precipitation. This is not seen for the F1+ filter, and this might be due to a higher number of possibilities for direct precipitation on filter grain surfaces.

For ICS and the other two metal salts there is a clear connection between effluent turbidity and metal residuals. This is an indication of particulate metal creating the turbidity in the effluent. All the experiments shown are performed at a pH of metal solubility below 0.1 mg Me/l.

When the cationic polymer chitosan is used an excellent effluent turbidity is achieved in both F1 and F2 and, of course, no residual metal problems arise. The TOC removal however, is less than for the metal based coagulants, about 40% for chitosan compared to about 70% with a metal coagulant. Chitosan contains about 60wt% TOC, and this has probably an effect on the TOC removal for this coagulant. ICS remove generally more TOC than the aluminium coagulants. No big differences in TOC removal is seen between the three filters.

The raw water containing colour and turbidity show generally lower effluent turbidity and lower metal residuals than the coloured, low turbidity raw water. The difference in effluent turbidity between F1 and F2 when AS is used, is much less than for the other raw water. This indicates that the flocs created with clay particles in the water are more filterable in the coarse Filtralite filter. When the effluent turbidity due to particulate metal using AS and PAC is lower so is also the metal residuals. It might be that the clay particles in the water enhance hydroxide precipitation, and that relatively more metal is precipitated with more turbidity in the raw water. For the iron coagulant the results for effluent turbidity and residual iron are more equal for filter F1 and F2 treating RW50/3 than RW50, as for the other coagulants, but the values are higher for RW50/3. This might be a result of the high organic matter removal efficiency for this coagulant, making it less effective for turbidity removal compared to the aluminium-based coagulants.

There are no big differences between the three filters in the removal of TOC, and the fractions removed are about the same for RW50/3 as for RW50. This indicates that clay turbidity has a much less coagulant demand than humic substances.

Figure 3 shows total head loss increase with time and distribution at breakthrough (filtrate turbidity >0.3 NTU) in the filter beds when PAC and chitosan (CHI) are used as coagulants with RW50.

The total head loss curves show the large difference in head loss build up with time for filter F1 and F2, while the deeper F1+ have almost similar development as F1. The highest value for each curve indicates termination of the run, and it is demonstrated that the runs with CHI have remarkable longer run times than the runs where PAC is used as coagulant.
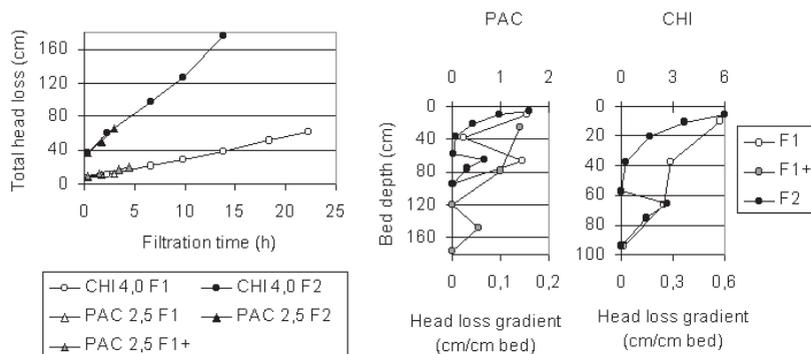

Figure 3 Total head loss and head loss gradient distributions for PAC and CHI treating RW50 (equal run times for each coagulant, filtration rate 7.5 m/h, F1 and F1+: bottom axis, F2: top axis)

This is a result of less suspended solids in the influent water, due to no hydroxide precipitation, and more filterable flocs/resistant deposits created with the polymer than with the metal coagulant. The steep slope for the anthracite/sand filter results in termination by head loss limitations when CHI is used, while all the other runs are terminated by breakthrough. Because the head loss build up with time is equal for both coagulants in each filter, the head loss increase per solids load is higher when using CHI. This implies that the deposits use more pore space to store equal amounts of solids (assuming none or equal concentration of suspended solids in filtrate).

The head loss gradient curves shown in Figure 3 give cm head loss increase per cm bed depth due to deposition, between the pressure measuring points in the filter column, when the initial gradient (clean bed) is subtracted. It gives a picture of the deposition pattern in the depth of the filter bed. However, deposition of 1 mg suspended solids in the top layer (anthracite or Filtralite NC) does not give the same head loss increase as deposition of 1 mg in the bottom layer (sand or Filtralite HC).

The size of the gradients are about ten times higher in F2 than in F1 and F1+. The fact that turbidity breakthrough occurs almost after the same run time in F1 and F2 when PAC is used, shows that the deposits in the fine-grained filter are able to resist much higher shear forces than the deposits in the coarse filters. The shape of the curve in F2 is almost identical for the two coagulants, with high gradients in the top section of each new layer dropping to zero in the bottom section of the layer. In F1 when CHI is used the deposits are stored more evenly in the filter bed, than when PAC is used.

Polymer as filter aid

A high molecular weight, non-ionic, synthetic polymer (polyacrylamide) was tested as a filter aid in order to see if the turbidity breakthrough could be postponed, when a metal salt was used with coloured water. Initial experiments indicated that a few minutes reaction time after coagulation before polymer addition was needed to get an effect of the filtration aid. In Figure 4 some results for experiments with aluminium sulphate, alone as well as AS together with polymer as filter aid, are shown for filter F1 and F2. The polymer dose used was 0.05 mg/l.

When a filter aid is used in combination with AS as primary coagulant, the effluent
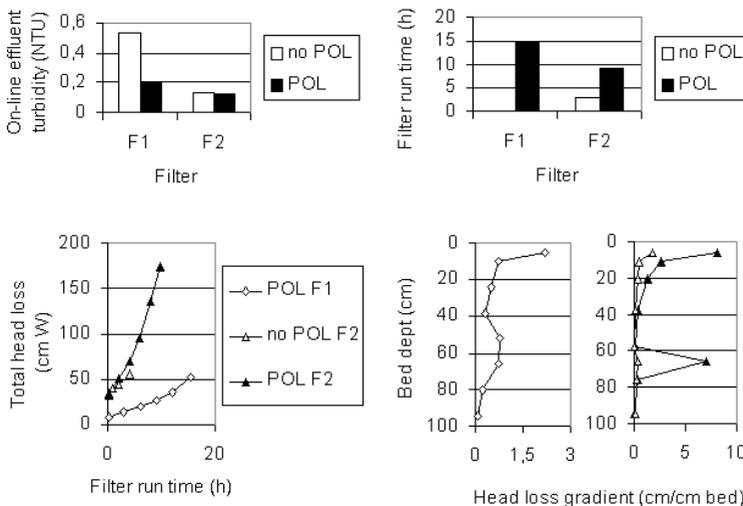

Figure 4 On-line effluent turbidity and length of filter run for experiments with and without polymer as filter aid treating RW50 (coagulant dose 3.0 mg Al/l as aluminium sulphate, polymer dose: 0.05 mg/l, head loss gradients are at run termination in each case)

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turbidity from the Filtralite filter (F1) is reduced to 0.2 NTU. The result of polymer addition was bigger flocs clearly visible in the water over the filter media. These flocs were much more filterable in the coarse filter grains of filter F1.

There is almost no reduction in effluent turbidity for the F2 filter when polymer is added. The filter run lengths, however, are considerably longer when the non-ionic polymer is used. The run length for F1 without polymer is not shown because the criteria for break-through termination (turbidity <0.3 NTU) was never obtained, but the period of stable effluent turbidity was about 3 hours (as for filter F1). A run length of 15 hours is obtained in F1 before breakthrough, and about 9 hours in F2 before head loss termination when adding polymer. The total head loss curve for F2 with polymer indicates that the polymer creates such big flocs that surface deposition occur, with an exponential head loss development with time. From the head loss gradient distribution, it can be seen that there are very high gradients in the top of the anthracite layer and in the top of the sand layer. The deeper sections of sand is almost not in use, because very few of the big flocs reach this layer. In comparison, the distribution of gradients in F2 for the experiment with only AS show much lower values, even though breakthrough occurs at that point in time shown.

The total head loss curve of filter F1 shows a more linear trend, implying that the size of the flocs are suitable for the filter grain size. The deposits are clearly distributed more even-ly in the filter bed in F1 than in F2, utilising the whole depth of the filter. When the run is terminated by breakthrough in F1, the possibility of increasing the polymer dose, resulting in extended run times, exists. In F2, the polymer dose could be decreased to optimise filter run length.

Head loss and breakthrough

Rebhun *et al.* (1984) suggested that deep bed filtration proceeds in working layers, imply-ing that in layers above the working layer there is no removal due to saturation, and in layers below the working layer there is no removal because the inlet concentration is close to zero. The head loss gradients for each section in a filter as a function of time was calculated. This resulted in curves like the ones shown in Figure 5, where the experiment treating RW50/3 with CHI in filter F2 is presented. The curves show mostly a linear trend with time for the sections of the anthracite layer, but the slope varies greatly with bed depth.
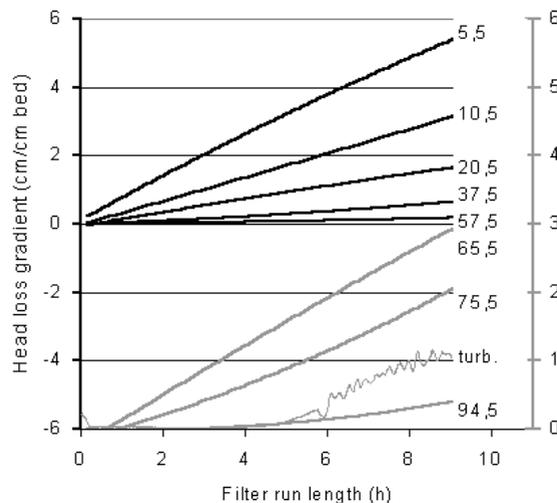

Figure 5 Effluent water turbidity and head loss gradients for each section of filter F2 (RW50/3, coagulant dose 4.0 mg/l CHI. Grey lines: sand gradients (right axis), black lines: anthracite gradients (left axis), num-bers indicate bed depths; slim grey line: effluent turbidity)

In the bottom anthracite section, there is almost no deposition of solids. The head loss increase for the sand layer starts with a little delay, and the delay is longer for sections further down in the sand. This is probably an indication of saturation with deposits in the above section of the bed. When a head loss gradient starts to develop in the bottom section of the sand, there is a breakthrough in turbidity at the same time. In relation to the working layer theory of Rebhun *et al.* (1984), the incipient head loss increase of a filter bed section is an indication of saturation conditions in the upper adjacent layer. From the figure it can be seen that almost all the sections of the anthracite layer start to develop head loss gradients just after start of the filtration run, showing that the working or effective layer is variable in depth. Other experiments like this also show the same thing, but with different head loss development with time. As seen the head loss continues to grow after saturation, and this can be a result of small flocs depositing at the same time as bigger clusters of flocs detach. This is experienced by several researchers (for example, Hunt *et al.*, 1993).

These results and others (Saltnes, 2002) obtained in this project but not shown here, indicate that the deep bed contact filtration process is much like an adsorption column process. The pore space of the filter media is saturated with deposits from the top downwards. Maximum saturation concentration of a section in the bed is dependent of coagulant and suspension as well as filter media grain size (porosity of filter bed). The controlling mechanism behind the saturation process is not clear.

Conclusions

The filter media grain size is an important filter bed characteristic, and can be the source of poor effluent water quality. When coagulating humic waters, flocs are generally weak and voluminous, creating low density deposits. When metal salts like aluminium sulphate and iron chloride sulphate are used, the coarse grain sizes of the Filtralite filter provides an insufficient barrier to turbidity, leading to high levels of residual metal. With increased turbidity in the raw water, the effluent turbidity and metal residuals were lower, and acceptable values were obtained in all three filters using aluminium coagulants. Poly-aluminium chloride obtained better results in the coarse Filtralite compared to aluminium sulphate. The cationic bio-polymer chitosan performed very well with the coarse filters, and residual metal is not a problem with this coagulant. By using chitosan, or a metal salt in combination with a polymer as filter aid, it is possible to utilise the large solids storage capacity of the Filtralite filter. The deeper Filtralite filter showed that it is possible to compensate for bigger grain sizes by increasing bed depth, but this is dependent of the raw water quality and coagulant used. Deep bed contact filtration of humic substances is believed to have similarities with an adsorption column process, with a saturation zone of deposits going further and further down in the bed.

Acknowledgements

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