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HOKKAIDO UNIVERSITY
DIRECT FILTRATION TO TREAT COLORED SURFACE WATER IN A COLD REGION

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INTRODUCTION

Direct filtration, as an attractive alternative to conventional water treatment, has been in successful application to treat water with low to moderate turbidities. However, there is little research in using direct filtration to treat water in the presence of relatively high concentration of naturally occurring color substances. In this study, to find out how to improve the removal of color substances, and to examine the treatability and the feasibility of using direct filtration to water sources with low turbidity and relatively high color content, the mechanisms of coagulation with two kinds of aluminum coagulants, i.e., polyaluminum chloride (PAC) and aluminum sulfate (alum), and three kinds of synthetic organic polymers (the cationic, nonionic and anionic polymers) by the aid of a modified jar test method were investigated, and the treatment efficiency of color substances contained in surface water of low temperature (about 0.3-8.0°C) by direct filtration with the widely used PAC or alum as the primary coagulant, or applying the cationic polymer as a coagulant aid was evaluated.

EXPERIMENT

Modified Jar Test Method

A modified jar test method was applied to compare the removal of color substances by various coagulants. This method was conducted in the following procedures. A designated dosage of coagulant was added to the tested water contained in a one liter beaker, the water pH adjustment being carried out with 0.5N-HCl or 0.5N-NaOH. After the solution was stirred at 180rpm for 5 minutes, the sample from each jar was filtered immediately through a pre-selected fiber filter with a pore size of 0.9/1 m, and through membrane filters with a pore size of 0.45, 1.0, 3.0 and 5.0 μm, respectively. Color levels of filtrate from the fiber filter were measured and used for the evaluation of color removal, and that from the membrane filters for the evaluation of floc size distribution. Differing from the standard jar tests, coagulant dosages were not selected based on the higher clarification efficiency as is the case in conventional water treatment applications, but on the formation of "pin point" flocs that were considered desirable for direct filtration applications. These modified procedures were effective particularly for low turbidity water because settleable flocs were not formed in most of the jar tests when the coagulant dosage was low.

Bench Scale Direct Filtration Apparatus and Method

A schematic diagram of the experimental apparatus is shown in Fig.1. It employed a direct sand filtration scheme and a two single medium filter arrangement. The filter columns had a sectional area of 49.6cm², inside of which was packed with silica sand (an effective size of 0.6mm and a uniformity coefficient of below 1.5) with a depth of 60cm, and the porosity was set at 44.3%. Both the jar tests and the bench scale direct filtration experiments were carried out in a water purification plant that used surface river water as the water resource, some important water quality parameters of which are summarized in Table 1.

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<th>Parameters</th>
<th>June - October</th>
<th>December - March</th>
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<tr>
<td>Water temperature (°C)</td>
<td>15.3</td>
<td>1.8</td>
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<tr>
<td>pH</td>
<td>7.1</td>
<td>7.0</td>
</tr>
<tr>
<td>Turbidity (mg/l)</td>
<td>20.1</td>
<td>7.4</td>
</tr>
<tr>
<td>Color (Pt-Co)</td>
<td>15.9</td>
<td>15.4</td>
</tr>
<tr>
<td>KCl⁻ Concentration (mg/l)</td>
<td>11.3</td>
<td>7.6</td>
</tr>
<tr>
<td>EROD (absorbance at 260nm)</td>
<td>0.288</td>
<td>0.251</td>
</tr>
<tr>
<td>THMFP (μg/l)</td>
<td>68.5</td>
<td>61.6</td>
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*THMFP-trihalomethane formation potential (pH 7.0±0.2, 20°C, 24hrs, residual chlorine 1-2mg/l)
RESULTS AND DISCUSSIONS

Jar Test
To evaluate the performance of various coagulants in treating water of low turbidity and relatively high color content, jar test experiments were first conducted. The water concerned had a temperature of between 18 and 20°C. In most of these experiments, because the turbid matter was effectively removed at the coagulant dosage and the water pH which were found to be most effective for the reduction of color substances, only the result concerning color removal is discussed here.

1. Effect of pH on Coagulation of Color Substances with PAC and Alum

Fig. 2 shows the effect of water pH on the coagulation of color substances with both the coagulants PAC and alum. Over the pH range of 2.5 to 5.0, PAC was more effective than alum in the removal of color substances. At the pH of around 7.0, which is representative of the pH of most river water, PAC showed a removal of about 36.5%, a removal rate of 28.5% higher than that for alum. The pH of around 5.5 was shown to be optimum for the coagulation of color substances contained in water with low turbidity for both PAC and alum. Although the removal tendencies were similar for both these coagulants, the profiles showing the relationship between the zeta potential and the pH were quite different, especially on the acidic side. The highest removal of color substances occurred at around the value of 2.5mv for alum, however, for the flocs formed with PAC, it was found that the highest removal was achieved at the value of around -7.0mv, not at the isoelectric point.

The difference of the removal of color substances at various pH values was attributed to the form of aluminum species in the solution. Table 2 shows the Al distribution when PAC and alum were dissolved in distilled water. At the pH of 2.5, the relatively higher removal of color closely corresponded to the region where nearly 100% of the total Al was soluble. Therefore, at this pH, the removal might be the result of the reaction of humic substances with the soluble Al species, i.e. sufficient cationic Al species coagulated with humic substances in the solution to form nuclear particles, and then because of the difference in charge character and/or the charge densities on the particle.

![Figure 2. Effect of pH on Coagulation with PAC and Alum](image1)

![Figure 3. Effect of pH on Size Distribution of Coagulated Color Substances at PAC Dosage of 16mg/l](image2)

![Figure 4. Effect of Dosages on Coagulation at Constant pH Values](image3)
surfaces, strong electrostatic interactions occurred, which led to the formation of flocs with larger size removable by the filter paper. Over the pH range of around 5.5, the soluble Al species decreased, and a concomitant increase of the insoluble Al(OH)_3 took place, so it was likely that simultaneous reactions between color substances and the soluble Al species and between ion neutralized particles and Al(OH)_3 occurred, which implied that the mechanisms of removal over this pH range were connected to the neutralization of polyanionic color substances with cationic soluble Al species, and then the bridging and/or direct adsorbing of these neutralized particles with Al(OH)_3. Over the pH range of around 7.0, with the increase of the insoluble Al species and the concurrent decrease of the soluble ones, the ion neutralization reaction did not perform sufficiently, as could be seen from the zeta potential, therefore it could be considered that the effect of adsorption of the insoluble Al species played the primary role in the removal of color substances over this pH range.

Fig. 3 shows the size distribution of flocs containing color substances formed in the coagulation with PAC at four different pH values. As indicated, at a pH of 5.5, the flocs with size larger than 5.0 μm occupied about 80% of the total color, leaving about 20% in the soluble situation (d<0.45 μm), which also verified that a pH range of around 5.5 was the optimum for coagulation of color substances as mentioned above.

2. Effect of PAC and Alum Dosages on Coagulation of Color Substances

The relationship between the color removal and the zeta potential with the coagulant dosages of PAC and alum is represented in Fig. 4. At a pH of 3.0, an increase in the coagulant dosage to 4 mg/l led to a rapid increase of the removal, and at this dosage, the zeta potential of flocs formed by coagulation with both these coagulants was about -10 mV. A further increase in the dosage from 4 to 24 mg/l did not cause a great improvement of the filtered water quality, reaching the removal of 35% for alum and 75% for PAC, with the corresponding zeta potential of around -7.0 and the isoelectric point, respectively. However, at a pH of 5.5, an increase in the coagulant dosage to about 20 mg/l resulted in a rapid increase of both the removal and the particle zeta potential, with the higher removal of color substances reaching about 85% at the dosage of 24 mg/l and the corresponding zeta potential being about -5 mV for alum and -3 mV for PAC. At a pH of 7.0, although an increase in the coagulant dosage brought about only a little increase of the zeta potential of particles in the solution, a rapid increase of the color removal was noticed. This further demonstrated that, over a pH range of around 7.0, the mechanism of removal greatly related to the adsorption of color substances onto the insoluble Al species, and to a little extent, to the ion neutralization. The corresponding size distribution, as indicated in Fig. 5, could also be applied to interpret the result concerning the color removal under these experimental conditions.

The different removal of color substances by PAC and alum was possibly attributable to the different distribution and the different molecule structures of the Al species when
these two coagulants were added to the water solution.

The results mentioned above indicated that the mechanisms of coagulation and removal of color substances differed with the pH and even with the coagulants used. To enhance the color removal, the coexistence of the soluble and insoluble Al species was necessary. The adjustment of water pH was found to be an effective way to reach this goal.

3. Color Removal with Organic Polymers

Fig. 6 shows the effect of polymer dosages on the coagulation of color substances at a constant pH value of 7.0. As indicated, the cationic polymer was much more effective than the anionic and nonionic polymers in the removal of color substances. For the water used in this experiment, the optimum dosage of the cationic polymer was around 3mg/l and the corresponding zeta potential was below the isoelectric point. Increasing the dosage over 3mg/l, on the contrary, led to the deterioration of the filtered water quality.

Concerning the floc size distribution at different dosages of the cationic polymer as shown in Fig. 7, it can be seen that, with the increase of the dosage, the soluble color substances (d<0.45μm) maintained a continuous decrease over the whole range of 0~6mg/l, which meant that, over this dosage range, color substances were constantly neutralised by the particles to form nuclear particles larger than 0.45μm; the particles within the size range of 0.45 to 3.0μm decreased to the minimum at the optimum dosage of 3mg/l, then increased with the increase of the dosage, corresponding to an increase and then a decrease of the color substances with particle size of larger than 5.0μm. This result indicated that somewhat overdosing did not impede the neutralization of polycationic color substances with polycation and the formation of flocs with the size larger than 0.45μm, but did influence the adsorption and bridging capabilities of polymers, hence inhibiting the formation of the flocs of sizes larger than 5.0μm.

It could be inferred that the important mechanisms of color removal with cationic polymers comprised the mechanism of mutual interaction/neutralization between the polycation and polyanion, and the mechanism of adsorption/bridging. The performance of both these mechanisms determined its effectiveness to treat water of low turbidity and relatively high color content. With the anionic and nonionic polymers, however, the removal could only be attributed to direct adsorption.

Direct Sand Filtration Experiments

1. Performance of Direct Sand Filtration with PAC or Alum as the Sole Coagulant.

Direct sand filtration with PAC as the sole coagulant without the adjustment of water pH was first conducted at filtration rates of 120 and 240m/d. The difference in filtration rates did not yield a considerable difference in the filtered water quality. Table 3 shows the results of filtration at the rate of 120m/d. As indicated, an increase in the PAC dosage caused a general increase of the removal of turbidity and color. To obtain filtered water with a color level below 5, a dosage of more than 30mg/l was necessary. However, filtration at this dosage led to a rapid development of head loss, reaching the set value of 250cm for a run time of 20 hours at 120m/d and 9 hours at 240m/d.

The effect of pH adjustment and
the treatment with PAC and alum were evaluated. The results at a filtration rate of 120 m/d, as shown in Table 4, indicated that the pH adjustment generally resulted in a marked improvement of the effectiveness in removing the color substances. However, compared to the results of filtration with PAC at both the pH of 5.5 and 7.0, it was found that alum was less efficient than PAC in treating water of low turbidity and relatively high color content when the water temperature was low. The lower removal rate with alum than that with PAC at the pH of 5.5, which was different from the jar test result concerning the color removal at this pH value as shown in Fig. 4, should be attributed to the water temperature, that is, the low temperature hindered the hydrolytic reaction of alum and the formation of flocs amenable to removal.

It should also be noticed that the pH adjustment brought about not only a substantial increase in the removal of turbid and color substances in the filter bed, but also a concomitant increase in head loss development.

Fig. 8 shows the corresponding relationship of the effluent turbidity and color with the filtration time in the filtration with PAC at a water pH of both 5.5 and 7.0 at a rate of 120 m/d obtained in part of the above mentioned experiments. Over the entire filtration period of 20 hours, with the dosage of 8 or 16 mg/l, filtration at the pH of 5.5 produced filtered water of a much better quality than did filtration at the water pH of 7.0. At the PAC dosage of 8 mg/l and the water pH of 5.5, except for the initial run period of 2 hours when the initial breakthrough of color substances occurred, filtered water with turbidities of below 1.0 and color levels of a little below 5.0 could be collected. At the PAC dosage of 16 mg/l and the pH of 5.5, a constant effluent with turbidities of below 0.5 and color levels of around 3.0 could be obtained from the beginning of the filter run.

2. Performance of Direct Sand Filtration with the Application of A Coagulant Aid

As mentioned above, the cationic polymer, as the sole coagulant, was very effective in treating naturally occurring organic color substances compared to the nonionic and anionic polymers, because of its higher neutralization capability. The results of a series of experiments using PAC as the primary coagulant and the cationic polymer as the coagulant aid also verified the effectiveness of applying the coagulant aid in direct filtration as could be seen from Table 5, which shows the results at the filtration rate of 120 m/d. However, to obtain filtered water that satisfied the quality requirements concerning water turbidity and color from the beginning of filter runs, the PAC dosage of 8 mg/l with the coagulant aid of 1.0 mg/l or the PAC dosage of 16 mg/l with the coagulant aid of 0.5 mg/l was necessary as
represented in Fig.9, which shows the corresponding relationship of the effluent turbidity and color with filtration time. It was also noted that applying the coagulant aid led to a rapid development of headloss in the single medium direct filtration. To solve this problem, dual medium direct filtration should be investigated.

To control the initial breakthrough of turbid and color substances caused by a relatively lower coagulant dosage, and to decrease the amount of coagulant aid applied, three kinds of coagulant dosing methods were tried. They were: 1) the cationic polymer with a dosage of 0.5mg/l was applied for the initial 4 hours only, and hereafter, the polymer dosing was stopped, 2) the polymer of 0.5mg/l was dosed for the initial 6 hours only and 3) the polymer of 0.5mg/l was applied for the initial 2 hours of the filter run, and then the dosage was decreased to 0.3mg/l. As shown in Fig.10, with the termination of the polymer dosing at an amount of 0.5mg/l during the filtration process, the filtered water suddenly degraded, especially in the experiment with method 1), in which the filtered water with color levels over 5.0 lasted for about 2 hours. However, with method 2) and 3), although the filtered water deteriorated to a certain extent, it could still meet the quality required. According to this result, another series of direct filtration experiments was conducted to evaluate which method was effective in handling the abrupt worsening of the raw water quality during the filtration process. The above mentioned methods 2) and 3) were compared with that of utilizing PAC as the sole coagulant at a relatively higher dosage of 24mg/l and that of decreasing the filtration rate. As indicated in Fig.11, the filtration at a lower rate of 60m/d, no matter whether and how the coagulant aid was added, could maintain filtered water with a little but negligible color change. However, considering filtration at a rate of 120m/d, only method 3) revealed the ability to handle the abrupt deterioration of raw water quality. The occurrence of this result was considered to be attributable to the difference in the strength and characteristics of flocs formed during the coagulation with and without the application of the coagulant aid.

CONCLUSIONS

The modified jar test procedures are effective and reliable in evaluating the performance of direct filtration to treat water with low turbidity and relatively high color content. The mechanism of color removal was directly related to the coagulant and the water pH. The most effective coagulation with Al coagulants occurred in a pH range of around 5.5, where both the mechanisms of ion neutralization and bridging took place efficiently, resulting in the higher removal of color substances. Direct filtration could be a feasible treatment method for water of low turbidity and relatively high color with pH modification or with the application of a cationic polymer as the coagulant aid when PAC was used as the primary coagulant. To deal with the abrupt deterioration of the raw water quality, applying the coagulant aid with a proper dosing method or decreasing the filtration rate appeared to be effective.

REFERENCES