## 입자분리효율을 높이기 위한 새로운 기술

# New Technologies for Enhancing Particles Separation Efficiency in Coagulation and Filtration

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#### **Abstract**

Polysilicato-iron coagulant (PSI) is receiving attention in Japan as a substitute for aluminum-based coagulants. In the first part of this article, coagulation, sedimentation and filtration experiments were carried out using kaolin clay particles as the turbidizing material and four types of PSI with various molar ratios of polysilicic acid to ferric chloride (Si/Fe ratio). Results demonstrate that use of a PSI with a high Si/Fe ratio can cause a more dramatic decrease in treated water turbidity but a higher suction time ratio (STR) than when PACI is used. However, optimization by increasing the rapid agitation strength GR is found to greatly improve the STR. In addition, the series of filtration experiments verified that optimization of GR is greatly effective in controlling rapid increases in filter head loss, and also formation of a thin aging layer in the upper part of the filter bed by slow-start filtration is effective in improving filtered water turbidity over the entire filtration process.

The second part of this article describes two innovative filtration techniques to increase the particle separation efficiency; (1) coagulant-coated filter medium by enhancing the electrical potential of the surface of the filter medium, and (2) coagulant dosing in influent by controlling the electrical potential of particles entering the filter layer. From the results of the various filtration experiments using a pilot plant, these two techniques were found to be very effective to reduce the effluent water turbidity from the start to the end of a filter run. Moreover, in the filtration experiments using these two methods simultaneously, higher removal efficiency of approximately 3-log (99.7%) was realized, resulting that the finished water turbidity was accordingly reduced to 0.004mg/L.

Key words: Polysilicato-iron coagulant, G value, coagulation, filtration, particle separation

주제어: 폴리실리케이토-철 응집, 속도경사, 응집, 여과, 입자분리

## 1. INCREASING PARTICLE SEPARATION EFFICIENCY THROUGH THE USE OF A NOVEL IRON-BASED COAGULANT PSI

## 1.1. INTRODUCTION

Polysilicato-iron coagulant (PSI) developed in Japan is prepared by adding polymeric silicic acid to ferric chloride and has an average molecular weight of approximately 200,000-500,000 Da. This coagulant has high coagulation ability, and forms greater and heavier floc particles than when poly-aluminum chloride (PACI) is used. In addition, the components that constitute the coagulant are safe, and the molar ratio between polymeric silicic acid and ferric chloride, the Si/Fe ratio, can be adjusted in accordance with the source water quality and the operating conditions. Although there have been numerous reports regarding the basic characteristics of this coagulant, very few reports are available on the treatability of the suspended particles when used in the rapid filtration systems.

The authors have proposed that any evaluation of the effects of a coagulation/sedimentation treatment should include material derived from the coagulant remaining in the treated water and not just the turbidity of the treated water. Specifically, evaluation should include the suction time ratio (STR), which is strongly correlated with the filter head loss. The authors also emphasized that the rapid agitation strength GR depends on the type of coagulant, and can be optimized to improve treatability during coagulation/sedimentation and rapid filtration.

The aim of the present study is to determine the suspended particles treatability, and elucidate methods for improving this treatability through the use of four types of PSIs conditioned by adding polymeric silicic acid.

### 1.2. Experimental Methods and Conditions

 Water source used in experiments and preparation of samples

The experiments were carried out using the

coagulation/sedimentation apparatus shown in Fig. 1, which has been newly developed to generate a larger stirring force than in common jar testers. The continuous flow experimental apparatus shown in Fig. 2 was also used, so as to include all processes of the rapid filtration system.

In the coagulation experiments, sample water was produced by adding prescribed quantities of kaolin to tap

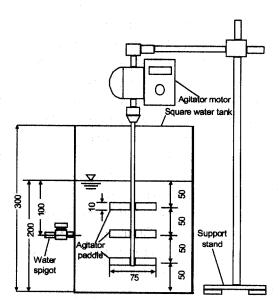


Fig. 1. Coagulation/sedimentation experimental apparatus.

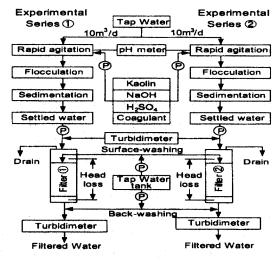


Fig. 2. Rapid filtration process experimental apparatus.

water from the Kitami Institute of Technology. Filtration experiments were carried out using sample water produced by adding 5mg/L of kaolin particles to tap water from the Kitami City water purification plant. All of the experiments were conducted at 20°C.

## 2) Coagulants

The coagulants were ferric chloride containing no polymeric silicic acid (PSI-0), and three types of PSI produced by introducing polymeric silicic acid at Si/Fe ratios of 1:0.5 (Fe<sub>2</sub>O<sub>3</sub> 2.9%, SiO<sub>2</sub> 1.1%; PSI-0.5), 1:1 (Fe<sub>2</sub>O<sub>3</sub> 2.9%, SiO<sub>2</sub> 2.2%; PSI-1) and 1:3 (Fe<sub>2</sub>O<sub>3</sub> 1.0%, SiO<sub>2</sub> 2.2%; PSI-3). For PSI-0, FeCl<sub>3</sub> solution (37.5%) was diluted beforehand with distilled water. For the other PSIs, solutions produced by diluting respective solutions to a prescribed concentration were all adjusted to pH 1.5 prior to use in the experiments.

## 3) Experimental apparatus and methods

The coagulation/sedimentation experiments were performed using a square water tank (length 200mm  $\times$  width 200mm  $\times$  height 300mm), and a stirrer (setting range: 10-800rpm). The main feature is that experiments were carried out at high stirring force unattainable with normal jar testers (maximum G value: 3900s<sup>-1</sup>). The stirring force G (s<sup>-1</sup>) during experiments was calculated by the following formula.

$$G = \left(\frac{C \cdot A \cdot v^3}{2 \cdot \gamma \cdot V}\right)^{0.5} \tag{1}$$

where C denotes the stirring coefficient (1.5), A exhibits the surface area of the stirring blades (m<sup>2</sup>), v is the average rate of stirring (m/s),  $\gamma$  stands for the dynamic viscosity coefficient (m<sup>2</sup>/s), and V represents the stirring tank volume (m<sup>3</sup>).

The coagulation experiment was carried out by introducing 8L of sample water into a water tank, and after injecting the prescribed amount of coagulant, stirring at high speed for coagulation ( $G_R = 60-1500s^{-1}$ ,  $T_R = 5$ min), stirring at low-speed for flocculation ( $G_S = 20s^{-1}$ ,

Ts = 20 min), and then resting for 40 min (over-flow rate = 0.25cm/min), followed by collection of the supernatant and water quality analysis. The coagulation pH value was set at pH 6.3  $\pm$  0.1, which was determined to be optimal in preliminary jar tests.

Experiments on the rapid filtration system were conducted using the apparatus shown in **Fig. 2**. The stirring conditions are presented below. The filtration rate was set at a nominal 240m/d, and the filtration time was 12h.

4) STR measurement and treated water quality analysis

The suction filtration test procedure involved measurement of the suction time for 500mL of sample using a suction device (vacuum container, filter folder, suction pump, evacuation level 2.7kPa). The pore diameter of the membrane filter was  $0.4\mu m$ . The STR is used as an index of filtration resistance, and is calculated using formula (2) below. The suction time for distilled water used as a standard was 63s at  $20 \pm 0.1^{\circ}$ C.

The concentration of polymeric silicic acid trapped on the membrane filter during STR measurement was determined using the molybdenum yellow method, and the iron concentration in the treated water was determined by absorption spectrometer using 1,10phenanthroline.

## 1.3. Results and Discussion

1) Treatability by coagulation/sedimentation and its improving

Fig. 3 shows the results of coagulation/sedimentation treatment using PSI at different Si/Fe ratios, while stepwise increment of the GR value from 150 to  $1500s^{-1}$ . The PSI dosage in these experiments was maintained at a constant  $2.0mg \cdot Fe/L$ .

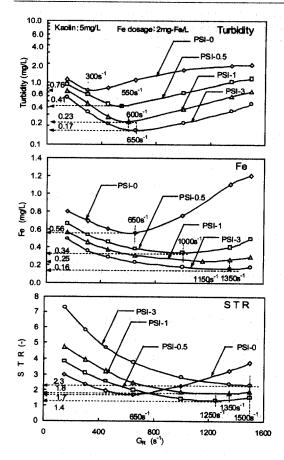


Fig. 3. Relationship between GR value and treatability.

The treated water turbidity decreased as the Si/Fe ratio of the coagulant increased, regardless of the  $G_R$  value. Consequently, in order to improve the efficiency of suspended solids treatment, it is important to use a coagulant with a high Si/Fe ratio. In addition, with regard to the change in treated water turbidity accompanying an increase in  $G_R$ , there was an initial decrease, followed by a shift towards increasing values after a minimum value had been reached, regardless of the coagulant. Specifically, regardless of which coagulant was used, there was a  $G_R$  value at which the treatment water turbidity reached a minimum, or in other words, an optimal  $G_R$  value. These values were achieved with stronger stirring for PSIs having higher Si/Fe ratios (about  $300s^{-1}$  for PSI-0, and about 550, 600 and  $650s^{-1}$  for PSI-0.5, -1 and -3

respectively). This means that for different Si/Fe ratios, the optimal GR value for the suspended solids treatment will be different.

Next, the change in iron concentration remaining in the treated water was considered. Although the change in concentration with increasing  $G_R$  and the change in treatment properties with increasing Si/Fe ratio exhibited the same trends as the treated water turbidity, the  $G_R$  value at which the iron concentration reached a minimum was different from the  $G_R$  value at which turbidity reached a minimum. Specifically, the optimal  $G_R$  value related to iron concentration appears in a range that is above the optimal  $G_R$  value with respect to turbidity. With PSI-0, -0.5, -1 and -3, the respective values were near 650, 1000, 1150 and 1350s<sup>-1</sup>. In addition, the iron concentrations at the optimum  $G_R$  values were 0.56 (72.0%), 0.34 (83.0), 0.25 (87.5) and 0.16mg/L (removal of 92.0%), respectively.

The optimal  $G_R$  value at which the STR reached a minimum accompanying an increase in the Si/Fe ratio appeared in a range that was above the range for turbidity and iron concentration. Specifically, when PSI-0 was used, the  $G_R$  value was 650s<sup>-1</sup>, and with PSI-0.5 and -1, the value was in the vicinity of 1350 s-1, whereas with PSI-3, the value appeared near 1500s<sup>-1</sup>. The need to focus on STR behavior is due to the fact that the treated water STR increases when a PSI with a high Si/Fe ratio is used. Specifically, the STR at the optimal  $G_R$  value of 650s<sup>-1</sup> for PSI-0 increased in the order 1.7, 2.0 2.5 and 3.7 with increasing Si/Fe ratio. Thus, the value for PSI-3 was as much as 2.2 times the value obtained for PSI-0. These results are due to the fact that although the suspended solids treatability in coagulation/sedimentation increases when a coagulant with high Si/Fe ratio is used, there is an increase in head loss in the subsequent filtration process. In other words, there is a decrease in filter run time.

Fig. 4 shows results for samples prepared with varying iron concentrations and silica concentrations in order to clarify the factors influencing the STR, and presents the relationship between silica concentration and STR measured immediately after stirring for 10min at a  $G_R$ 

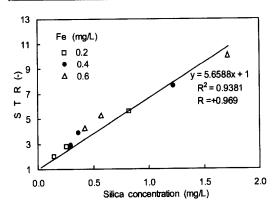


Fig. 4. Relationship between increasing silica concentration in PSI and STR.

value of 150s<sup>-1</sup>. In this figure, even at the same iron concentration, the STR increases if the silica concentration increases. For example, even if the iron concentration is constant at 0.6mg/L, the STR value changes from 4.3 to 10.1 for silica concentrations of 0.37 and 1.75, representing a 2.4 times increase in the STR. From these results, it was determined that when a coagulant having a large Si/Fe ratio is used (e.g., in Figure 3), the turbidity and iron concentration of the treated water decrease, whereas the STR increases, due to the increase in the amount of residual silica contained in the coagulant.

## 2) Treatability by rapid sand filtration

The results of experiments carried out using the experimental rapid filtration apparatus in Fig. 2 are presented in Tables 1 and 2. In these experiments, a  $G_R$  value of 150 s-1 was used, and optimal  $G_R$  values for the turbidity treatment pertaining to each coagulant were used.

Initially, the head loss after filtration for 12h was kept low as a result of decreasing STR due to optimization of the  $G_R$  value, regardless of the coagulant. When PSI-3 having the best head loss decrease was used, the  $G_R$  value increase from 150 to  $650 \, \mathrm{s}^{-1}$  resulted in the head loss decrease from 183.5 to 122.7cm (improvement of 33.1%). However, when a PSI having a high S/Fe ratio was used, there was a large amount of intrinsic head loss generated

**Table 1.** Effect of coagulation / sedimentation treatment using experimental rapid filtration apparatus

•	•			
Coagulant	$G_R(s^{-1})$	Turb. (mg/L)	Fe (mg/l)	STR (-)
	150	2.86	1.29	3.0
PSI-0	300	2.08	0.72	2.6
		(27.3)	(44.2)	(13.3)
	150	1.24	0.89	2.8
PSI-0.5	550	1.03	0.69	2.0
		(16.9)	(22.5)	(28.6)
	150	1.11	0.69	3.6
PSI-1	600	0.88	0.52	2.2
		(20.7)	(24.6)	(38.9)
	150	0.75	0.58	7.2
PSI-3	650	0.46	0.26	4.3
		(38.7)	(55.2)	(40.3)

The values in parenthesis indicate improving rate (%) on the basis at  $G_R$  150s<sup>-1</sup>

even when the sedimentation treatment was carried out at the optimal  $G_R$  value. As can be understood based on this STR behavior, this is because the polymeric silicic acid derived from PSI contributes greatly to the production of head loss. Consequently, although the turbidity or Fe concentration of the settled water is sufficiently reduced, Si derived from the coagulant remains in large quantities, making it difficult to significantly decrease head loss. It is thus necessary to take some countermeasure at the stage of sand filtration.

Improvement of settled water quality was also attempted using  $G_R$  values from  $150s^{-1}$  to the optimal  $G_R$  value, for all coagulants. However, the peak turbidity increased slightly when settled water was filtered at the optimal  $G_R$  value.

The average turbidity over 2-12h of filtration decreased when the  $G_R$  value was at the optimized level, regardless of the coagulant used, in contrast to peak turbidity. On the other hand, when PSI-3 having the highest settled water STR was used, the terminal break-through of turbidity was confirmed in as little as 12h of filtration. However, PSI-0.5 and PSI-1 both had favorable results with regard to average turbidity and average Fe.

From the above results, it is concluded that the use of a coagulant PSI with high Si/Fe ratio in suspended solids

Table 2. Effect of filtration on coagulation / sedimentation treated water (effective medium size: 0.60mm)

		0	~ 2hrs	2 -	- 12hrs	Head Loss
		Peak	Avg.	Avg	Avg.	(after 12hr
Coagulant	G <sub>R</sub> (s <sup>-1</sup> )	Turb.	Turb.	Turb.	Fe	(cm)
		(mg/L)	(mg/L)	(mg/L)	(mg/L)	
	150	0.041	0.019	0.057	0.08	54.3
PSI-0	300	0.093	0.028	0.024	0.03	52.1
	(-126.8)	(-47.4)	(57.9)	(63.6)	(4.1)	0.009
	150	0.040	0.013	0.009	0.01	68.5
PSI-0.5	550	0.065	0.012	0.002	0.01	60.7
		(-62.5)	(7.7)	(77.8)	(0.0)	(11.4)
	150	0.022	0.012	0.016	0.02	111.8
PSI-1	600	0.041	0.011	0.003	0.01	77.6
		(-86.4)	(8.3)	(81.3)	(22.2)	(30.6)
	150	0.021	0.014	0.050	0.02	183.5
PSI-3	650	0.027	0.011	0.013	0.01	122.7
		(-28.6)	(21.4)	(74.0)	(50.0)	(33.1)

The values in parenthesis indicate improving rate(%) on the basis at GR 150s<sup>-1</sup>

treatment is effective for treatments up to coagulation/ sedimentation, whereas the use of a coagulant with small Si/Fe ratio is effective when carrying out filtration after coagulation/sedimentation.

Control of head loss when switching to a filter with large medium size

Although the treatment efficiency of suspended solids and Fe increases when coagulation/sedimentation treatment is carried out with a PSI with a high Si/Fe ratio, an increase in the STR of the treated water cannot be avoided. It was found that, even if the STR is reduced by optimization of  $G_R$ , STR increases naturally. Filtration in such cases leads to an increase in head loss and a shortening of the filter run time.

Thus, when using PSI coagulants, it is necessary to establish a method whereby filtration head loss is controlled. One conceivable method for controlling filter head loss is to reduce the suspended solids blocking probability, or specifically, to increase the diameter of the filter medium. This way, head loss reducing effects are ensured by increasing the effective medium size of the silica sand from 0.60 to 0.84 mm. **Table 3** shows the results of performing filtration on settled water using PSI-

1 and PSI-3 in **Table 2** while varying the effective medium size. Regardless of the coagulant used, when a large effective medium size was employed, the average turbidity over 2-12h of filtration was substantially improved, but the peak turbidity and average turbidity at 0-2 h of filtration, as well as the Fe concentration at 2-12 h of filtration, were slightly increased.

Table 4 shows the head losses generated above the filter layer at completion of filtration (after 12h of filtration) for each filter medium. From this table, it can be seen that head loss is reduced by about half when a large filter medium is used, regardless of whether PSI-1 or -3 is used. In addition, the head loss ratio was lower for a filter medium size of 0.84mm between 0 and 10cm of the filter layer, and for a filter medium size of 0.60mm

Table 3. Effect of silica sand size on filtration

Coagulant	Effective	0~2	hrs	2~12 hrs		
	diameter (mm)	Peak Turb. (mg/L)	Avg. Turb. (mg/L)	Avg. Turb. (mg/L)	Avg. Fe (mg/L)	
PSI-1	0.60	0.041	0.011	0.003	0.01	
	0.84	0.122	0.036	0.003	0.04	
PSI-3	0.60	0.027	0.011	0.013	0.01	
	0.84	0.094	0.028	0.007	0.03	

Coagulant	Effective diameter	Head Loss	The	The depth of filter layer(cm)				
	(mm)	0-60 (cm)	0-5	5-10	10-15	15-20		
	0.60	77.6	33.9	34.0	5.6	2.2		
			(43.7)	(43.8)	(7.2)	(2.8)		
PSI-1	0.84	36.3	13.7	14.4	4.3	1.5		
			(37.7)	(39.7)	(11.8)	(4.1)		
	0.60	122.7	57.5	56.2	5.3	1.4		
PSI-3			(46.9)	(45.8)	(4.3)	(1.1)		
	0.84	63.6	28.7	25.7	5.4	1.3		
			(45.1)	(40.4)	(8.5)	(2.0)		

The values in parenthesis indicate the rate (%) to head loss

between 10 and 20cm. This demonstrates that the suspended solids are distributed throughout the filter layer when the effective medium size is increased from 0.60 to 0.84mm.

It was thus found that water quality slightly deteriorates if the effective medium size is increased, but that a dramatic decrease in head loss can be obtained.

## Suppression of filtered water turbidity through formation of a thin aging layer

When the filter medium size is increased in order to reduce head loss during filtration, the filtered water turbidity increases at initiation of filtration (0-2h). The second part of this paper reports that the coagulant coating method and coagulant injection method are effective in suppressing suspended solids leakage during initial filtration. Here, one more attempt was made to reduce peak turbidity during initial filtration, which increases with increased GR and filter medium size, by introducing an aging layer prior to filtration. This technique is also expected to provide efficiency equivalent to the other methods.

Fig. 5 shows the change in turbidity during the initial leakage period for rapid filtration at a filtration rate of 240 m/d (effective medium sizes of 0.60mm and 0.84mm) and aged layer filtration (effective medium size of 0.84mm) using settled water treated using PSI-0.5 and PSI-1. In the latter case, water was supplied for 20min at a filtration rate

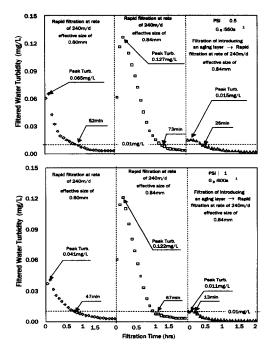


Fig. 5. Decrease in finished water turbidity with the introduction of an aging layer.

of 40m/d prior to initiation of filtration to form a thin aging layer on top of the filter layer.

As shown in the same figure, when filtration was carried out after introducing an aging layer, the peak turbidity was suppressed regardless of the PSI used. Namely, compared to the peak turbidity in filtrations using an effective medium size of 0.60mm, 0.050mg/L (improvement of 57.1%) with PSI-0.5, and 0.030mg/L (improvement of 76.9%) with PSI-1 was reduced. The average turbidity at initial filtration was reduced to 0.002 mg/L with PSI-0.5 (improvement of 50.0%), and 0.001 mg/L with PSI-1 (improvement of 66.7%). These results make clear that, although the turbidity increases in the initial filtration period when the filter medium size is increased in order to reduce head loss, the average turbidity over the initial filtration period and the peak turbidity can be greatly reduced by forming an aging layer prior to filtration.

### 1.4. Concluson

The findings obtained by the present experiments are summarized below.

- 1) Treatability in coagulation/sedimentation varies greatly with the Si/Fe ratio of the PSI and the  $G_R$  value used in the experiments. When a PSI with a high ratio is used at the low  $G_R$  value of 150s<sup>-1</sup>, the treated water turbidity and Fe concentration are reduced, though the treated water STR is correspondingly too high for filtration.
- 2) In order to reduce the water STR after coagulation/sedimentation treatment, it is essential to optimize  $G_R$  values by increasing the agitation strength. The optimal  $G_R$  values at which the STR reached a minimum were 650, 1250, 1350 and 1500s<sup>-1</sup> when PSI-0, -0.5, -1.0 and -3.0 were used, respectively.
- 3) Head loss can be greatly reduced by increasing the filter medium size when settled water with a high STR is processed by sand filtration. In addition, although turbidity during the initial leakage period and stabilization period increases slightly in this case, the filtered water turbidity can be improved by forming a thin aging layer by low-speed filtration for a short period of time during initial filtration.

## 2. IMPROVING THE FINISHED WATER TURBIDITY DURING SAND FILTRATION BY COAGULANT-COATED MEDIUM AND COAGULANT DOSING IN INFLUENT

## 2.1. Introduction

The particle removal efficiency of current rapid sand filtration systems is about 2.5-log when influent turbidity is round 20-30mg/L. In order to solve the various issues related to water quality, this efficiency needs to be increased to 4.0-log, which is 31.6 times higher than current levels. In order to increase the process efficiency by such a large degree, efforts need to be taken to investigate and improve the overall rapid sand filtration system, starting with the selection of coagulant and

coagulation methods that are suitable for particular raw water conditions, through to the improvement of separation efficiency by focusing on filtration in the final stage of the particle removal process, as well as on coagulation and sedimentation.

In light of the present situation, the objective of the second part of this article is to examine the effectiveness of two new techniques in the filtration process; (1) coating the filter medium with coagulant PACl to increase separation efficiency in the early stages of filtration by increasing the electrical potential of the filter medium, and (2) coagulant dosing in the influent in order to increase the particle separation efficiency by increasing the electrical potential of incoming particles.

## 2.2. Materials and Methods

## 1) Filtration experiments

Fig. 6 is a schematic of the experimental apparatus. This apparatus is a direct sand filtration line consisting of the following components; (1) raw-water conditioning, (2) filtration/filter-washing, (3) head-loss measurement, (4) turbidity measurement and particle counts and (5) coagulant coating. The filter column has a cross-sectional area of 52.0cm², in which silica sand was packed to a depth of 60cm. The silica medium has an effective size of

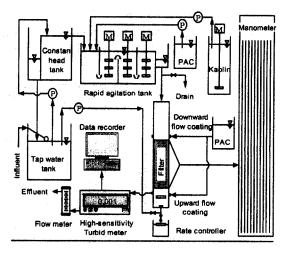


Fig. 6. Schematic diagram of experimental apparatus.

0.61mm, a uniformity coefficient of 1.39, and a specific gravity of 2.60. The raw water used in the experiments was tap water from the Kitami Institute of Technology (KIT), to which reagent-grade kaolin was added as suspended particles at a rate of 1mg/L of influent. PACl (Poly Aluminum Chloride, [Al<sub>2</sub>(OH)<sub>n</sub>Cl<sub>6-n</sub>]<sub>m</sub>, here,  $1 \le n \le 5$ ,  $m \le 10$ , 10 to 12% Al<sub>2</sub>O<sub>3</sub>) was added as the coagulant at the specified concentration.

Turbidity measurements and particle counts were made using a turbidity meter with a particle count function. The average turbidity of the tap water without the addition of kaolin and PACl was 0.033mg/L, and the number of particles in the size range of 0.5-1.0, 1.0-3.0, 3.0-7.0 and  $\geq 7.0 \mu m$  was shown in Table 1 for the influent with PACl added at 0-3mg/L. The zeta potentials of the particles were determined through the electrophoretic mobility method using a microscope and a Briggs' glass cell. The values measured for kaolin particles ranged from -25 to -30mV at pH 7.0.

In the coagulant-coated experiments, the sand filter medium was coated with PACL by downward flow of a PACl solution of specified concentration prior to filtration. In the coagulant dosing experiments, a PACl solution was injected continuously at a given rate into the influent at the inlet of the rapid agitation tank. The duration of all filtration experiments was set at 20h at a filtration velocity of 120m/d. The turbidity measurements and particle counts were made 10 times every minute, and the mean value per every minute was used for evaluation of the phenomena. As the maximum head loss after 20h

Table 5. Particle numbers of KIT tap water with PACI dosage

PACI	Turb.	Particle Size (μm)				
(mg/L)	(mg/L)	0.5	1.0	3.0	≥	
-		~1.0	~3.0	~7.0	7.0	
0.0	0.033	47,642	1,525	137	11	
	0.033	96.83	2.88	0.27	0.02	
0.0	0.034	52,424	1,602	139	6	
0.2	0.034	97.00	2.74	0.25	0.01	
4.0	0.000	57,818	1,392	133	3	
1.0	0.036	97.64	2.13	0.22	0.01	
	0.044	66,605	1,461	130	5	
3.0	0.041	97.82	1.98	0.19	0.01	

**Table 6.** Turbidity and particle counts by particle size for influent containing 1mg/L kaolin and dosed with PACI

PACI	Turb.	Particle Size (μm)					
(mg/L)	(mg/L)	0.5	1.0	3.0	≧		
		~1.0	~3.0	~7.0	7.0		
	4.040	1,527,029	58,630	7,549	326		
0.0	1.343	95.83	3.68	0.47	0.02		
	4.000	1,597,964	59,565	7,656	393		
0.2	1.399	95.94	3.58	0.46	0.02		
	4.004	1,508,355	58,162	7,440	323		
1.0	1.321	95.81	3.70	0.47	0.02		
	4.000	1,499,096	58,625	7,547	290		
3.0	1.303	95.75	3.75	0.48	0.02		

Upper portion: particle number (counts/mL) Lower portion: particle distribution(%)

filtration was very low (as little as 11.8cm), head loss will not be addressed in this article.

Particle counts and zeta potential measurements for coagulant-dosed influent

**Table 6** shows the results of turbidity measurements and particle counts for the influent containing 1 mg/L kaolin and 0-3mg/L PACl, both were injected at the inlet of the rapid agitation tank as shown in **Fig. 6**. Even with the addition of the maximum PACl concentration of 3mg/L, there was no significant increase in the number of particles. This result implies that particle agglomeration did not occur in these cases because the rapid agitation conditions employed here (velocity gradient of  $160 \text{s}^{-1}$  and agitation time of 4.4min) did not reach agglomeration onset time  $τ_1$  as proposed by Matsui et al.

Fig. 7 shows the zeta potentials of particles measured through the electrophoretic mobility method. The zeta potentials increased with increasing PACl addition by approximately 6.5mV with the addition of 1.0mg/L PACl, 11.5mV with 3mg/L PACl, and 14.0mg/L with 5mg/L PACl, indicating the gradual approach to the coagulation range proposed by Tambo. It was also noted that the increase in the potential per unit PACl dosage was higher at lower dosage. From these results, it was found that the addition of PACl to the influent did not change number

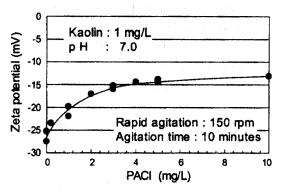


Fig. 7. Zeta potential of kaolin particle after PACI dosing.

of particles significantly, but the zeta potential of particles was significantly improved.

## 3) Distribution of Al and zeta potential inside the filter after PACl coating

In case of filter medium coating, PACI solutions of twice the filter porosity volume with specified concentration and pH of 5.0 were introduced into the filter column through a vinyl hose from a storage tank. The flow rate of the PACl solution was set at 80m/d: coating was therefore completed within 10min. After that, the water was drained from the filter, the acrylic filter cover was opened, and sand samples of 1cm depth (about 78g) were extracted from designated filter levels. The zeta potential measurements of the extracted sand samples were made using the streaming potential analyzer as shown in Fig. 8. That is, the sand sample was packed into a cylindrical cell ( $\phi$ 14  $\times$  50mm), and then the streaming potentials (E1, E2) and pressures (P1, P2) across the cell were measured in N2-gas ambient. These values were then substituted into the following Helmholtz-Smoluchowski formula to determine the zeta potential of the sand sample.

$$= 1.15 \times 1010 \frac{\mu}{\varepsilon} \times \frac{\kappa}{M} \times \frac{E_1 - E_2}{P_1 - P_2} \tag{1}$$

in which,  $\zeta$  is the zeta potential (mV),  $\mu$  is the viscosity coefficient of the fluid (g-mass · sec/cm),  $\varepsilon$  is the dielectric

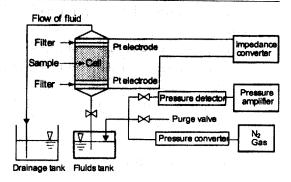


Fig. 8. Streaming potential analyzer.

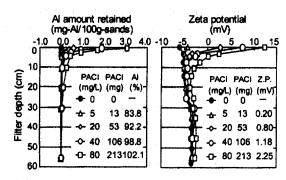


Fig. 9. Distribution of retained Al and zeta potential inside filter media.

constant of water (-),  $\kappa$  is the cell constant of electric conductivity (cm<sup>-1</sup>), and M is the resistance of water ( $\Omega$ ).

The distributions of Al coating and zeta potential inside the filter bed are shown in Fig. 9. The amount of Al retained represents the maximum value at the top of the filter bed, and rapidly decrease with filter depth. As an example, in the case of using 80mg/L PACl solution for filter coating, the amount of Al retained was approximately 3.0mg in the first 1cm to the filter bed, but was negligible small at filter depths exceeding 30cm.

On the other hand, the zeta potential at the top of the filter, where the sand is the finest with corresponding maximum surface area per unit volume, before coating was -6.1mV. This value gradually increased to -3.4mV in the deepest part of the filter, where the unit surface area is the smallest. After coating, the amount of Al retained in the upper layer had increased in proportion to the concentration of the PACl solution applied, with a

corresponding positive shift in zeta potential in this section.

Using a PACl solution with a concentration of 80mg/L, the zeta potential in the first 1cm of the filter was greatly increased by 18.6mV from -6.1 to +12.5mV, although the average zeta potential for the whole filter was increased by 2.25mV.

#### 2.3. Results and Discussion

## 1) Effects of coagulant-coated filter medium

Fig. 10 shows the turbidity of finished water for the PACl-coated filter systems. In the non-coated filter, the initial removal rate is as low as 55% (turbidity 0.5-0.6mg/L). However, the removal rate improves with filtration time to about 85% (0.2mg/L) after 5h, thereafter improving very gradually with time. As shown in Table 7, the average finished water turbidity and removal efficiency were 0.111mg/L and 91.7% at the best levels in the 15 to 20h period. The average removal rate over the entire 20 h filtration period was 86.2%. In this filtration experiments, the dominant particle removal mechanism at the start of this filtration process is mechanical support of kaolin particles at the upper filtration front in the filter media. As filtration progresses, however the effect of secondary capture in the concavo-convex field formed around particles trapped on the filter media is considered to become predominant. The retention of larger particles is

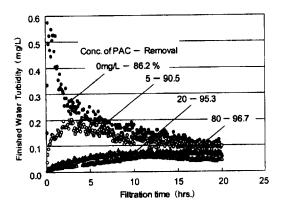


Fig. 10. Finished water turbidity for PACI coated filtration.

thought to be primarily due to physical entrapment by constriction formed between the filter media.

The removal efficiency of the coagulant-coated filtration was significantly improved. There was a clear improvement when using the 5mg/L PACl solution, and the performance was markedly improved at the 20 and 80mg/L of PACl levels. As indicated in **Table 7**, the average removal rates over the 20 h filtration period were increased to 90.5, 95.3 and 96.7% for coating with the PACl solutions of 5, 20 and 80mg/L, respectively.

The most remarkable feature of this coagulant-coated filtration is that the finished water turbidity at the start of filtration was extremely suppressed. In particular, in the case of coating with the 80mg/L PACl solution, the removal efficiency in the 1st hour was as high as 99.0%, compared to 65.5% for non-coated filtration, representing an approximately 35-fold reduction in finished water turbidity. However, the turbidity of the finished water increases slightly with time as the quantity of trapped particles increases. Even so, the turbidity of the finished water in the 15 to 20 h period is more than 2.3 times lower than that for the non-coated filtration.

Comparing these findings with the results in Figures 4 and 5, the reduction in the initial and long-term turbidity of the finished water appears to be proportional to the increase in the amount of Al retained on the surface of the filter media, or the increase of the zeta potential. The

**Table 7.** Finished water turbidity(mg/L) and removal rates(%) for PACI coated filtration

Filtration	Conc. of PACI coating solution (mg/L						
time (h)	0	5	20	80			
0 – 1	0.463	0.111	0.021	0.013			
	(65.5)	(91.7)	(98.4)	(99.0)			
0 – 5	0.310	0.158	0.040	0.022			
	(76.9)	(88.2)	(97.0)	(98.4)			
45 00	0.111	0.096	0.072	0.048			
15 – 20	(91.7)	(92.9)	(94.6)	(96.4)			
0-20	0.185	0.127	0.063	0.044			
	(86.2)	(90.5)	(95.3)	(96.7)			

The values in parenthesis indicate removal rates (%)

extension of the high zeta potential zone deeper into the filter may also contribute to the increase in performance. Based on these results, we consider that the negatively charged kaolin particles are trapped with high efficiency in the upper part of the filter where the zeta potential is positively high as a result of coagulant coating. The filter media then eventually becomes coated with kaolin particles.

Coagulant coating therefore dramatically increases the performance of the filtration system for low-turbidity influent, and the performance increase is maintained for long periods of filtration. Therefore, this technique of pre-coating the filter medium with PACl is expected to become very important for the highly efficient treatment of low-turbidity influent, for which the filter ripening period is typically very long.

## 2) Effects of coagulant dosing in influent

Fig. 11 shows the finished water turbidity for the PACl-dosing filtration. There was an initial brief period of turbidity breakthrough in all these filtration experiments, similar to the untreated case. As pointed out by Ebie et al., the degree of initial breakthrough is reduced with an increase of coagulant dosing, and the period of high finished water turbidity is also shortened. However, the initially higher turbidity levels cannot be suppressed completely. These results demonstrate that an untreated filter with negative zeta potential always

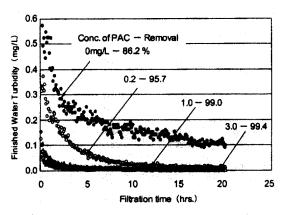


Fig. 11. Finished water turbidity for PACI dosing filtration.

requires the coating with coagulant before the start of filtration.

As filtration proceeds, the finished water turbidity with PACI dosing decreases sharply from the initial high values, thereafter decreasing gradually with filtration time. Although the effect of coagulant dosing during the initial period of filtration is smaller than that of coagulant coating, the notable feature of this method is the trend toward the complete removal of particles as filtration proceeds.

Table 8 shows the finished water turbidity and the removal efficiencies for coagulant dosing. As shown in Fig. 7, the zeta potential of the influent particles is increased according to the PACl dose. At 0.2mg/L PACl dosing, the removal efficiency in the first 5 h was 88.1%, increasing to 99.0% in the 15 to 20 h period, which represents an almost 12-fold decrease in finished water turbidity, compared to 0.111mg/L for non-dosed filtration. The turbidity is 8.5 times lower than for the non-dosed filtration in the same period, and 12.3 times lower than that in the first 5h. The removal rate also was 91.7% in the 15 to 20 h period in the non-dosed filtration, increasing to 99.8% at 3.0mg/L PACl dosing. The effectiveness of coagulant dosing has been shown to increase with filtration time, though the initial filtration efficiency for the coagulant-coated filter system is initially extremely high and gradually decreases with time.

**Table 8.** Finished water turbidity (mg/L) and removal Rates (%) for PACI dosing filtration

Filtration	PACI dosage (mg/L)					
time (hrs)	0	0.2	1.0	3.0		
0 4	0.463	0.272	0.068	0.042		
0-1	(65.5)	(79.7)	(94.9)	(96.9)		
0-5	0.310	0.160	0.031	0.018		
	(76.9)	(88.1)	(97.7)	(98.7)		
15 00	0.111	0.013	0.005	0.003		
15 – 20	(91.7)	(99.0)	(99.6)	(99.8)		
0-20	0.185	0.058	0.013	0.008		
	(86.2)	(95.7)	(99.0)	(99.4)		

The values in parenthesis indicate removal rate(%)

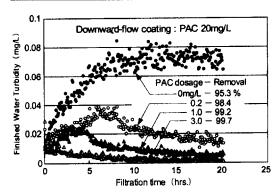


Fig. 12. Finished water turbidity in filtrations using both PACI coating method and PACI dosing method.

 Combined use of PACl-coated filter medium and PACl dosing in influent

Fig. 12 and Table 9 show the finished water turbidity and removal efficiency for a filtration incorporating both coagulant-coated filter media and continuous coagulant dosing in the influent. The removal rates are significantly improved over the use of coagulant coating or coagulant dosing alone, with remarkably low turbidity values over all periods of filtration. The combined effects of the two techniques can be clearly recognized in the figure; the rise in finished water turbidity with filtration time seen for

Table 9. Finished water turbidity (mg/L) and removal rates (%) for combined techniques

Filtration		PACI dosage (mg/L)						
time (hrs)	0	0.2	1.0	3.0				
0 1	0.021	0.017	0.013	0.011				
0 – 1	(98.4)	(98.7)	(99.0)	(99.2)				
	0.040	0.023	0.018	0.008				
0-5	(97.0)	(98.3)	(98.7)	(99.4)				
15 – 20	0.072	0.015	0.006	0.002				
15-20	(94.6)	(98.9)	(99.6)	(99.9)				
0 00	0.063	0.022	0.011	0.004				
0-20	(95.3)	(98.4)	(99.2)	(99.7)				

The values in parenthesis indicate removal rates (%)

coagulant coating (Fig. 10) is completely suppressed by the effect of coagulant dosing, and the initial breakthrough of turbid matter characteristic of coagulant dosing (Fig. 11) is largely prevented by the initial high performance of coagulant coating.

Specifically, the removal efficiency in the first hour for coagulant dosing with 3mg/L PACl was 96.9%, and that for coagulant coating with a 20mg/L PACl solution was 98.4%. The combined effect was 99.2% for this period as shown in **Table 9**. This 0.8% increase in efficiency

Table 10. Particles counts (counts/mL) and removal rates (%) for 0-1 hour filtration period

At-	PACI	PACI	Turrb.		Particle Size (μm)	)					
No.	Coating (mg/L)	Dosing (mg/L)	(mg/L)	0.5 – 1.0	1.0 – 3.0	3.0 - 7.0	>7.0				
			0.463	66,388	27,315	3,869	82				
1	<del>-</del>	-	65.5	95.7	53.4	48.7	74.8				
		,	0.021	21,178	1,037	81	0				
2	2 20	-	98.4	98.6	98.2	98.9	100.0				
_	3 80						0.013	12,672	649	33	0
3		-	99.0	99.2	98.9	99.6	100.0				
		4.0	0.068	60,945	4,685	617	5				
4	_	1.0	94.9	96.0	92.0	91.8	98.5				
			0.042	38,455	2,690	326	4				
5	-	3.0	96.9	97.5	95.4	95.7	98.8				
_			0.012	15,399	739	72	0				
6	6 20	1.0	99.1	99.0	98.7	99.0	100.0				
_			0.011	11,895	647	19	0				
7	7 20	3.0	99.2	99.2	98.9	99.7	100.0				

corresponds to a 50% reduction in the turbidity of the finished water. In the 15 to 20h period, the removal rate for coagulant coating with a 20mg/L PACl solution was 94.6%, and that for coagulant dosing of 3mg/L PACl was 99.8%.

By combining these two methods, we have achieved a maximum turbidity removal rate of 99.9%, or 3 log in the 15 to 20h filtration period, corresponding to a turbidity of 0.002mg/L.

Table 10 shows the particle counts and removal rates for filtration in the 1st hour, when coagulant coating is dominant, and Table 11 lists the results for the 15 to 20h period when the effect of coagulant dosing is greater. Firstly, from these tables, a strong correlation can be seen between the particle counts and the turbidity in the finished water.

As an example, the effect of coagulant coating and coagulant dosing in the first hour of filtration can be seen in the particle count for sizes between 0.5 and 1.0 $\mu$ m (run 7 in **Table 10**) (PACl coating: 20mg/L, PACl dosing: 3mg). The effect of coagulant coating can be seen by comparing run 7 with run 5; the particle count for the 0.5-1.0 $\mu$ m particles is reduced by 26,560 (38,455  $\rightarrow$ 

11,895) counts/mL, representing a reduction of 69.1%. The effect of coagulant dosing can be seen by comparing run 7 with run 2; the particle count is reduced by 9,283  $(21,178 \rightarrow 11,895)$ , representing a 43.8% reduction.

Making the same comparison for the 15 to 20h period, coagulant coating reduces the particle count by 1,182 counts/mL (32.3%), and the combined effect is a reduction of 36,336 counts/mL (93.6%). Therefore, the effectiveness of the combined use of coagulant coating and coagulant dosing is confirmed.

It has therefore been verified that the use of a coagulant-coated filter medium is more effective in the initial periods of filtration, and coagulant dosing in the influent becomes more effective as filtration progresses. It is expected that the turbidity reduction efficiency can be further improved by increasing the amount of coagulant used in each process. The implementation of this technology will depend on the minimum coagulant doses required to achieve a desired level of water quality with respect to the development of head loss and Al contamination of the finished water.

Table 11. Particles counts (counts/mL) and removal rates (%) for 15-20 h filtration period

Ma	PACI	PACI	Tumb.		Particle	Size (μm)	
No.	Coating (mg/L)	Dosing (mg/L)	(mg/L)	0.5 – 1.0	1.0 – 3.0	3.0 – 7.0	>7.0
4	The state of the s		0.111	68,211	7,671	1,210	12
ı	<del>-</del>	<del>-</del>	91.7	95.5	86.9	84.0	96.3
•			0.072	38,809	4,121	576	3
2	20	:	94.6	97.5	93.0	92.4	99.1
•	3 80		0.048	22,579	2,461	301	1
3		<del>-</del>	96.7	98.5	95.8	96.0	99.7
4	de del el esta esta en esta en la esta en el esta el es Esta el esta en el esta el esta el esta el e	- 1.0	0.005	6,949	362	17	0
4	-		99.6	99.5	99.4	99.8	100.0
			0.003	3,655	204	6	0
5	-	3.0	99.8	99.8	99.7	99.9	100.0
	20	4.6	0.006	5,579	299	13	0
. 0	6 20	1.0	99.6	99.6	99.5	99.8	100.0
			0.002	2,473	132	4	0
1	7 20	3.0	99.9	99.8	99.8	99.9	100.0

#### 2.4. Conclusion

In order to enhance the particle removal efficiency of rapid sand filtration systems, two new filtration techniques were examined. As a result, the following findings were obtained from the filtration experiments using a pilot plant.

- 1) The coagulant coating of the filter medium has been shown to suppress the initial breakthrough of particles in the period of filter ripening by increasing the zeta potential of the medium, allowing negatively charged influent particles to be trapped efficiently.
- Coagulant dosing in influent was also shown to increase the removal efficiency of the filter progressively with filtration time.
- 3) The combination of these techniques results in a filtration system exhibiting extremely high removal efficiency over the entire filtration period.
- 4) As an example, in the case of using 20mg/L PACl solution for filter coating and 3mg/L PACl dosing in the influent, the average removal rate over the entire 20h filtration period was 99.7%, corresponding to a finished water turbidity of 0.004mg/L.

Finally, we expect that rapid sand filtration systems incorporating new technologies such as those presented in this article will be widely utilized in many countries.

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