Enhancement of *Cryptosporidium* Oocyst Removal by Coagulation and Sedimentation with Poly-Silicate Iron (PSI)

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Abstract

The improvement of *Cryptosporidium* oocysts removals is an urgent need in drinking water treatment and one of the possible solutions is to use high-performance coagulant such as poly-silicate iron coagulant (PSI) instead of conventional coagulants like poly aluminum chloride (PAC). The efficiency of synthetic *Cryptosporidium* oocysts (S-Crypto) removal using PSI was evaluated by both jar tests and pilot plant experiments. The residual concentration of S-Crypto could be reduced even though the coagulation was operated under the optimum conditions for turbidity removal. The removal efficiencies of S-Crypto using PSI were up to 42% higher than those using PAC in the pilot plant. The higher performance of PSI is brought about by the presence of ferric species in the coagulant which promotes better sedimentation and not by the higher flocculation performance of the coagulant. In addition, the performance of PSI was independent of temperature, and S-Crypto removal by ferric chloride (FC) was not stable in cold raw water. It was suspected that the bound polymerized silica of PSI increases the stability of coagulation in cold water.

Introduction

Coagulation and sedimentation followed by rapid sand filtration is the most common drinking water treatment process in Japan. Coagulation is a process whereby colloid materials and small particles coalesce to form larger aggregates, thereby facilitating their removal during the following sedimentation and filtration stages. Chemicals conventionally used for coagulation are aluminum sulfate (alum), poly aluminum chloride (PAC), ferric sulfate and ferric chloride (FC). PAC is the most widely used coagulants in drinking water treatment plants in Japan.

Cryptosporidium oocysts are known to cause outbreaks of Cryptosporidiosis not only in Japan (Yamamoto *et al.*, 2000) but in the whole world (Fox and Lytle, 1996; Kramer *et al.*, 1996). The 1996 outbreak of cryptosporidiosis in Ogose (Japan) forced water treatment authorities also to reduce the concentration of *Cryptosporidium* in wastewater. Therefore, their removal or inactivation is necessary in drinking water treatment (Corona-Vasquez *et al.*, 2002). The better removal of *Cryptosporidium* oocysts in drinking water treatment process, especially in the coagulation and sedimentation stage, is urgently needed to reduce the possibility of leakage into tap water. Many studies (Dolejs, 1993; Bustamante *et al.*, 2001; Huck *et al.*, 2002; States *et al.*, 2002; Wang *et al.,* 2002) reported the effects of treatment conditions on the removal efficiency of *Cryptosporidium* oocysts and the comparison among some coagulants. Although the conventional drinking water treatment methods are effective for the removal of *Cryptosporidium* oocysts (Hsu and Yeh, 2003), the removal performance is not perfect and leakage into treated water could occur (Hashimoto *et al.,* 2002). Therefore enhancing removal efficiency is desired.

Poly-Silicate Iron coagulant (PSI) is a new coagulant approved by the Center of Quality Certification of Japan Water Works Association in 2001. PSI is made from polymerized silica and iron, and has an average molecular weight of around 500,000 Da. It has been proven in previous studies that it has a higher performance for turbidity and phytoplankton removals than PAC (Hasegawa *et al.*, 1991, Wang *et al.*, 2002). However, its performance for the removal of *Cryptosporidium* oocysts has not been studied yet.

Some researches (Dugan *et al.,* 2001; States *et al.*, 2002) reported that ferric coagulant had the same or higher performance for the removal of *Cryptosporidium* oocysts compared with alum and polymer coagulants. Bustamante *et al.* (2001) suggested that *Cryptosporidium* oocyst was removed primarily by sweep coagulation (flocculation) using FC and some researchers showed high affinity of iron species for

phytoplanktons (Jiang and Graham, 1998; Ma and Liu, 2002a). PSI may have a higher performance for *Cryptosporidium* oocyst removal compared to other coagulants because it is a ferric coagulant and contains polymerized silica which promotes sweep coagulation.

In this study, the performance of PSI for the removal of *Cryptosporidium* oocyst from an eutrophic reservoir water in Higashi-Hiroshima city (Japan) was investigated using a jar test and a pilot plant. A synthetic *Cryptosporidium* oocyst was used as surrogate. The performance of PSI was compared with those of PAC and FC, and the reason of enhancement was studied.

1. Experimental

1.1 Coagulants

PSI was produced by the following procedure. Aqueous glass solution (sodium silicate solution) was introduced into a sulfuric acid solution with rapid mixing and was heated at 70°C to induce polymerization. After the polymerization, ferric salt was added into the solution to produce PSI. The average molecular weight of PSI produced was about 500,000 Da (Wang *et al.*, 2002; Hasegawa, 2005). The silica-to-iron molar ratio was adjusted to 3-to-1 in this study. PAC and FC were obtained from Central Glass Co., Ltd. and Tsurumi Soda Co., Ltd., respectively. The information of each coagulant are shown in **Table 1**.

1.2 Raw water and synthetic *Cryptosporidium* oocyst

Raw water was sampled from an eutrophic reservoir in Hiroshima prefecture, Japan. The volume of the reservoir is approximately 2,600,000 m^3 and the sample was taken from a depth of approximately 7 m at the middle of the reservoir.

S-Crypto (Synthetic *Cryptosporidium* oocyst; Crypto-Tracer, Japan Water Research Center) is made from poly-methyl methacrylate which contains a fluorescent material to facilitate the analysis. The characteristics of kaolin, phytoplankton, *Cryptosporidium* oocyst and S-Crypto are shown in **Table 2**. The characteristics of S-Crypto are similar with real *Cryptosporidium* oocysts. Hundred million cells/mL of the original S-Crypto solution was diluted into 4 ~ 1,140 cells/mL for each experiment.

Table 1

Table 2

1.3 Jar test experiment

A jar test was carried out to study S-Crypto removal and sedimentation characteristics. Eight liters of raw water was coagulated in a 10 L jar and the sample was taken from 3 cm below the surface of water. Coagulants were added at the optimum dose and the solution was agitated using impellers at 100 rpm for 2 min. Then the mixing speed was reduced to 30 rpm and was kept for 15 min. The pH was kept at the optimum for turbidity during coagulation by adding hydrochloric acid and sodium hydroxide. Temperature was kept 20 ± 2°C.

1.4 Pilot plant experiment

The performance of each coagulant for turbidity, phytoplankton and S-Crypto removal was evaluated in a real flow system. The experiment for S-Crypto removal could be conducted 3 times in the pilot plant while 12 ~ 34 experiments could be conducted for turbidity and phytoplankton removal, because of the difficulty of wastewater treatment for residual S-Crypto. The schematic flow sheet and the details of the pilot plant are shown in **Figure 1** and in **Table 3**, respectively. The plant consisted of rapid mixing, flocculation (slow mixing) and sedimentation tanks with mixing speeds of 360, 18 and 0 rpm, respectively, and hydraulic retention times of 3, 30 and 90 min, respectively.

The pilot plant was basically operated every two weeks at the optimum conditions of each coagulant for turbidity removal. The optimum conditions, i.e. coagulant dose and coagulation pH, were determined and decided every time before each pilot plant experiment. Samples for analyses were taken after one day of operation. The optimum coagulant doses for PSI, FC and PAC experiments were 6.0 ± 0.6 mg-Fe/L, 5.6 ± 0.4 mg-Fe/L and 5.9 ± 0.4 mg-Al/L, respectively, whereas the optimum coagulation pH was 5.5, 5.5 and 6.0, respectively. pH was adjusted before the rapid mixing tank by an automatic pH controller with sulfuric acid. All tanks were washed every two months.

Figure 1

Table 3

1.5 Analytical methods

Residual turbidity, expressed as "mg-kaolin/L", was determined using a turbidimeter (ANA-148, Tokyo Photoelectric) and pH was determined by a pH meter (F-8, Horiba Ltd). The number of phytoplanktons was

determined by counting (10 fields and 6 times) using a microscope (magnifications of 400×) after the sample was filtered through a membrane filter (RIGO Co., Ltd：Plankton-Net HD1 with 1 μm pore size). S-Crypto was counted using a fluorescence microscope with UV irradiation because the S-Crypto contains fluorescent material in it. Dissolved organic carbon (DOC) was determined by total organic carbon analyzers (TOC-500 and TOC-5000, Shimadzu Crop). Alkalinity was determined by titration using an automatic titrator (APB-410, Kyoto Electronics Manufacturing Co., Ltd.).

2. Results and Discussion

2.1 Turbidity and phytoplankton removal

Coagulation is basically used for the turbidity removal in drinking water. In this study at first, the coagulation capacity for turbid materials and total planktons was evaluated for one year. The properties of raw water used in this study are summarized in **Table 4**. Water temperature ranged from 5.0 to 29.8°C. Turbidity was around 20 mg-kaolin/L with the highest turbidity observed during summer and the lowest during winter. DOC ranged from 2.0 to 3.7 mg-C/L and alkalinity ranged from 32 to 45 mg-$CaCO_3$/L. The common phytoplankton species found in the reservoir were *Cyclotella* species (plural) (spp.), *Melosira* spp., *Synedra* spp. and *Nitzschia* spp. and *Scenedesmus* spp..

Table 5 shows the summary of the removal of turbidity and total phytoplanktons by coagulation and sedimentation using the three coagulants throughout the one year study period. A part of these results are shown in a previous paper (Wang *et al.*, 2002). Coagulation with PSI followed by sedimentation (Run-PSI) resulted to a residual turbidity of around 0.9 mg-kaolin/L and could maintain levels lower than those with PAC and FC ("Run-PAC" and "Run-FC", respectively) almost throughout the year. The P-values of t-test between Run-PSI and Run-PAC were less than 0.01 for both turbidity and total planktons, indicating that the coagulation performance of PSI for both parameters was higher than that of PAC at over than 99% probably level. The average residual numbers of total phytoplankton were 43, 63 and 96 cells/mL for Run-PSI, Run-FC and Run-PAC, respectively. The P-values for t-test between Run-PSI and Run-FC were less than 0.01 and 0.25 for turbidity and total planktons removal, respectively, indicating that the coagulation performance between these two coagulants statistically significantly different for turbidity removal but were not significant for total planktons removal.

Table 4

Table 5

2.2 S-Crypto removal in the pilot plant

Figure 2 shows the residual concentrations of S-Crypto in the pilot plant experiments using PSI and PAC as mentioned before. The pilot plant experiments were conducted 3 times. Initial concentration of S-Crypto was adjusted to around 10 (4 ~ 14) cells/mL and the coagulation conditions (pH and coagulant dosage) were adjusted for optimum turbidity removal at that period. Initial turbidity was 8.7, 11.3 and 13.7 in the 3 experiments. On September 2000, Run-PSI and Run-PAC could reduce the number of S-Crypto to 0.49 and 0.84 cells/mL, respectively, from 13.3 cells/mL. The residual number of S-Crypto in Run-PSI was 58% of those in Run-PAC. The differences in the removal S-Crypto removal between Run-PSI and Run-PAC were 18 ~ 42% in the 3 experiments. It was found that PSI has a higher removal performance for S-Crypto than PAC even when the coagulation was operated for optimum turbidity removal.

Figure 2

Akiba *et al.* (2002) suggested that *Scenedemus quadricauda* can be used as a suitable surrogate for *Cryptosporidium* oocysts. **Figure 3(a)** and **(b)** show the initial and residual *Scenedemus* spp., respectively, in the pilot plant study conducted throughout a year. Lower residual number of the *Scenedemus* spp. was observed with Run-PSI than with Run-PAC irrespective of season. It confirmed that PSI has a better performance than PAC for the removal of *Cryptosporidium* oocysts, and the difference in performance is not influenced by water conditions through the year. In this pilot plant experiment with *Scenedemus* spp. indicator, the comparison of PSI with FC was also conducted to clarify the effect of the bound polymerized silica in PSI. The residual concentration of *Scenedemus* spp. in Run-FC was similar to that of Run-PSI almost throughout the year, becoming higher only during the winter season (November to January). Kang and Cleasby (1995) reported that cold water had detrimental effects on flocculation kinetics of FC, which explains lowest performance of Run-FC in the winter. The decline in the coagulation performance of FC was also detected in the removal of turbid material and phytoplankton during winter. The performance of PSI, however, was independent of the temperature variation even though PSI is made from iron. The bound

polymerized silica would reduce the disadvantage of FC, because PSI is a complex of FC and the polymerized silica (Hasegawa *et al.*, 1991).

Figure 3

2.3 S-Crypto removal in jar test

The removal efficiency of PSI and PAC for S-Crypto was also studied using jar test experiments in order to evaluate their flocculation performance and sedimentation performance. The initial concentration of S-Crypto was adjusted to around 1,100 cells/mL, which was much higher than that in the pilot plant experiments. Higher residual concentrations were necessary in the jar test in which smaller amounts of samples were provided compared to these in the pilot plant experiments. **Figure 4 (a)** shows the number of residual S-Crypto at various settling times in the jar test experiment with the three coagulants. It was confirmed from the pre-experiment that less than 10% of S-Crypto was precipitated in 180 min by the sedimentation without any coagulant. Generally, the coagulation performances of the 3 coagulants were compared at 20 ~ 60 min of sedimentation in the jar test (Hasegawa *et al.*, 1991; Jiang and Graham, 1998; Ma and Liu, 2002a, 2002b; Exall and Loon, 2003). The residual number in Run-PSI was always lower than those in Run-PAC and FC during the sedimentation period.

Sedimentation characteristics were also evaluated in order to elucidate the differences in the removal performance between PSI and the other two coagulants on the basis of Figure 4 (a). The flocculation performance (amount of particles coagulated or flocculated) of each coagulant was evaluated from the final (at 180 min) residual number of S-Crypto. The sedimentation performance of particles and flocs produced was evaluated on the basis of sedimentation velocity. There was no significant difference in the final concentration of residual S-Crypto at 180 min for the three coagulants. This indicates that the 18 ~ 42% difference between Run-PAC and Run-PSI, obtained in the pilot plant experiments is not caused by the difference in their flocculation performance.

The comparison of Run-PSI with Run-FC was also conducted for S-Crypto removal to study the mechanism of enhancement of coagulation performance. There was a big difference between Run-FC and Run-PSI in residual number of S-Crypto at the initial stage (up to 20 min), indicating the presence of larger amount of big or high-density flocs, which quickly settled within 20 min. Hasegawa *et al.* (1991) found that coagulation with PSI produced bigger flocs than with PAC, alum and FC because of the bound polymerized silica of PSI. When PSI is used, the production of bigger flocs might also be effective for S-Crypto.

Figure 4 (a) also showed the large differences in the sedimentation velocity of S-Crypto when PSI or PAC was used. For example, the number of residual S-Crypto reached between 1.0 cells/mL at around 30 in Run-PSI and at 120 min in Run-PAC. Clearly, there was big difference between the sedimentation performance of Run-PSI and Run-PAC. This difference brought about the difference in their performance in S-Crypto removal. Thus, the difference in flocculation performance between PSI and PAC was not the reason for the difference in S-Crypto removal.

The number of residual S-Crypto reached 1.0 cells/mL at around 60 min in Run-FC, it was higher than around 120 min in PAC. This show the higher sedimentation performance in FC and the higher sedimentation performance of PSI compared to PAC would be caused by the ferric species of coagulant because PSI and FC showed higher sedimentation performance than PAC.

Figure 4

Generally, high sedimentation performance is caused by higher floc density or bigger floc size. Bustamante *et al*. (2001) reported that alum (aluminum sulfate) and PAC neutralized the zeta potential of *Cryptosporidium* oocyst during coagulation, but FC didn't neutralize it. It suggested that the coagulation of *Cryptosporidium* oocyst by the ferric coagulant was mainly caused by sweep coagulation. Generally, more coagulants can adsorb onto S-Crypto in the case of sweep coagulation compared to charge neutralization. In addition, since the atomic weight of iron (about 56) is higher than that of aluminum (about 27), the increment of the density and/or size of S-Crypto floc would be higher in Run-PSI, resulting to higher sedimentation performance.

Figure 4 (b) shows the residual turbidity at various settling times during the experiment for turbidity removal. The trend observed was almost the same as that of S-Crypto removal. On the basis of these data, the relationships between residual turbidity and S-Crypto for PSI, PAC and FC were summarized in **Figure 5**. A straight relationship between residual turbidity and S-Crypto was observed for each coagulation, indicating that most of flocs were complexes of turbid material and S-Crypto. However, the relationship was not clear in low residual concentrations (60 ~ 180 min in Figure 4) at which S-Crypto would exist mainly as particles, not as flocs. It suggests the sedimentation performances of residual particles during sedimentation are independent among turbid materials and S-Crypto particles.

Figure 5

Conclusions

The purpose of this study was to investigate the performance of PSI in removing *Cryptosporidium* oocysts using S-Crypto. The specific conclusions derived from this research are as follows:

1. PSI had a higher performance for S-Crypto removal than PAC, achieving a 42% reduction in residual S-Crypto.
2. The high removal performance of PSI was attributed to the better sedimentation of the particles and flocs formed and not to the flocculation performance of the coagulant. The higher performance of PSI could be due to its ferric species.
3. The performance of PSI was independent of temperature variation while that of FC was not stable for *Scenedemus* spp., removal during winter. The bound polymerized silica in PSI reduce this disadvantage of FC.

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Tables

Table 1 Information for the three coagulants used in this study

Coagulant	Abbreviation	Concentration of stock solution (g-Metal/L)	Reference
Poly-silicate iron	PSI	7.33	Fe : Si ＝1 : 3 (mol)
Ferric chloride	FC	17.60	$FeCl_3$
Poly aluminum chloride	PAC	31.23	$(Al_2(OH)_n\ Cl_{6-n})m$; $1 \leqq n \leqq 5$, $m \leqq 10$

Table 2 Characteristics of kaolin, phytoplankton, *Cryptosporidium* oocysts and synthetic *Cryptosporidium* oocyst (S-Crypto)

	Kaolin	Phytoplankton <*Synedra* acus>	*Cryptosporidium parvum* oocysts	S-Crypto
Particle size [μm]	4.8 (d_{50})	Central value ; 1 ~ 50 <4.5 ~ 300>	4.2 ~ 5.4	4.5 ~ 5.5
Density [g/cm^3]	Central value ; 2.6	Central value ; 1.2 <1.1>	1.05 ~ 1.10	1.2
References	Besra *et al.*, 2000	Han and Kim, 2001; Jun *et al.*, 2001	USEPA, 1995; Bustamante *et al.*, 2001; JWWA, 2001	Japan Water Research Center

Table 3 Details of the pilot plant used in this study

	Rapid mixing tank	Flocculation tank	Sedimentation tank
Shapes	Column (downflow)	Cubic (upflow)	Cubic
Widths [cm] <I.D. [cm]>	<30>	70	70
Mixing speeds [rpm]	360	18	0
Hydraulic retention times [min]	3	30	90 (with plats)

Table 4 The properties of raw water

Properties	Unit	Range of value
Turbidity	mg-kaolin/L	11.4 ~ 28.3
Temperature	℃	5.0 ~ 29.8
Total Planktons	cells/mL	1,259 ~ 10,166
Alkalinity	mg-$CaCO_3$/L	32 ~ 45
DOC	mg-C/L	2.0 ~ 3.7
pH	-	7.3 ~ 7.8
Experimental period		2000.05 ~ 2001.07

Table 5 Results of t-tests for residual turbidity and total planktons at 48 hours operation

	for Turbidity				for Total planktons			
	Sample number	Average [cells/mL]	Dispersion	Significance (P-value)	Sample number	Average [cells/mL]	Dispersion	Significance (P-value)
Run-PSI	33	0.89	0.029	-	34	42.7	671.4	-
Run-PAC	20	1.02	0.012	<0.002	21	96.4	2471.4	<0.001
Run-FC	14	1.45	0.166	<0.001	13	63.1	3463.1	0.248

Figures

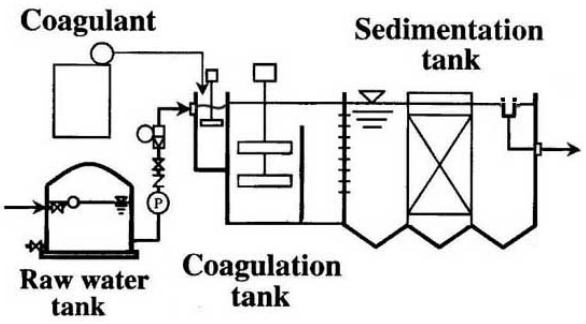


Fig. 1 Schematic flow sheet of the pilot plant

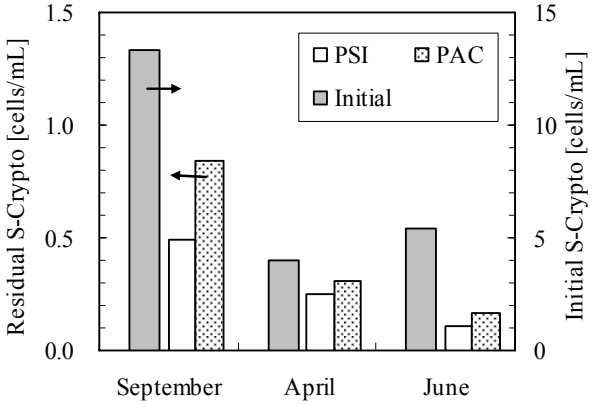
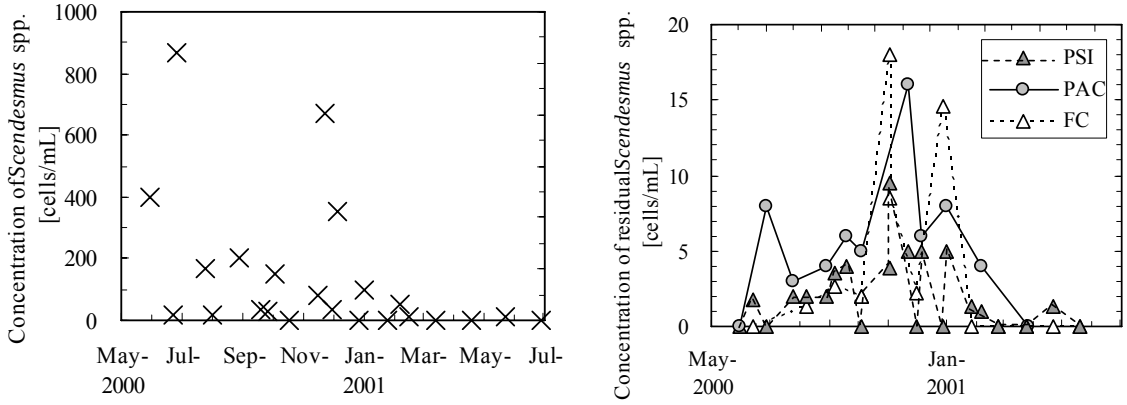
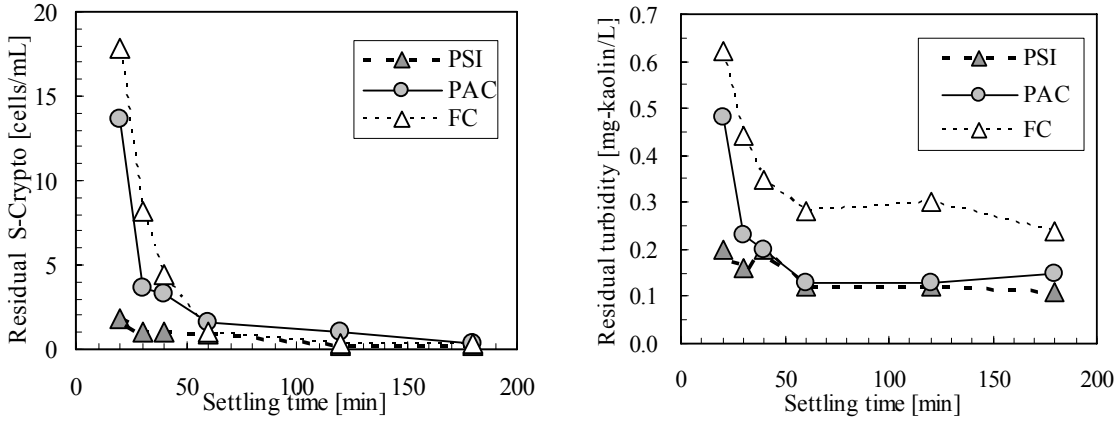


Fig. 2 Performance of three coagulants for the removal of S-Crypto in pilot plant experiments



a) Initial concentrations b) Residual concentrations

Fig. 3 Performance of three coagulants for the removal of *Scenedemus* species (plural) in pilot plant experiments



a) S-Crypto b) Turbidity

Fig. 4 Residual of S-Crypto and turbidity versus settling time (Initial turbidity, 11.7 mg-kaolin/L; Initial number of S-Crypto, 1,140 cells/mL)

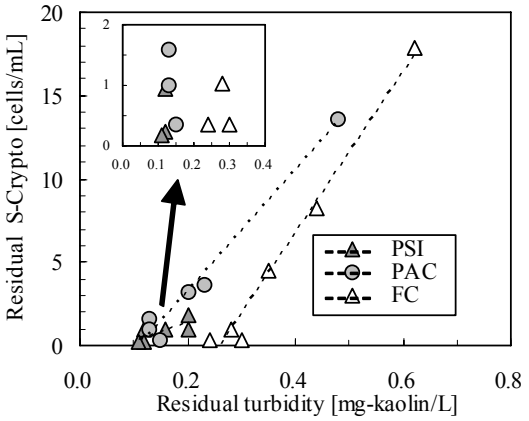


Fig. 5 Relationship between the residual turbidity and concentration of residual S-Crypto (from the data in Figure 4)

Figure Captions

Table 1 Information for the three coagulants used in this study

Table 2 Characteristics of kaolin, phytoplankton, *Cryptosporidium* oocysts and synthetic *Cryptosporidium* oocyst (S-Crypto)

Table 3 Details of the pilot plant used in this study

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Fig. 2 Performance of three coagulants for the removal of S-Crypto in pilot plant experiments

a) Initial concentrations b) Residual concentrations

Fig. 3 Performance of three coagulants for the removal of *Scenedemus* species (plural) in pilot plant experiments

a) S-Crypto b) Turbidity

Fig. 4 Residual of S-Crypto and turbidity versus settling time (Initial turbidity, 11.7 mg-kaolin/L; Initial number of S-Crypto, 1,140 cells/mL)

Fig. 5 Relationship between the residual turbidity and concentration of residual S-Crypto (from the data in Figure 4)