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Author(s)	Hideo, Maruyama; Hideshi, Seki
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Effect of Soy Protein-Based Flocculant on Flocculation and Filtration of Diatomite and Kaolin Suspensions

Hideo Maruyama* and Hideshi Seki

Division of Marine Bioresources, Graduate School of Fisheries Sciences,

Hokkaido University

Minato-cho 3-1-1, Hakodate 041-8611, Hokkaido, JAPAN

Tel:+81-138-40-8813; E-mail: maruyama@fish.hokudai.ac.jp

* Correspondence concerning this paper should be addressed to H. Maruyama

1 ABSTRACT

2
3 The effect of ethylated soy protein-based bioflocculant (EtSP) as a filter aid
4 reagent was investigated. The efficiency of EtSP as a filter aid was evaluated in
5 terms of the specific cake resistance, α , and was compared with chitosan and
6 polyaluminum chloride (PAC). Diatomite and kaolin were used as model
7 particles. Total filtration resistance, R , decreased with increasing flocculant
8 dosage (wt.%, flocculant/particle) and was almost constant in the range of 1
9 wt.% or more for both particles. The α value was significantly decreased from
10 1.01×10^{11} to 9.01×10^{10} m/kg for diatomite and from 5.11×10^{10} to 5.20×10^9 m/kg
11 for kaolin by the addition of EtSP in the case of 1.0 wt.%. The α value for cakes
12 formed by EtSP was much smaller than that formed by chitosan and PAC. In the
13 case of diatomite, in the dose range of 0.5-1.0 wt.%, the α value for cakes
14 formed by EtSP and chitosan was almost the same. However, at the excess dose
15 of 2.0 wt.% over, the α value formed by chitosan abruptly increased. In the case
16 of kaolin, in the dose range of 1.0-2.0 wt.%, the α values of chitosan and PAC
17 were mostly the same, however, these values were larger ca. nine times than that
18 of EtSP.

19
20 Keywords: flocculation filtration, filter-aid flocculant, soy protein,
21 biodegradable flocculant, specific cake resistance, ethylated protein

1 **HIGHLIGHTS**

2

3 The proposed filtration model was verified by experimental results.

4

5 Evaluation of the efficiency of EtSP as a filter aid by the specific cake resistance,

6 α .

7

8 The α value by EtSP was much smaller than that by PAC at proper dosages.

9

1 INTRODUCTION

2
3 Removal of suspended solids by filtration plays an important role in
4 wastewater treatment (Nishi *et al.* 2012), tap water production (Zouboulis &
5 Katsoyiannis 2002; Fiksdal & Leiknes 2006; Jeong & Vigneswaran 2013), and
6 downstream processes (Khouni *et al.*, 2020) in various industries. Sometimes,
7 during the filtration process, small particles penetrate deeply into the filter media
8 and cause premature turbidity breakthrough. This requires more frequent
9 backwashing of the filter and the use of large volumes of backwash water to be
10 able to remove the particles that have penetrated deeply into the filter media. To
11 avoid the clogging of filter media with small particles, the use of filter aid is one
12 of the most effective methods. When a flocculant is used as a filter aid, the size
13 of particles can be increased by interparticle bridging.

14 Indeed, the use of filter aids has demonstrated multi-benefits, namely, an
15 improvement of filtrate quality, a resistance to early breakthrough, and a
16 reduction in the magnitude and duration of the filter ripening sequence. It has
17 been reported that the addition of polyelectrolytes as a filter aid could drastically
18 improve filtration performance of suspended solids in the effluent stream from
19 recirculating aquaculture systems (Ebeling *et al.* 2003; Sharrer *et al.* 2009;
20 Guerdat *et al.* 2013). In other industrial fields, flocculants/filter aids have been
21 used in the dewatering of digested sewage sludge (Qi *et al.* 2011) and ultrafine
22 coal (Tao *et al.* 2000), and pre-treatment to reverse osmosis desalination (Johir
23 *et al.* 2009). Polyelectrolytes were also used as a membrane performance

1 enhancer to prevent membrane fouling in a membrane bioreactor process (Yoon
2 *et al.* 2005; Wu *et al.* 2006; Ji *et al.* 2008).

3 On the other hand, the harmfulness of conventional flocculants has been
4 pointed out by many researchers. Synthesized high-molecular-weight polymers,
5 such as polyacrylamide, are proved to be toxic especially to aquatic organisms
6 (Dearfield *et al.* 1988; Hasegawa *et al.* 1990; Bolto & Gregory 2007), while
7 others, such as aluminum compounds are known to negatively impact plant
8 growth (Jones & Kochian 1995; Balcelo' & Poschenrieder 2002). Therefore,
9 effective and eco-friendly bioflocculants, that are easily biodegraded and
10 produce no secondary pollution, have been a topic of intense research and have
11 gained much wider attention in recent years. However, for practical applications,
12 high production costs have been a major problem in bioflocculant production by
13 using microorganisms due to the relatively expensive conventional substrates
14 such as glucose, fructose, sucrose, and so on (Fujita *et al.* 2000; Shih *et al.* 2001;
15 Deng *et al.* 2003; He *et al.* 2004).

16 Based on the strong demand for safe, eco-friendly, and low-cost
17 bioflocculants, we investigated the use of general proteins, such as chicken egg
18 white albumin, cow's milk casein, and soy protein as raw materials for
19 bioflocculant. We have reported that bioflocculants prepared by methyl
20 esterification of proteins exhibited much higher flocculation performance for
21 diatomite, kaolin, and kanto loam suspensions than polyaluminum chloride
22 (PAC) and chitosan (Seki *et al.* 2003, 2004, 2009, 2010; Liu *et al.* 2012).
23 However, methanol is toxic and hazardous to human health. So, we considered

1 that the use of ethanol for esterification is more suitable for the safety of human
2 health, the manufacturing field, and the environment than the use of methanol
3 (Seki *et al.* 2009).

4 In the previous study (Seki *et al.* 2009), we demonstrated that the ethylated
5 egg albumin was a high-performance bioflocculant in comparison with
6 flocculation by PAC by using diatomite suspension. As described in the above
7 paragraph, it has been necessary to be biodegradable and eco-friendly for filter
8 aid reagent or flocculant. In addition, in our previous study (Seki *et al.* 2010; Liu
9 *et al.*, 2012), soy protein was also an alternative potential protein-based
10 bioflocculant as same as egg albumin.

11 Therefore, in this study, ethylated soy protein (EtSP) was applied to the
12 flocculation filtration of diatomite and kaolin suspension. Diatomite and kaolin
13 particles were selected as typical model substances for soil particles and clay
14 particles in common soil. The gravity filtration experiments were carried out by
15 using EtSP, chitosan, and polyaluminum chloride (PAC) as flocculants or filter
16 aids. The effect of EtSP on the flocculation filtration of diatomite and kaolin
17 suspensions was compared with chitosan and PAC in terms of specific cake
18 resistance as a filter aid performance indicator.

19

1 MATERIALS AND METHODS

3 Materials

5 Protein powder (soybean, ca. 90 %), ethyl alcohol (99.5 %), ammonia
6 solution (25 %), HCl solution (36 %), NaOH (97 %), and NaHCO₃ (99.5 %)
7 were purchased from Wako Pure Chemical Industries, Japan. Protein powder
8 was of practical grade and other chemicals were of reagent grade, respectively.
9 Polyaluminum chloride (PAC) solution (10.2 ± 0.2 wt.% as Al₂O₃) was
10 purchased from Taki Chemical Co., Japan. Chitosan which has an average
11 molecular weight of about 1000 kDa was purchased from Hokkaido Soda, Japan.
12 The chitosan powder was dissolved in the same weight of acetic acid and diluted
13 to 5.0 kg/m³ with distilled water. They were used without further purification.

14 Diatomite (diatomaceous earth) and kaolin were purchased from Kanto
15 Chemical Co. Inc., Japan, and Wako Pure Chemical Industries. Ltd., Japan,
16 respectively. According to the data from the supplier, diatomite was composed
17 of about 90% SiO₂ and kaolin was composed of about 45% SiO₂ and 40% Al₂O₃.
18 Both powders were dried at 80°C for 24 h and stored in a desiccator. The size
19 distribution of particles was measured using a laser diffraction particle size
20 distribution analyzer (LA-300, Horiba Ltd., Japan). The median diameter and
21 mode diameter of diatomite were 11.1 and 16.2 μm, respectively. The density of
22 diatomite measured by a pycnometer was 2510 kg/m³. In the same manner, the
23 median diameter and mode diameter of kaolin were 2.7 and 2.8 μm, respectively.

1 The density of kaolin was 2960 kg/m³.

3 **Preparation of bioflocculant (EtSP)**

4
5 Soy protein was ethylated according to the method reported by
6 Fraenkel-Conrat and Olcott (1945). Soy protein powder (20 g) was dissolved in
7 0.001 M NaOH (1.0 L) and then precipitated by the addition of 0.1 M HCl
8 solution at about pH 4.5. The soy protein was separated from the aqueous phase
9 by filtration and was washed twice with ethyl alcohol. It was suspended in 1.0 L
10 of ethyl alcohol containing HCl (0.1 M) and was stirred for 48 h at room
11 temperature. After neutralization with ammonia solution, the ethylated soy
12 protein (EtSP) was separated by centrifugation at 3000 rpm for 10 min and was
13 air-dried at room temperature with occasional grinding for 1 day. The dried EtSP
14 was pulverized in a high-speed mixer and was stored in a desiccator. The dry
15 powder of EtSP was dissolved in 5% ethanol solution under ultrasonication (20
16 kHz, 30 W, 2 min) before use. The degree of esterification of soy proteins by
17 ethanol was determined from the change in the number of carboxylic groups
18 before and after esterification. The number of carboxylic groups was determined
19 by potentiometric titration. The procedure was mostly the same method of
20 previous studies. The detail information is referred to in the previous study (Seki
21 et al., 2003). The degree of esterification of EtSP in the present study was ca.
22 90 %. The electrostatic properties of EtSP were mostly the same as those of
23 MeSP (methylated soy protein) used in the previous study. The detail

1 information would be referred to in the literature (Liu *et al.* 2012).

3 Filtration experiments

5 A Schematic diagram of
6 the experimental setup is
7 shown in Figure. 1. Gravity
8 filtration experiments were
9 performed in a clear acrylic
10 cylindrical tube with an inner
11 diameter of 32 mm and a
12 height of 250 mm. A filter
13 holder was attached to one end
14 of the tube. The outlet diameter
15 of the filter holder was 50 mm
16 and the effective filtration area

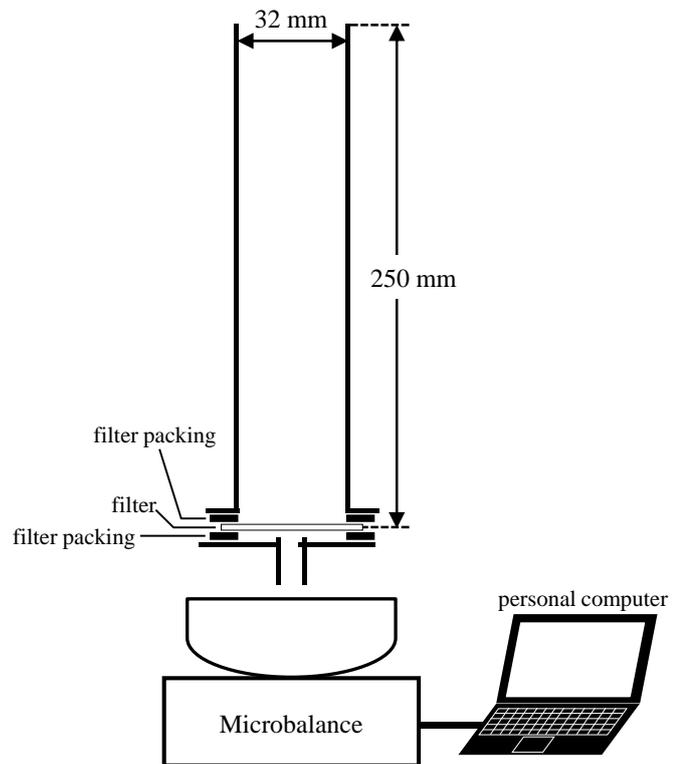


Figure 1 Schematic drawing of the experimental setup for flocculation filtration system.

17 was $8.0 \times 10^{-4} \text{ m}^2$ (hereafter this setup will be referred to as "filtration apparatus").
18 To ensure a clear filtrate in all experiments, a filter paper which consists of 99%
19 α -cellulose and has a cut-off size of 5 μm (Qualitative filter paper No. 2,
20 ADVANTEC MFS, INC., Japan) was used as the filter medium. The filtration
21 apparatus was placed above an electronic balance and the filtrate was collected
22 in a 300 mL plastic beaker placed on the electronic balance. The filtrate outlet of
23 the filtration apparatus was closed with a silicone rubber stopper until the

1 filtration was started.

2 Diatomite or kaolin suspensions were prepared by dispersing dry powder in
3 distilled water in a 300 mL glass beaker. The pH of the suspension was adjusted
4 to 7.0 ± 0.2 with the addition of 0.1 mL of 0.01 M NaHCO_3 solution. The
5 suspension was stirred with a propeller stirrer for more than 12 h to establish the
6 equilibrium state. A certain amount of flocculant solution was added to the
7 suspension. The total volume of the suspension at this point was 150 mL. It was
8 stirred rapidly at 150 rpm for 13 min, followed by slow stirring at 50 rpm for 2
9 min with a jar tester.

10 The suspension was poured into the filtration apparatus and settled for 30
11 min to ensure the formation of the cake layer. The flocs formed by EtSP settled
12 on the filter medium within 5 min, and a clear liquid upper phase was obtained.
13 After the settling, the silicone rubber stopper was removed and the clear liquid
14 was filtered through the cake layer. The weight of filtrate collected in the beaker
15 was recorded every 5 seconds by using a personal computer. After each
16 experiment, the total weight of filtrate was measured to obtain the density of the
17 filtrate. Based on the density of water, the weight of filtrate at each time point
18 was converted into the volume of the filtrate. In the case of the control
19 experiment without flocculant, the suspension was settled for about 24 h to
20 ensure the formation of the cake layer. A filtration experiment without
21 suspended solids or with distilled water was also conducted to determine the
22 filter medium resistance. The experiments were repeated at least three times, and
23 the average values were used for data analysis.

24

1 THEORETICAL

3 *Proposed filtration model for estimation of specific cake resistance, α*

5 According to Darcy's law, a general equation for a filtration process is
6 expressed as:

$$8 \quad \frac{dV}{dt} = \frac{A\Delta P}{\mu R} \quad (1)$$

9
10 where A is the filtration area, ΔP is the pressure drop across the filter, R is the
11 total filtration resistance, t is the filtration time, V is the filtrate volume at t , μ is
12 the viscosity of the fluid, respectively. In the case of batch-type gravity filtration,
13 the pressure drop across the filter is given as:

$$15 \quad \Delta P = \rho g(h_0 - V/S) \quad (2)$$

16
17 where, S is the cross-sectional area of the filtration apparatus, h_0 is the initial
18 height of the suspension, ρ is the density of suspension, and g is the gravitational
19 acceleration, respectively. The total filtration resistance (R) is the sum of the
20 cake resistance (R_c) and the filter medium resistance (R_m). Thus, Equation (1) is
21 written as:

$$\frac{dV}{dt} = \frac{A\rho g(h_0 - V/S)}{\mu R} = \frac{A\rho g(h_0 - V/S)}{\mu(R_c + R_m)} \quad (3)$$

The filter medium resistance is usually considered to be a constant for a given medium. As for the cake resistance in our study, the diatomite particles were settled on the filter medium before the filtration, and the mass of the cake was constant through the filtration process. By assuming that the cake is incompressible, the cake resistance can also be considered to be constant for each run of the filtration experiments. Thus, by integrating Equation (3) assuming that R_c and R_m are constant, the following equation is obtained:

$$\begin{aligned} \frac{\rho g A}{\mu S} t &= (R_c + R_m) \ln\left(\frac{h_0 S}{h_0 S - V}\right) \\ &= (R_c + R_m) \ln\left(\frac{V_T}{V_T - V}\right) \end{aligned} \quad (4)$$

where V_T is the total volume of the diatomite suspension (150 mL). According to Equation (4), the plot of the $\rho g A t / \mu S$ vs. $\ln[(V_T / (V_T - V))]$ gives a straight line through the origin, and the total filtration resistance ($R = R_c + R_m$) can be obtained from the slope value of the line. The cake resistance (R_c) was obtained by subtracting the filter medium resistance from the total filtration resistance. The cake resistance can be expressed by the following equation assuming that the cake is incompressible:

1
$$R_c = R - R_m = \frac{\alpha C V_T}{A} \tag{5}$$

2

3 where, α is the specific cake resistance and C is the solids concentration in the
4 suspension, respectively. Consequently, the specific cake resistance can be
5 obtained from Equation (5).

6

RESULTS AND DISCUSSION

Determination of specific cake resistance

The filter medium resistance (R_m) was determined from the flow rate of distilled water through the filter medium. In the case of the filtration of distilled water, the cake resistance can be set to zero. Thus, the slope of the line obtained from the plot of Equation (4) gives the value of R_m . The value of R_m was found to be $7.55 \times 10^8 \text{ m}^{-1}$

for the filter paper used in this study. The plot of Equation (4) showed a good linear relationship with a correlation factor of 0.997 (data were not shown).

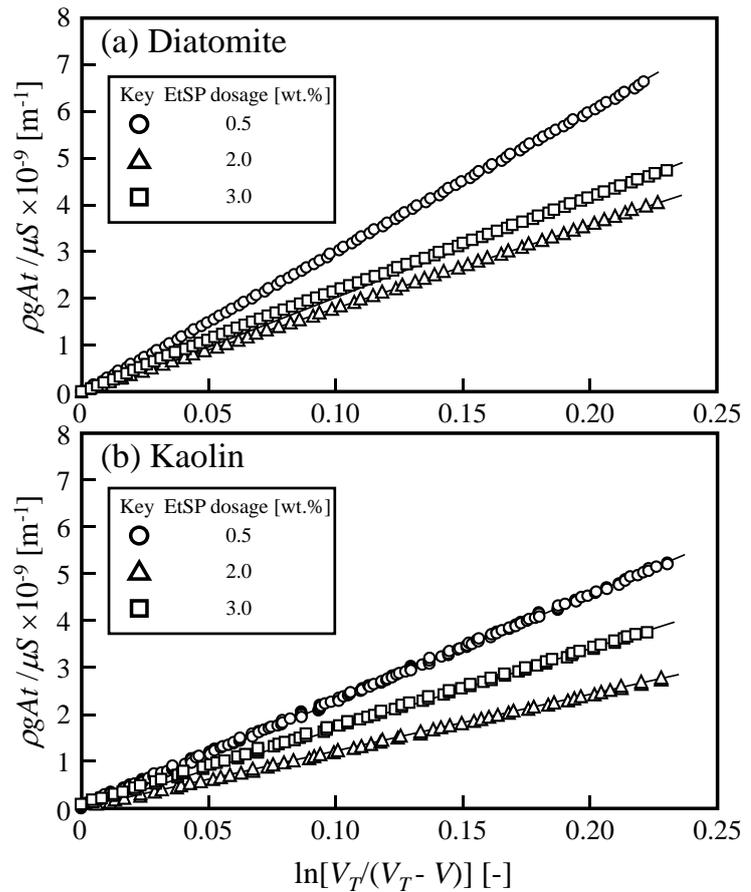


Figure 2 Determination of total filtration resistance, R , by fitting of the data to Equation (4) for (a) diatomite and (b) kaolin. The concentration of the particles was 20 kg/m^3 . The value of the slope for the regression line (straight line) of the data corresponds to the total filtration resistance.

1 The linear equation represented by Equation (4) was applied to the
 2 experimental data to determine the total filtration resistance. The typical results
 3 were presented in Figure 2. The data used in Figure 2 were obtained from the
 4 gravity filtration experiments conducted at a diatomite concentration of 5.0
 5 kg/m^3 . The dosages of EtSP were 0.5 - 3.0 wt.% of diatomite and kaolin. The
 6 unit of wt.% corresponds to the percentage of the mass of flocculant added per
 7 unit mass of particles. The results showed good linear relationships with
 8 correlation coefficients greater than 0.995, thus, the total filtration resistance
 9 could be obtained from the slope of each line.

10

11 **Effect of suspended particle**
 12 **concentration**

13
 14 Figure 3 shows the effect
 15 of the suspended particle
 16 concentration on the total
 17 filtration resistance at different
 18 dosages of EtSP. The dosage
 19 was expressed as the
 20 percentage of EtSP relative to
 21 particles on a dry weight basis.

22 The total filtration resistance at
 23 the suspended particle

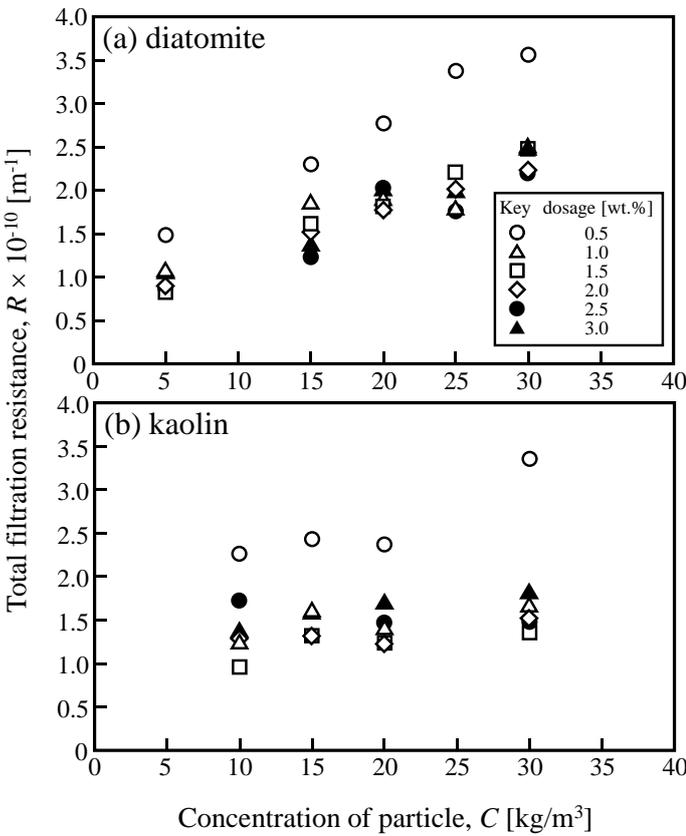


Figure 3 Effect of particle concentration on the total filtration resistance, R , at different EtSP dosages for (a) diatomite and (b) kaolin. The symbol in Figure 3b is same as Figure 3a.

1 concentration of 0 kg/m^3
 2 corresponds to the filter medium
 3 resistance, $R_m (7.55 \times 10^8 \text{ m}^{-1})$.
 4 The filter medium resistance
 5 was much smaller than the total
 6 filtration resistance in the
 7 presence of diatomite or kaolin.
 8 The total filtration resistance
 9 increased proportionally with
 10 the increase in the diatomite
 11 concentration. Assuming that

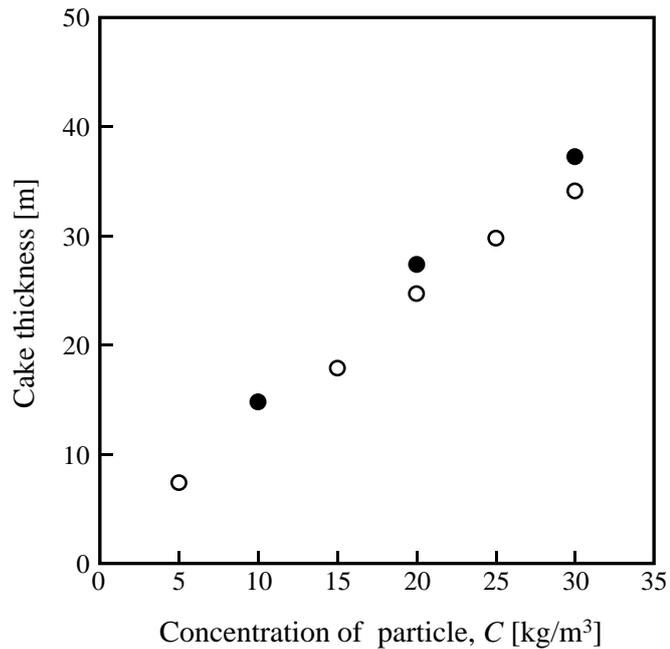


Figure 4 Effect of particle concentration, C , on the cake thickness for diatomite (open circle) and kaolin (solid circle). Dosage of EtSP was 1.5 wt.% of both the particles.

12 the cake is incompressible, the thickness of the cake increases proportionally
 13 with the mass of diatomite settled on the filter medium. Thus, it is considered
 14 reasonable that the cake resistance increased proportionally with diatomite
 15 concentration. In the case of diatomite, the total filtration resistance increased
 16 proportionally with the increase in the diatomite concentration or the mass of
 17 diatomite. In this study, almost all of the suspended particles are settled on the
 18 filter before the filtration. So, when the cake is incompressible, the thickness of
 19 the cake should proportionally increase with the mass of the suspended particle
 20 or the mass of sediment on the filter. Thus, it is considered reasonable that the
 21 total filtration resistance proportionally increases with the concentration of
 22 suspended particles. On the other hand, in the case of kaolin, expect for 0.5
 23 wt.%, the value of R became almost constant at the higher dosage (1.0-3.0

1 wt.%).

2 Figure 4 shows the cake thickness
3 as a function of particle concentration.
4 The EtSP dosage was 1.5 wt.% of
5 each particle. The cake thickness
6 increased proportionally with particle
7 concentration. Judging from the
8 results, the cakes of both particle floc
9 formed by EtSP were considered to be
10 incompressible.

11 Figure 5 shows the effect of

12 suspended particle concentration on the specific cake resistance, α , at different
13 EtSP dosages. The specific cake resistance was determined from Equation (5).
14 As expected the α value decreased as the void volume in the cake was larger. In
15 the absence of EtSP, the specific cake resistances were almost constant at the
16 particle concentrations of 5.0 – 30.0 kg/m³. The average value of specific cake
17 resistances of diatomite and kaolin were 1.03×10^{11} and 4.05×10^{10} m/kg,
18 respectively. At the same dosage of EtSP, the specific cake resistances were
19 almost constant at the particle concentration higher than 15 kg/m³ for diatomite
20 and kaolin, respectively. The specific cake resistances of diatomite and kaolin at
21 15 kg/m³ were significantly reduced from 1.03×10^{11} to 0.12×10^{11} m/kg and from
22 4.05×10^{10} to 0.79×10^{10} m/kg by the addition of 1.0 wt.% of EtSP, respectively.
23 In other words, according to Equation (3), the filtration rate was increased about

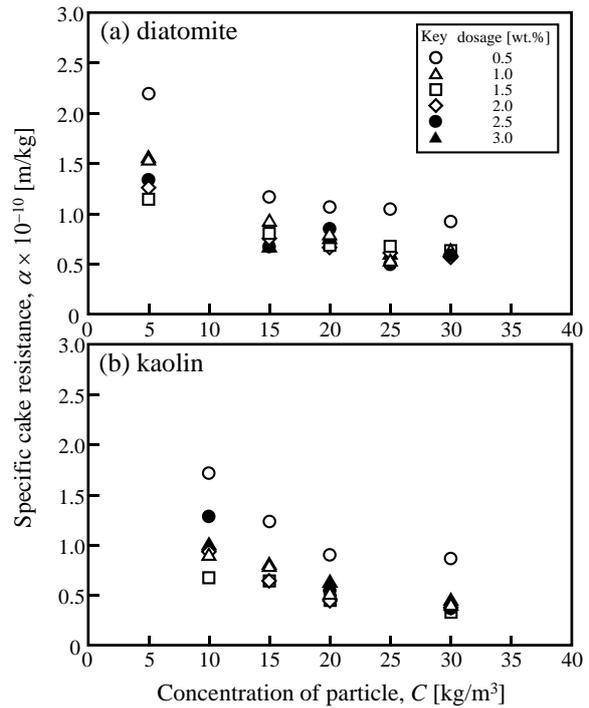


Figure 5 Effect of diatomite concentration on the specific cake resistance, α , at different EtSP dosages for (a) diatomite and (b) kaolin. The symbol in Figure 5b is same as Figure 5a.

1 ten times for diatomite and
 2 about five times for kaolin by
 3 the addition of EtSP.

5 Effect of flocculant dosage

7 Figure 6 shows the effect
 8 of EtSP dosage on the specific
 9 cake resistance at different
 10 particle concentrations. The
 11 specific cake resistance of
 12 both particles was significantly
 13 decreased by the addition of
 14 0.5 wt.% of EtSP. At the same

15 particle concentration, the specific cake resistances were almost constant at the
 16 EtSP dosages of 1.0–2.5 wt.%. The specific cake resistances at 15–30 kg/m³
 17 were almost the same at the EtSP dosages of 0.5–2.0 wt.% for both particles.

18 In visual observation, large flocs were formed in the flocculation process by
 19 adding EtSP before the filtration process. EtSP should be considered as
 20 macromolecular and polyvalent flocculant. These flocculants are known to
 21 bridge between particles by chemical binding forces and to form large flocs
 22 having high settling velocities. Ruehrwein and Ward (1952) first proposed the
 23 basic principle of bridging flocculation in 1952, presenting a model in which a

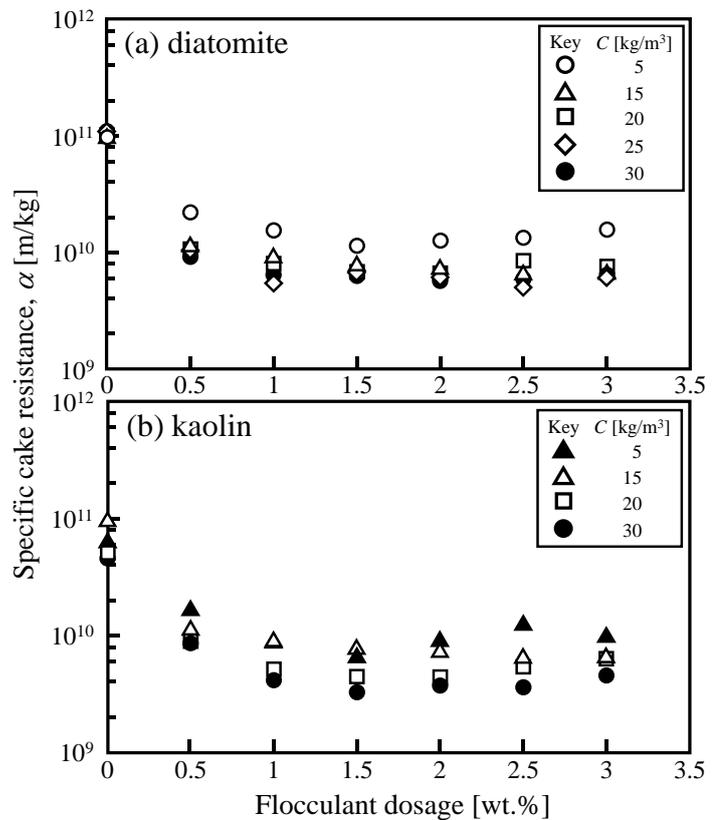


Figure 6 Influences of EtSP dosage and particle concentration, C , on the specific cake resistance, α , for (a) diatomite and (b) kaolin.

1 single polymer chain was
 2 bridging between two or more
 3 particles. Some more vigorous
 4 studies about bridging
 5 flocculation have been
 6 developed (Smellie and La
 7 Mer, 1958; Healy and La Mer,
 8 1964; La Mer, 1966; Fler and
 9 Lyklema, 1974; Gregory,
 10 1988; Biggs *et al.* 2000;
 11 Nyström *et al.* 2003). The
 12 flocculation mechanism in the
 13 present study could be
 14 bridging flocculation.

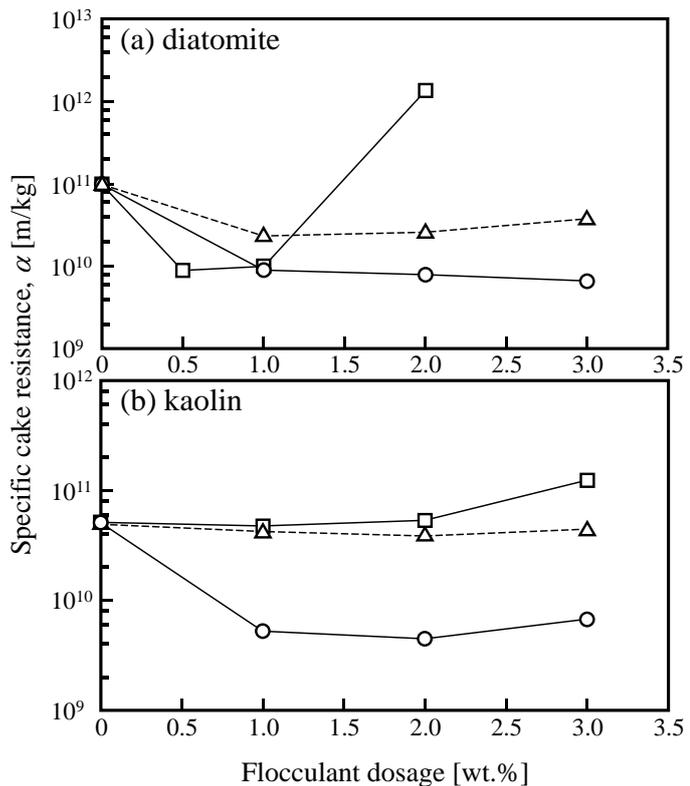


Figure 7 Comparison of the specific cake resistances, α , of (a) diatomite and (b) kaolin floes formed by EtSP (circle), chitosan (square) and PAC (triangle) as a function of flocculant dosage. The particle concentration is 20 g/L.

16 Comparison of the efficiency of EtSP with chitosan and PAC

18 In Figure 7, the specific resistances of cakes formed by EtSP, chitosan, and
 19 PAC are compared as a function of flocculant dosage. In the case of PAC, no
 20 significant improvement in the specific cake resistance was observed for both
 21 particles. The average of the specific cake resistances of diatom and kaolin at the
 22 dosages of 1.0–3.0 wt.% were 7.87×10^{10} and 5.45×10^9 m/kg, respectively. The
 23 specific resistances of cakes formed by EtSP were almost lower than those

1 formed by chitosan and PAC for diatomite and kaolin. As seen in Figure 7a, the
2 abrupt increase in α of diatomite adding chitosan at 2.0 kg/m^3 might be caused
3 by becoming viscous in the suspension. As a result, EtSP is found to be the
4 most effective flocculant in comparison with chitosan and PAC in the present
5 experimental range.

6 Wei *et al.* (2018) summarized the dewatering performance of practical
7 sludge by using several kinds of flocculants, thus, inorganic salt, organic
8 synthetic polymeric, natural polymeric, and so on. Specific resistance to
9 filtration (SRF) was employed to evaluate optimum conditions in each
10 dewatering process. The magnitude of these values was mostly ranged in $10^{12} -$
11 10^{13} m/kg , which were somewhat larger value than the values shown in the
12 present study although simple comparison might be inadequate because the
13 objective suspension is not the same one.

14 In most studies on bioflocculants, especially extracellular or intracellular
15 produced substances, flocculation ability was estimated by clarification of
16 turbidity (Fujita *et al.* 2000; Shih *et al.* 2001; Krentz *et al.* 2006; Chen *et al.*
17 2011). Some investigators have reported dewatering performances of
18 bioflocculants by using practical sludge. A few studies have dealt with
19 evaluating biodegradable flocculants' ability as a filter aids by the specific
20 resistance as the evaluation parameter. Mohtar *et al.* (2019) recently reported
21 flocculation kinetics and dewatering by using a bioflocculant, which is
22 quaternized cellulose derived from oil palm empty fruit bunches (q-EFBC) and
23 evaluated the specific resistance to filtration (SRF) with kaolin suspension. They

1 reported that the optimum dosage was 62.5 mg/L and the value of SRF was 1.49
2 $\times 10^{10}$ kg/m, which corresponded to ca. 34 % of SRF at the blank. The dosage
3 could be translated 4.4 wt.% according to their experimental conditions. The
4 degree of decrease in the specific cake resistance by adding flocculant in the
5 present study is larger and the optimum dosage in the present study is lower than
6 their values, however, the comparison might be difficult because Mohtar *et al.*
7 (2019) used kaolin as a model suspension and the difference in particles and
8 each experimental condition should affect the value of the specific resistance.

9

1 CONCLUSION

2
3 The effect of soy protein-based bioflocculant, EtSP, on the flocculation
4 filtration of diatomite and kaolin suspensions was examined. Both particles were
5 flocculated and settled on the filter medium to form the cake layer. Then the
6 clear liquid phase was filtered through the cake layer of flocculated diatomite
7 and kaolin particles.

8 The experimental data followed the modified Darcy's equation very well.
9 The total filtration resistance, the filter medium resistance, and specific cake
10 resistance were obtained from the data analysis. The cake thickness and the cake
11 resistance of both particle flocs formed by EtSP increased proportionally with
12 the increase of diatomite. Thus, the cake of diatomite and kaolin flocs formed by
13 EtSP was considered to be incompressible. In the absence of flocculant, the
14 specific cake resistance of diatomite and kaolin were 1.01×10^{11} m/kg and
15 5.11×10^{10} m/kg, respectively. The specific cake resistance significantly reduced
16 to ca. 8.9×10^9 m/kg and 6.6×10^9 m/kg for diatomite and kaolin by the addition
17 of 1.0 wt.% of EtSP.

18 The performance of EtSP as the filter aid was compared with a well-known
19 bioflocculant chitosan and a widely used commercial flocculant, polyaluminum
20 chloride (PAC) in terms of the specific cake resistance. The specific resistances
21 of cakes formed by EtSP and chitosan were much lower than those formed by
22 PAC. In the case of diatomite, the specific resistances of cakes formed by EtSP
23 were almost the same as those formed by chitosan at the dosages of 1.0–2.0

1 wt.%. However, at the dosages of 2.5 and 3.0 wt.%, the specific resistances of
2 cakes formed by EtSP were lower than those formed by chitosan. In the case of
3 kaolin, the specific cake resistance was mostly lower than those formed by
4 chitosan and PAC in the dosage range of 1.0-3.0 wt.%.

5 From a practical point of view, the scale-up of this process should be
6 important. In scale up, especially the mixing and the agitation parameters would
7 play an important role in the flocculation process before the filter process. In a
8 further study, it would be necessary to reveal the influences of the stirring and
9 the mixing on the filtration efficiency and the specific cake resistance in the
10 flocculation filtration process.

11 In further study we will conduct flocculation and filtration experiments with
12 a real wastewater by using EtSP.

13

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2

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5

1 NOMENCLATURE

2

3 A = filtration area [m²]

4 V = filtrate volume [m³]

5 t = time [min]

6 ΔP = pressure drop across filter [Pa]

7 R = total filtrate resistance [m⁻¹]

8 R_c = cake resistance [m⁻¹]

9 R_m = medium resistance [m⁻¹]

10 S = cross sectional area of filtration apparatus [m²]

11 h_0 = initial height of suspension [m]

12 g = gravitational acceleration [m/min²]

13 V_T = total volume of suspension [m³]

14 C = solid concentration in suspension [kg/m³]

15 α = specific cake resistance [m/kg]

16 μ = viscosity of the fluid [kg/(m s)]

17 ρ = density of suspension [kg/m³]

18

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9

1 **Figure captions**

2 **Figure 1** Schematic drawing of the experimental setup for flocculation
3 filtration system.

4

5 **Figure 2** Determination of total filtration resistance, R , by fitting of the data
6 to Equation (4) for (a) diatomite and (b) kaolin. The concentration of the
7 particles was 20 kg/m^3 . The value of the slope for the regression line (straight
8 line) of the data corresponds to the total filtration resistance.

9

10 **Figure 3** Effect of particle concentration on the total filtration resistance, R ,
11 at different EtSP dosages for (a) diatomite and (b) kaolin. The symbol in Figure
12 3b is same as Figure 3a.

13

14 **Figure 4** Effect of particle concentration, C , on the cake thickness for
15 diatomite (open circle) and kaolin (solid circle). Dosage of EtSP was 1.5 wt.%
16 of both the particles.

17

18 **Figure 5** Effect of diatomite concentration on the specific cake resistance, α ,
19 at different EtSP dosages for (a) diatomite and (b) kaolin. The symbol in Figure
20 5b is same as Figure 5a.

21

22 **Figure 6** Influences of EtSP dosage and particle concentration, C , on the
23 specific cake resistance, α , for (a) diatomite and (b) kaolin.

1

2 **Figure 7** Comparison of the specific cake resistances, α , of (a) diatomite and
3 (b) kaolin flocs formed by EtSP (circle), chitosan (square) and PAC (triangle) as
4 a function of flocculant dosage. The particle concentration is 20 g/L.