HOKKAIDO UNIVERSITY

Title	Surface-Modified Waste Scallop Shell as a Flocculant and an Optical Method for Evaluating Flocculation Performance
Author(s)	Maruyama, Hideo
Citation	Separations, 13(4) https://doi.org/10.3390/separations13040116
Issue Date	2026-04-14
Doc URL	https://hdl.handle.net/2115/100435
Rights	© 2026 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license.
Rights(URL)	https://creativecommons.org/licenses/by/4.0/
Type	journal article
File Information	Surface-Modified Waste Scallop Shell as a Flocculant and an Optical Method for Evaluating Flocculation Performance.pdf

Hokkaido University Collection of Scholarly and Academic Papers : HUSCAP

Surface-modified waste scallop shell as a flocculant and an optical method for evaluating flocculation performance

Hideo MARUYAMA*

Division of Marine Biosciences, Graduate School of Fisheries Sciences, Hokkaido University

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

Telephone: +81-138-40-8813, Facsimile: +81-138-40-5048, E-mail: maruyama@fish.hokudai.ac.jp

* corresponding to the author

Abstract

Surface-modified waste scallop shells were investigated as a solid flocculant for removing suspended particles, and a light transmission method was examined as a simple approach for evaluating flocculation behavior. Kaolin suspensions (3, 5, 10 g/L, pH 6.95–7.05) were used as model wastewater. Temporal changes in transmitted light intensity were monitored using a white LED–sensor optical system after agitation of the suspension was stopped. The transmitted light intensity I was normalized by the intensity measured for particle-free water (I_0), and an optical extinction index $A = -\log_{10}(I/I_0)$ was used to describe the attenuation of light in the suspension. An apparent clarification rate (rate of change in optical extinction) v was defined from the initial decrease in the optical extinction index and used as an operational kinetic parameter for comparing flocculation behavior under identical conditions. The results showed that the surface-modified scallop shell particles exhibited measurable flocculation activity toward kaolin suspensions, although the performance was lower than that of commercial polymer flocculants. The optical transmission method enabled continuous monitoring of the flocculation process and provided a practical index for comparing flocculation performance of different materials.

KEYWORDS: Flocculation, Solid particle flocculant, Waste scallop shell, Illuminance, Clarification efficiency

1. Introduction

The removal of suspended solids (SS) is an essential separation process in industrial wastewater treatment, such as food wastewater [1,2] and refinery wastewater [3,4]. The removal of suspended solids (SS) in wastewater is usually performed in two stages, that is, flocculation-sedimentation and filtration [5,6]. SS particles smaller than a few micrometer cannot be separated by filtration [7]. Therefore, a flocculation-sedimentation method is used as a pretreatment to improve the efficiency of filtration, in which flocculants are added to SS-containing wastewater to flocculate fine SS particles into larger flocs for sedimentation.

The inorganic and organic SS have been generated in large-scale construction and quarrying/mining fields [8,9], and food industries and sewage treatment processes, respectively [10,11]. In flocculation-separation processes, in general, flocculants are added to suspension to cause the small SS particles to form larger aggregates that settle liquid phase out.

Flocculants used in flocculation-separation can be categorized into two kinds of flocculant. First, inorganic flocculant have been used for flocculation such as polyaluminum chloride and ferric chloride [12-14]. Second synthetic organic polymer flocculant have also used for flocculation such as polyacrylamide [15-16] and poly(dimethylaminoethyl methacrylate) [17-18].

In general, synthetic polymer flocculants are more effective than inorganic flocculants, however, they are resistant to degradation and can persist in the natural environment for long periods of time. In addition, acrylamide degradated from polyacrylamide has been associated with carcinogenic and neurotoxic effects [19-24]. As a result, Japan strictly regulates the concentration of these substances under the “Ministerial Ordinance on Technical Standards for Waterworks Facilities” [25]. According to this regulation, the maximum allowable concentration for substances introduced into drinking water by treatment chemicals is 50

mg/L. This strict regulatory requirement has motivated the search for alternative flocculants derived from environmentally benign materials.

Judging from this background, the development of safer flocculants has been actively improved. Flocculants made from bio-resources such as starch [26-28], cellulose [29-30], chitosan [31-32], protein [33-34], and tannin [35, 36, 37, 38] have been developed and reported as environmentally kind flocculants.

In our laboratory, we have been studying on the development of flocculants using reasonable and non-expensive bio-resources, such as soybean protein [39], hen egg white albumin [40], milk casein [41-42], and alginate-protamine complex [43].

On the other hand, scallop shell is an important fisheries production in Hokkaido prefecture, Japan, however, the waste seashells have also been produced, for example, in 2021, about 130,000 tons of the waste seashells have been produced in Hokkaido prefecture [44]. The waste seashells have been used in various applications, such as catalysts [45-46], adsorbents for phosphate and heavy metal ions adsorption [47-48], antimicrobial materials [49], and concrete materials [50-51].

In our laboratory, we have successfully developed a method to use seashell particles as metal ion adsorbents through mechanochemical reactions using a ball mill with the addition of dicarboxylic acids. [52]. This utilization of seashell particles lead to the idea, which can potentially be used as a flocculant as same as the surface modified seashell based adsorbent. There have been few reports about application of solid particles to flocculants. We attempted to develop surface modified seashell particles as a flocculant for a new utilization of waste seashells.

A reactive surface was generated by grinding the particles together with a solution containing a substance with two or more ionizable dissociative groups using a ball mill. It is thought that the surface of these shells can stably attach coexisting substances in a chemically stable state. The adsorption performance and flocculation performance of the modified scallop

shell particles were tested.

On the other hand, as to the estimation method of the flocculation efficiency, the turbidimetry or the relative turbidity have been employed by many researchers in literature. [40,43,53-54].

In the turbidimetry, in general, the turbidity was determined from the absorbance of the sampling suspension, which was collected from a certain position under the suspension surface at a certain time after adding flocculant. This estimation method is relatively effective for abrupt flocculation systems, although the sampling position and time affect the estimation profiles. In addition, it is considered that this turbidimetry is not effective for slow flocculation systems due to necessity of long time for estimation.

In past study, a light transmission method was employed for analysis of aggregation phenomena of small particles suspended in liquid phase [55]. We think that utilization of light transmission is effective for estimation of relatively slow and gradual flocculation systems. This light transmission method could be considered to have advantages of successive measurement and non-necessity of sampling.

In conventional water treatment studies, turbidity measurements are widely used to evaluate clarification performance. However, turbidity measurements mainly reflect the final degree of particle removal and provide limited information about the temporal dynamics of the flocculation process. In contrast, monitoring the change in transmitted light intensity during the settling process enables the observation of the temporal evolution of suspension clarity. This approach allows the estimation of flocculation kinetics and provides additional insight into the aggregation behavior of suspended particles.

The flocculation mechanism of solid particles such as modified shell powders is expected to differ from that of conventional polymer flocculants. Possible mechanisms include hetero-aggregation between kaolin particles and shell particles, particle bridging effects, and partial surface charge neutralization. In the present study, the influence of these mechanisms

is evaluated indirectly through the temporal behavior of optical transmission during the flocculation process.

In this study, we newly developed a surface modified waste seashell based solid particle flocculant. In addition, we applied the light transmission to estimation of clarification efficiency. We conducted with flocculation experiments using two commercial organic flocculant and the seashell particle flocculant. The flocculation efficiencies were also estimated by the turbidimetry for comparison with the estimation results obtained by light transmission method. The purpose of this study was to develop a flocculant using waste shells and to evaluate its performance. We would like you to understand that this research is highly incipient and nascent step, however, innovative one.

2. Materials and methods

2.1. Waste scallop seashell

The scallop shells were mostly obtained from markets in Hakodate City, Japan. They were mainly captured in Funka Bay (Hokkaido Prefecture, Japan).

2.2. Reagents

Phenylalanine, kaolin, hydrochloric acid were purchased from Kanto Chemical Co. Inc. (Tokyo, Japan). Sodium hydroxide was purchased from Fujifilm Wako Pure Chemical Co. Inc. (Osaka, Japan). Kuriflock PA-362 (polyacrilyc acid type) and Kuriflock PN-161 (polyacrilamide type) were supplied from Kurita Water Industries Ltd. (Tokyo, Japan). According to the information from the supplier, the molecular weight of both of them is $12{,}000\times10^3$ g/mol, respectively.

2.3. Experimental procedure

2.3.1. Preparation of seashell flocculant

After washing, the shells were dried and mechanically ground. The obtained powder was sieved to obtain a specific particle size fraction. The shell particles were then subjected to surface modification using the reagent described above. The modification treatment was conducted under controlled conditions of reagent concentration and treatment time. After the modification process, the particles were washed with distilled water to remove residual reagents and then dried before use. The shells were crushed into pieces approximately 2-3 cm in size using a hammer and then used for ball milling. 5 g of shell fragments (this amount corresponds to 0.046 mol of calcium.), 3.8 g of phenylalanine and 150 g of water were placed into the ball mill. These amounts of shell and phenylalanine corresponded to the molar ratio

of calcium and phenylalanine as 1 : 0.5. Alumina balls (2 cm) were added, and grinding was carried out at 120 rpm for 24 hours. The pod used for grinding was 500 mL in volume (outer diameter 10 cm, height 10.5 cm, made of alumina, Takishin Co., Japan) , and the ball mill stand was used as AV-1 (ASAHI-RIKA SEISAKUSHO CO., LTD., Chiba, Japan). After milling, the contents of the pods were removed into a 1000-mL beaker, and the suspension solution was separated by centrifugation (3000 rpm, 10 min). Then the particles were suspended in distilled water and were washed, and then the suspension solution was separated by centrifugation (3000 rpm, 10 min) again. This operation was repeated three times. The particles were then collected by centrifugation and dried in a dryer (60°C). After drying, the particles were dispersed using an agate mortar, placed in polystyrene containers. These particles were used as a seashell flocculant (SF).

The particle size distribution was measured using a laser particle size analyzer (HORIBA LA-300, Kyoto, Japan). IR spectra of adsorbents were measured using a Fourier transform infrared spectrometer (FT/IR-4600AC, JASCO Co., Tokyo, Japan). Samples for the measurements were prepared by the tablet method using KBr. The particle morphology was observed using a scanning electron microscopy (SEM, JSM6010LA, JEOL Ltd., Tokyo, Japan).

2.3.2. Clarification method

100 mL of kaolin at a specified concentration was poured into a 100 mL volumetric flask. The volumetric flask was placed on top of the stirrer. A flocculant prepared at the specified concentration was added to the volumetric flask containing kaolin. After stirring for 1 minute at 500rpm, the mixture was allowed to stand for 1 minute, and a sample was taken using a pipette from 2 cm below the surface of the water. The absorbance of the sample at 700 nm was measured using a spectrophotometer (JASCO V-630, JASCO Corporation, Tokyo, Japan). The clarification efficiency was evaluated using the relative optical extinction as, $1– A/A_0$,

where, A represents the absorbance of the suspension after the addition of the flocculant, and A_0 represents the absorbance of the suspension in the absence of the flocculant. All clarification experiments were conducted within the pH range of 6.95–7.05. The amount of flocculant added to the suspension was expressed as the mass ratio of flocculant to kaolin (g-flocculant / g-kaolin). The required volume of the flocculant suspension was calculated based on the solid concentration of the flocculant dispersion.

2.3.3.. Light transmission experiment

The schematic drawing of the experimental setup is shown in Fig. 1. A light emitting/receiving apparatus was made of cylindrical paper with 24 cm in height, 4 cm in outside diameter, and 1.5 mm in thickness. Ten white LED lights (emitting part) were vertically installed at 16 mm intervals (in 14.5 cm), and six optical fibers were vertically installed at 20 mm intervals (in 10cm) on the opposite side of the emitting part. The top of the light emitting/receiving apparatus was covered with vinyl chloride tape to prevent from letting in natural light. The six optical fibers were connected to an illuminometer and the illuminometer was connected to a personal computer to record signals at time intervals of two seconds. The optical system was enclosed to minimize the influence of external light, and measurements were conducted under controlled laboratory illumination conditions in order to suppress stray light effects. The kaolin suspension used in the flocculation experiments was prepared at a concentration of 1.0 g/L. The pH of the suspension during the optical measurements was approximately neutral (pH 6.9–7.0). The transmitted light intensity (I) was normalized by the intensity measured for particle-free water (I_0), which was used as the reference light intensity in the optical measurements. This light emitting/receiving apparatus was used to cover a flocculation glass graduated cylinder to detect change in light intensity.

In the case of using SF, A certain amount of SF and distilled water were taken into a 100 mL beaker. SF suspension was stirred with a magnetic stirrer and ultrasonic wave was

irradiated using an ultrasonic disperser (20 kHz, 25 W, Powersonic Model 50, POWERSONICS, Inc., CT, US) to disperse SF particles. The concentration of SF was set in 10 g/L. The preparation method of the kaolin suspension was mostly the same as the method described in section 2.3.2. Then, a certain amount of flocculant was added into the kaolin suspension using a macro pipette. Within 1 minute after adding flocculant, the light emitting/receiving apparatus was covered the cylinder as soon as possible. After 1 min adding flocculant, the stirring was stopped and the recording signals were also started.

In this study, the moment at which stirring was stopped was defined operationally as the initial time (time zero) of the measurement. The recording of the illuminance signal was started immediately after stopping agitation. Although the operations were conducted manually, repeated measurements confirmed that the overall temporal trends were reproducible within small variations. Each flocculation experiment was repeated two times under identical conditions, and the reproducibility of the measured optical signals was confirmed.

3. Results and discussion

3.1. Characterization of surface modified waste scallop shell particle

Fig. 2 shows the measurement results of the particle size distribution (Fig. 2a), the FI-IR spectra (Fig. 2b), and the SEM images (Figs. c1 and c2). The particle size distribution was ranged from approximately 0.6 μm to 80 μm, and the average particle diameter was 8.3 μm. As seen in Figs. c1 and c2, a porous structure was not observed at the surface of the particles. These results were mostly the same as our previous observations reported in the previous paper [52].

Li et al reported Electrochemical and FTIR studies of L-phenylalanine adsorption at the Au(III) electrode and they have described that the transmission peak at 1562 cm^{-1} and 1625 cm^{-1} were originated by NH^{3+} symmetric deformation vibrations and NH^{3+} asymmetric deformation vibrations of phenylalanine [56]. Although the peaks were very small in our measurements, we were able to confirm the presence of three peaks. Based on this finding, we hypothesize that the observed spectral change may be related to the presence of phenylalanine associated with the shell particle surface. However, the peaks were relatively weak, and therefore the FTIR results provide only qualitative evidence suggesting possible surface modification.

3.2. Influence of flocculant dosage on time course of light intensity and clarification efficiency

Typical experimental results of the time course of change in the illuminance were shown in Fig. 3. In the present experiments, time zero was operationally defined as the moment when agitation of the suspension was stopped and the illuminance measurement was initiated.

The ordinate represents the intensity of the illuminance [lx]. A rapid increase in the illuminance indicates clarification of the suspension caused by floc formation and sedimentation of particles.

In the case of adding organic flocculants, PA362 (polyacrilyc acid type) and PN161 (polyacrilamide type), the increase in the intensity were observed in the ranges of from 0 to 30 sec, and from 0 to 10 sec for PA362 and PN 161, respectively.

On the other hand, in the case of using SF, the increase in the intensity was more gently than those of PA362 and PN161. The range of the increase in the intensity was observed in the range of from 0 to ca. 15 min. As for SF flocculant, it took a very long time to increase the intensity of the illuminance to increase in comparison with the organic polymer flocculants PA362 and PN161. In the case of using FS, the abrupt change in the intensity of the illuminance is observed between 120 and 480 sec, which are 4-16 times longer than in the case of using PA362 and PN161. However it was confirmed that the SF flocculant and kaolin particles were definitely formed and settled in the cylinder. When the kaolin suspension was allowed to stand without adding flocculant and without stirring, the intensity of the illuminance changed almost linearly from 8.61 lx to 15.5 lx in 7 hours for the 3 g/L kaolin suspension. This can be converted to a change of 0.0164 lx per minute (data not shown). This fact, together with the results shown in Fig. 3c, clearly indicates that the surface-modified shell particles proposed in this study have flocculation performance.

In the case of PA362 and SF, the much increase in the dosage the higher values of the intensity were shown. On the other hands, in the case of adding PN161, the much increase in the dosage the lower values of the intensity was shown. This tendency could be considered that in this case the dosage of 0.3 mL was much closer value for the optimum dosage than adding 0.8 and 1.2 mL. The detail reason of the fact will be explained for Fig. 6 in the section 3.3. When we measured the intensity of the illuminance, the operation to stop the stirring in the cylinder, the operation to start measurement with the illuminometer, and the operation to

start recording the signal on the PC were all performed manually, resulting in a slight difference in the intensity of the illuminance at time zero. In particular, when PN161 was used, PN161 has the highest coagulation ability, and rapid sedimentation occurred as soon as the stirring was stopped, so that a slight time difference (1 or 2 seconds) in the operation to start recording greatly affected the value of the intensity of the illminance. The PN362 and SF did not show a large difference in the irradiance intensity at time zero, because the settling was not as rapid as for PN161.

According to the Lambert-Beer's law [57], the following equation was obtained.

$$A = -\log_{10}(I/I_0) \quad (1)$$

Where A represents the apparent optical density (optical extinction) derived from the illuminance ratio, and I and I0 represent the transmitted and incident light intensities, respectively.

Adopting this relationship to our experimental system, I corresponds to the intensity of the illuminance through the suspension and I0 corresponds to the intensity of the illuminance through water without particles. In the present system using a white LED source, the attenuation of transmitted light mainly reflects optical extinction caused by scattering and absorption by suspended particles. Therefore, the value calculated using Eq. (1) should be regarded as an approximate optical density parameter rather than a strict spectroscopic absorbance.

Typical results of the clarification efficiency, $1–A/A_0$, were shown in Fig. 4. The values of clarification efficiency ($1–A/A_0$) of the ordinate of Fig. 4 were obtained by converting from the data of the intensity of the illminance shown in Fig. 3. The influence of the dosage of the each flocculant could be considered to be more clearly expressed when converted to clarification efficiency.

3.3. Influences of flocculation dosage and concentration of kaolin suspension on time rate of change in absorbance and clarification efficiency.

Based on a plot similar to Fig. 3, we determined the time rate of change of optical extinction value with time, *v*, as an indicator of the apparent clarification rate (rate of change in optical extinction). The method is as follows. We identified the data point where become the maximum value of the difference with the before and the next points. The data range for determination of the slope value was defined as the five data series included the two before and the two next data points. The slope value of the five data series was determined by a least squares method. The determined slope value was defined the time rate of change in optical extinction.

In conventional flocculation studies, the performance of flocculants is generally evaluated by turbidity measurements or sedimentation tests, which mainly provide information on the final clarification state after a certain settling period. In contrast, the method proposed in the present study evaluates the temporal change in transmitted light intensity immediately after the cessation of stirring. Therefore, the method enables evaluation of the kinetic behavior of flocculation during the initial stage of particle aggregation.

The time rate of change in optical extinction, *v*, defined in this study represents the apparent rate of change in optical attenuation associated with particle aggregation and subsequent settling. Although the optical signal is influenced by scattering effects and the geometry of the optical system, the temporal variation of transmitted light intensity provides a practical indicator for comparing the relative flocculation behavior among different flocculants under identical experimental conditions.

Compared with conventional turbidity measurements, which often provide only discrete values before and after sedimentation, the present optical monitoring method allows

continuous observation of the flocculation process. This characteristic is useful for distinguishing differences in the kinetics of flocculation among different types of flocculants.

Typical results of relationship between the time rate of change of optical extinction value (A) with time, v, and the flocculant dosage per unit mass of kaolin particles were shown in Fig. 5. In many previous studies on water treatment, polymeric flocculants have been widely used to enhance aggregation of suspended particles through mechanisms such as charge neutralization and polymer bridging. These flocculants generally produce large flocs rapidly, which leads to a rapid decrease in turbidity or optical attenuation of the suspension.

In the present study, the commercial polymer flocculants PA362 and PN161 showed rapid increases in transmitted light intensity, indicating fast flocculation kinetics. In contrast, the surface-modified scallop shell particles (SF) exhibited a slower change in transmitted light intensity. This difference can be attributed to the different flocculation mechanism. For polymeric flocculants, particle aggregation occurs mainly through polymer bridging between suspended particles, whereas for SF particles the aggregation is considered to occur mainly through heteroaggregation and collision between kaolin particles and the modified shell particles. Although the aggregation kinetics of SF particles were slower than those of polymer flocculants, the results demonstrate that the proposed optical monitoring method can clearly distinguish differences in flocculation behavior among different types of flocculants. The unit of the abscissa of Fig. 5 represents the mass (gram) of flocculant dose per unit mass (gram) of kaolin particles. The ordinate of Fig. 5 is the time rate of change in A, v The concentration of the kaolin suspension was employed as 4 g/L. In the case of using organic polymer flocculant (PA362 (0.04g/L) and PN161 (3 g/L) in Fig. 5a), the maximum point of v were confirmed at ca. 1×10^{-4} g-flocculant/g-kaolin, and ca. 2×10^{-3} g-flocculant/g-kaolin for PA362 and PN161,respectively. The dosage that gives the maximum value of v may be considered as the optimal dosage. The value of v increases toward the maximum value as the dosage increases until the dosage that gives the maximum value of v. In the region of higher dosages than the

dosage that gives the maximum value of v, conversely, the value of v decreased. Dosing flocculants in excess of the optimal dosage is thought that the bare surface of the kaolin particles, which is not adsorbed with the flocculant necessary for flocculation, is reduced, making it more difficult for flocculation to occur. Liu et al. (2024) have reported the flocculation experiments using gallic acid-grafted cationic chitosan as a flocculant and kaolin as test solid particles [53]. Borchert et al. (2021) have reported the flocculation experiments using polydiallyldimethylammonium chloride, polyacrylamide with trimethylammonium chloride, and chitosan as flocculant and silica and china-clay particles as test solid particles [57]. Their report described a similar flocculation phenomenon with unique optimal dosages. Similar phenomena have been observed in our previous studies [39, 43, 58]. On the other hand, in the case of using SF (10 g/L in Fig. 5b), no maximum value of v indicating the optimum addition was identified. It is considered that the optimum amount of dosage was not reached within the range of this experiment. Further improvement of the flocculation performance of SF may be achieved by controlling the particle size distribution, improving the dispersion of the particles by pre-wetting or stronger mixing, and exploring higher effective collision frequencies between particles. It was difficult to adjust the SF suspension to a high concentration and further SF addition was not practical.

The results of the clarification experiments were shown in Fig. 6 as the relationship between the relative optical extinction, A/A_0 and the the dosage (grams) of flocculant per unit mass (grams) of kaolin particles. The clarification experiments were conducted with varying the concentration of the kaolin suspension as 3, 5, and 10 g/L for PN362 (Fig. 6a), PN161 (Fig. 6b), and SF (Fig. 6c) flocculants, respectively. The relative optical extinction, A/A_0, reflects the residual rate of kaolin particle at certain depth from the surface of the suspension. The lower the value of A/A_0 means the better the clarification efficiency. In the case of using PA362 and PN161, Figs. 6a and 6b show that there is no significant difference in the effect of flocculant dosage per unit mass of kaolin on A/A_0, regardless of the concentration of the

kaolin suspension. There was also a minimum value of A/A_0, which was ca. 1×10^{-4} g-flocculant/g-kaolin and ca. 1.5×10^{-3} g-flocculant/g-kaolin for PA362 and PN161, respectively. These minimum values are almost identical to the dosage of the flocculant per unit mass of the kaolin particles that gives the maximum value of v seen in Fig. 5a.

In the case of using SF, the dosage of the flocculant per unit mass of the kaolin particles that gives the maximum value of v was not observed in Fig. 5. It was considered that SF is a solid particle and, unlike the dissolved molecules of PA362 and PN161, requires an increase in the frequency of particle collisions. However, as described above, for physical reasons, it was quite difficult to achieve the dosage of the flocculant higher than 1.7 g-flocculant/g-kaolin per unit mass of the kaolin particles in the present experimental system. The results obtained by using SF in Fig. 6c show that the A/A_0 values are much higher than those of PA362 and PN161. The reason for this high A/A_0 value could be largely due to the fact that in the clarification experiment, after the addition of the flocculant, the sample was stirred (500 rpm) for 1 min, then allowed to stand for 1 min and collected from 2 cm below the water surface. Therefore, the decrease in A/A_0 in this method was about 0.6 at the lowest in the range where the experiment was conducted. In the case of using SF flocculant, the trend was different for the concentration of the kaolin suspension of 10 g/L and lower concentrations of 3 g/L and 5 g/L. In the case of 10 g/L of the concentration of the kaolin suspension, the decrease in A/A_0 was observed up to the dosage of ca. 0.2 g-flocculant/g-kaolin of flocculant per unit mass of the kaolin particles, but the value of A/A_0 remained constant at about 0.7 at the dosage of ca. 0.2 g-flocculant/g-kaolin or the higher dosage. On the other hand, in the case of kaolin suspension concentrations of 3 and 5 g/L, there was almost no decrease in A/A_0 up to a dosage of approximately 0.24 g-flocculant/g-kaolin, but a decrease in A/A_0 values was observed at doses of 0.24 g-flocculant/g-kaolin or the higher dosage. The flocculant is compared to a common polymer flocculant, polyacrylamide combined with aluminum hydroxide [60]. In this experiment,

flocculants were dosed against 2.5 g/L of kaolin suspension. The clarification efficiency in this study was about 90%. This issue would be considered in future studies.

3.4. Relationship of the time rate of change in optical extinction and clarification efficiency

Based on Figs. 5 and 6, the relationships between the time rate of change in optical extinction, *v*, and the relative optical extinction, A/A_0 are shown in Fig. 7. The abscissa is the relative optical extinction, A/A_0, and the ordinate is time rate of change in the optical extinction, *v*. In the case of PA362, ca. 0.2 for the value of A/A_0 corresponds to the dosage of the flocculant per unit mass of the kaolin particles of ca. 1×10^{-4} g-flocculant/g-kaolin. In the case of PN161, ca. 0.04-0.1 for the value of A/A_0 corresponds to the dosage of the flocculant per unit mass of the kaolin particles of ca. $1\text{-}2\times10^{-3}$ g-flocculant/g-kaolin. Moreover, in the case of SF, ca. 0.6 for the value of A/A_0 corresponds to the dosage of the flocculant per unit mass of the kaolin particles of ca. 0.2 g-flocculant/g-kaolin. In addition, it could be seen that v also finely reflected the difference in the degree of flocculation speed in the value of A/A_0 in the range of 0.1-0.2 for PA362 and 0.04-0.1 for PN161 where rapid flocculation was occurring. In the case of SF, where flocculation occurred more slowly than in the case of PA362 and PN161, the degree of faster or slower flocculation speed could also be expressed by *v* value. The usage of *v* value as the indicator could express fine differences in the degree of apparent clarification rate.

The turbidimetric method (clarification experiment in this study) is admittedly easy and available evaluation method of flocculation for everyone. However, the sample could not be correctly evaluated unless the appropriate sampling position and sampling time were set, which requires trial and error in advance. On the other hand, the measurement of apparent clarification rate by the optical transmission method takes time depending on the state of flocculation. However, considerations of the sample collection position and time as

turbidimetric method are not necessary. The present method shown in this study is effective in that it can be used as an indicator of the degree of flocculation by evaluating the apparent clarification rate based on changes in the illuminance of the entire system in which flocculation and sedimentation occur.

4. Conclusions

In this study, we developed a surface-modified particle flocculant derived from waste scallop shells (SF). The flocculation performances were compared with those of typical polymer organic flocculants. The flocculation performance was evaluated using the relative optical extinction value based on the widely used clarification method and the time rate of change in optical extinction obtained from the measurements of time change in illuminance. Although the flocculation performance of SF was inferior to that of typical organic polymer flocculants, flocculation sedimentation with kaolin particles was observed in the system, and it was confirmed that SF has flocculation ability even with solid particles. However, since SF is not dissolved, a fairly high concentration of SF particle suspension was required. Evaluation by relative optical extinction value requires time to set the appropriate sampling time and sampling position, but is widely used because it is a simple method. Although the measurements of time change in illuminance proposed in this study requires the use of special equipment, it is an excellent method for sensing the aggregation state of the entire system. Comparison of the two evaluation methods indicates that the method using the time rate of change in optical extinction can quantify more detailed evaluation indices. In the further study, the author is going to prepare more effective solid particle for flocculation by using solid or soft-material particles.

Acknowledgments

Author gratefully acknowledges financial support provided by The South Hokkaido Science Promotion Foundation for a part of this study. Author gratefully appreciates Dr. Shiro Takahashi and Dr. Tomoaki Sugawara, Hokkaido Industrial Technology Center, for his technical support regarding SEM instrument. Author also appreciates Dr. Harumi Matsui, National Institute of Technology, Hakodate College for her technical guidance about measurement by FT-IR instrument. Author also appreciates Mr. Shotaro Ishiguri, a former student of the department of Bioresources chemistry, Faculty of Fisheries, Hokkaido University, for his experimental effort.

Rerefences

[1] A Tenca, R.D. Cusick, A. Schievano, R. Oberti, B.E. Logan, Evaluation of low cost cathode materials for treatment of industrial and food processing wastewater using microbial electrolysis cells, International Journal of Hydrogen Energy, 38, 1859-1865 (2013). http://dx.doi.org/10.1016/j.ijhydene.2012.11.103.

[2] V. Leifeld, T.A. Takashina, J.C. Zeni, M.A. Ribeiro, L. Igarashi-Mafra, Sequential coagulation treatment and processes based on ozone (O3 , O3 /UV, O3 / UV/H2 O2) for remediation of agro-food industrial wastewater, Desalination and Water Treatment, 106, 80-89 (2018). DOI: 10.5004/dwt.2018.22007

[3] Y. Sun, Y. Liu, J. Chen, Y. Huang, H. Lu, W. Yuan, Q. Yang, J. Hu, B. Yu, D. Wang, W. Xu, H. Wang, Physical pretreatment of petroleum refinery wastewater instead of chemicals addition for collaborative removal of oil and suspended solids, Journal of Clearer Production, 278, 123821 (2021). https://doi.org/10.1016/j.jclepro.2020.123821

[4] C.E. Santo, V.J.P. Vilar, A. Bhatnagar, E. Kumar, C.M.S. Botelho, R.A.R. Boaventura, Biological treatment by activated sludge of petroleum refinery wastewaters, Desalination and Water Treatment, 51, 6641–6654 (2013). DOI: 10.1080/19443994.2013.792141

[5] N. Alam, O. Ozdemir, M. Hampton, A. Nguyen, Dewatering of coal plant tailings: Flocculation followed by filtration, Fuel, 90, 26-35 (2011). DOI:10.1016/j.fuel.2010.08.006

[6] Y, Nakazawa, T, Abe, Y, Matsui, K, Shinno, S, Kobayashi, N, Shirasaki, T. Matsushita, Differences in removal rates of virgin/decayed microplastics, viruses, activated carbon, and kaolin/montmorillonite clay particles by coagulation, flocculation, sedimentation, and rapid sand filtration during water treatment, Water Research, 203, 117550(2021). https://doi.org/10.1016/j.watres.2021.117550

[7] O. Samuel, M.H.D. Othman, R. Kamaludin, O. Sinsamphanh, H. Abdullah, M.H. Puteh, T.A. Kurniawan, T. Li, A.F. Ismail, M.A. Rahman, J. Jaafar, T. El-badawy, S.C. Mamah, Oilfield-produced water treatment using conventional and membrane-based technologies for beneficial reuse: A critical review, 308, 114556(2022). https://doi.org/10.1016/j.jenvman.2022.114556

[8] G.A. Ekama, M.C. Wentzel, S.W. Sotemann, Tracking the inorganic suspended solids through biological treatment units of wastewater treatment plants, Water Research, 40,

3587-3595 (2006). DOI:10.1016/j.watres.2006.05.034

[9] L. He, F. Ji, X. He, W. Zhou, X. Xu, M. Lai, Validation of accumulation models for inorganic suspended solids of different particle size in an activated sludge system, Bioresource Technology, 149, 51–57 (2013). http://dx.doi.org/10.1016/j.biortech.2013.09.042

[10] C. Ratanatamskul, T. Siritiewsri, A compact on-site UASBeEGSB system for organic and suspended solid digestion and biogas recovery from department store wastewater, International Biodeterioration & Biodegradation, 102, 24-30 (2015). http://dx.doi.org/10.1016/j.ibiod.2015.04.002

[11] K.H. Rosenwinkel, D. Weichgrebe, H. Meyer, and D. Wendler, Suspended Solids from Industrial and Municipal Origins, Ecotoxicology and Environmental Safety, 50, 135-142 (2001). DOI:10.1006/eesa.2001.2082

[12] D. Wang, W. Sun, Y. Xu, H. Tang, J. Gregory, Speciation stability of inorganic polymer flocculant–PACl, Colloids and Surfaces A: Physicochemical and Engineering Aspects, 243, 1-10 (2004). DOI:10.1016/j.colsurfa.2004.04.073

[13] D. Wang , H. Tang, Modified inorganic polymer flocculant-PFSi: Its preparation, characterization and coagulation behavior, Water Research, 35, 3418-3428 (2001). https://doi.org/10.1016/S0043-1354(01)00034-3

[14] H. Chen, X. Wang, H. Liang, Z. Yan, Z. Ma, Z. Wang, C. Du, Preparation, oil removal and flocculation efficiency evaluation of a PAC-P (AM-BA) hybrid polymer flocculant based on response surface method, Journal of Environmental Chemical Engineering, 12, 114503 (2024). https://doi.org/10.1016/j.jece.2024.114503

[15] Q. Wang, W. Wang, K. Zhang, Z. Wang, K. Su, Molecular simulation of polyacrylamide types on flocculation performance in oily wastewater, Chemical Physics Letters, 867, 142023 (2025). https://doi.org/10.1016/j.cplett.2025.142023

[16] C. Castillo, P. Fawell, A. Costine, Optimising the activity of acrylamide-based polymer solutions used to flocculate mineral processing tailings suspensions – A review, Chemical Engineering Research and Design, 199, 214-237 (2023).

[17] V.P. Beyer, J. Blockx, A.P. Maciel, S. Tyagi, I. Demir-Yilmaz, C´. Formosa-Dague, K. Muylaert, W. Thielemans, Use of PDMAEMA as a non-toxic synthetic flocculant for

harvesting microalgae and evaluation of the influence of polymer size on flocculation behaviour, Journal of Environmental Chemical Engineering, 11, 111094 (2023). https://doi.org/10.1016/j.jece.2023.111094

[18] N.R.H. Rao, V. Beyer, W. Thielemans, K. Muylaert, Techno-economic comparison of flocculation combined with dissolved air flotation versus sedimentation for microalgae harvesting, Algal Research, 81, 1038581 (2024). https://doi.org/10.1016/j.algal.2024.103581

[19] Y. Arryanto, L.S. Bark, Reineckate ions as a precipitant for the determination of trace amounts of polyacrylamides in potable waters, Analytica Chimika Acta, 263, 119-129 (1992). https://doi.org/10.1016/0003-2670(92)85433-7

[20] B. Bolto, J. Gregory, Organic polyelectrolytes in water treatment, WATER RESEARCH, 41, 2301-2324 (2007). DOI:10.1016/j.watres.2007.03.012

[21] K.L. Dearfield, C.O. Abernathy, M.S. Ottley, J.H. Brantner, P.F. Hayes, Acrylamide: its metabolism, developmental and reproductive effects, genotoxicity, and carcinogenicity, Mutation Research, 195, 45-77 (1988). https://doi.org/10.1016/0165-1110(88)90015-2

[22] R. Hasegawa, K. Naitoh, Y. Kawasaki, Y. Aida, J. Momma, M. Saitoh, Y. Nakaji and Y. Kurokawa, Acute and subacute toxicity studies on 2,3-dichloropropionic acid and chlorinated polyacrylamide in rats, Water Research, 24, 661-666 (1990). https://doi.org/10.1016/0043-1354(90)90200-P

[23] S. Lattemann, T. Hopner, Environmental impact and impact assessment of seawater desalination, Desalination, 220, 1-15 (2008). DOI:10.1016/j.desal.2007.03.009

[24] C. Ruden, Acrylamide and cancer risk – expert risk assessments and the public debate, Food and Chemical Toxicology, Toxicol, 42, 335-349 (2004). DOI:10.1016/j.fct.2003.10.017

[25] The Ministry of Health, “Ministerial Ordinance Establishing Technical Standards for Water Supply Facilities” 2000.

[26] R. Shogren, Flocculation of kaolin by waxy maize starch phosphates, Carbohydrate Polymers, 76, 639-644 (2009). DOI:10.1016/j.carbpol.2008.11.027

[27] Q. Du, H. Wei, A. LI, H. Yang, Evaluation of the starch-based flocculants on flocculation of hairwork wastewater, Science of The Total Environment, 601-602, 1628-1637 (2017). http://dx.doi.org/10.1016/j.scitotenv.2017.06.029

[28] D. Sun, J. Zeng, D. Yang, X. Qiu, W. Liu, Full biomass-based multifunctional flocculant from lignin and cationic starch, International Journal of Biological Macromolecules, 253(2023)127287. https://doi.org/10.1016/j.ijbiomac.2023.127287

[29] K. Kemppainen, T. Suopajarvi, O. Laitinen, A. Ammala, H. Liimatainen, M. Illkainen, Flocculation of fine hematite and quartz suspentions with anionic cellulose nanofibers, Chemical Engineering Science, 148, 256-266 (2016). http://dx.doi.org/10.1016/j.ces.2016.04.014

[30] N. Yudong, Z. Tao, W. Haihua, C. Haixing, Upcycling harmful algal blooms into short-chain organic matters assisted with cellulose-based flocculant, Bioresource Technology, 397, 130425 (2024). https://doi.org/10.1016/j.biortech.2024.130425

[31] R. Rojas-Reyna, S. Schwarz, G. Heinrich, G. Retzold, S. Schutze, J. Bohrisch, flocculation efficiency of modified water soluble chitosan versuscommonly used commercial polyelectrolytes, Carbohydrate Polymers, 81, 317-322 (2010). DOI:10.1016/j.carbpol.2010.02.010

[32] J. Zheng, X. Chw, S. Zhang, T. Huang, H. Zheng, B. Sun, Relationship between the structure of chitosan-based flocculants and their performances in the treatment of model azo dyeing wastewater, Chemosphere, 247, 125920 (2020). https://doi.org/10.1016/j.chemosphere.2020.125920

[33] N. Amaly, S. Harrison, J.S. Tumuluru, G. Sun, P.K. Pandey, Development and application of a polycationic soybean protein-based f locculant for enhanced flocculation and dewatering of dairy manure, 371, 144050 (2025). https://doi.org/10.1016/j.chemosphere.2024.144050

[34] A. Aryan, P. Pal, S. Singh, T. Bhoutika, S.K. George, S. Sinha, J.P. Pandey, G. Sen, Development of grafted pea protein (PP-g-PAM): An innovative derivative as flocculant towards mineral suspensions, Industrial Crops & Products, 222, 119685 (2024). https://doi.org/10.1016/j.indcrop.2024.119685

[35] W. Liao, Y. Bian, C. Liu, M. Liu, W. Liang, Z. Gao, Y. Gao, J. Xie, A new flocculant tannic acid for improving separation efficiency and lowering gas emissions from liquid

and solid fractions of dairy slurry, Journal of Cleaner Production, 494, 145040 (2025). https://doi.org/10.1016/j.jclepro.2025.145040

[36] Y.T. Hameed, A. Idris, S.A. Hussain, N. Abdullah, H. Che Man, Effect of pre-treatment with a tannin-based coagulant and flocculant on a biofilm bacterial community and the nitrification process in a municipal wastewater biofilm treatment unit, J. Environ. Chem. Eng., 8, 103679 (2020). https://doi.org/10.1016/j.jece.2020.103679

[37] J. Sánchez-Martín, M. González-Velasco, J. Beltrán-Heredia, Surface water treatment with tannin-based coagulants from Quebracho (Schinopsis balansae), Chem. Eng. J., 165, 851-858 (2010). https://doi.org/10.1016/j.cej.2010.10.030

[38] R. Singh, S. Kumar, M. Garg, Domestic wastewater treatment using tanfloc: a tannin based coagulant, Geostatistical and Geospatial Approaches for the Characterization of Natural Resources in the Environment, Springer, Cham. https://doi.org/10.1007/978-3-319-18663-4_53

[39] H. Seki, H. Maruyama, Y. Shoji, Flocculation of diatomite by a soy protein-based bioflocculant, Biochemical Engineering Journal, 51, 14-18 (2010). DOI:10.1016/j.bej.2010.04.009

[40] H. Seki, A. Suzuki, Flocculation of diatomite by methylated egg albumin, Journal of Colloid and Interface Science, 263, 42-46 (2003). DOI:10.1016/S0021-9797(03)00288-1

[41] H. Seki, A. Suzuki, M. Shinguh, H. Maruyama, Flocculation of diatomite by methylated milk casein in seawater, Journal of Colloid and Interface Science, 270, 359-363 (2004). DOI:10.1016/j.jcis.2003.09.025

[42] H. Maruyama, H. Seki, Y. Satoh, Removal kinetic model of oil droplet from o/w emulsion by adding methylated milk casein in flotation, Water Research, 46, 3094-3100 (2012). DOI:10.1016/j.watres.2012.03.016

[43] H. Maruyama, H. Seki, A. Igi, Flocculation of quartz and kaolin by alginate-protamine complex, Biochemical Engineering Journal, 162, 107713 (2020). https://doi.org/10.1016/j.bej.2020.107713

[44] Hokkaido Government ;”Survey on the Generation of Fisheries-Related Waste and Its Amount” 2021.

[45] N. Chaihad, I. Kurnia, A. Yoshida, C. Watanabe, K. Tei, P. Reubroycharoen, Y. Kasai, A. Abudula, and G. Guan, Catalytic pyrolysis of wasted fishing net over calcined scallop shells: Analytical Py-GC/MS study, Journal of Analytical and Applied Pyrolysis, 146, 104750 (2019). https://doi.org/10.1016/j.jaap.2019.104750

[46] S. Sirisomboonchai, M. Abuduwayiti, G. Guan, C. Samart, S. Abliz, X. Hao, K. Kusakabe, A. Abudula, Biodiesel production from waste cooking oil using calcined scallop shell as catalyst, Energy Conversion and Management, 95, 242–247 (2015). http://dx.doi.org/10.1016/j.enconman.2015.02.044

[47] S.H. Yeom, K. Jung, Recycling wasted scallop shell as an adsorbent for the removal of phosphate, Journal of Industrial and Engineering Chemistry, 15, 40-44 (2009). DOI:10.1016/j.jiec.2008.08.014

[48] T. Chenet, G. Schwarz, C. Neff, B. Hattendorf, D. Günther, A. Martucci, M. Cescon, A. Baldi, L. Pasti, Scallop shells as biosorbents for water remediation from heavy metals: Contributions and mechanism of shell components in the adsorption of cadmium from aqueous matrix, Heliyon, 10, e29296 (2024). https://doi.org/10.1016/j.heliyon.2024.e29296

[49] A. Tsuruma, Y. Nomoto, M. Nishio, M. Ishikawa, J. Sawai, Efficacy of sorbitol-coated heated scallop-shell powder for the antimicrobial treatment of fresh vegetables, Food Control, 110, 106972 (2020). https://doi.org/10.1016/j.foodcont.2019.106972

[50] P.P. Shetty, A.U. Rao, B.H.V. Pai, M.V. Kamath, Performance of High-Strength Concrete with the Effects of Seashell Powder as Binder Replacement and Waste Glass Powder as Fine Aggregate, Journal of Composites Science, 7, 92 (2023). https://doi.org/10.3390/jcs7030092

[51] Z. Deng, J. Lin, N. Li, A review on recycling seashells as aggregates and binders for mortar and concrete in China: Production, engineering properties and new applications, Sustainable Materials and Technologies, 43, e01242 (2025). https://doi.org/10.1016/j.susmat.2025.e01242

[52] H. Maruyama, Preparation of Adsorbent from Mechanochemical Reaction-Based Waste Seashell with Sodium Oxalate and Its Application in Pb Ion Adsorption, surfaces, 7, 208-224 (2024). https://doi.org/10.3390/surfaces7020014

[53] J. Liu, H. Zhao, M. Wang, W. Ban, X. Lu, B. Yan, The preparation of gallic acid-grafted

cationic chitosan as effective salt-tolerant flocculants, RSC Advances, 14, 15021-15030 (2024). DOI: 10.1039/d4ra02046d

[54] S. Pandey, A. Kumar, Krishna Kumar, S. Krishnamoorthi, K. Tungala, Fabrication, characterization and flocculation properties of starch, chitosan or cellulose based graft copolymers: A review, Bioresource Technology Reports, 29, 102023 (2025). https://doi.org/10.1016/j.biteb.2025.102023

[55] J. Gregory, Monitoring particle aggregation processes, Advances in Colloid and Interface Science, 147-148, 109-123 (2009). DOI:10.1016/j.cis.2008.09.003

[56] H.Q. Li, A. Chen, S.G. Roscoe, J. Lipkowski, Electrochemical and FTIR studies of L-phenylalanine adsorption at the Au(III) electrode, Journal of Electroanalytical Chemistry, 500, 299-310 (2001). https://doi.org/10.1016/S0022-0728(00)00391-0

[57] G. Su, F. Deng, C. Sun, M. Su, A study on hybrid light extinction model and inversion based on Lambert-Beer Law, 577, 131342 (2025). https://doi.org/10.1016/j.optcom.2024.131342

[58] K.B.L. Borchert, C. Steinbach, S. Schwarz, D. Schwarz, A Comparative Study on the Flocculation of Silica and China Clay with Chitosan and Synthetic Polyelectrolytes, Marine Drugs, 19, 102 (2021). https://doi.org/10.3390/md19020102

[59] H. Maruyama, H. Seki, Influence of methylated milk casein flocculant dosage on removal rate of oil droplet removal from o/w in flotation, Biochemical Engineering Journal, 159, 107584 (2020). https://doi.org/10.1016/j.bej.2020.107584

[60] W.Y. Yang, J.W. Qian, Z.Q. Shen, A novel flocculant of Al(OH)3–polyacrylamide ionic hybrid, Journal of Colloid and Interface Science, 273, 400-405 (2004). DOI:10.1016/j.jcis.2004.02.002

Credit authorship contribution statement

Hideo Maruyama: Planning the experiments of this research, Conducting the experiments, Analysis of the experimental data, Writing and correcting manuscript.

Declaration of Competing Interest

The author declares that I have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

Data will be made available on request.

Figures captions

Fig. 1. Schematic diagram of the experimental setup for measurement of the change in the intensity of the illuminace. (1) Personal computer, (2) illminometer, (3) optical fiber, (4) LED light, (5) glass graduated cylinder, (6) light emitting/receiving apparatus, (7) magnetic stirrer, (8) power supply.

Fig. 2. Results of measurements concerning the characterization of the modified shell particle: Particle size distribution (a), FT-IR spectra (b), and SEM images (C1 (×50) and C2 (×2,000)).

Fig. 3. Influences of the dosage of flocculant on the time course of change in the intensity of the illuminance through the flocculation cylinder in the case of using 0.04 g/L of PA362 (a), 3 g/L of PN161 (b), and 10 g/L of SF (c). The concentration of the kaolin suspension was 3 g/L.

Fig. 4. Influence of the dosage of flocculant on the time course of change in the clarification efficiency in the case of using PA362 (a), PN161 (b), and SF (c). The experimental conditions were the same as in Fig. 3.

Fig. 5. The relationship between the time rate of the change in optical extinction, v, and the dosage of the flocculants per unit mass of the kaolin particles for PA362 (open square), PN161 (open circle), and SF (open triangle). The experimental conditions were the same as in Fig. 3.

Fig. 6. The relationship between the relative optical extinction, A/A_0, and the dosage of the flocculants per unit mass of the kaolin particles for PA362 (a), PN161 (b), and SF (c). The experiments were conducted with using 3, 5, and 10 g/L of the concentration of the kaolin suspensions.

Fig. 7. The relationship between the time rate of the change in optical extinction, v, and the relative optical extinction, A/A_0, for PA362 (a), PN161 (b), and SF (c). The experiments were conducted with using kaolin suspensions of 3, 5, and 10 g/L.

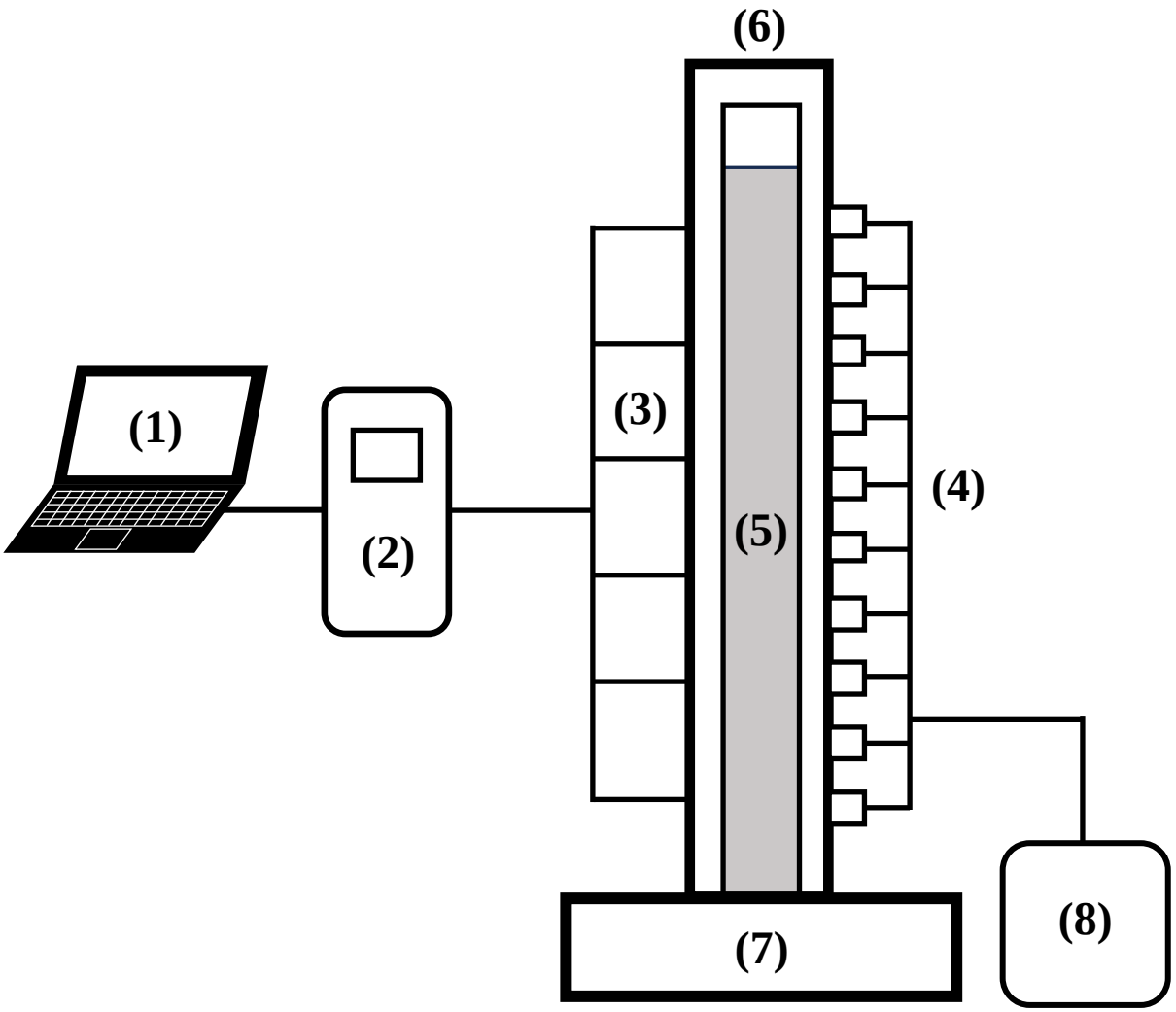


Fig. 1. Schematic diagram of the experimental setup for measurement of the change in the intensity of the illuminance. (1) Personal computer, (2) illuminometer, (3) optical fiber, (4) LED light, (5) glass graduated cylinder, (6) light emitting/receiving apparatus, (7) magnetic stirrer, (8) power supply.

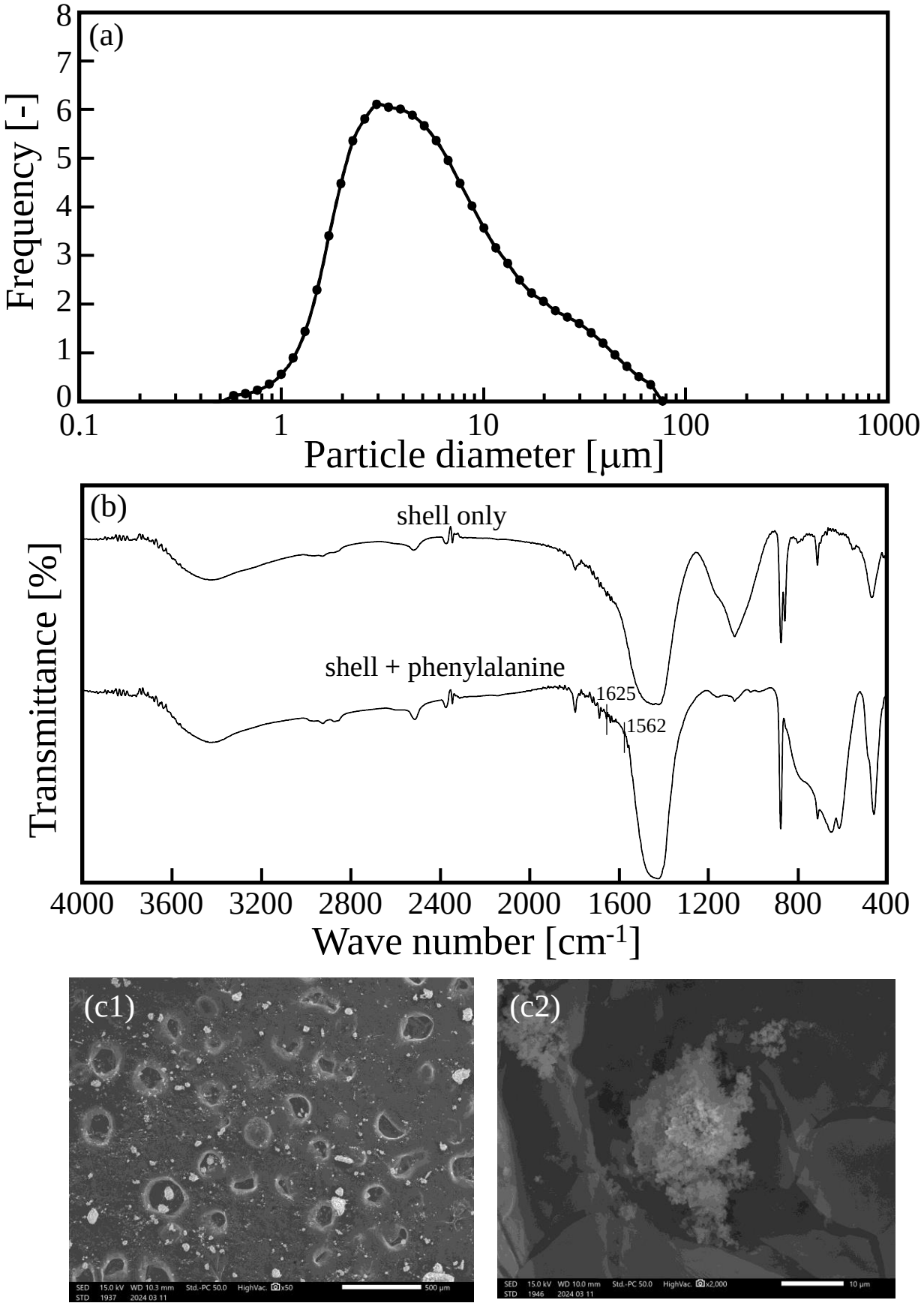


Fig. 2. Results of measurements concerning the characterization of the modified shell particle: Particle size distribution (a), FT-IR spectra (b), and SEM images (C1 (×50) and C2 (×2,000)).

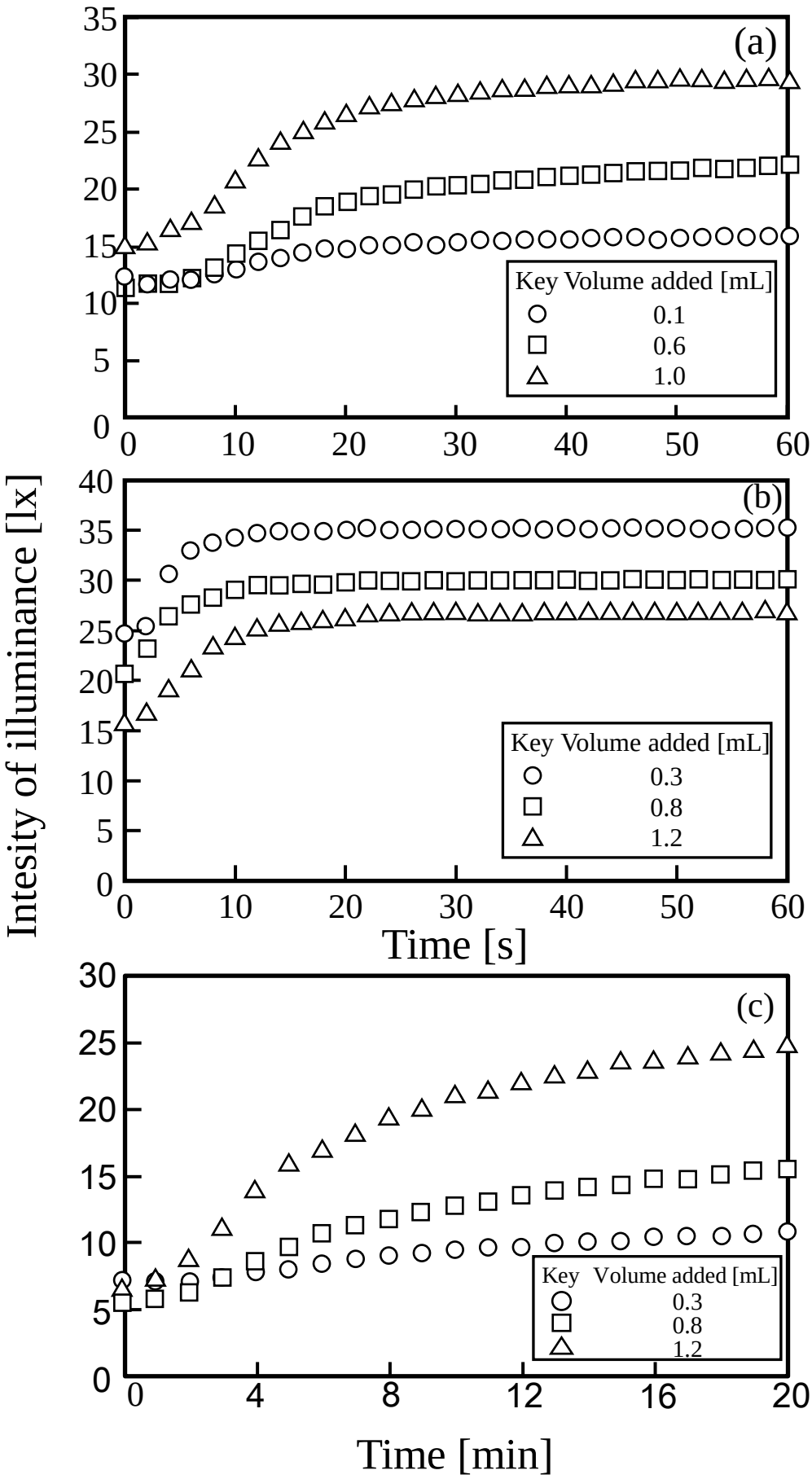


Fig. 3. Influences of the dosage of flocculant on the time course of change in the intensity of the illuminance through the flocculation cylinder in the case of using 0.04g/L of PA362 (a), 3g/L of PN161 (b), and 10g/L of SF (c). The concentration of the kaolin suspension was 3g/L.

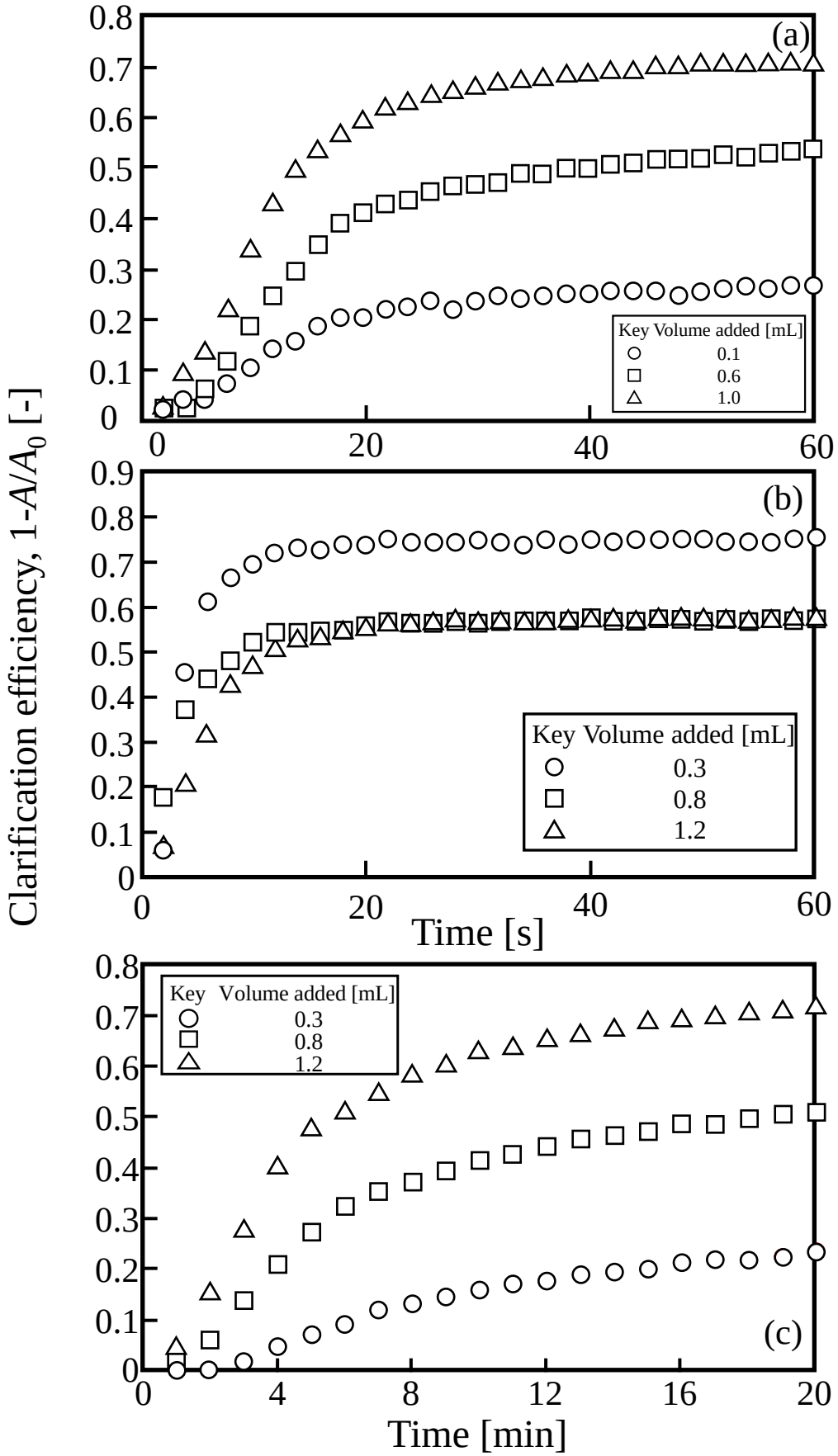


Fig. 4. Influence of the dosage of flocculant on the time course of change in the clarification efficiency in the case of using PA362 (a), PN161 (b), and SF (c). The experimental conditions and symbols were the same as in Fig. 3.

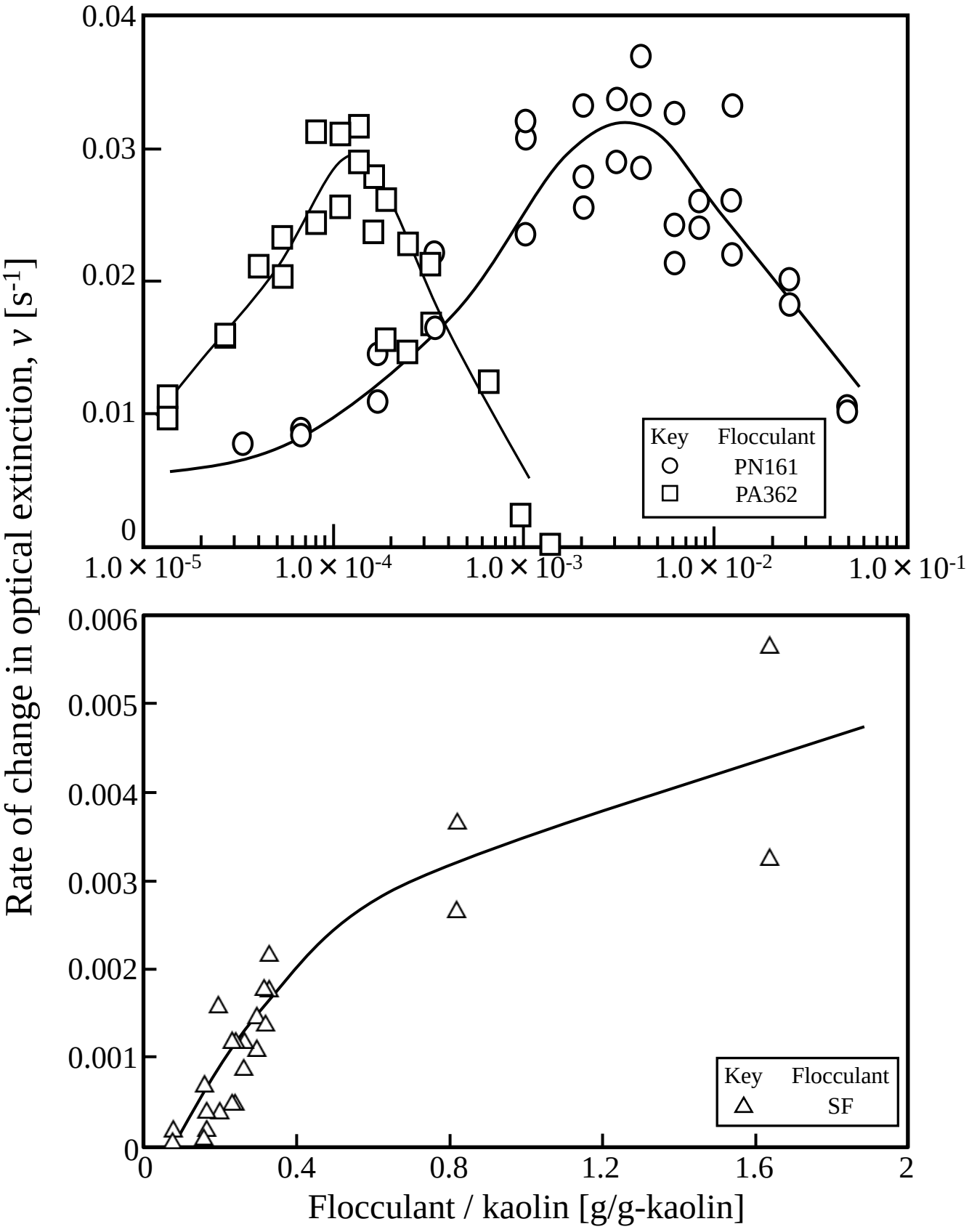


Fig. 5. The relationship between the same rate of the change in absorbance, *v*, and the dosage of the flocculants per unit mass of the kaolin particles for PA362 (open square), PN161 (open circle), SF (open triangle). The experimental conditions were the same as in Fig. 3.

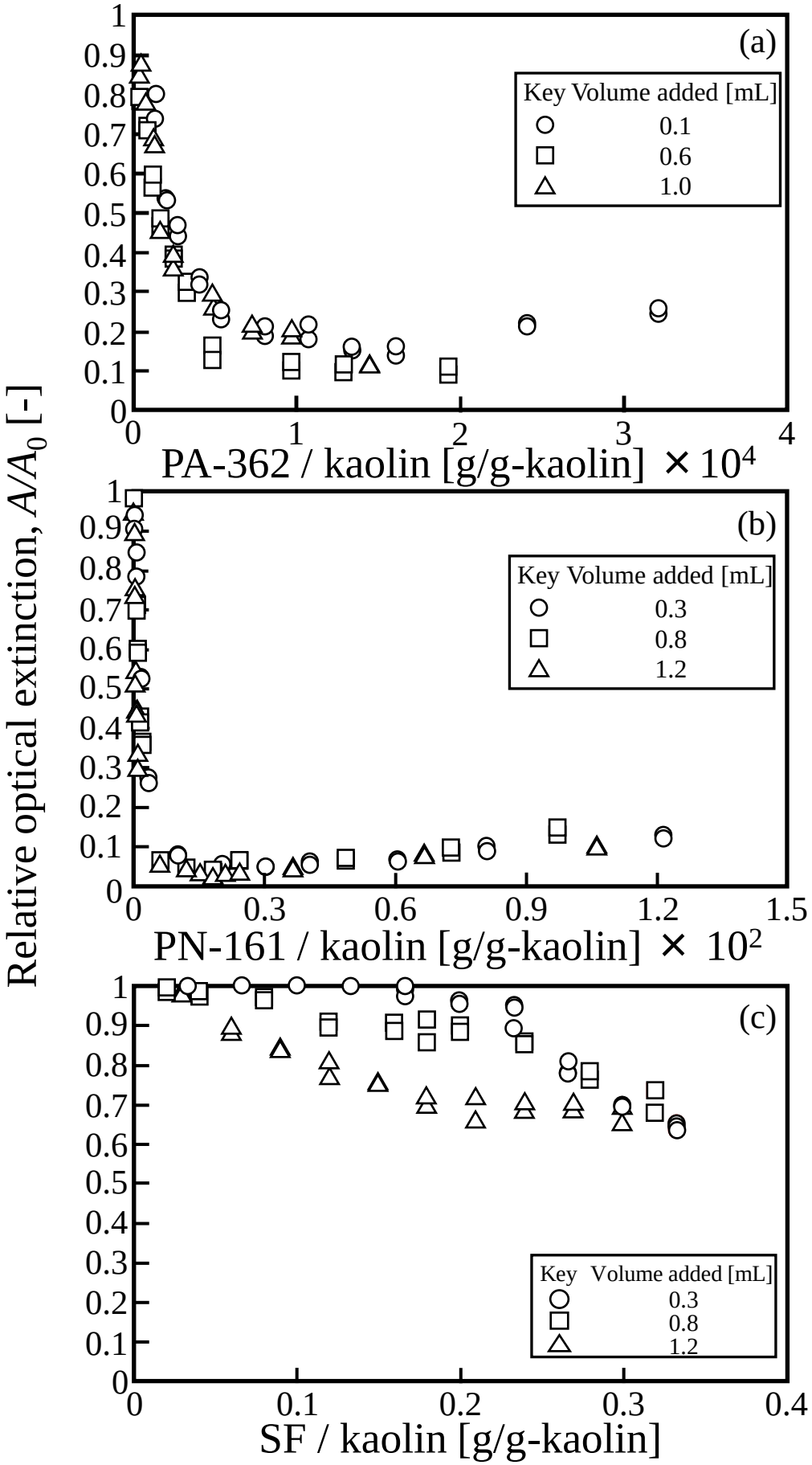


Fig. 6. The relationship between the relative optical extinction, A/A_0 and the dosage of the flocculants per unit mass of the kaolin particles for PA362 (a), PN161 (b), and SF (c). The experiments were conducted with using kaolin suspensions of 3, 5, and 10g/L.

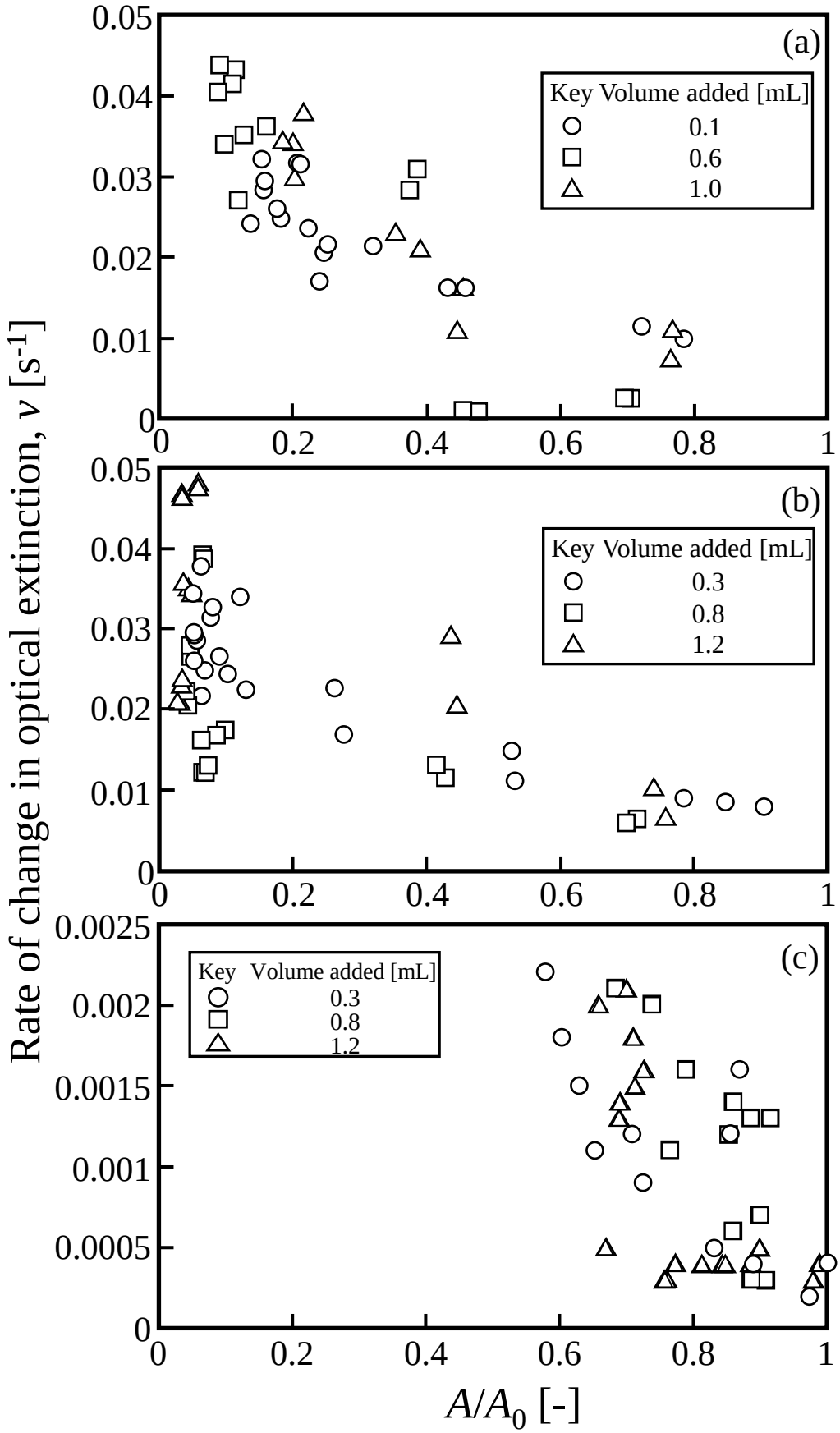


Fig. 7. The relationship between the time rate of change in absorbance, v, and the relative optical extinction, A/A_0, for PA362 (a), PN161 (b), and SF (c). The experiments were conducted with using 3, 5, and 10g/L of the concentration of the kaolin suspensions.