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# Separation of Suspended Solids in Cow Manure Effluent Particles by Electrocoagulation as a Pre-treatment

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## Abstract

Aluminum coagulants and cation polyacrylamide polymer coagulants (CPAM) have been used in wastewater treatment. But high concentrations of coagulant use have both health and environmental impacts, and CPAM is still much costly even it plays a large flocs bridging. In this study, electrocoagulation (EC) treatment with a platinized titanium anode and a steel cathode was carried out as a pre-treatment of cow manure effluent at 35 V, 0.55 A/cm$^2$, and 15 min operation time. An electrolysis apparatus is assembled to neutralize the negative charges of cow manure effluent particles owing to an oxidation reaction promoted on a non-consumable anode, by applying a device and surface pretreatment on a steel cathode to retard the cathodic reduction. After the treatment, removal rates of 99.1 % for turbidity and 90.7 % for COD were achieved with 200 ppm of AlCl3 and 80 ppm of CPAM. This apparatus for particle oxidation prevents contamination of the treated retentate by dissolved metal ions.

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Cow manure effluent, Electrocoagulation, Surface charge, Wastewater treatment

## Introduction

Toward the establishment of a recycling-oriented agricultural society, we recognize that livestock manure is not simply disposed of as waste, but is a raw material for compost, liquid fertilizer, and other products. The form of livestock manure to be treated depends on the type of livestock and the way the farm itself is operated, and it can be classified into three types: solid, slurry, and sewage, which are converted and effectively used as compost, liquid manure, and energy such as methane. Cow manure is an important source of inorganic and organic nutrients and may be used in agriculture for soil fertilization and improvement [1,2].

Aluminum (Al) coagulants were commonly used in the treatment of dairy manure effluent, palm oil mill effluent, and paper and pulp [3-5]. When Al coagulants are added into water, Al ions hydrolyze rapidly and form a range of metal hydrolysis species. These cationic species adsorb onto the negatively charged particles and neutralize the charge. In this mechanism, particles get destabilized, and aggregation occurs [6]. However, Al coagulants increase Al concentration in water which may become toxic either to plants, humans or to the aquatic environment [7,8]. Polyacrylamide has been also used in the treatment of dairy manure wastewater, paper and pulp wastewater, and beverage wastewater [9-11]. Cationic polyacrylamide (CPAM) is one of the most effective flocculants for the separation of colloidal suspensions [12,13]. CPAM removes pollutants and organic matter by (1) neutralizing the negatively charged colloidal particles, (2) the destabilized colloidal particles are wrapped and connected to form large flocs that dewater easily and quickly settle down under the effect of bridging and sweeping [14]. Cationic degree and relative molecular weight (limiting viscosity) are closely related to electrical neutralization and bridging effect performance, respectively [15]. Although polyacrylamide demonstrates advantages such as inertness to pH changes, high efficiency with low dosage, form bigger and stronger flocs generating less volume of sludge, however their application as flocculants market cost is higher [16]. Therefore, use of high Polyacrylamide concentration dosage should be limited to avoid cost efficiency.

Alternatively, electro-flotation (EF) may not only achieve similar performance of coagulation and flocculation but help overcome their drawbacks such as removing pollutants using none, or negligible amount of chemicals [17,18]. Experimental scales of EF have been used in the treatment of domestic wastewater paper and pulp [19,20]. EF method removed pollutant by the electrical generation in situ of coagulant material (metallic ions) by dissolution from a sacrificial metal anode to form a range of aqueous species and precipitates (primarily metal oxides and hydroxides) that can operate via destabilization of a colloidal suspension and aggregation of suspended particles. Although EF requires electrical energy, raising the overall costs of the process, its application in cow manure effluent treatment for agricultural purposes as fertilizers may be important, because it uses metallic ions and gas bubbles, not chemical coagulants. Hence reducing the use of chemical coagulants and avoiding environmental hazards. This study aims at demonstrating and confirming the hypothesis that electrocoagulation method (EC) using platinized titanium and steel anode as an anode and cathode electrode, respectively.

## Methodology

### Sample collection

The cow manure effluent was collected from a discharge point of a dairy farm, Enomoto-Bokujo, located in Ageo-City, Saitama Japan (35o 57’46”N, E139o 32’26”E). A manager translated raw materials into a polyethylene tank and then immediately transported to Applied Ecological Engineering laboratory in Saitama University (Figure 1). The cow manure effluent was later diluted until the upper limit of measurement and sieved using a 250-micron mesh sieve before experiment (Figure 2). Characteristics of the cow manure effluent are given in Table 1.

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| Component | Value |
|---|---|
| Turbidity, NTU | 4,000 |
| pH | 7.2 |
| Zetal potential, mV | -16 |
| CODcr, mg/L | 3,650 |

Table 1: Characteristic of cow manure effluent used in this study.

Figure 1: Sample collection of cow manure effluent, Enomoto-Bokujo, Ageo-City, Saitama Japan.

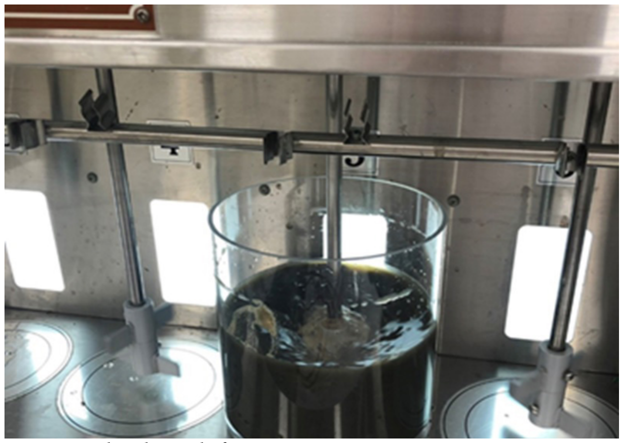

Figure 2: Diluted sample for experiment.

Where, the turbidity was determined using a turbidimeter (2100N, Hach, USA). The solution pH and ORP were measured with a glass electrode (TOA-DKK, Tokyo, Japan). Zeta potential was measured with Zeta-potential & particle size analyzer (ELSZ-2000 series, Otsuka Electronics Co., Ltd., Tokyo, Japan). CODcr was determined using Spectroquant® COD cell test (Merck KGaA, Darmstadt, Germany).

### Experimental setup

A cylindrical platinized titanium anode (12cm diameter x 12cm high) and a cylindrical steel cathode (14cm diameter x 17cm high) were installed in a 3-L cylindrical cell with distances of 1 cm and 2 cm from the lower edge of the anode to the bottom of the cell to ease flowing (Figure 3a and 3b). The cathode surface was covered with 0.2-mm pore size filter paper (ADVANTEC, No.5C, Tokyo, Japan) to retard the diffusion of the catholyte and avoid direct contact with caw manure particles; tiny holes were made at the top of the covered filter paper for effluent hydrogen gas. The anode and cathode were connected to a galvanostatic rectifier of 35V–2A (Kikusui, Yokohama, Japan). For electrolysis, the anode current density was 4–6 $mA/cm^2$ [21].

### Coagulant agent

Coagulation and flocculation experiments were performed on a jar test apparatus (VT-4P, Sugiyamagen Inc., Tokyo, Japan). Aluminum chloride (AlCl3) coagulant and cationic polyacrylamide flocculant (CPAM, commercial name: C-512, MT Aqua Inc., Tokyo Japan) were used as a chemical coagulant. The cationic polyacrylamide polymer was prepared from radical polymerization of acrylamide monomers, with characteristic given in Table 2.

### Experiment conditions

### Case-1: Coagulation method

Coagulation experiments were performed to determine the effect of $AlCl_3$ in cow manure effluent treatment and optimal $AlCl_3$ concentration. Firstly, predetermined $AlCl_3$ concentration at 50, 100, 200, 400, 600 and 800 ppm were added into 1L of cow manure effluent, respectively and rapidly stirred at 230 rpm for 3 min to fully collide $AlCl_3$ coagulants and suspension particles. Finally, the formed flocs freely settled for 10 mins and the supernatant turbidity, COD and

Zeta potential at depth 2 cm below the water surface were analyzed for $AlCl_3$ coagulation performance.

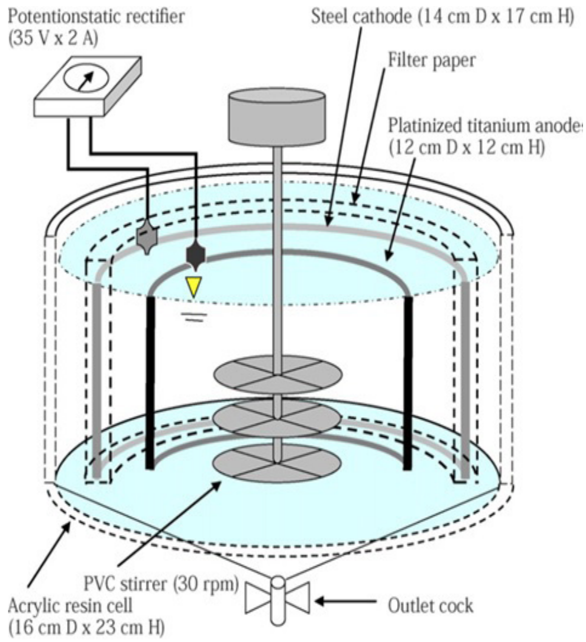


Figure 3a: Schematic diagram of the experimental setup for electrolytic treatment.

Figure 3b: Photo of the experimental setup for electrolytic treatment.

| | |
|---|---|
| Type | Cation |
| Principal component | Polyacrilic acid ester |
| Bulk specific gravity | 0.65 ± 0.1 |
| pH in water | 2.5-5.5 |
| Molecular weight | 230 mPa.s |

Table 2: Characteristic of cationic polyacrylamide flocculant (CPAM) used in this study, C-512.

**Case-2: coagulation -flocculation method**

Coagulation-flocculation experiments were performed to determined optimal CPAM concentration when a fixed amount of $AlCl_3$ concentration is added. Another reason was also to determine the effect and advantage of adding $AlCl_3$ to CPAM. Firstly, 200 ppm $AlCl_3$ coagulant was added into each beaker containing 1L of the cow manure and rapidly stirred for 1 min. Immediately predetermined CPAM concentration at 20, 40, 80, 120, 160 and 200 ppm were added, respectively and continued stirring at 230 rpm for 2 min. Finally, the formed flocs free settled for 10min. Coagulation-flocculation performances were evaluated by analyzing the supernatant turbidity, COD and Zeta potential at depth 2 cm below the water surface.

**Case-3: EF-coagulation-flocculation method**

EF-coagulation-flocculation experiments were performed to determine optimal CPAM concentration when a fixed amount of AlCl3 concentration and EF operation time is used with it. Another reason was to determine the advantage of using electrocoagulation method in this process. During each run, 1L of cow manure effluent was added to a reactor having platinized titanium anode and steel cathode. The reactors were placed under a jar test stirrer apparatus with the speed held constant at 120 rpm during each electro-flotation run that was performed at 15 mins electrolysis time. Next all electrodes were removed for coagulation-flocculation test which was done with jar test apparatus. Firstly, 200 ppm AlCl3 coagulant concentration was added into each beaker containing 1L of the pretreated cow manure without change of pH and rapidly stirring at 230 rpm for 1 min. Immediately predetermined CPAM concentration at 20, 40, 80, 120, 160 and 200 ppm were added, respectively and continued stirring for 2min at 230 rpm. Finally, the formed flocs were allowed to settle free for 10min. EF-coagulation-flocculation performances were evaluated by analyzing the supernatant turbidity, COD and Zeta potential at depth 2 cm below the water surface.

## Results and Discussion

**Effect of $AlCl_3$ concentration (Case-1)**

From the coagulation experiment, it was observed that increased $AlCl_3$ concentration enhanced pollutant and organic matter removal from cow manure effluent. Figure 4a shows that increased $AlCl_3$ concentration increased turbidity and COD removal. Optimum $AlCl_3$ concentration 800 ppm gave the highest removal rate of turbidity (99.8%) and COD (84.4%), respectively. In relation to pH, increase $AlCl_3$ concentration decreased the cow manure effluent pH at 6.1. Increase pollutant and organic matter removal after the addition of $AlCl_3$ concentration to cow manure effluent was because $AlCl_3$ coagulant hydrolyzed and formed positive charge aluminum ions, which neutralized the negative charges on the colloidal particles resulting to an increase Zeta potential value at -6.8 mV, thus promoting destabilization of the colloid dispersion and agglomeration of the resulting individual colloidal particles (Figure 4b) [4].

**Effect of CPAM concentrations with fixed 200ppm AlCl3 concentration (Case-2)**

Coagulation for Case-1 on cow manure treatment showed that high concentration of $AlCl_3$ was needed to give an appropriate result which could lead to high concentration of Al in the treated wastewater. It may threaten human life, plants, and aquatic environment [5-6]. Here, 200ppm $AlCl_3$ was applied with CPAM to enhance organic

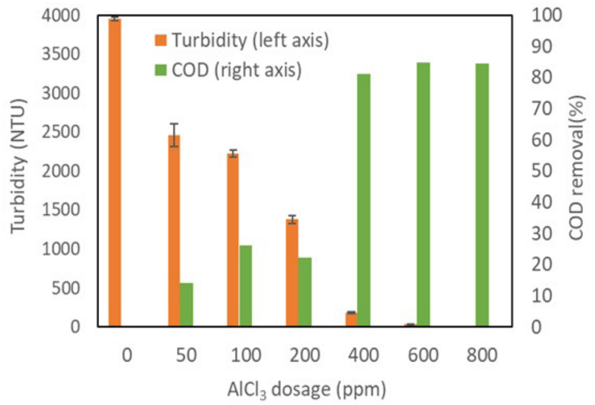


Figure 4a: Turbidity and COD removal rate results by adding AlCl3 on cow manure effluent treatment (Case-1).

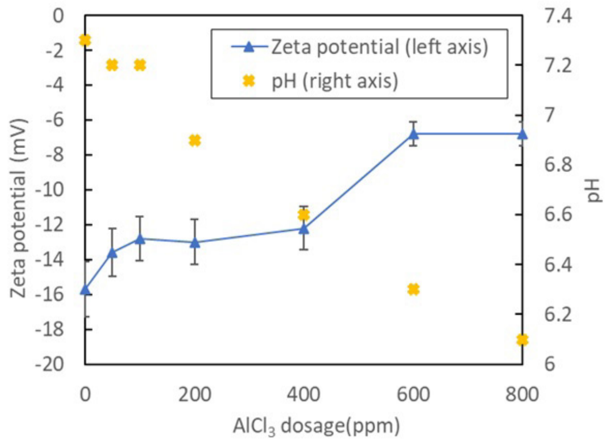


Figure 4b: Zeta potential and pH results by adding AlCl3 on cow manure effluent treatment (Case-1)

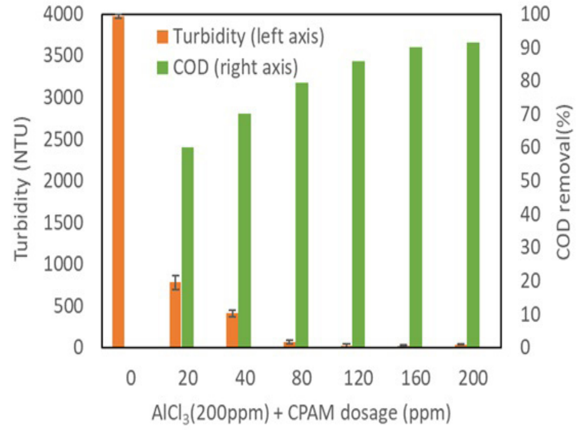


Figure 5a: Turbidity and COD removal rate results by adding $AlCl_3$ (200 ppm) and CPAM dosage on cow manure effluent treatment.

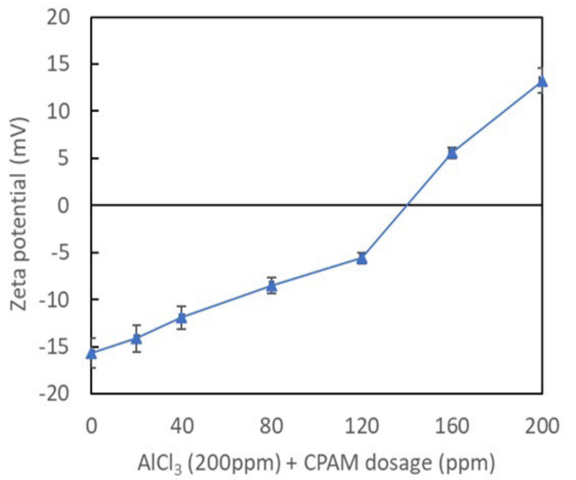


Figure 5b: Zeta potential result by adding AlCl3 (200 ppm) and CPAM dosage on cow manure effluent treatment (Case-2)

matter removal. Figure 5a shows turbidity and COD removal rate results by adding CPAM on cow manure effluent treatment under 200ppm $AlCl_3$ addition. Optimum CPAM concentration 200 ppm enhanced turbidity and COD removal rate by 99.1% and 91%, respectively. A slight increase in pH due to low $AlCl_3$ addition was also observed. Increase pollutant and organic matter removal after increased CPAM concentration under low $AlCl_3$ addition was because increased CPAM concentration enhanced the neutralization effect and bridging effect by destabilizing, increasing organic matter floc size and removal [14]. A charge neutralization is modified by both from positive charge Al ions and CPAM increased Zeta potential reached at −5 mV corresponding to 120 ppm of CPAM under 200 ppm of $AlCl_3$ concentration (Figure 5b). Further increase to 200 ppm of CPAM concentration resulted in a much higher zeta potential (+13mV) and turbidity removal tended to slight decrease.

### Effect of Electrocoagulation on $AlCl_3$ with CPAM (Case-3)

From the coagulation-flocculation experiment, it was noticed that fixed 200 ppm of $AlCl_3$ concentration enhanced organic matter removal on cow manure treatment when used with CPAM flocculant. The aim of this process was to enhance pollutant and organic matter removal and reduce chemical coagulant concentration. To achieve this aim, a fixed EC operation time 15 min was used with fixed 200 ppm of $AlCl_3$ concentration and different concentration of CPAM. Since increased electrocoagulation time results in an increased energy consumption, only 15 min EC operation time was applied [22,23]. Figure 6a shows that the optimum CPAM concentration at 80 ppm was noticed which gave 99.1% and 95% of removal rates for turbidity and COD, respectively. Trend is almost same as Case-2 and a difference is lower pH inner anode and aggregation is enhanced in EC system. The positive charge aluminum ion from $AlCl_3$ and CPAM cations increased the Zeta potential at -9.48 mV corresponding to 80 ppm of CPAM concentration (Figure 6b). Further increase to 200 ppm CPAM concentration with 15 min EC operation time resulted in a much higher zeta potential at +11.82 mV. While carrying out the water electrolysis, oxygen gas bubble is generated inner the anode which may play the leading role in the EC process, however, the aggregated sludge did not float (Figure 7a).

### Other characters of electrocoagulation method as a pre-treatment: effect of filter paper cover on cathode

Our study used filter paper cover on cathode to retard the diffusion of the catholyte and avoid direct contact with cow manure particles; tiny holes were made at the top of the covered filter paper for effluent hydrogen gas [24]. Figures 7a and Figure 7b show different

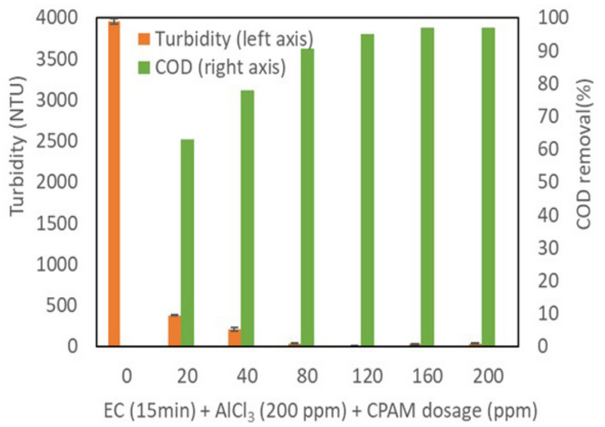


Figure 6a: Turbidity and COD removal rate results by electrocoagulation, EC (15 min) as a pre-treatment, and by adding AlCl3 (200 ppm) and CPAM dosage on cow manure effluent treatment (Case-3).

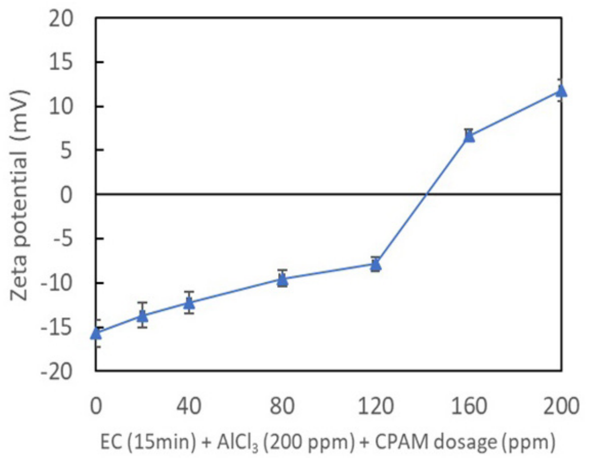


Figure 6b: Zeta potential result by electrocoagulation, EC (15 min) as a pre-treatment, and by adding AlCl3 (200 ppm) and CPAM dosage on cow manure effluent treatment (Case-3).

results between (a) with and (b) without covered filter paper. If we used filter covered, aggregated floc of cow manure sink which is the normal aggregation phenomenon. The pH of inner anode tended to be acid which are advance for coagulation. If we do not used filter covered, on the other hand, they became float due to hydrogen gas mixing produced on the cathode surface and pH was neutral due to mixing. Each final turbidity and COD removal rates were almost the same. Platinum coated titanium was utilized as an anode electrode and released oxygen gas from the surface. This is a different point from the previous study for dairy wastewater treatment [25]. An electrolysis apparatus is assembled to neutralize the negative charges of cow manure effluent particles owing to an oxidation reaction promoted on a non-consumable anode, by applying a device and surface pre-treatment on a steel cathode to retard the cathodic reduction. This apparatus for particle oxidation prevents contamination of the treated retentate by dissolved metal ions.

## Conclusions

We demonstrated electrocoagulation method as pre-treatment of cow manure effluent wastewater. An electrolysis apparatus is assembled to neutralize the negative charges of cow manure effluent particles owing to an oxidation reaction promoted on a non-consumable anode. This apparatus for particle oxidation prevents contamination of the treated retentate by dissolved metal ions and succeeded to reduce chemical coagulant. Also, it can be used as flotation method if we do not filter paper cover on cathode. Further experiment may be needed for more practical stage.

## Competing Interests

The authors declare that they have no competing interests.

## Author Contributions

Conceptualization, T.F.; methodology, E.Y.F. and T.F.; data analysis, E.Y.F.; writing—original draft preparation, E.Y.F.; writing—review and editing, T.F; visualization, T.F.; supervision, T.F.

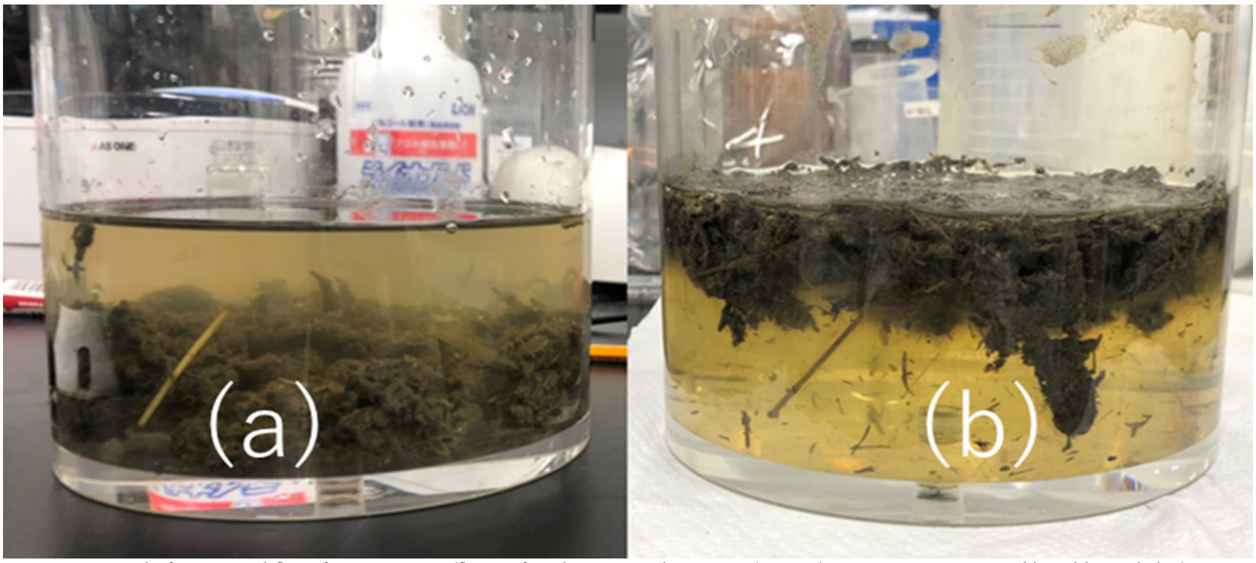


Figure 7: Control of aggregated floc of cow manure effluent after electrocoagulation, EC (15 min) as a pre-treatment, and by adding AlCl3 (200 ppm) and CPAM (80 ppm) dosage, (a) with filter paper covered, and (b) without filter paper covered.

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