Membrane filtration retentate thickening efficiency using electrolytic ion products instead of chemical coagulants

J.Y. Huang*, T. Uchiburi** and K. Fujita***

* Center Research Institute, Maezawa Industries, Inc., 5-11 Nakachuo, Kawaguchi, Saitama, 332-8556 Japan (E-mail: huang@maezawa.co.jp)
** Department of Environment Science and Human Engineering, Graduate School of Science and Engineering, Saitama University, 255 Shimo-Okubo, Urawa, Saitama, 338-8570 Japan
*** Japan Water Research Center, 2-8-1 Toranomon, Minato, Tokyo, 105-0001 Japan

Abstract Electrolytic coagulation has advantages compared with conventional chemicals coagulation treatment. First of all, the addition of alkaline agent is necessary in chemicals coagulation treatment as the addition of acid agents in coagulation of dense wash water makes pH decline. While not much change of pH is observed with electrolytic coagulation, where alkaline agent is not demanded. Secondly, it is easy to control the dose of additional coagulants. In this study, electrolytic coagulation sedimentation treatment is adopted for membrane retentate from membrane filtration treatment without using coagulants. As electricity can control the addition of coagulation, an automation method is applied in the experiment. The lower the iron as well as aluminum dissolved, the higher the sludge density becomes; especially aluminum has this tendency. It is possible to achieve 400 kg/m³ of condensed sludge density with artificial wash water and more than 80 kg/m³ for real retentate, in terms of electrolytic coagulation of iron anode. The sludge density of aluminum anode is higher, but its filtrate density is lower than iron anode.

Keywords Electrolytic coagulation; ion; membrane retentate; sedimentation; sludge; thickening

Introduction Nowadays, membrane filtration has already been adopted in water purification treatment. Normally, membrane water treatment processes do not use any coagulant in order to reduce cost and sludge production. Five to ten per cent of raw water is discharged as concentrate containing one to several hundred mg/L of suspended solids (Fujita and Takizawa, 1994). In fact, the thickening of suspended solids in concentrate is not satisfactory in a conventional gravity thickener without chemical flocculation (GMTD, 1997). Consequently, membrane retentate from membrane filtration treatments has the same problem as sludge from conventional purification systems (Huang et al., 1999, 2001).

The difficulty of sludge treatment is in the addition of substances in the purification and sludge treatment process that increase the capacity and moisture content of sludge, and make it difficult to dehydrate and cause large variation of pH value as well. This also causes the problem of transportation and strength of soil, which increased pollution of soil and groundwater by sludge content. In order to solve the above problems and improve the efficiency of treatment, it is important to maintain natural circulation and environment. That is why we try to find a feasible treatment process for membrane retentate, which contains colloids and metals for the adaptation of the membrane filtration process to large-scale plants.

In this study, electrolytic coagulation treatment is adopted to treat condensed turbid water of membrane retentate without using coagulants. In terms of electrolytic coagulation, it has the following benefits compared with conventional chemicals coagulation.

First of all, no need to add alkaline agent. As the addition of acid agent in coagulation of dense wash water makes pH decline, it is necessary to use alkaline agent. While not much
change of pH is observed with electrolytic coagulation, and alkaline agent is not demanded.

Secondly, it is easy to control the addition of coagulant dose. An automation method is feasible in the experiment, as electricity can control the addition of coagulation.

**Materials and methods**

The characteristics and concentration of sludge with this method is investigated compared with those of chemical flocculation. The main condensed wash water is artificially kaolin turbid and part of the wash water is real wash water from Yokohama Kosuzume membrane water treatment processes (Huang et al., 2000). A schematic diagram of the experimental setup is shown in Figure 1.

In this electrolytic treatment, aluminium and iron are used for the positive electrode. In order to coagulate formatted ion, NaClO was added in the treated water before iron was electrolyzed. In the chemical treatment, alum was compared with Fe$^{2+}$ sulfate. Electrolytic ion density was measured and recorded based on an electric circuit and each electrolysis treatment (Uchiburi, 1999). The specifications of each electrolytic coagulation experiment are as follows:

**Batch experiment**

Electrolytic bath size: 142 Ø × 250 depth transparent acrylic fiber,
Capacity: 3 × 10^{-3} m^{3}, Stirrer: 40 w,
Electrode measure: anode: 140 Ø × 170 depth, valid area: 5.272 × 10^{-2} m^{2},
Cathode: 120 Ø × 117 depth, valid area: 4.41 × 10^{-2} m^{2},

After electrolytic coagulation of sham retentate or real membrane retentate in the electrolysis bath, it was retained for 1 to 24 hours, and filtrate turbidity was measured. When filtrate was separated, condensed liquid was then withdrawn to a mess cylinder and kept for 24 hours. Then, sludge capacity as well as suspended substance density was measured.

**Continuous experiment**

Electrolysis bath, electric power set-up, the same as batch experiment.
Sediment condensed bath: Size: 300Ø × 950 height, treatment capacity: 0.3 L/min,
Surface area loading: 4.2 mm/min, storage time: 3.27 hours.

All the water quality parameters were measured according to the *Standard Methods for the Examination of Water* (JWWA, 1993) and the *Standard Methods for the Examination of
Water and Wastewater (APHA/AWWA/AEF, 1995). Furthermore, artificial wash water continuously flowed to the electrolysis bath and outflow liquid was withdrawn to the sediment condensed bath. The sludge was regularly picked out of the bottom of the condensed bath and suspended solids (SS) density was measured.

**Results and discussion**

**Batch experiment (Run 1)**
The electrolytic coagulation experiment was carried for 1,000 mg/L artificial wash with 1,000 mg/L kaolin and 400 mg/L in NTU. The density of condensed concentration sludge and filtrated turbidity are shown in Table 1, under the conditions of iron anode and aluminium anode comparatively.

In the case of iron anode coagulation, the peak of sludge density was observed when iron solution reached 20 mg/L. It should be noted that all the absolute value of density was more than 200 kg/m³ which is a high value. Moreover, the filtrate turbidity was over 10 NTU.

In the case of aluminium anode coagulation, sludge density decreased along with the increase of aluminium solution. Moreover, the sludge density was only 50% of sludge density by iron anode coagulation. Filtrate turbidity was about 1 NTU, which is much lower compared with iron coagulation.

On the whole, little change of pH is observed before or after electrolysis with iron or aluminium electrolytic coagulation.

**Coagulation effect for retentate from Yokohama Kosuzume membrane water treatment processes wash water (Run 2)**
The results of the same experiment are shown in Table 2, with membrane wash water from real river water. In the case of iron coagulation, sludge density was 80 kg/m³ and irrelevant with iron solution. Although filtrate turbidity decreased along with the increase of iron solution, it still remained as high as 170 NTU. In comparison with artificial wash water, it is a higher value. However, filtrate turbidity showed a similar tendency with real or artificial wash water in the case of aluminium coagulation. Besides, its SS density was below 20 kg/m³, and its filtrate density only reached 2 NTU.

| Ion products concentration mg/L Retentate Electrolytic concentration Turbidity NTU (24 hr) Sludge capacity mL (24 hr) | SS g | Dissolved material g | Sludge concentration kg/m³ |
|---|---|---|---|---|---|---|---|
| Fe, 10 400 | 6.29 | 11.8 | 0.005 | 255.4 |
| Fe, 20 400 | 6.10 | 11.5 | 2.9306 |
| Fe, 30 400 | 20.1 | 10.0 | 2.8862 |
| Fe, 40 400 | 25.8 | 10.5 | 0.0048 |
| Fe, 50 400 | 8.31 | 14.0 | 3.0471 |
| Fe, 60 400 | 5.44 | 15.0 | 0.0075 |
| Fe, 70 400 | 16.6 | 15.0 | 202.6 |
| Fe, 80 400 | 12.2 | 16.5 | 205.3 |
| Al, 10 400 | 2.76 | 21.5 | 0.0067 |
| Al, 20 400 | 2.43 | 21.0 | 144.1 |
| Al, 30 400 | 1.92 | 33.0 | 0.0095 |
| Al, 40 400 | 1.63 | 30.0 | 102.8 |
| Al, 50 400 | 3.40 | 43.0 | 3.0938 |
| Al, 60 400 | 1.49 | 41.5 | 0.0147 |
| Al, 70 400 | 0.82 | 57.5 | 77.3 |
| Al, 80 400 | 0.79 | 56.0 | 59.7 |
The electrolytic coagulation was carried out for the artificial wash water similar to real wash water. The same experiment was conducted for 250 NTU turbidity. The result shown in Table 3 was between the values of the above two experiments and showed a similar tendency.

From these experiments, it is observed that the condensation speed of coagulation particles was so quick that only after 1 hr condensation data was obtained, as shown in Table 4.

### Continuous experiment

Since thick condensed concentration sludge was obtained by iron coagulation in the batch experiment, the continuous experiment was conducted only for iron anode coagulation. The artificial wash water was made near 400 mg/L NTU everyday. With times being, turbid condensation in the retentate and the turbid of wash water verified.

When electrolytic iron condensed was done at 0.5 mg/L and operated continuously for 20 days, filtrate turbidity was as high as 30 to 80 mg/L because of low iron density and sludge density even reached 400 kg/m³.

### The characteristics of condensed suspended liquid

Condensed suspended liquid is different from the normal thin colloid suspended liquid. On
the basis of artificial kaolin suspended wash water or membrane filtration retentate, the fact that the lower quantity of metals, the higher sludge density is that suspended solids contain a lot of particles easy to sediment. The lower the coagulation of metal, the higher the density of the sludge becomes. The maximum density is obtained when metal quantity is zero. This is probably corresponded to metal oxides with low density deposited on sludge in terms of density formed by electrolysis. As a result, the density of filtrate separated from sedimentation decreases by electrolysis coagulation. In this experiment, low density is correlated with high metal dissolved; the lowest density appears in the case of the aluminium anode. The electrolysis operation is conducted under steady-state sludge concentration conditions, when filtrate is fed back to the membrane treatment process without affecting the main process treatment.

Electrolytic material
In comparison of the iron anode and aluminium anode, the density of condensed concentration sludge of iron is higher than aluminium, while the density of filtrate is lower with the
aluminum anode, this is because of smaller water oxide produced by aluminium. In the case of iron anode, not all iron becomes $3^+$ iron ($\text{Fe}^{3+}$) but remains as $2^+$ iron ($\text{Fe}^{2+}$), which contributes to incomplete condensation of iron.

The density of condensed concentration sludge
In the experiment of artificial wash water with iron anode, as high as 400 kg/m$^3$ condensed sludge density is due to high density of particles without coagulant in kaolin water. The same situation occurred in membrane filtration retentate of river raw water without using coagulant, as the density of electrolytic coagulation condensation sludge reached 80 kg/m$^3$, higher than normal density. It is easier for membrane filtration retentate to concentrate than coagulation condensed sludge.

The density of filtrate
As for the density of filtrate, the aluminium coagulation shows a better result, while the iron anode gives a better result in the case of concentration sludge density.

pH
Almost no change of pH is observed before or after electrolytic coagulation.

Conclusions
The following results are achieved in the electrolytic coagulation sediment concentration of artificial wash water and real wash water; the lower iron as well as the aluminium dissolved, the higher the sludge density becomes; especially aluminium has this tendency. It is possible to achieve 400 kg/m$^3$ of condensed sludge density with artificial wash water and more than 80 kg/m$^3$ for real retentate, in terms of electrolytic coagulation of iron anode. The sludge density of aluminium anode is higher, but its filtrate density is lower than iron anode. Little change of pH is observed before or after electrolysis with the method of electrolytic coagulation.

References


