Removing fluoride from hot spring wastewater by an electrolysis system with a perforated plate as a diaphragm

Yuki Imai and Tomonori Kawakami

Abstract: A continuous electrolysis system using an electrolysis reactor cell separated into an anode cell and a cathode cell by a perforated plate as a diaphragm was operated in a laboratory to remove fluoride (F\(^-\)) from hot spring wastewater to meet the Japanese national minimum effluent standard for F\(^-\) of 8 mg/L. The perforated plate was 5 mm thick and had 740 holes with a diameter of 1.2 mm. F\(^-\) was removed by co-precipitation with magnesium hydroxide (Mg(OH)\(_2\)) produced in the cathode cell. Water from Gero Hot Spring containing 17.0 mg/L of F\(^-\) was used as a model wastewater. Since Gero Hot Spring water does not contain magnesium (Mg\(^{2+}\)), magnesium carbonate (MgCO\(_3\)) was added as a form of slurry into the anode cell with the hot spring water. The hot spring water containing Mg\(^{2+}\) passed through the perforated plate and flowed into the cathode cell, where Mg(OH)\(_2\) was co-precipitated with F\(^-\) due to hydroxide ion (OH\(^-\)) produced by electrolysis. Alkalinity (Alk), which interferes with the precipitation of Mg(OH)\(_2\) by forming MgCO\(_3\) in the cathode cell, was removed in the anode cell by hydrogen ion (H\(^+\)) produced by electrolysis. Various combinations of the added Mg\(^{2+}\) concentration (Mgi) and the applied current (Ia) were tested to elucidate the effect of these parameters on F\(^-\) removal (\(\Delta F\)). It was revealed that, less than 8 mg/L of the F\(^-\) concentration in treated water (F\((\text{treated})\)) was achieved when the Mgi was between 85 and 110 mg/L and Ia was more than 90 mA.

Subjects: Industrial Chemistry; Separation Processing; Water Engineering

ABOUT THE AUTHOR
Yuki Imai is a student of the Graduate School of Engineering at Toyama Prefectural University, Japan. The focus of her research is to develop a technology to remove fluoride and arsenic from wastewater to levels below the minimum effluent standard. She has experimented with removing fluoride and arsenic by co-precipitation with magnesium hydroxide. Magnesium hydroxide is produced in a cathode cell of an electrolysis system. Her novel idea is to apply a perforated plate as a diaphragm that separates the cathode cell from the anode cell. The advantages of the perforated plate are described in the manuscript.

PUBLIC INTEREST STATEMENT
Many hot springs in Japan contain high concentrations of fluoride. In consideration of impact on the environment, the Japanese Ministry of the Environment has set the minimum effluent standards for fluoride and arsenic in wastewater at 8 mg/L and 100 µg/L, respectively. Thus far, however, no suitable treatment technologies are available from the viewpoint of removal efficiency and cost. This paper presents clues on the development of a novel treatment system to remove fluoride and arsenic from wastewater from hot spring facilities. It is possible to apply this technique not only to hot spring wastewater but to industrial wastewater containing fluoride and arsenic.
Keywords: Fluoride; electrolysis; magnesium; perforated plate; co-precipitation

1. Introduction

Hot springs are popular Japanese tourist attractions, and numerous resort towns are centered on hot springs. Dissolved matter in hot springs is believed to provide health benefits when people soak in a hot spring. However, hot springs sometimes contain materials not good for the environment, such as fluoride (F\textsuperscript{−}). In 2001, the Japanese Ministry of Environment set the national minimum effluent standard for fluoride at 8 mg/L. (Ministry of the Environment Government of Japan, 2018) However, a provisional standard was applied to hotels with hot springs since no appropriate treatment techniques were available. (Ministry of the Environment Government of Japan, 2016) The provisional standard has a period of three years; however, even by 2016, no effective technology had been found even though the Japanese Ministry of Environment had invited private companies to compete to develop treatment techniques for wastewater. Accordingly, the provisional standard of 15-50 mg/L is still applied to hotels with hot springs. (Ministry of the Environment Government of Japan, 2018, 2016)

To remove F\textsuperscript{−} for industrial wastewater treatment, calcium ions are often added to produce insoluble precipitation of calcium fluoride; however, obtaining 8 mg/L of F\textsuperscript{−} requires the addition of much more calcium than that calculated from stoichiometry. (Saha, 1993) A high amount of the source of calcium as well as the produced sludge causes cost rising, which cannot be afforded by the hot water spring industries. In addition to chemical precipitation, adsorption, ion exchange, and reverse osmosis (RO), electrodialysis and electrocoagulation techniques can be used for removing F\textsuperscript{−} from water. (Maurice, 2006) However, co-existing contaminants in hot springs degrade the effectiveness of adsorption and ion exchange. (Amit, Kumar, & Sillanpää, 2011; Paripurnanda, Vigneswaran, Kandasamy, & Naidu, 2013) Bone char is a well-known adsorbent that selectively adsorbs F\textsuperscript{−}; however, its low adsorption capacity (5 mg/g) means that a large amount of bone char is needed. (Fawell et al., 2006; Herath, Sunali, Tomonori, & Masamoto, 2018; Medellin-Castillo et al., 2014; Rojas-Mayorga et al., 2013) When we consider treatment of Gero Hot Spring, one of the most famous hot springs in Japan and the focus of this paper, the volume of hot spring water is 3300 m\textsuperscript{3}/day with a F\textsuperscript{−} concentration of 16.5 mg/L. (xxxx, 0000) This means that, 5600 kg/day of bone char is required to achieve the F\textsuperscript{−} concentration of 8 mg/L, and it is not realistic to prepare this amount of bone char every day. Kato et al. proposed to use a waste gypsum apatite to remove F\textsuperscript{−} and showed 9.1 mg/g of adsorption capacity, (Katoh, Hada, Kitagawa, Terao, & Sato, 2014) which would require 3100 kg/day of waste gypsum apatite. This is not realistic because it requires too much apatite and produces too much sludge.

RO and electrodialysis are available for fluoride removal of drinking water. However, in wastewater treatment, the rejected water from RO and electrodialysis containing high concentrations of fluoride should be treated by an additional method, since the entire volume of water should be treated. (Sachin et al., 2015) In the electrocoagulation system, sacrificial electrodes, usually aluminum plates, are used for anodes. The aluminum eluted by electrolysis forms a complex with F\textsuperscript{−} and precipitates. (Mameri et al., 1998; Guzmán, Nava, Coreño, Rodriguez, & Gutiérrez, 2016) The cost of the sacrificial electrodes and of the treatment of the sludge are not affordable.

The technology to remove F\textsuperscript{−} from a solution by an electrolysis system has been developed by the authors. (Tomonori et al., 2018; Yuki, Konishi, & Kawakami, 2019; Yuki, Yanagawa, Konishi, & Kawakami, 2017) The electrolysis system is a method in which a solution containing magnesium (Mg\textsuperscript{2+}) is electrolyzed in an electrolysis cell separated by a diaphragm into an anode cell and a cathode cell. When the electrolysis is performed, the following reactions take place in the cathode (Equation (1)) and in the anode (Equation (2)) to produce hydroxide ion (OH\textsuperscript{−}) and hydrogen ion (H\textsuperscript{+}), respectively.

\begin{align*}
2\text{H}_2\text{O} + 2\text{e}^- &\rightarrow 2\text{OH}^- + \text{H}_2 & (1) \\
2\text{H}_2\text{O} &\rightarrow 4\text{H}^+ + \text{O}_2 + 4\text{e}^- & (2)
\end{align*}
As the pH increases in the cathode cell due to OH\(^-\) produced by Equation (1) when the electrolysis is performed, Mg\(^{2+}\) contained in the solution precipitates as magnesium hydroxide (Mg(OH)\(_2\)); at that time, F\(^-\) is co-precipitated with Mg(OH)\(_2\) to be removed. (Tomonori et al., 2018)

In the electrolysis system, alkalinity (carbonate and bicarbonate) must be removed in advance, since it suppresses the production of Mg (OH)\(_2\) by forming magnesium carbonate (MgCO\(_3\)). (Yuki et al., 2017) Well water (drinking water) containing Mg\(^{2+}\) and F\(^-\) was treated by a batch-type electrolysis system, using a membrane filter for the diaphragm, and it was shown that, the electrolysis system effectively removed F\(^-\). (Tomonori et al., 2018) When the treatment system is applied to drinking water, it is enough to treat part of the water volume; on the contrary, when it comes to hot spring wastewater, the entire volume should be treated. Therefore, electrolysis treatment by a continuous flow reactor with a diaphragm made of an unglazed plate, which is more durable and cheaper than a membrane filter, was performed. (Yuki et al., 2019) It was confirmed that, the system achieved a concentration of less than 8 mg/L of F\(^-\). However, in a long-term operation, plugging of the unglazed plate caused a gradual increase in power consumption due to increased electrical resistance, requiring increased voltage. Therefore, in this study, as a novel system for removing F\(^-\), an electrolysis system using a perforated plate as a diaphragm was tested, and its performance was evaluated.

2. Material and methods

2.1. The principle of the perforated plate

The perforated plate used for a diaphragm of the electrolysis system in the current study was a plastic plate 5 mm thick having holes with a diameter of 1.2 mmφ with 3 mm interstices. Figure 1(a,b) show a schematic diagram and a photo of the perforated plate, respectively. Small holes with a diameter of 1.2 mmφ suppress eddy diffusion, which bears most of the mass transfer in water. Since the molecule diffusion transports much less mass transfer in water, (Marion, 2008) the solutions separated by the perforated plate do not mix easily. On the other hand, this perforated plate can be used as a diaphragm, since electrons and ions can pass through holes when there is a potential gradient.

2.2. Outline of experimental equipment

The water of Gero Hot Spring in Japan’s Gifu Prefecture was used as a model wastewater. Table 1 shows the water quality of Gero Hot Spring from literature (Gifu Research Center for Public Health, 2013) and as measured. The literature value and the measured value coincided well with each other; the F\(^-\) concentrations were 16.5 mg/L and 17.0 mg/L, respectively. Figure 2(a,b) show an overview of the electrolysis reactor when a perforated plate was used for the diaphragm. Figure 2(a) is a schematic diagram of the electrolytic cell, which had dimensions of 134 mm (W) * 74 mm (D) * 87 mm (H) for each cell. The perforated plate had dimensions of 134 mm (W) * 87 mm (H) and had 740 holes. Figure 2(b) is a top view of the electrolysis cells. The raw water (Gero Hot Spring water) was allowed to flow into the anode cell at 7 mL/min, which was equivalent to 10 L/day. The retention time
in each cell was 100 minutes. Since Gero’s hot spring water does not contain Mg$^{2+}$, as is shown in Table 1, MgCO$_3$ was added to the anode cell as a form of slurry with a concentration of 50 g/L. The magnesium concentration to be added was adjusted by changing the flow rate of the MgCO$_3$ slurry. For example, to obtain a magnesium concentration of 100 mg/L, the flow rate of the MgCO$_3$ slurry was set to 0.056 mL/min.

The electrode used a platinum-coated titanium plate for the anode and a stainless steel wire for the cathode. In the anode cell, H$^+$ produced according to Equation (2) reacts with MgCO$_3$ to produce carbon dioxide (CO$_2$). Consequently, alkalinity is removed, and Mg$^{2+}$ dissolves into the solution (Equation (3)).

\[
\text{MgCO}_3 + 2\text{H}^+ \rightarrow \text{Mg}^{2+} + \text{CO}_2 + \text{H}_2\text{O}
\]  

Aeration was performed in the anode cell to remove CO$_2$ from the solution. The raw water from which alkalinity was removed in the anode cell passes through the holes in the perforated plate to the cathode cell. In the cathode cell, the pH rises by the OH$^-$ produced by Equation (1) to precipitate Mg(OH)$_2$, which co-precipitates F$^-$. 

The electrolysis was performed under a constant current. A constant current power supply (TAKASAGO 2X-S-400M) was used for a DC power supply.

**2.3. Experimental conditions**

In this study, the effect of the Mg$^{2+}$ concentration and the applied current on F$^-$ removal was investigated. An increase in the concentration of Mg$^{2+}$ could increase the amount of Mg(OH)$_2$ precipitation in the cathode, leading to increased co-precipitation with F$^-$. An increase in the applied current could increase the amount of Mg(OH)$_2$ precipitation in the cathode cell when adequate Mg$^{2+}$ is available. In addition, it could reduce the excessive alkalinity in the cathode
cell, according to Equation (3). On the contrary, too much current could cause excessive H\(^+\), which suppress the increase of pH in the cathode cell after all alkalinity is consumed in the anode cell. Therefore, the combination of Mg\(^{2+}\) and the applied current (I\(_a\)) is an important factor of the system. When the Mg\(^{2+}\) concentration was 50 mg/L, I\(_a\) varied as 50, 80, and 100 mA. When the Mg\(^{2+}\) concentration was 100 mg/L, I\(_a\) varied as 80, 100, and 120 mA. When the Mg\(^{2+}\) concentration was 150 mg/L, I\(_a\) varied as 80, 100, 120, and 150 mA. A total of 10 experiments were conducted. In the current electrolysis system, I\(_a\) of 50, 80, 100, 120, and 150 mA were equivalent to charge loadings of 430, 690, 860, 1040, and 1300 Coulomb/L, respectively.

2.4. Analyses
When the system was stabilized after more than 20 hours of operation, the water in the anode cell and the treated water were sampled. These samples were filtered for analysis through a membrane filter with a pore size of 0.45 µm (Cobetter Lab, Unichro Syringe Filter, Type PES).

The ion concentrations of F\(^-\), chloride ion (Cl\(^-\)), nitrate ion (NO\(_3^-\)), sulfide ion (SO\(_4^{2-}\)), sodium ion (Na\(^+\)), potassium ion (K\(^+\)), ammonium ion (NH\(_4^+\)), Mg\(^{2+}\), and calcium ion (Ca\(^{2+}\)) were analyzed using an ion chromatograph (Cation: Thermo ICS1500, Separation column IonPac CS12A, Eluent Methanesulfonic acid 30 mmol/L, Suppressor CERS 500), (Anion: Thermo ICS2000, Separation column IonPac AS18, Eluent KOH 23–40 mmol/L (gradient), Suppressor AERS 500).

The pH was measured with a BECKMAN ∅32 pH Meter (electrode: BECKMAN 511,070). The alkalinity was determined by taking the difference between the total cation charge and total anion charge obtained by the ion chromatograph and the pH measurement.

2.5. Model calculation
An empirical model was established to estimate the F\(^-\) concentration in the treated water F (treated) using the experimental results with various combinations of the initial Mg\(^{2+}\) concentration (Mgi) and I\(_a\) in the operation of the electrolysis system. As mentioned previously, F\(^-\) removal (\(\Delta F\)) is affected by the alkalinity in the anode cell (Alk) and the concentration of Mg precipitated (\(\Delta Mg\)), since F\(^-\) is removed by co-precipitation with Mg(OH)\(_2\). Therefore, Alk and \(\Delta Mg\) were the important parameters for estimating \(\Delta F\).

The empirical model established first predicts Alk and \(\Delta Mg\) from I\(_a\) and Mgi, and then it predicts \(\Delta F\) from the predicted Alk and \(\Delta Mg\).

Alk was estimated by a polynomial equation, as shown in Equation (4), where a and b are the model constants. The values of a and b were varied to obtain the best fit of the estimated value to the measured value. After obtaining the model constants of a and b, a correction was made to Alk when Alk was 0 or less by Equation (4). In such a case, Alk was regarded as 0, as shown in Equation (5), since even when Alk had a negative value, the carbonate in the solution that affects the precipitation of Mg(OH)\(_2\) was regarded as 0.

\[
\text{Alk} = a \times \text{Mgi} + b \times \text{Ia} \quad (4)
\]
\[
\text{Alk} = 0 \quad (\text{Alk} \leq 0), \quad (5)
\]

where Mgi is the initial concentration of Mg (mg/L),

I\(_a\) is the applied current (mA), and

Alk is the alkalinity in the anode cell (µeq/L).

The Mg concentration in the treated water (Mg (treated)) was estimated by a polynomial equation, as shown in Equation (6). In the equation, a’ and b’ are the model constants. The values
of a’ and b’ were varied to obtain best fit of the estimated value to the measured value. $\Delta M_g$ was calculated by Equation (7). Since it is reported that $F^-$ is not co-precipitated with Mg(OH)$_2$ when the concentration of magnesium ions is less than 20 mg/L, (Yuki & Kawakami, 2019) $\Delta M_g$ should not be taken into account when the Mg (treated) is less than 20 mg/L. Equation (8) shows the correction when the Mg (treated) was less than 20 mg/L.

$$M_g(treated) = a'M_g + b'I_a$$
(6)

$$\Delta M_g = M_g - M_g(treated)$$
(7)

$$\Delta M_g = M_g - 20 \quad (M_g(treated) < 20 \text{ mg/L})$$
(8)

where $\Delta M_g$ is the concentration of Mg precipitated (mg/L), and Mg (treated) is the Mg concentration in the treated water (mg/L).

As the next step, $\Delta F$ was estimated by Equation (9), where $a''$ and $b''$ are the model constants. The values of $a''$ and $b''$ were varied to obtain best fit of the estimated value to the measured value. $F$ (treated) was calculated by Equation (10).

$$\Delta F = a''Alk + b''\Delta M_g$$
(9)

$$F_{(treated)} = F_{(raw \text{ water})} - \Delta F$$
(10)

where $F_{(treated)}$ is the $F^-$ concentration in the treated water (mg/L), and $F_{(raw \text{ water})}$ is the $F^-$ concentration in the raw water, here 17.0 mg/L.

3. Results and discussion

3.1. Electrolysis system with a perforated plate as a diaphragm

Figures 3 and 4 show the relationship between $I_a$ and $F$ (treated) and the relationship between $I_a$ and Alk, respectively, in different Mgi. When compared at the same Mgi, smaller $I_a$, higher $F$ (treated), and higher Alk were observed. With the small $I_a$ in the high Mgi, Alk was not sufficiently removed, leading to less $\Delta F$. The relationship between Alk and $F$ (treated) is shown in Figure 5. It clearly shows that when $I_a$ was small, the alkalinity remaining in the anode cell hindered $\Delta F$ by producing MgCO$_3$ in the cathode cell, which interfered with the precipitation of Mg(OH)$_2$. However, when comparing $F$ (treated) in the different Mgis, lower $F$ (treated) can be seen when Mgi was higher at the same Alk. For example, according to Figure 5, $F$ (treated) were 3.8 mg/L, 7.7 mg/L, and 13.3 mg/L for 150 mg/L, 100 mg/L, and 50 mg/L of Mgi, respectively, at Alk of 1500 µeq/L. According to Figure 4, $I_a$s that gave Alk of 1500 µeq/L were 50 mA, 90 mA, and 150 mA for Mgis of 50 mg/L, 100 mg/L, and 150 mg/L, respectively. Figure 6...
shows the relationship between $I_a$ and $\Delta \text{Mg}$. When $I_a$s that gave Alk of 1500 $\mu$eq/L are applied to Figure 6, $\Delta \text{Mgs}$ of 36 mg/L, 90 mg/L, and 148 mg/L were obtained for Mgis of 50 mg/L, 100 mg/L, and 150 mg/L, respectively. Therefore, higher $\Delta F$ was achieved by the higher $\Delta \text{Mg}$ when Mgi was higher at the same Alk.

### 3.2. Model calculation

From Mgi and $I_a$, Alk was estimated by a regression analysis as Equation (11), based on Equation (4).

$$\text{Alk} = 52.6 \times \text{Mgi} - 39.0 \times I_a$$  \hfill (11)
The positive coefficient for Mgi is caused by the alkalinity from the added MgCO$_3$. The negative coefficient for Ia is caused by the reduction of alkalinity by H$^+$ produced by Equation (2).

Figure 7 shows a good relationship between the measured Alk with the estimated Alk in the anode cell by Equations (11) and (5).

From Mgi and Ia, Mg (treated) was estimated by a regression analysis, as shown in Equation (12), based on Equation (6).

$$\text{Mg (treated)} = 0.496 \times \text{Mgi} - 0.385 \times \text{Ia}$$  \hspace{1cm} (12)

The positive coefficient for Mgi indicates that more Mg$^{2+}$ remained in the cathode solution when more MgCO$_3$ was added to the anode cell, and the negative coefficient for Ia indicates that more current produced more precipitation of Mg.

Figure 8 shows a comparison of the measured $\Delta$Mg and estimated $\Delta$Mg by Equations (12), (7), and (8). The estimated $\Delta$Mg had a good correlation with the measured $\Delta$Mg.

It was revealed that Alk and $\Delta$Mg could be estimated from Mgi and Ia. As a next step for estimating $\Delta$F, a regression analysis was done by using the estimated values of Alk by Equations (11) and (5) and $\Delta$Mg by Equations (12), (7), and (8). From the estimated Alk and $\Delta$Mg, $\Delta$F$^-$ was estimated by a regression analysis, as shown in Equation (13), based on Equation (9).

$$\Delta F = -0.0018 \times \text{Alk} + 0.144 \times \Delta \text{Mg}$$  \hspace{1cm} (13)

The negative coefficient for Alk reflects alkalinity’s interference with F$^-$ removal, and the positive coefficient for $\Delta$Mg reflects F$^-$ removal by co-precipitation with Mg(OH)$_2$.

Figure 9 shows the measured F (treated) and estimated F (treated) according to Equations (13) and (10). F (treated) was successfully estimated by the model.
By applying this model, the combinations of Mgi and Ia were calculated so that, the effluent standard of 8 mg/L of F (treated) was achieved. The results are shown in Figure 10. When the Ia was more than the combination in Figure 10, less than 8 mg/L of F (treated) would be achieved. However, when the Mgi was less than 83 mg/L, F (treated) less than 8 mg/L was not achieved, even when a high current was applied. This is because the ΔMg was not sufficient to decrease F$^{-}$ by coprecipitation. According to Figure 10, more than 110 mg/L of Mgi caused an increase in the current, indicating that, the best combination of Mgi and Ia rises between 83 mg/L and 110 mg/L of Mgi. Appropriate Ia should be chosen by a cost comparison of MgCO$_3$ and electricity.

The production of sludge can be estimated by assuming 110 mg/L as Mgi and 9 mg/L as ΔMg to meet the minimum standard for F$^{-}$ of 8 mg/L, since the hot spring water of Gero contained 17.0 mg/L of F$^{-}$. According to the model, 19 mg/L of magnesium remained in the treated water, indicating 91 mg/L of magnesium, which is equivalent to 220 mg/L of Mg(OH)$_2$ precipitates. When 3300 m$^3$/day is taken into consideration as the volume of the wastewater, the amount of sludge would be 730 kg/day, which is much less than that produced by adsorption systems.

4. Conclusions

A continuous electrolysis system using an electrolysis reactor cell separated into an anode cell and a cathode cell by a perforated plate as a diaphragm was operated to remove F$^{-}$ from hot spring wastewater to meet the national minimum effluent standard for fluoride, set at 8 mg/L. Water from Gero Hot Spring, which contained 17.0 mg/L of F$^{-}$ concentration, was used as a model wastewater. Since water from Gero Hot Spring does not contain magnesium, MgCO$_3$ was added to the hot spring water as a Mg$^{2+}$ source.

The electrolysis system was operated with various combinations of Mgi and Ia to elucidate the effect of these parameters on ΔF. ΔF was affected negatively by Alk in the anode cell and positively by the precipitated Mg$^{2+}$ (ΔMg) in the cathode cell. Alk interfered with the precipitation of Mg(OH)$_2$.
when Ia was not enough to remove Alk. Based on the experimental results, F (treated) was estimated from MgI and Ia by an empirical model. The model could well reproduce measured F (treated). The model indicated that less than 8 mg/L of F (treated), which is the Japanese national minimum standard for wastewater, would be achieved when the MgI was between 85–110 mg/L, and more than 90 mA of Ia was applied.

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