



Continuous electrolytic decarbonation and recovery of a carbonate salt solution from a metal-contaminated carbonate solution

Kwang-Wook Kim*, Yeon-Hwa Kim, Se-Yoon Lee, Eil-Hee Lee, Kyusuk Song, Kee-Chan Song

Korea Atomic Energy Research Institute, 150 Deokjin, 1045 Daedeok-daero, Yuseong, Daejeon Republic of Korea

ARTICLE INFO

Article history:

Received 4 February 2009

Received in revised form 4 May 2009

Accepted 10 June 2009

Available online 18 June 2009

Keywords:

Continuous

Electrolytic

Decarbonation

Recovery

Stacked-cell

Carbon dioxide

ABSTRACT

This work studied the characteristic changes of a continuous electrolytic decarbonation and recovery of a carbonate salt solution from a metal-contaminated carbonate solution with changes of operational variables in an electrolytic system which consisted of a cell-stacked electrolyzer equipped with a cation exchange membrane and a gas absorber. The system could completely recover the carbonate salt solution from a uranyl carbonate complex solution in a continuous operation. The cathodic feed rate could control the carbonate concentration of the recovered solution and it affected the most transient pH drop phenomenon of a well type within the gas absorber before a steady state was reached, which caused the possibility of a CO₂ gas slip from the gas absorber. The pH drop problem could be overcome by temporarily increasing the OH⁻ concentration of the cathodic solution flowing down within the gas absorber only during the time required for a steady state to be obtained in the case without the addition of outside NaOH. An overshooting peak of the carbonate concentration in the recovered solution before a steady state was observed, which was ascribed to the decarbonation of the initial solution filled within the stacked cells by a redundant current leftover from the complete decarbonation of the feeding carbonate solution.

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1. Introduction

A great deal of interest has recently been shown on a uranium-leaching from spent nuclear fuel in a high alkaline carbonate media instead of an acid media to enhance its safety, economic competitiveness, and to minimize the generation of secondary waste streams [1–5]. When a carbonate solution is used to treat nuclear spent fuel or uranium ore, a large amount of carbonate salt such as M₂CO₃ (M = Li⁺, K⁺, Na⁺, NH₄⁺) is used. Therefore, if the carbonate salt is not recycled in the process using carbonate solutions, an enormous amount of carbonate salt would be accumulated, which weakens the advantages and the environmental friendliness of the process using a carbonate solution. The carbonate solutions used in those systems may contain metal-carbonate complex ions or be mixed with several impurity metal ions.

Our previous works studied the batch operation characteristics of an electrolytic system consisting of a cation exchange membrane-equipped cell and a gas absorber to recover the carbonate salt solution for recycle from a uranyl peroxo carbonate complex solution [6]. In the system, the uranium-complexed carbonate solution was acidified in the anodic chamber by H⁺ ions generated through

the water split reaction to release CO₂ gas from the solution, while the uranium ions were left in the anodic chamber. The released CO₂ gas was recovered as a carbonate salt solution again by the OH⁻ ions generated through the water split reaction in the cathodic chamber and by the migration of the cations composed of the carbonate salt, which remained after the decarbonation in the anodic chamber. In our previous work, the decarbonation was elucidated to be proportional to the total electric charge supplied to the system, and the constant current operation was found to be better than the constant voltage operation from an energy consumption point of view.

In order to apply an electrolytic recycling system into actual cases [1,2,5,6], the continuous operation characteristics of the system should be studied, but experiments for that have not been carried out yet. Accordingly, in this work, a once-through decarbonation and a recovery of carbonate salt solution from a uranyl carbonate complex solution by using a system which consisted of a multi-cell-stacked electrolytic system equipped with cation membrane and a gas absorber were studied with several variables of the applied current density, anodic and cathodic flow rates, and so on.

2. Experimental

A schematic diagram of the electrolytic system used in this work for the electrolytic decarbonation and recycling of a carbonate salt from a uranium-containing carbonate solution is shown in Fig. 1. The electrolytic system consisted of a three cell-stacked

* Corresponding author at: Korea Atomic Energy Research Institute, 1045 Daedeok-daero, Yuseong, Daejeon, Republic of Korea. Tel.: +82 42 868 2044; fax: +82 42 868 2351.

E-mail address: nkwkim@kaeri.re.kr (K.-W. Kim).

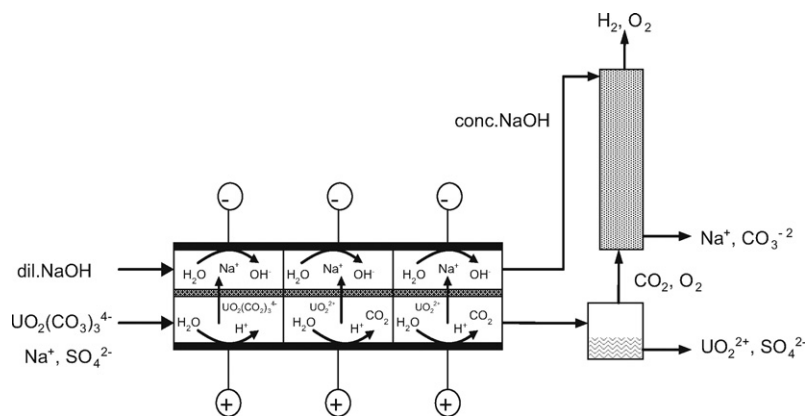


Fig. 1. Schematic diagram of an apparatus used for electrolytic decarbonation and recycling of a sodium carbonate salt from uranyl peroxo carbonate complex solution.

electrolyzer and a CO₂ gas absorption column for a gas–liquid contact. In the electrolytic system, the anolyte and catholyte flowed independently in series through the respective anodic and cathodic chambers of the stack. The volumes of the anodic and cathodic chambers of each cell were about 22 ml. A line for discharging gas from a gas–liquid separator, where the final anolyte coming out of the electrolyzer went into, was connected to the bottom end of the CO₂ gas absorber mounted outside the electrolyzer, and the final catholyte coming out of the electrolyzer was introduced to the top end of the CO₂ gas absorber. Each cell of the cell-stacked electrolyzer had an anode of IrO₂ and a cathode of Ti and was divided by a cation exchange membrane (Nafion 242). The size of each electrode was W 2 cm × H 4 cm × T 0.1 cm of a Madras type with a mesh size of 6 mm × 12 mm. The gas absorber was prepared by packing a Pyrex glass tube of 40 cm in length and 2.5 cm in diameter with silica beads of 1 mm in diameter. The void fraction of the packed column was about 40%. In our preliminary experiments, the column was confirmed to be able to completely absorb the CO₂ gas released from the anodic chamber.

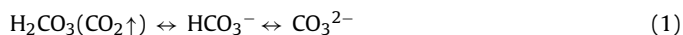
For the experiments of an electrolytic decarbonation and recovery of the carbonate salt solution, a 0.1 M NaOH solution and a 0.5 M Na₂CO₃ solution (or a uranyl tri-carbonato complex ion (UO₂(CO₃)₃⁴⁻) solution containing 1.0 g/l uranium in 0.5 M Na₂CO₃) mixed with 0.1 M Na₂SO₄ were used as the catholyte and anolyte solutions to be fed into the electrolyzer, respectively. The Na₂SO₄ in the anolyte was used as a supporting electrolyte to prevent an operational cut-off caused by a rapid increase of the cell voltage, which occurs because the conductivity of an anolyte, bearing only a carbonate salt, decreases rapidly as its electrolytic decarbonation is being progressed [6]. The uranyl carbonato complex ion solution was prepared by dissolving UO₂ powder in a 0.5 M Na₂CO₃ solution for a sufficient dissolution time. The pH values of the recovered carbonate and anolyte solutions leaving the gas absorber and the gas–liquid separator mounted outside the final anodic chamber of the electrolyzer were recorded on-line by pH-meters and a data-logger. The solutions in the anodic and cathodic chambers were sampled at regular intervals for their analyses.

The carbonate concentration was analyzed by an inorganic carbon analyzer (Shimadzu TOC-V CSH/TNM-1). The concentrations of the sulfate, sodium, and hydroxyl ions were analyzed by using an auto-titrator and an ion chromatography (Dionex ICS 90). The concentration of U was analyzed by a colorimetric method using Arsenazo III. All the chemicals used in this work were reagent grade, and they were dissolved, as received, in demineralized water of 18.2 MΩ prepared by a double distillation and an ion-exchanger (Milli-Q plus). All the experiments were carried out at room temperature.

3. Results and discussion

3.1. Background for electrolytic decarbonation of a uranyl carbonate solution and recovering a carbonate salt solution

This work is for an investigation of the characteristic changes during a continuous electrolytic decarbonation and recovery of a carbonate salt solution from a used carbonate solution containing metal ions such as uranyl carbonato complex solution. Therefore, it is necessary to understand the reactions relevant to the carbon species and the electrochemical reactions occurring in the electrolytic system, which are described in detail elsewhere [5–7]. Carbonate species are interchangeable among the carbonic acid of H₂CO₃, bicarbonate of HCO₃⁻, and carbonate of CO₃²⁻ species depending on the solution pH as shown in Eq. (1). Only carbonate ions exist in the solution at pH of more than 12, and only carbonic acid (CO₂ gas is released from the solution) does at pH of less than 4. In the range between pH 4 and 8, the carbonate ions, bicarbonate ions, and carbonic acid coexist in different mole fractions of their species, depending on the solution pH [6]. Uranyl ion of UO₂²⁺ is known to form a uranyl tri-carbonato complex ion of UO₂(CO₃)₃⁴⁻ in carbonate solution as shown in Eq. (2).



From the above equations, the uranyl ions embedded in the carbonato complex ions can be considered to be separated from the carbonate species by controlling the solution pH. Fig. 2 shows the changes of the uranium concentrations in the solutions according to the pH-adjustments of the uranium solutions which were prepared by dissolving UO₂ powder in a carbonate solution of 0.5 M Na₂CO₃. An inset box of Fig. 2 shows the absorbance spectra of the prepared uranyl carbonato complex ion solution at pH 11 and the uranium solution at pH 2 after by pH-controlling the uranyl carbonato complex solution by using HNO₃. The spectra of the solution at pH 11 and 2 reveal the typical characteristic absorbance peaks of a UO₂(CO₃)₃⁴⁻ solution at 435, 448, and 462 nm, and those of a UO₂²⁺ solution at 404, 414, and 426 nm, respectively [1,6]. When the prepared uranyl carbonato complex solution was acidified with HNO₃, the uranium concentration in the solution did not change from pH 11 to 1 except for around pH 4 where a slight precipitation was observed. The solubilities of the uranyl ions in the solution with bicarbonate or carbonate ions at pH of more than 5, and in the solution free of carbonate ions at pH of less than 3 were higher than the uranium concentration of the initial carbonate solution, so that there is no change of the uranium concentration in those pH ranges. However, the solubility of uranyl ions in the solution at

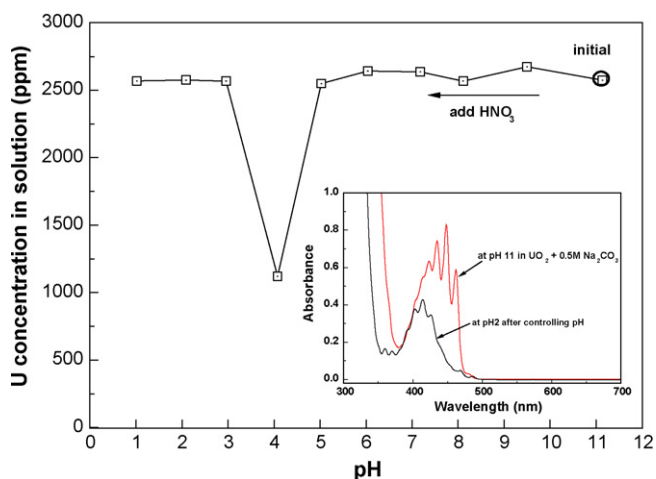
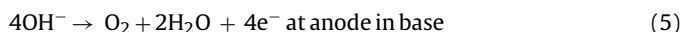
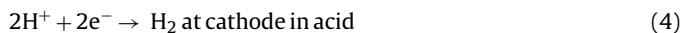
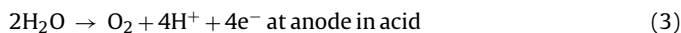


Fig. 2. Changes of the uranium concentrations in a uranyl carbonate complex solution by its pH-adjustment and absorbance spectra of the solution at pH 2 and 11.

pH 4 becomes lower showing a precipitation of uranium because of the hydrolysis of the uranyl ions. These results mean that the uranyl carbonate complex solution can be separated easily into uranyl ions and CO_2 gas by a sufficient acidification of its solution. In this work, a continuous acidification of a uranyl carbonate complex solution and a recovery of the released CO_2 as a carbonate salt solution were carried out by using an electrolytic system without adding any chemicals into the solution system.

The water splitting reactions at the anode and cathode in a divided cell to be used for the electrolytic decarbonation of uranyl carbonate complex solutions and recovering a carbonate salt are known to change, depending on the solution pH. The evolutions of oxygen and hydrogen are expressed by Eqs. (3) and (4) in an acidic condition and by Eqs. (5) and (6) in a basic condition [6,8]. The water splitting reactions imply that the anodic solution and cathodic solution can be acidified and alkalinized in a cell.



Accordingly, when a sodium carbonate solution with uranyl carbonate complex ions is put into an anodic chamber, proton ions generated in the anodic chamber through the water splitting reaction of Eqs. (3) or (5) convert the carbonate species embedded in the uranyl carbonate complex ions to CO_2 according to Eq. (1). As the CO_2 gas is released out of the solution, the remaining cations of Na^+ in the anodic chamber should be migrated into the cathodic chamber through the cation exchange membrane to meet the electric neutrality of the anodic solution, as depicted in Fig. 1. On the other hand, a NaOH solution is generated in the cathodic chamber with the migrated Na^+ ions and with the OH^- ions generated through the water splitting reaction of Eqs. (4) or (6). When the CO_2 gas contacts the cathodic solution with NaOH within the gas absorber, it is converted to a carbonate salt solution again through Eq. (1). Finally a uranyl ion solution free of carbonate and a recovered carbonate salt solution leave from anodic and cathodic chambers of the electrolytic system, respectively.

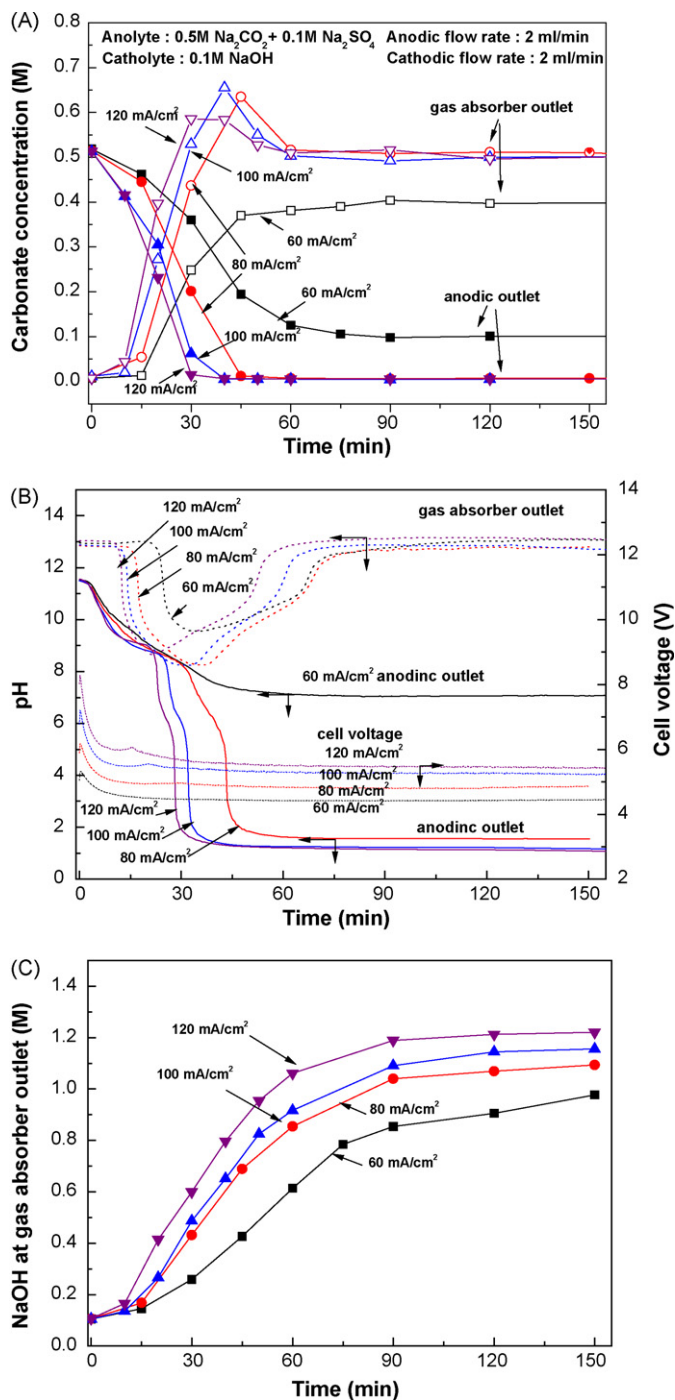


Fig. 3. Changes of carbonate concentrations (A), and pHs at anodic outlet and gas absorber outlet, and cell voltages (B), and NaOH concentrations at gas absorber outlet (C) with electrolysis time at several current densities.

3.2. Characteristics of decarbonation of a uranyl carbonate solution and recovering a carbonate salt in an electrolytic continuous system

Fig. 3 shows the changes of the carbonate concentrations (A), and the pHs at the anodic outlet and the gas absorber outlet, and the cell voltages (B), and NaOH concentrations at gas absorber outlet (C) during the electrolysis time at several current densities of 60, 80, 100, 120 mA/cm^2 in the system of Fig. 1. The feeds to the system were a uranyl carbonate complex solution of 1000 ppm uranium in 0.5 M Na_2CO_3 mixed with 0.1 M Na_2SO_4 as an anolyte, and a

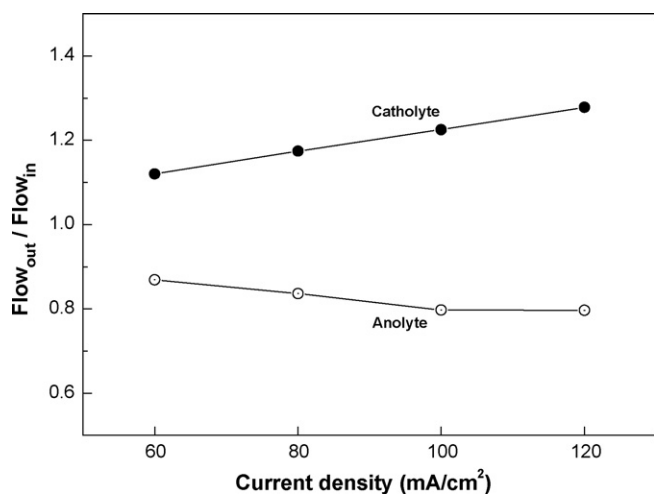


Fig. 4. Flow ratios at inlet and outlet of in anodic and cathodic solutions with applied current in Fig. 3.

0.1 M NaOH as a catholyte, respectively. The feeds were at the same flow rate of 2 ml/min. In Fig. 3(A), as the electrolysis progressed, the anodic carbonate concentrations reached a steady state of zero within 30–40 min at 80, 100, and 120 mA/cm², while the times required to reach the steady states were shortened with the applied current. In the case of 60 mA/cm², the carbonate concentration was about 0.1 M at the steady state. The recovered carbonate salt solution at the gas absorber outlet reached their steady state values while showing overshooting peaks before the steady states in all the cases except for 60 mA/cm². During the electrolysis, the water in the anodic chamber was observed to move into the cathodic chamber by an osmotic phenomenon due to the migration of the Na⁺ ions from the anodic chamber. Fig. 4 shows the change of the flow ratios at the inlet and the outlet of the anodic and cathodic solutions at the steady state at the applied current densities. In the figure, the flow ratio of 1 at inlet and outlet means that there is no transfer of water through the membrane during the electrolysis. The result shows that the transferred water increases with the applied current. The carbonate concentrations in the following figures as well as Fig. 3(A) are the concentrations evaluated after taking into consideration the water transfer. The recovered carbonate concentration at the gas absorber in the steady state was the same as the initial carbonate concentration of the anodic reservoir. These results imply that the CO₂ released from the anodic chamber can be completely recovered into the cathodic solution without any gas slip from the CO₂ gas absorber. At this time, the UO₂²⁺ ions detached from the uranyl carbonate complex ions and the SO₄²⁻ ions used as a supporting electrolyte were experimentally observed to remain in the anolyte solution after the acidification without moving into the cathodic chamber. Metal ions with a higher valance than +1, polyatomic ions like UO₂²⁺, and anions are known to be hardly migrated through the cation exchange membrane used in this work [6]. Above results imply that the electrolytic system used in this work can continuously separate and recover the carbonate salt solution from a used carbonate salt solution contaminated with metal ions without chemicals being added into the system. In Fig. 3(B), the pH values at the gas absorber were constant at the initial time, and then they revealed a transient drop like a well, and finally reached a steady state. On the other than, the pH at the anodic outlet decreased to a steady state in the acidic condition, revealing two typical inflection points at about pH 8 and 4 by a titration of the carbonate solution with the proton ions produced through the water splitting reaction. The cell voltages became stable after a rapid decrease at the initial time, and the cell voltages at their steady states increased

with the applied current density. In Fig. 3(C), the NaOH concentration at the gas absorber outlet increased with the applied current. When a 0.5 M carbonate solution is decarbonated in this system, the theoretical NaOH concentration measured at the final cathodic chamber should be 1.1 M, because a feed cathodic solution of 0.1 M NaOH was used and because 1.0 M Na⁺ ions are migrated from the anodic chamber after the decarbonation. The migration is known to be proportional to the cell voltage applied between the anode and cathode [9]. The NaOH concentration in the case of 80 mA/cm² was about 1.1 M. However, that in the case of 120 mA/cm² reached about 1.2 M. The reason is considered to be because additional Na⁺ ions from the supporting electrolyte of Na₂SO₄ were migrated into the cathodic chamber at 120 mA/cm² showing a higher cell voltage (as seen in Fig. 3(B)), compared with the case of 80 mA/cm².

In a flow-through electrolytic system like the system used in this work, the relation of the applied current and the concentration change of a material during an electrolysis can be expressed by a simple equation as shown in the following equation [10,11].

$$i = nFf(C_{in} - C_{out}) \quad (7)$$

where i , f , C , n , and F are the current (A), flow rate (l/s), concentration (mol/l), and number of electrons relevant to electrolysis reaction, and the Faraday constant (96,487 C), respectively. In this electrolytic decarbonation system, the decarbonation in the anodic chamber occurs by the proton ions generated due to the water split reaction and two mole electrons are necessary to convert one mole of CO₃²⁻ to CO₂ gas. By using Eq. (7), the theoretical current required to completely decarbonate a feed solution of 0.5 M Na₂CO₃ at 2 ml/min is calculated to be 3.21 A. Total currents supplied in the three cells-stacked electrolyzer used in this work at 60, 80, 100, and 120 mA/cm² were 3.06, 4.08, 5.1, and 6.12 A, respectively. Accordingly, only in the case of 60 mA/cm², because the total current supplied to the system was insufficient, its pH at the anodic outlet was not decreased below pH 4 where only carbonic acid exists in the solution. It resulted in an incomplete decarbonation in this flow-through system.

The phenomena of an overshooting peak of the carbonate concentration and a transient pH drop at the gas absorber outlet before a steady state can be explained as follows. The overshooting peak of the carbonate concentration in the cases of 80, 100, and 120 mA/cm² is considered to be because the initial solution filled within the stacked cells was additionally decarbonated by the redundant current leftover after the complete decarbonation of the feed carbonate solution before a steady state. Therefore, in the case of 60 mA/cm² which is insufficient to completely decarbonate the feed solution, this overshooting phenomenon was not observed. The transient pH drop phenomena is considered to be because the OH⁻ ions existing within the gas absorber were immediately consumed by the CO₂ gas released from the anodic chamber before the amount of OH⁻ generated in the cathodic chamber arrived at the gas absorber. The time delay for the OH⁻ generated in the cathodic chamber to reach the gas absorber is firstly due to the migration time of the Na⁺ ions from the anodic chamber and secondly due to the time taken for the generated NaOH solution to arrive at the gas absorber through the connecting line. From the results of Fig. 3(B), the prevention of these pH drop phenomena at the gas absorber outlet are considered to be necessary. If the solution pH decreases below 8, a CO₂ gas slip can occur from the gas absorber without recovering all the CO₂ gas generated in the anodic chamber. H₂CO₃, which releases CO₂ gas easily, begins to coexist with HCO₃⁻ in a solution below pH 8 [6]. This will be discussed in Fig. 7 more in detail.

Fig. 5 shows the changes of the carbonate concentrations (A) and pHs at the anodic outlet and the gas absorber outlet (B), and

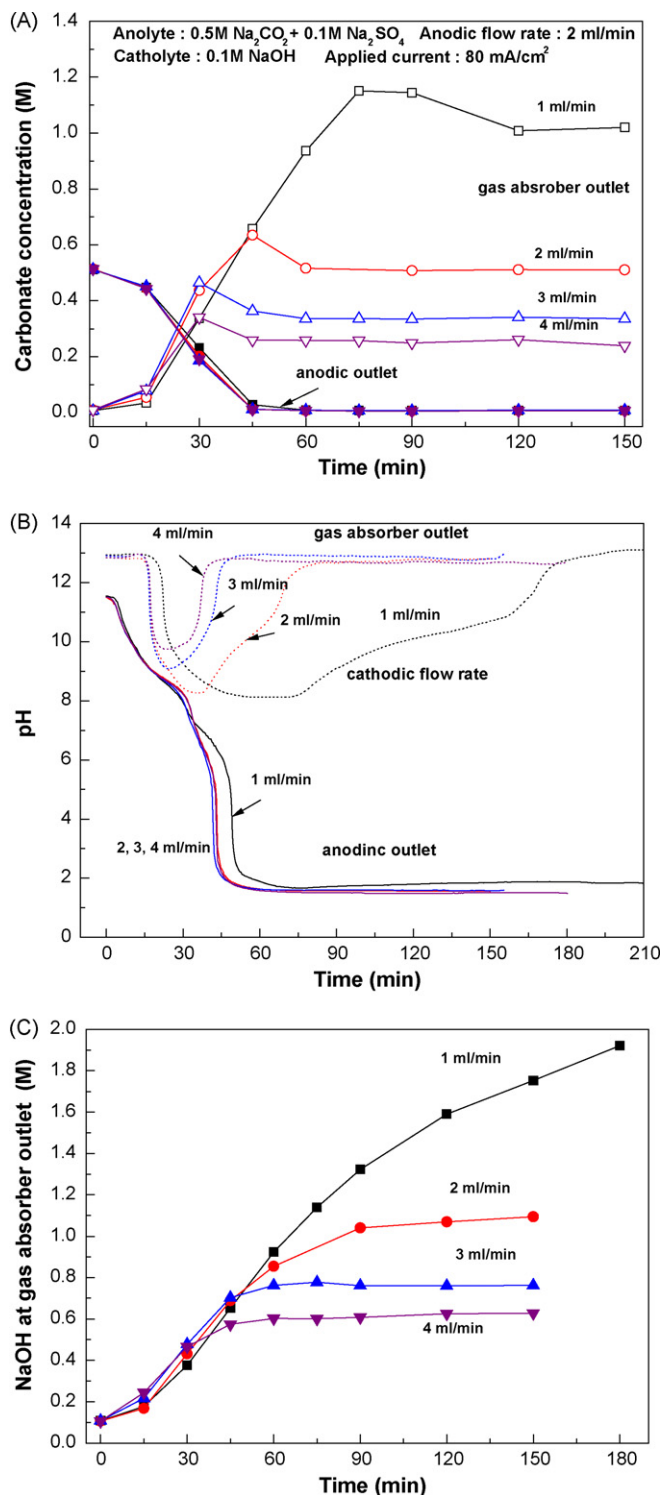


Fig. 5. Changes of carbonate concentrations (A), and pHs at anodic outlet and gas absorber outlet (B), and NaOH concentrations at gas absorber outlet (C) with electrolysis time at several cathodic feed rates.

the NaOH concentrations at the gas absorber outlet (C) during the electrolysis time at a constant current density of 80 mA/cm² with a change of the cathodic feed rates of 1, 2, 3, and 4 ml/min. This cathodic feed rate is an important variable to control the concentration of a recovered carbonate salt solution and to affect the gas–liquid contact in the gas absorber. In Fig. 5(A), the recovered carbonate concentration at the gas absorber was reversely proportional to the cathodic feed rate at the steady state where all the CO₂

gas generated from the acidification in the anodic chamber could be recovered in the cathodic solution. At this time, the concentration change of the carbonate solution at the anodic outlet was almost the same regardless of the feed rate. In Fig. 5(B), the pH at anodic outlet did not reveal a distinct change with the cathodic feed rate, as well. However the pH at the gas absorber revealed a considerable change with the cathodic flow rate before a steady state. The pH drop became bigger and wider with decrease of the cathodic feed rate. The reason is considered to be because the time for the OH⁻ generated in the cathodic chamber to arrive at the gas absorber becomes longer with decrease of the cathodic flow rate. Accordingly, in Fig. 5(C), the time taken for the NaOH concentration at the gas absorber outlet to reach a steady state where a sufficient NaOH solution flowed down within the gas absorber to completely absorb the CO₂ from the anodic chamber became retarded at a slower cathodic feed rate. During this retarded time, the pH in the gas absorber can become lower to below pH 8 where a CO₂ gas slip can occur.

Fig. 6 shows the changes of the carbonate concentrations (A) and the pHs at the anodic outlet and the gas absorber outlet (B), and the NaOH concentrations at the gas absorber outlet (C) during the electrolysis time at a constant current density of 80 mA/cm² with a change of the anodic feed rates of 1, 2, and 3 ml/min. In Fig. 6, when the applied current into the electrolytic system was lower than the currents calculated by Eq. (7) at the different anodic feed rates to achieve a complete decarbonation of the feed carbonate solutions, the pH and the carbonate concentration at the anodic outlet could not become acidic enough, and could not become zero, respectively. In the case of an anodic feed flow of 0.5 M Na₂CO₃ at 3 ml/min, the necessary current is 4.83 A to complete decarbonation of the feed solution. However, the actual total current applied in the system at 80 mA/cm² was 4.08 A. Therefore, the decarbonation was not complete, and the solution pH at the anodic outlet stopped at 7.3. However, in the cases of 1 and 2 ml/min where the theoretically necessary currents were lower than the applied currents, the concentration of the recovered carbonate solution at the gas absorber was proportional to the anodic flow rate, being the same as the theoretical evaluation.

3.3. Overcoming the initial transient disadvantageous phenomena in the continuous electrolytic system

A way to prevent the pH drop problem with a possibility of CO₂ slip before a steady state at the gas absorber, which is due to the retardation of the OH⁻ solution generated in the cathodic chamber to the gas absorber, should be taken into consideration, when the electrolytic continuous system is applied for an actual case. The prevention of a CO₂ slip can be achieved by using a NaOH solution with a higher concentration as a cathodic feed solution, or introducing additional NaOH solution into the cathodic feed only until the time taken for a steady state in the case without using the additional NaOH to appear, or by a pre-electrolysis prior to starting the decarbonation electrolysis to make the OH⁻ concentration of the cathodic solution higher. Fig. 7 shows an example to prevent the pH drop problem by adding a 10 M NaOH solution at 0.2 ml/min into the normal cathodic feed solution of 0.1 M NaOH before the time taken for a steady state to appear. There are no distinct changes of the carbonate concentrations at the anodic and gas absorber outlets in Fig. 7(A) regardless of the additional NaOH solution. However, the pH drop phenomena at the gas absorber outlet almost disappeared in the case of using the additional NaOH solution, compared with the case of using only a cathodic feed solution of 0.1 M NaOH in Fig. 7(B). This is considered to be because the OH⁻ concentration within the gas absorber was enough to immediately convert the CO₂ gas coming from the anodic chamber into the carbonate

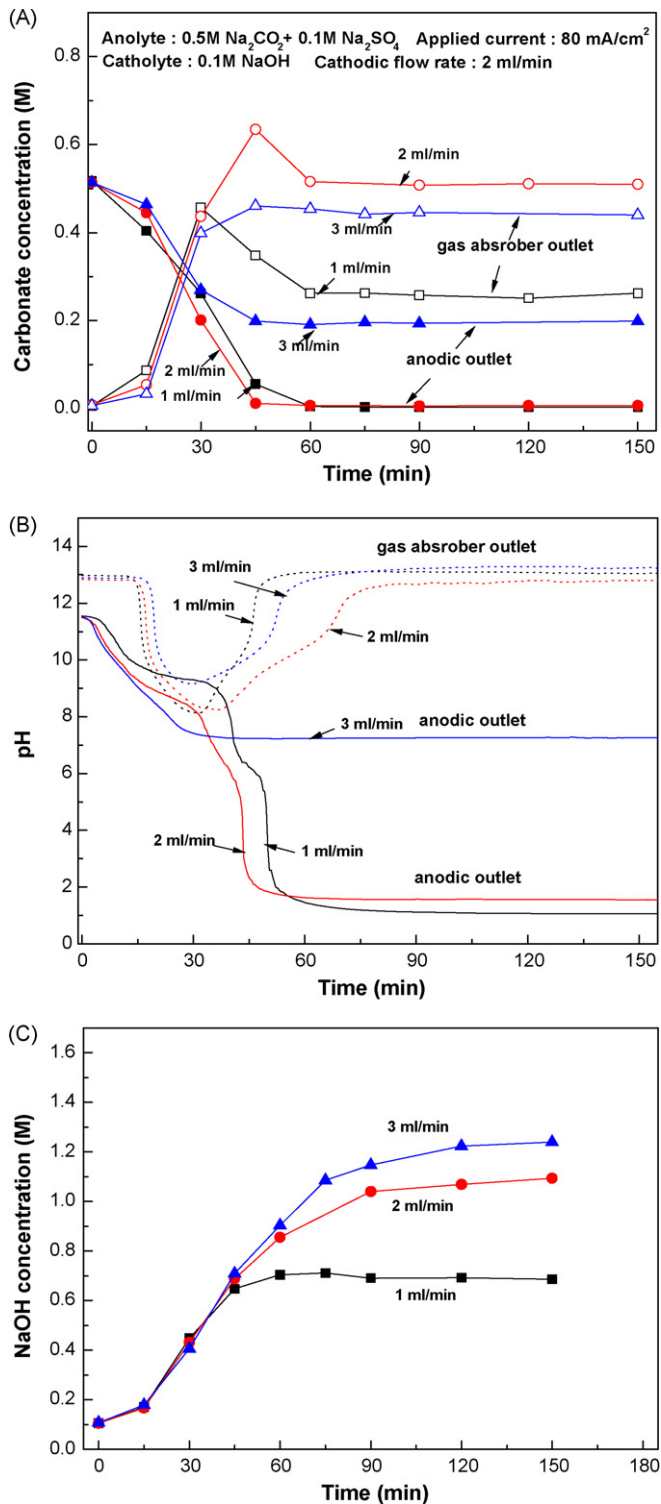


Fig. 6. Changes of carbonate concentrations (A), and pHs at anodic outlet and gas absorber outlet (B), and NaOH concentrations at gas absorber outlet (C) with electrolysis time at several anodic feed rates.

salt solution before the NaOH solution generated in the cathodic chamber reached the gas absorber. In Fig. 7(C), the NaOH concentration at the gas absorber outlet during the addition of the NaOH solution increased, but after stopping the introduction of the additional NaOH solution, it went back to the same concentration as that without the additional NaOH solution and then reached a steady state.

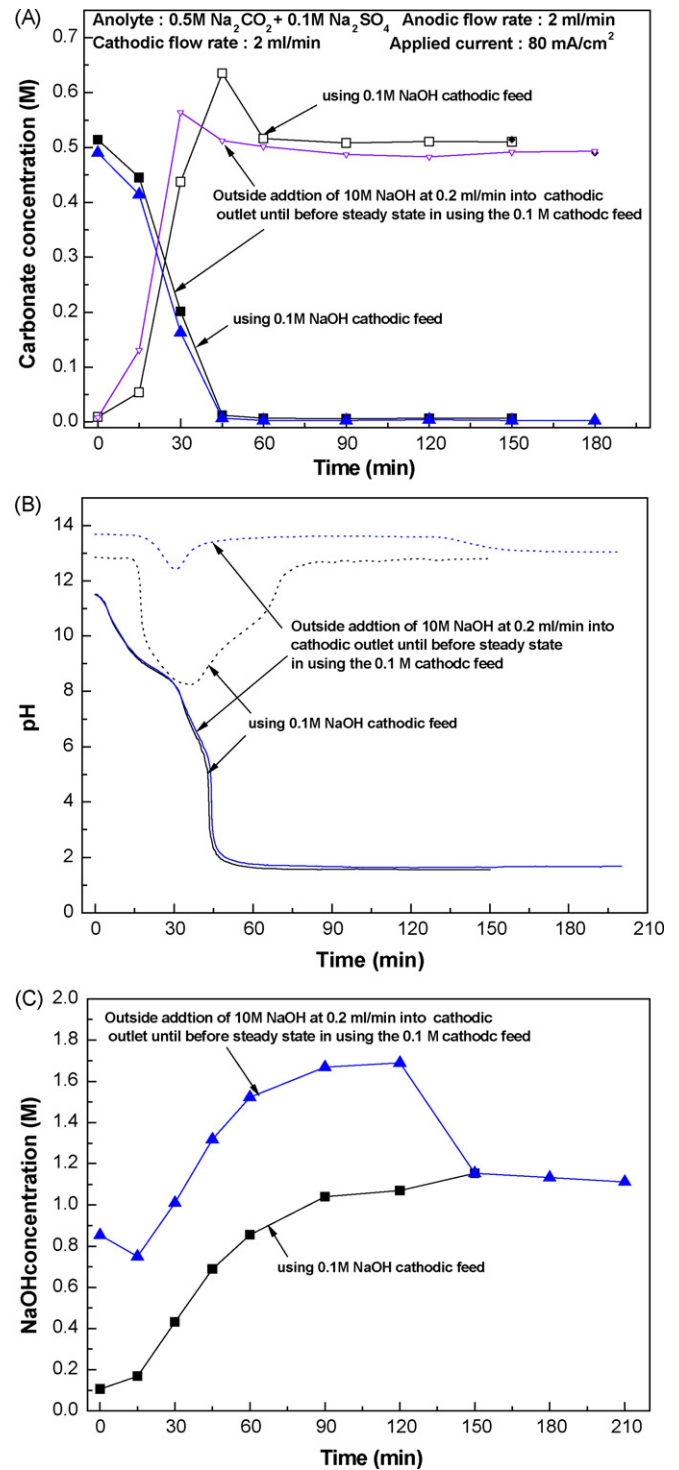


Fig. 7. Changes of carbonate concentrations (A), and pHs at anodic outlet and gas absorber outlet (B), and NaOH concentrations at gas absorber outlet (C) with electrolysis time with and without outside addition of 10M NaOH at 0.2 ml/min into the cathodic feed solution.

4. Conclusions

The continuous electrolytic system devised in this work, which consisted of a cells-stacked electrolyzer equipped with a cation exchange membrane and a gas absorber, could continuously decarbonate and recover a carbonate salt solution from a uranyl carbonato complex solution, the uranyl ion solution being separated from the system. The cathodic feed rate could control the carbonate

concentration of the recovered solution and it affected considerably the transient pH drop phenomena of a well type within the gas absorber before a steady state, which caused a possibility of a CO₂ gas slip from the gas absorber. The pH drop problem could be overcome by temporarily increasing the OH⁻ concentration of cathodic solution within the gas absorber only during the time required for a steady state to be obtained in the case without the addition of outside NaOH. The overshooting peak of the carbonate concentration in the recovered solution before a steady state was ascribed to the decarbonation of the initial solution filled within the stacked cells by a redundant current leftover from the complete decarbonation of the feeding carbonate solution.

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