

Experimental Investigation on Hypochlorous Acid Water Production using Electrode Plates without a Barrier Membrane (Part I: Production conditions for high-concentration Hypochlorous acid water)

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Abstract: Available chlorine has been widely used for the sterilization and disinfection of the tap water supply. The development of a device is awaited which will enable the generation of available chlorine at high concentrations with high efficiency. The purpose of this paper is to propose optimum operation conditions for available chlorine production from the standpoint of high concentration. The experiment is conducted using a flow-type reactor with narrow and parallel electrode plates, even though it lacks a barrier membrane between the plates. The governing factors are the electrode plate interval and the flow rate of sodium chloride solution from the viewpoint of hydrodynamics, and the concentration of sodium chloride of the medium and current density supplied to the electrode plates from the standpoint of chemical reactions. The concentration of available chlorine was measured using a residual chlorine meter. The influence of operation conditions on the concentration of available chlorine is shown. Moreover, the governing factors are examined using experimental design because many experimental conditions are expected for the above conditions. As a result, the high concentration production of available chlorine is strongly affected by the current density as well as sodium chloride concentration. These results will be useful for producing chlorinated water, called hypochlorous acid water.

Keyword: Hypochlorous acid water, Concentration, Available chlorine, Parallel electrode plates, Experimental design, Experiment

I. INTRODUCTION

The supply of safe drinking water is very important for sustainable human life. An abundant supply of drinking water from a faucet remains insufficient in many developing countries. Unpurified water leads to infection and disease. Hypochlorous acid water, which contains available chlorine, is useful for the sterilization and disinfection of the tap water supply [1, 2]. In addition, disinfected water is in demand from a medical standpoint because it is used in measures dealing with natural calamities [3]. In this case, a highly concentrated disinfectant or the on-site production of disinfected water is desired in order to reduce transport capacity. As a sterilization disinfectant, it is widely used to disinfect devices and various apparatuses, e.g., in the manufacturing process of various foods including the disinfection of drinking water, fruits, vegetables, and dairy products [4]. The bactericidal effects of available chlorine are useful to the marine products industry as well as the food industry. As an example, the disinfection of water for the aquaculture of seed production is very important for preventing the spread of infectious disease [5]. In addition, the production efficiency of available chlorine is also important from the standpoint of energy consumption for production. Therefore, the development of a simple device is needed which makes it possible to produce available chlorine at a high concentration with a high efficiency. The characteristics of available chlorine and related chemical substances have been investigated in detail [6-11].

The purpose of this paper is to examine parameters influencing the production of hypochlorous acid water with emphasis on obtaining a high concentration [12, 13]. In the experiment, hypochlorous acid water is produced by the electrolysis of sodium chloride solution. Hypochlorous acid water with gas flows between the anode and cathode electrode plates, and with no membrane separating the flow between the electrode plates. Gas and liquid two-phase flow in narrow and parallel electrode plates is examined, where gas is generated during the experiment by chemical reactions and becomes an obstacle for the chemical reactions. The concentration of hypochlorous acid is evaluated by changing the electrode interval, electric current density, flow rate and concentration of sodium chloride solution.

II. HYPOCHLOROUS PRODUCTION MECHANISM

2.1 Available chlorine

Available chlorine is a generic name for chlorine that shows bactericidal action. Free chlorine and combined chlorine are both included in available chlorine. The free chlorines are hypochlorous acid (HClO), hypochlorous acid ion (OCl⁻) and chlorine gas (Cl₂) [14-16]. These three kinds of free chlorine change their respective existence ratios based on pH, as shown in Fig. 1. In this experiment, almost all of the free chlorine in the solution is HClO, because the produced solution becomes around pH 5 [16]. Table 1 shows the principal characteristics of HClO [9]. The order of antiseptic capability is Cl₂ < OCl⁻ < HClO. HClO shows the strongest bactericidal activity in free chlorines. Free chlorine destroys the cell films and the cell walls of microorganisms and viruses using its strong oxidation, and sterilization or disinfection effects are demonstrated by the degeneration of an internal protein and nucleic acid. Moreover, HClO exists only as an aqueous solution, and it can be obtained in a solution of up to 25%. It is readily susceptible to heat, and the decomposition of HClO to hydrochloric acid HCl, oxygen O₂ and chloric acid HClO₃ are promoted at water temperatures of over 44 °C [17]. Therefore, it is necessary to note the temperature of the aqueous solution.

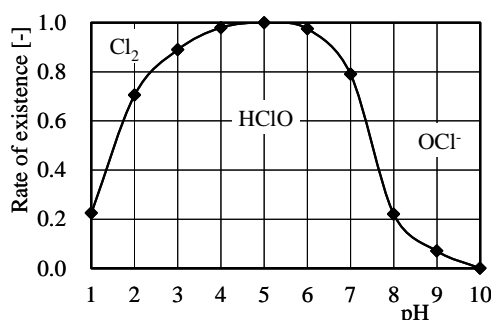


Figure 1. Forms of free chlorine

Table 1. Properties of HClO

Structural diagram	H - O - Cl
Dissociation constant K_{HClO}	2.95×10^{-8} mol/l ($HClO \rightarrow H^+ + ClO^-$)
Molecular weight M_{HClO}	52.45 g/mol
Degradation temperature	44 °C
Oxidation number	+1

2.2 HClO production mechanism

It is necessary to add chloride to water as an electrolysis auxiliary agent in order to produce HClO. Sodium chloride, NaCl, is used as an electrolysis auxiliary substance in this experiment because its treatment is safe and easy. Figure 2 shows a production process for HClO that uses no diaphragm electrolyzer [12, 13]. NaCl is dissociated into chloride ion Cl⁻ and sodium ion Na⁺ in water. At the anode, H₂O is decomposed into O₂ and H⁺ by oxidation action, and Cl₂ is produced from Cl⁻ also by oxidation action. The production of Cl₂ occurs preferentially over H₂O decomposition because Cl⁻ is a halogen ion. Although almost all produced Cl₂ is released outside as a gas, part of it is dissolved into H₂O and produces HClO. On the other hand, at the cathode, an electron is given to H₂O by a reducing process, and H₂ and hydroxide ion OH⁻ are produced. Sodium ion Na⁺ produced at the cathode combines with hydroxide OH⁻, and then sodium hydroxide NaOH is produced. In addition, Na⁺ combines with HCl and hypochlorous acid HClO, then NaCl and sodium hypochlorite NaClO are also produced in the solution, too. NaClO also exhibits bactericidal action [18-21].

Figure 3 shows the relation of electric potential and voltage in a reactor consisting of electrode plates with electrolytic solution [22-26]. The electrified state of the electrode plates can be controlled by the application of voltage to the plates. The distribution of ions which have an opposite charge to the electrode plates will change in proportion of the electrified state. The electric potential is divided into three types that are caused at each layer, such as the Helmholtz double layer, the diffuse electric double layer and other solution regions, as shown in Fig. 3. The electric double layer, and especially the Helmholtz double layer, has a property of accumulating electricity. The electric potential of the solution resistance becomes lower as the concentration of ions in the electrolyte solution becomes higher. Generally, almost all impressed voltage to the electrode plates is consumed at the Helmholtz double layer.

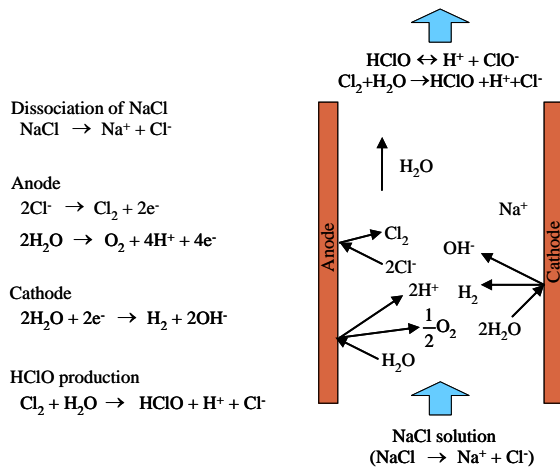


Figure 2. HClO production process

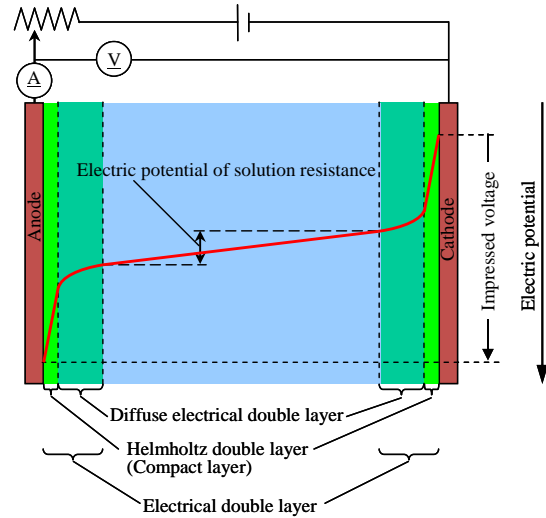


Figure 3. Relation of electric potential and voltage

III. EXPERIMENTAL APPARATUS AND PROCEDURE

3.1 Experimental apparatus

The experimental apparatus employed in this experiment is shown in a decomposed state in Fig. 4. The system consists of the test section, tubing pump, power supply for electrolysis, data logger for recording applied voltage and local temperature, clamping meter for the current measurement and the beaker for inspection [12, 13]. In this system, NaCl solution passes between the narrow electrode plates, that is, a flow-type electrolysis device without a membrane separator. The interval between the titanium electrode plates, d , is changed using spacers of several thicknesses. The aspect ratio of the electrode is 100 mm x 50 mm in height and width, respectively. In addition, the reaction area, S , of each electrode plate is the same. A sight glass is placed at the exit region of the reactor in order to observe the behavior of the bubbles created by the chemical reaction. An electrode made by a titanium plate coated with a thin layer of platinum is applied [27]. The thickness of the plate is 0.5 mm and the platinum is 50 μm . The thermocouples are fitted on the back side of the titanium plate to observe the plate temperatures which are then recorded by a data logger. Each titanium electrode plate is fixed to a 30 mm thick acrylic plate, with the titanium electrode plates fixed parallel to one another. The joint surface of each element of the device is sealed by O-rings. A small chamber with parallel thin tubes is set at the entrance region of the test section in order to have a uniform stream over the inlet cross section.

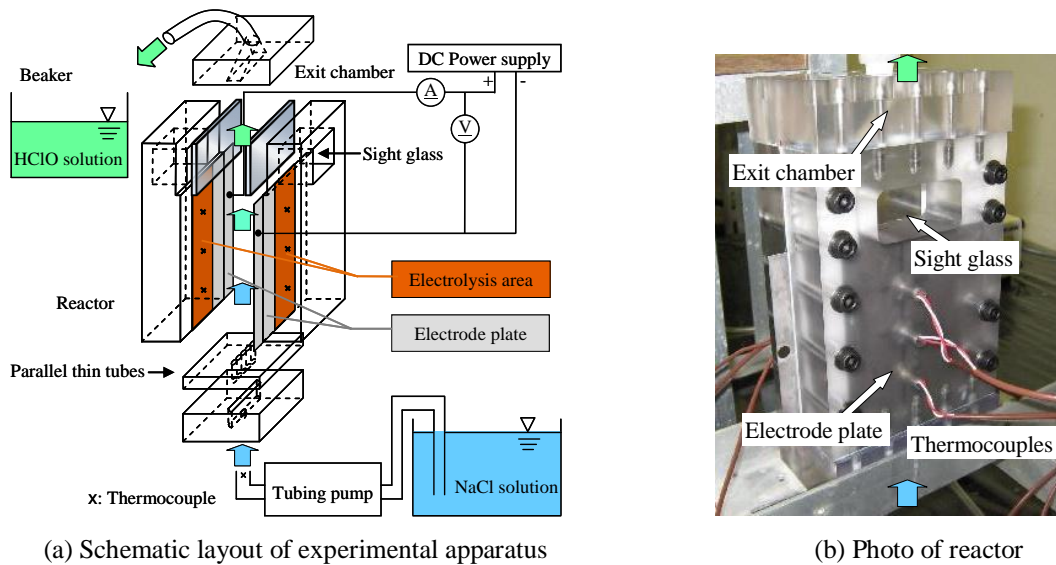


Figure 4. Experimental apparatus employed in this experiment

3.2 Experimental procedure

Firstly, the decomposition voltage of the NaCl solution is measured for each NaCl concentration, electrode plate interval and flow rate to find a minimum impressed voltage. Then, the experiment is started. At

the beginning of the experiment, the flow temperature is unstable as the electrode plates are heated because of Joules' law. After the steady state condition is achieved, HClO solution, local temperatures, impressed voltage and current are measured.

A residual chlorine meter is introduced to measure the concentration of available chlorine by means of the absorptiometric method using a DPD (Diethyl-Phenylene-Diamine) reagent. The concentration of collected available chlorine is too dense to measure using a residual chlorine meter. Therefore, the collected solution is diluted to a predetermined magnification using purified water in order to obtain the optimum range of concentration for the meter. The measured value is multiplied corresponding to the above magnification factor to provide a true dimension.

3.3 Theoretical decomposition voltage of NaCl solution

In order to produce HClO from a NaCl solution by electrolysis, Cl₂ has to be produced at the anode side, as shown in Fig. 2. The minimum voltage to produce Cl₂ should be confirmed theoretically before the experiment. First, the standard electrode potentials of each electrode plate are estimated. The electrolysis will begin over this electrode potential. The standard electrode potential is derived using standard Gibbs energy of formation. The standard Gibbs energy of formation ΔG° is emitted from each electrode plate as an electric energy.

$$-\Delta G^\circ = nF\Delta V \tag{1}$$

Here, *n* is the electron count that participates in the reaction, *F* is a Faraday's constant $F = 9.65 \times 10^4$ C/mol, and Δ*V* is the electric potential difference.

The reaction at an anode plate, Cl₂ is produced from Cl⁻ as shown in Eq. (2).



Therefore, the electric potential difference, Δ*V*_{Cl₂}, required to produce Cl₂ is estimated by Eq. (1) because 1 mole of electron reacts with 1 mole of Cl⁻.

$$\begin{aligned} \Delta V_{\text{Cl}_2} &= -\frac{\Delta G^\circ(\text{Cl}^-)}{nF} \\ &= -\frac{-131.23 \times 10^3}{1 \times (9.65 \times 10^4)} = 1.36 \text{ V} \end{aligned} \tag{3}$$

Here, Δ*G*°(Cl⁻) = -131.23 kJ/mol at 25 °C, 1 atmosphere [28].

On the other hand, at a cathode plate, H₂ is produced from H₂O as shown in Eq. (4).



The electric potential difference, Δ*V*_{H₂}, required to produce H₂ is estimated by Eq. (1) as follows because 2 moles of electron reacts with 1 mole of H₂.

$$\begin{aligned} \Delta V_{\text{H}_2} &= -\frac{\Delta G^\circ(\text{OH}^-)}{nF} \\ &= -\frac{-157.24 \times 10^3}{2 \times (9.65 \times 10^4)} = 0.81 \text{ V} \end{aligned} \tag{5}$$

Here, Δ*G*°(OH⁻) = -157.24 kJ/mol at 25 °C, 1 atmosphere [28]. Therefore, the overall reaction needed to produce HClO is shown in Eq. (6).



As a result, the electric potential difference between anode and cathode electrode plates is estimated as follows.

$$\begin{aligned} \Delta V_{\text{HClO}} &= \Delta V_{\text{Cl}_2} + \Delta V_{\text{H}_2} \\ &= 1.36 + 0.81 = 2.17 \text{ V} \end{aligned} \quad (7)$$

The electric potential difference, called the decomposition voltage, $\Delta V_{\text{HClO}} = 2.17 \text{ V}$ is required theoretically to produce HClO. The decomposition voltage will increase slightly in the experiment because the electrolytic solution includes infinitesimal impurities and because of overvoltage at the electrode plates.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

4.1 Current-voltage characteristics of electrode plates

In order to confirm the characteristics of the electrode plate, the current-voltage relationship is important to setup an experimental condition. Figure 5 shows a current-voltage relation for the concentration of NaCl $C_{\text{NaCl}} = 50,000 \text{ mg/l}$ and volume flow rate $\dot{Q} = 0.0551 \text{ ml/s}$ as an example. In this experiment, the decomposition voltage for the reaction is estimated to be around 2.2 Vdc. Therefore, the voltage between electrode plates is set at over 2.2 Vdc in this experimental condition.

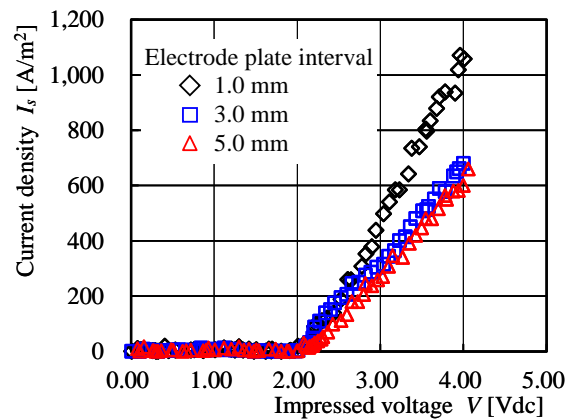


Figure 5. Example of current-voltage relation ($C_{\text{NaCl}} = 50,000 \text{ mg/l}$, $\dot{Q} = 0.0551 \text{ ml/s}$)

4.2 Index for high-concentration production

The purpose of this study is to produce HClO at a high-concentration. The production at a high efficiency is also important. However, this is next to impossible to carry out simultaneously. Therefore, the concentration and efficiency of the produced HClO are evaluated individually.

The basic balance condition may be expressed as follows:

$$\frac{\dot{Q} \times C_{\text{NaCl}}}{M_{\text{NaCl}}} \times 10^{-6} = \frac{I_s \times S}{F} \quad (8)$$

Here, C_{NaCl} is the concentration of the NaCl solution [mg/l], \dot{Q} is the flow rate of the NaCl solution [ml/s], M_{NaCl} is the molecular weight of the NaCl [g/mol], F is the Faraday's constant [C/mol], I_s is the current density supplied to the electrode [A/m^2], and S is the reaction area [m^2]. This is configured to be equal to the number of moles of both Cl^- and e^- . The supplied current density on the electric plates is estimated by Eq. (8). Then, after the influence of concentration of NaCl solution on the available chlorine concentration is recorded, the current density and flow rate of the NaCl solution will be changed for the electrode plate intervals.

There are many experimental conditions in this experiment. Therefore, the narrowing of experimental conditions is introduced in this experiment in order to obtain the influence of these experimental parameters. Firstly, the influence of the NaCl concentration in the solution, C_{NaCl} , on the available chlorine concentration, C_{AC} , is evaluated for plate intervals with a constant flow rate. Next, the influence of the current density, I_s , supplied to the electrode plate on the C_{AC} is examined for the C_{NaCl} where the higher C_{AC} was obtained. Then, the influence of the flow rate of solution on the C_{AC} is examined based on the optimum C_{NaCl} and I_s . Finally, the influence of the plate interval, d , on the C_{AC} is examined based on the optimum amounts of C_{NaCl} , I_s and \dot{Q} . The electrode plate intervals are varied in each experiment because the bubbles generated by the chemical reaction affect the effective reaction area of the electrode plates.

4.3 Dependence of NaCl concentration of solution flowing into a reactor

The NaCl concentration in the solution flowing into a reactor, C_{NaCl} , is varied to show the influence of Cl^- on available chlorine production. Table 2 and Fig. 6 show the experimental results for a constant flow rate and several plate intervals. As the NaCl concentration increases and the electrode plate interval becomes larger, the available chlorine concentration, C_{AC} , increases. The reason is that the increase in the abundance of Cl^- will increase the molar density of Cl^- close to the anode. It makes reactions at the anode surface easily, and the available chlorine concentration will increase. The gradient of C_{AC} resulting from $d = 1.0$ mm in Fig. 6 is smaller than in other results. It may be considered that the bubbles generated by the reaction will reduce the contact surface area between the liquid and electrode plates. As a result, the bubbles prevent a reaction between narrow electrode plates. Higher generation of C_{AC} is obtained at $C_{NaCl} = 50,000$ mg/l in this experiment.

In the low C_{NaCl} conditions, the range of C_{AC} distribution is small compared to the highest C_{NaCl} conditions, as shown in Fig. 6. In this condition, Cl^- production may be small and Cl_2 may react sufficiently in each plate interval. As a result, C_{AC} does not heavily depend on the plate interval in the lower C_{NaCl} . On the other hand, in the high C_{NaCl} conditions, the difference obtained from $d = 1.0$ mm to 3.0 mm is smaller than that of $d = 3.0$ mm to 5.0 mm. It may be considered for $d = 1.0$ mm that the bubbles created between the narrow parallel plates fill up the reaction surface of the plates and prevent the reactions.

Table 2. Experimental results at a constant flow rate ($\dot{Q} = 0.0551$ ml/s)

Electrode interval d [mm]	Concentration of NaCl C_{NaCl} [mg/l]	Current density I_s [A/m^2]	Voltage V [Vdc]	Concentration of available chlorine C_{AC} [mg/l]
1.0	10,000	182	3.12	720
	20,000	363	3.27	2,180
	30,000	545	3.36	3,190
	40,000	727	3.50	4,240
	50,000	909	3.65	4,910
3.0	10,000	182	3.46	610
	20,000	363	3.67	2,550
	30,000	545	3.59	4,060
	40,000	727	3.61	4,660
	50,000	909	3.75	6,040
5.0	10,000	182	3.65	680
	20,000	363	3.83	3,120
	30,000	545	3.89	4,290
	40,000	727	3.98	5,710
	50,000	909	4.03	6,840

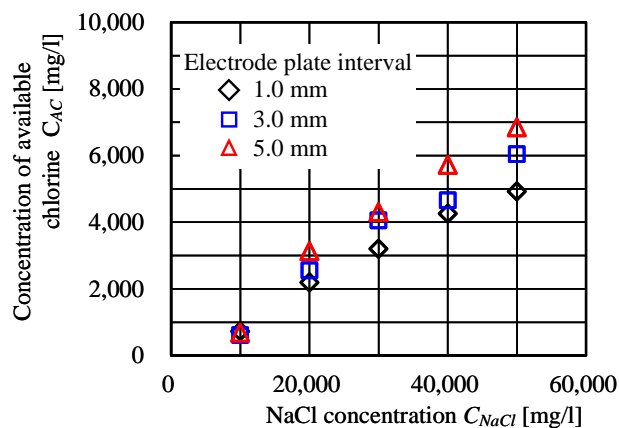


Figure 6. Concentration of available chlorine ($\dot{Q} = 0.0551$ ml/s)

4.4 Dependence of current density

Current density supplied to the electrode plate is varied. However, the flow rate is fixed at $\dot{Q} = 0.0551$ ml/s, and the NaCl concentrations of the medium chosen here are $C_{NaCl} = 30,000, 40,000$ and $50,000$ mg/l, because higher concentration production of the available chlorine were achieved in these conditions during the preliminary experiment.

The experimental results are shown in Table 3 and Fig. 7. The concentration of available chlorine C_{AC} increases as the current density increases. The mobility of ions and electrons become active as the current density increases. Here, the mobility of Cl^- becomes active and the reaction of Cl^- increases. As a result, C_{AC}

increases as the current density increases. On the other hand, the mobility of Cl^- becomes slower when the current density is smaller. In this case, the electron will be consumed in the H_2O reducing reaction. The fluid temperature between the electrode plates rises with higher current densities and HClO will be decomposed at just over 44°C . A higher C_{AC} was obtained at $I_s = 800 \text{ A/m}^2$ in this experiment.

Table 3. Experimental results at several current densities ($\dot{Q} = 0.0551 \text{ ml/s}$)

Electrode interval d [mm]	Concentration of NaCl C_{NaCl} [mg/l]	Current density I_s [A/m^2]	Voltage V [Vdc]	Concentration of available chlorine C_{AC} [mg/l]
1.0	30,000	200	3.04	1,960
		400	3.26	3,260
		600	3.45	3,590
		800	3.60	4,060
	40,000	200	2.65	2,000
		400	3.18	3,490
		600	3.40	3,920
		800	3.59	4,590
	50,000	200	2.96	2,280
		400	3.19	3,380
		600	3.34	3,840
		800	3.51	4,460
3.0	30,000	200	3.03	1,390
		400	3.34	3,030
		600	3.65	3,790
		800	3.87	4,770
	40,000	200	2.55	2,700
		400	3.35	3,700
		600	3.57	5,210
		800	3.78	5,540
	50,000	200	2.56	3,080
		400	3.27	4,120
		600	3.53	5,180
		800	3.70	6,280
5.0	30,000	200	3.24	2,240
		400	3.68	3,960
		600	3.97	4,690
		800	4.22	5,200
	40,000	200	3.11	2,910
		400	3.58	4,700
		600	3.82	5,340
		800	4.00	6,450
	50,000	200	2.64	3,150
		400	3.44	4,540
		600	3.75	5,830
		800	3.93	6,840

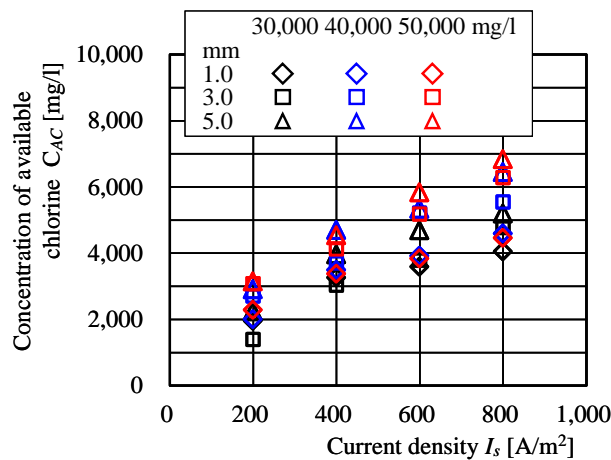


Figure 7. Concentration of available chlorine versus current density ($\dot{Q} = 0.0551 \text{ ml/s}$)

4.5 Dependence of solution flow rate

The flow rate of the NaCl solution is varied and the NaCl concentration is fixed at $C_{NaCl} = 50,000$ mg/l. Current densities are set at $I_s = 600$ and 800 A/m² in order to achieve a sufficient reaction. The results are shown in Table 4 and Fig. 8. The concentration of available chlorine C_{AC} decreases as the flow rate increases. As the flow rate increases, the volume flow rate of H₂O increased and the reaction of Cl₂ on the electrode plate is reduced because Cl₂ has fewer chances to undergo the desired reaction. As a result, Cl₂ does not sufficiently dissolve into H₂O and then flows out. In order to produce a high concentration of available chlorine, a lower flow rate may be desired.

Table 4. Experimental results at several flow rates ($C_{NaCl} = 50,000$ mg/l)

Electrode interval d [mm]	Flow rate \dot{Q} [ml/s]	Current density I_s [A/m ²]	Voltage V [Vdc]	Concentration of available chlorine C_{AC} [mg/l]
1.0	0.0358	600	3.31	4,620
	0.0551		3.34	3,840
	0.0775		3.40	3,800
	0.1140		3.41	3,080
	0.0358	800	3.47	5,220
	0.0551		3.51	4,460
	0.0775		3.58	4,630
	0.1140		3.56	3,390
3.0	0.0358	600	3.51	5,100
	0.0551		3.53	5,180
	0.0775		3.61	4,540
	0.1140		3.61	4,010
	0.0358	800	3.66	6,640
	0.0551		3.70	6,280
	0.0775		3.75	5,520
	0.1140		3.82	4,870
5.0	0.0358	600	3.74	6,070
	0.0551		3.75	5,830
	0.0775		3.77	5,220
	0.1140		3.79	4,390
	0.0358	800	3.87	7,030
	0.0551		3.93	6,840
	0.0775		3.98	6,010
	0.1140		4.01	5,140

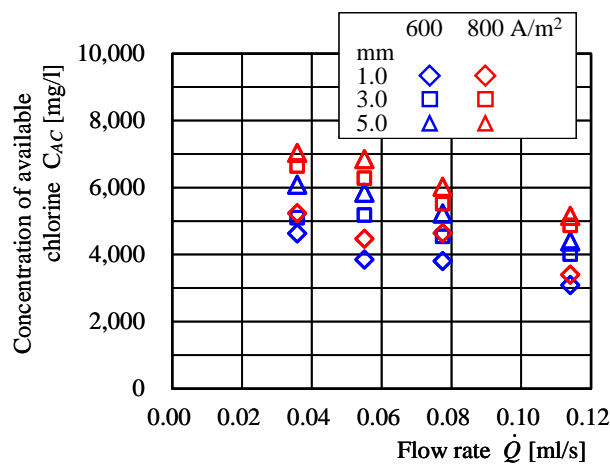


Figure 8. Concentration of available chlorine versus flow rate ($C_{NaCl} = 50,000$ mg/l)

4.6 Dependence of electrode plate interval

Gas and liquid two-phase flow in the narrow and parallel flat plates has very complex flow characteristics. Especially, the generated gas in the flow field may fill up the narrow space and prevent further reactions. The concentration of available chlorine, C_{AC} , increases as the plate interval increases, as shown in Figs. 6-8. As the plate interval increases, the liquid has a chance to make contact with the electrode plates without small bubbles, and the flow velocity becomes low for a constant volume flow rate. In this case, the reaction time of the NaCl solution increases. As a result, Cl₂ is easy to dissolve into the solution at low flow velocity, and thus C_{AC} increases.

4.7 Overall evaluation for high-concentration production

In this experiment, the effects of the experimental parameters on the concentration of available chlorine, C_{AC} , are examined. Geometrical influence and flow conditions are considered as experimental parameters in this experiment. The electrode plate interval is varied for the first mentioned of the two, and the flow rate, current density and NaCl concentration of the solution are varied for the latter. However, the overall evaluation is very important for an actual device design. An experimental design is introduced in this evaluation [29, 30]. This method clarifies the respective importance of each parameter. Table 5 shows the results estimated using an experimental design method for an obtained C_{AC} mentioned above. As a result, current density strongly contributes to C_{AC} production. These results agree well with the experimental results mentioned above.

Table 5. Contribution of parameters to the formation of available chlorine

Factor	Rate of contribution [%]
Current density I_s	56.6
Flow rate \dot{Q}	2.7
Concentration of NaCl C_{NaCl}	23.2
Error	17.5
Total	100.0

V. CONCLUSION

The high concentration production of available chlorine is investigated experimentally using a narrow and parallel electrode plates without a barrier membrane. The flow between the plates is gas and liquid two-phase flow, and the bubbles in the flow field prevented the chemical reaction. In this experiment, the NaCl concentration of the solution, current density supplied to the electrode, electrode plate interval and volume flow rate are all taken into account as experimental parameters. The high concentration production of C_{AC} is affected strongly by the current density and the NaCl concentration. However, bubbles between the electrode plates prevent the chemical reaction. These results will be useful to produce chlorinated water, called hypochlorous acid water, with a high efficiency, too.

Nomenclature

C	= Concentration [mg/l]
d	= Electrode plate interval [mm]
F	= Faraday constant (9.65×10^4 C/mol)
ΔG°	= Standard Gibbs energy of formation [J/mol]
I	= Current density [A/m^2]
K	= Dissociation constant [mol/l]
M	= Molecular weight [g/mol]
n	= Electron count [-]
\dot{Q}	= Volume flow rate [ml/s]
S	= Reaction area [m^2]
V	= Voltage [V]
ΔV	= Electric potential difference [V]

Subscripts

AC	= Available chlorine
$NaCl$	= Sodium chloride
s	= Electrode surface

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