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

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Abstract: *Available chlorine has an effect on the sterilization and disinfection of a water supply, especially for drinking water. In order to obtain available chlorine industrially, it is important to generate available chlorine at high-concentrations with high-efficiency. However, it is difficult to simultaneously attain high concentration with high-efficiency. In this paper, the optimum operation conditions for available chlorine production are proposed from the standpoint of high-efficiency. The experiment was 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 production efficiency of the available chlorine was estimated by the ratio of actually reacted available chlorine to the ideally reacted available chlorine. The governing factors were examined based on the experimental results. As a result, the production of available chlorine with high-efficiency is strongly affected by the flow rate as well as the current density. These results will be useful for producing chlorinated water, called hypochlorous acid water.*

Keywords: *Hypochlorous acid water, Production efficiency, Available chlorine, Parallel electrode plates, Experiment*

I. Introduction

Hypochlorous acid water, which contains available chlorine, is useful for the sterilization and disinfection of safe drinking water supplies[1, 2]. The applications of these kinds of water for medical and industrial purposes have been introduced in many studies[3-5]. The characteristics of available chlorine and related chemical substances have been investigated in detail[6-11]. If hypochlorous acid water, which contains available chlorine, can be produced with high-efficiency, the energy consumption and operation cost may be reduced. In addition, high-concentration production of available chlorine is also important from the standpoint of energy consumption for their transportation. A flow-type reactor with narrow and parallel electrode plates without a barrier membrane between the plates were introduced and examined[12-14]. The operation conditions, such as the electrode plate interval, electric current density, flow rate and concentration of sodium chloride solution, have been investigated for high-concentration production in these literatures. Therefore, it is necessary to develop a simple device which makes it possible to produce available chlorine with high-efficiency as well as a high concentration.

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

II. Hypochlorous Production Mechanism

A production mechanism and the characteristics of available chlorine have already been presented in reference[12]. Here, only the characteristics which affect the production efficiency are shown.

The free chlorines, that is one of the available chlorines, are hypochlorous acid (HClO), hypochlorous acid ion (OCl^-) and chlorine gas (Cl_2)[15-17]. In this experiment, almost all of the free chlorine in the solution is HClO, because the produced solution reaches about pH 5[17]. Table 1 shows the principal characteristics of

HClO[9]. It is readily susceptible to heat, and the decomposition of HClO to hydrochloric acid HCl, oxygen O_2 and chloric acid $HClO_3$ are promoted at water temperatures of over 44 ºC[18]. Generally, the electrode plates and aqueous solution are made to generate heat by Joule's heat. Therefore, it is necessary to note the temperature of the aqueous solution under 44 ºC.

Figure 1 and Eqs. (1)-(5) show a production process for HClO that uses no diaphragm electrolyzer [12-14]. NaCl is dissociated into chloride ion Cl^- and sodium ion Na^+ in water, and flows into the reactor. At the anode, H_2O is decomposed into O_2 and H^+ by oxidation action, and Cl_2 is produced from Cl^-, also by oxidation action. The production of Cl_2 occurs preferentially over H_2O decomposition because Cl^- is a halogen ion. On the other hand, at the cathode, an electron is given to H_2O by a reducing process, and H_2 and hydroxide ion OH^- are produced. During the reaction, Cl_2 and H_2 are generated as gases. Although almost all produced Cl_2 is released outside as gas, part of it is dissolved into H_2O and produces HClO. Therefore, NaCl solution at the inlet of the reactor becomes gas and liquid two-phase flow during the fluid pass through the reactor.

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 ºC
Oxidation number	+1

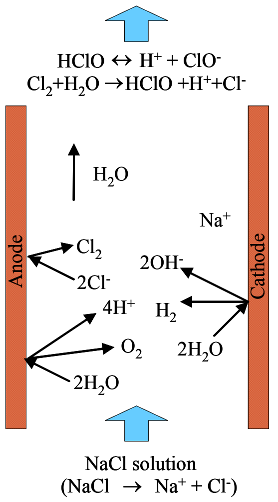


Figure 1. HClO production process in a flow-type electrolysis device without a membrane separator

Dissociation of NaCl

$$NaCl \rightarrow Na^+ + Cl^- \tag{1}$$

Anode:

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \tag{2}$$

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{3}$$

Cathode:

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \tag{4}$$

HClO production:

$$Cl_2 + H_2O \rightarrow HClO + H^+ + Cl^- \tag{5}$$

III. Experimental Apparatus and Procedure

3.1 Experimental apparatus

A flow-type reactor with narrow and parallel electrode plates lacking a barrier membrane between the plates is introduced in this experiment. Figure 2 shows the experimental apparatus employed in this experiment [12-14]. 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.

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 electrode plates, *d*, is changed using spacers of several thicknesses. The dimension 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 composed of a titanium plate coated with a thin layer of platinum is applied[19]. 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 plates fixed parallel to one another. 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.

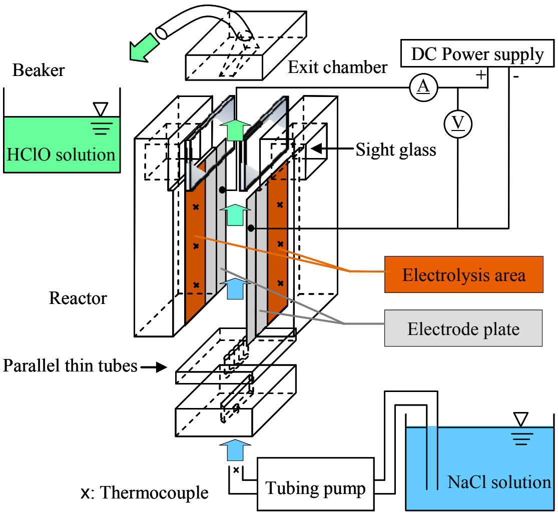


Figure 2. Experimental apparatus employed in this experiment

3.2 Experimental procedure

It is important to find the decomposition voltage of the NaCl solution. The chemical reaction occurs over this voltage. The voltage is theoretically estimated to be 2.17 Vdc[12]. Then the voltage is measured experimentally for each NaCl concentration, electrode plate interval and flow rate. 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 experiment. The decomposition voltage will increase slightly in the experiment because the electrolytic solution includes infinitesimal impurities even though purified water is used. At the beginning of the experiment, the fluid temperature is unstable as the electrode plates are heated because of Joules' law. After the steady state condition is achieved, HClO concentration, local temperatures, impressed voltage and current are measured.

IV. Experimental Results and Discussion

4.1 Index for high-efficiency production

The production efficiency of the available chlorine, η_{AC}, is defined by Eq. (6).

$$\eta_{AC} = \frac{N_{AC}}{N_e/2} \tag{6}$$

It expresses the ratio of the number of moles of produced available chlorine N_{AC} to estimated charge amount N_e for reaction, since the mole balances are expressed by Eqs. (3) and (5). Where, N_{AC} and N_e are expressed by Eqs. (7) and (8).

$$N_{AC} = \frac{C_{AC} d S}{M_{AC}} \times 10^{-3} \tag{7}$$

$$N_e = \frac{I_s S t}{F} \tag{8}$$

Here, C_{AC} is the concentration of available chlorine in [mg/l], d is the electrode plate interval in [mm], S is the reaction area in [m^2], M_{AC} is the molecular weight of HClO in [g/mol], I_s is the current density acting on the electrode in [A/m^2], t is the reaction time in [s], and F is the Faraday constant in [C/mol]. Reaction time t is estimated by the flow rate $\dot{Q}$ [ml/s] as expressed by Eq. (9).

$$t = \frac{Sd}{\dot{Q}} \times 10^{3} \tag{9}$$

Accordingly, the production efficiency of the available chlorine η_{AC} is estimated by Eq. (10).

$$\eta_{AC} = \frac{C_{AC}\dot{Q}}{\frac{1}{2} \cdot \frac{M_{AC} I_s S}{F}} \times 10^{-6} = \frac{\dot{m}_{AC}}{\frac{1}{2} \cdot \frac{M_{AC} I_s S}{F}} \times 10^{-3} \tag{10}$$

Here,

$$\dot{m}_{AC} = C_{AC}\dot{Q} \times 10^{-3} \tag{11}$$

is the production amount in [mg/s].

4.2 Dependence of NaCl concentration of solution flowing into a reactor

NaCl concentration in the inflow medium was varied to show the influence of Cl^- on the production efficiency. Table 2 and Fig. 3 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. However, production efficiency of the available chlorine does not increase despite the increase in the NaCl solution, as shown in Fig. 3. The efficiency has a maximum value around C_{NaCl} = 20,000 mg/l. The efficiency does not change remarkably around C_{NaCl} = 20,000 mg/l to 50,000 mg/l, and the C_{AC} have higher values around these range. The current density is subordinate to the NaCl concentration. Cl_2 should be solved into H_2O for HClO production. It will be more difficult to dissolve Cl_2 into H_2O as the Cl^- concentration becomes higher. Then, Cl_2 may flow out from the outlet section without reaction. As a result, the production efficiency of available chlorine production will become lower in higher C_{NaCl} concentrations.

4.3 Dependence of current density

Current density supplied to the electrode plate is varied. As an example of experimental results, the flow rate is fixed and the NaCl concentrations of the medium chosen here are C_{NaCl} = 30,000, 40,000 and 50,000 mg/l, because the efficiency does not change remarkably at around these ranges. It is effective to examine the effect of current density on the efficiency directly. In addition, higher efficiency with higher concentration production is achieved in these conditions. The experimental results are shown in Table 3 and Fig. 4. The production efficiency becomes low as the current density increases even though concentration of available chlorine increase. Since the mobility of Cl^- increases close to the electrode plates, Cl^- becomes insufficient in the solution. As Cl^- decreases in the vicinity of the electrode plates, H_2O is decomposed and Cl_2 dissolves in the solution at a lower rate.

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]	Concentration of available chlorine C_{AC} [mg/l]	Production efficiency η_{AC} [-]
1.0	10,000	182	720	0.160
	20,000	363	2,180	0.243
	30,000	545	3,190	0.237
	40,000	727	4,240	0.236
	50,000	909	4,910	0.219
3.0	10,000	182	610	0.136
	20,000	363	2,550	0.284
	30,000	545	4,060	0.301
	40,000	727	4,660	0.259
	50,000	909	6,040	0.269
5.0	10,000	182	680	0.150
	20,000	363	3,120	0.349
	30,000	545	4,290	0.319
	40,000	727	5,710	0.318
	50,000	909	6,840	0.305

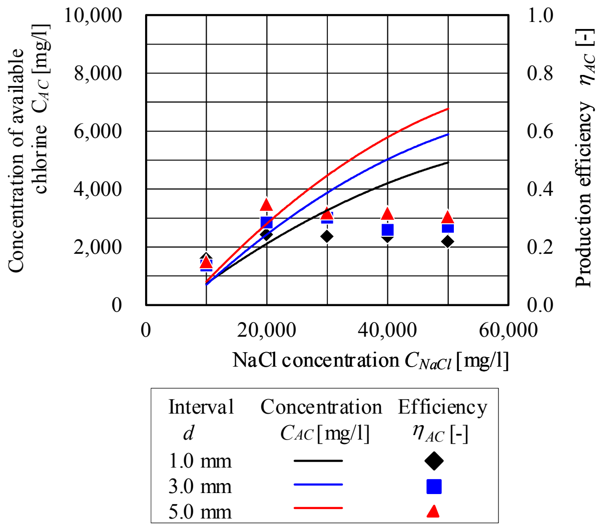


Figure 3. Production efficiency and concentration of available chlorine versus NaCl concentration ($\dot{Q}$ = 0.0551 ml/s)

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

Electrode interval d [mm]	Concentration of NaCl C_{NaCl} [mg/l]	Current density I_s [A/m^2]	Concentration of available chlorine C_{AC} [mg/l]	Production efficiency η_{AC} [-]
1.0	30,000	200	1,960	0.397
		400	3,260	0.330
		600	3,590	0.243
		800	4,060	0.205
	40,000	200	2,000	0.405
		400	3,490	0.353
		600	3,920	0.264
		800	4,590	0.232
	50,000	200	2,280	0.462
		400	3,380	0.342
		600	3,840	0.259
		800	4,460	0.226
3.0	30,000	200	1,390	0.281
		400	3,030	0.307
		600	3,790	0.256
		800	4,770	0.241
	40,000	200	2,700	0.547
		400	3,700	0.375
		600	5,210	0.352
		800	5,540	0.280
	50,000	200	3,080	0.624
		400	4,120	0.417
		600	5,180	0.350
		800	6,280	0.318
5.0	30,000	200	2,240	0.453
		400	3,960	0.401
		600	4,690	0.317
		800	5,200	0.263
	40,000	200	2,910	0.589
		400	4,700	0.476
		600	5,340	0.360
		800	6,450	0.326
	50,000	200	3,150	0.637
		400	4,540	0.460
		600	5,830	0.394
		800	6,840	0.346

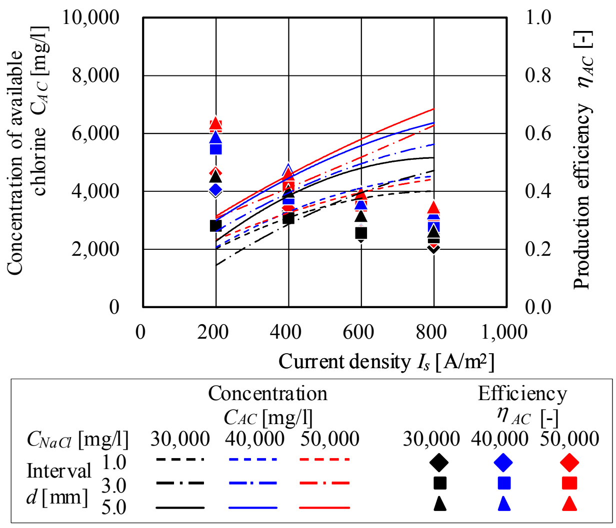


Figure 4. Production efficiency and concentration of available chlorine versus current density ($\dot{Q}$ = 0.0551 ml/s)

4.4 Dependence of solution flow rate

Flow rate of the NaCl solution is varied and the NaCl concentration is fixed as C_{NaCl} = 50,000 mg/l, for example. Current density is set at I_s = 600 and 800 A/m^2 in order to achieve a sufficient reaction. And if the I_s = 600 and 800 A/m^2 are chosen, the efficiency does not strongly depends on the C_{NaCl} and d, as shown in Fig. 4. Therefore, the effects of the flow rate on C_{AC} and η_{AC} can be evaluated easily. The results are shown in Table 4 and Fig. 5. The production efficiency becomes high as the flow rate increases even though the concentration of available chlorine C_{AC} decreases. Production efficiency means the ratio of the number of moles of produced available chlorine to estimated charge amount for reaction. The efficiency depends on the production amount $\dot{m}_{AC}$, that is, flow rate $\dot{Q}$, estimated from Eq. (10). Therefore, the solution should pass through the narrow parallel plates rapidly and reaction time becomes short. In this case, Cl$^-$ will react most efficiently.

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

Electrode interval d [mm]	Flow rate $\dot{Q}$ [ml/s]	Velocity u_{NaCl} [m/s]	Current density I_s [A/m^2]	Concentration of available chlorine C_{AC} [mg/l]	Production amount $\dot{m}_{AC}$ [mg/s]	Production efficiency η_{AC} [-]
1.0	0.0358	0.716x10^{-3}	600	4,620	0.165	0.202
	0.0551	1.102x10^{-3}		3,840	0.212	0.259
	0.0775	1.550x10^{-3}		3,800	0.295	0.361
	0.1140	2.280x10^{-3}		3,080	0.351	0.431
	0.0358	0.716x10^{-3}	800	5,220	0.187	0.172
	0.0551	1.102x10^{-3}		4,460	0.246	0.226
	0.0775	1.550x10^{-3}		4,630	0.359	0.329
	0.1140	2.280x10^{-3}		3,390	0.386	0.355
3.0	0.0358	0.239x10^{-3}	600	5,100	0.183	0.224
	0.0551	0.367x10^{-3}		5,180	0.285	0.350
	0.0775	0.517x10^{-3}		4,540	0.352	0.431
	0.1140	0.760x10^{-3}		4,010	0.457	0.561
	0.0358	0.239x10^{-3}	800	6,640	0.238	0.218
	0.0551	0.367x10^{-3}		6,280	0.346	0.318
	0.0775	0.517x10^{-3}		5,520	0.428	0.393
	0.1140	0.760x10^{-3}		4,870	0.555	0.511
5.0	0.0358	0.143x10^{-3}	600	6,070	0.217	0.266
	0.0551	0.220x10^{-3}		5,830	0.321	0.394
	0.0775	0.310x10^{-3}		5,220	0.405	0.495
	0.1140	0.456x10^{-3}		4,390	0.500	0.613
	0.0358	0.143x10^{-3}	800	7,030	0.252	0.231
	0.0551	0.220x10^{-3}		6,840	0.377	0.346
	0.0775	0.310x10^{-3}		6,010	0.466	0.428
	0.1140	0.456x10^{-3}		5,140	0.586	0.539

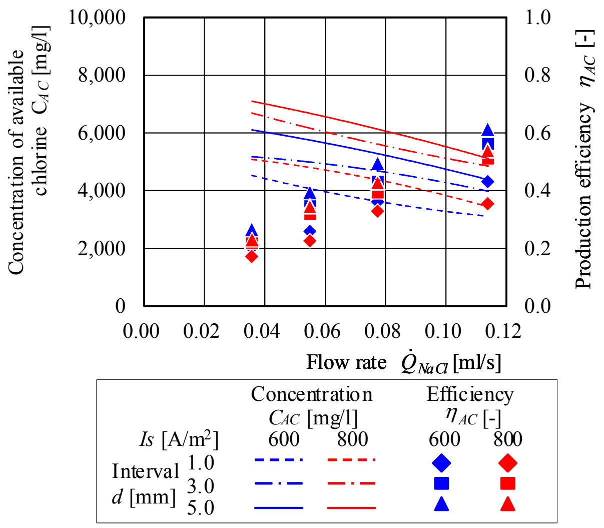


Figure 5. Production efficiency and concentration of available chlorine versus flow rate (C_{NaCl} = 50,000 mg/l)

4.5 Dependence of electrode plate interval

Gas and liquid two-phase flow in the narrow and parallel flat plates has very complex flow characteristics. The gas phase in the flow field may occupy the narrow space and prevent reactions. The experiments are conducted using the same conditions for the respective plate interval. Generally, the production efficiency becomes high as the plate interval becomes greater as shown in Figs. 3-5. Therefore, since the C_{AC} concentration increases as the plate interval becomes greater, the production efficiency is also considered to increase.

4.6 Dependence of flow velocity

From the above results, production efficiency η_{AC} increases as the NaCl concentration C_{NaCl} and plate interval d increase. However, η_{AC} decreases at higher current density I_s. Figure 6 shows that the relationship between η_{AC} and flow velocity u_{NaCl}. u_{NaCl} is derived from flow rate $\dot{Q}$ and cross section of flow path, also shown in Table 4. u_{NaCl} decreases as the plate interval increases for constant flow rate. η_{AC} increases as velocity decreases, and then gradually approaches the fixed value at higher velocity. Therefore, the solution should pass through the narrow reaction area rapidly like the result obtained from flow rate.

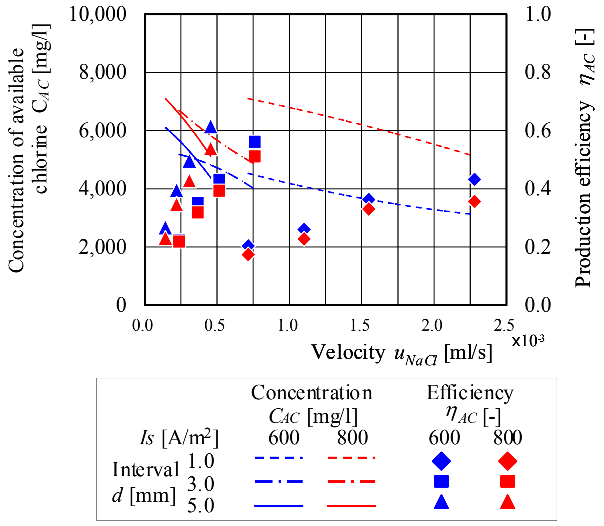


Figure 6. Production efficiency and concentration of available chlorine versus flow velocity (C_{NaCl} = 50,000 mg/l)

4.8 Energy consumption for high-efficiency production

Energy consumption, that is electricity consumption, is also important for the actual device design. Therefore, the molar production efficiency, ζ_{AC}, based on the input power and molar volume is defined as Eq. (11).

$$\zeta_{AC} = \frac{\frac{\dot{m}_{AC}}{M_{AC}} \times 2}{P} \times 10^{-3} \qquad (12)$$

Here, P is electricity supplied to the electrode plates in [W].

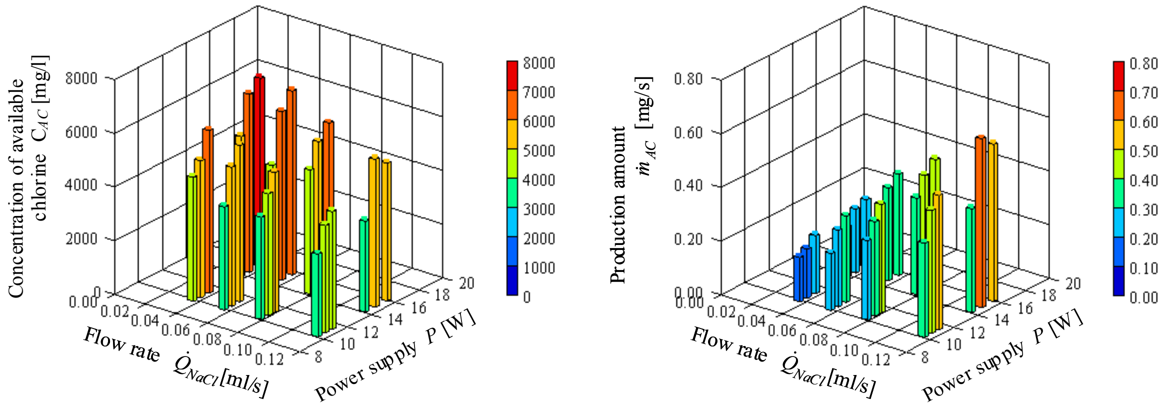
Table 5 and Fig. 7 show the results estimated by Eqs. (10)-(12). C_{AC} gradually approaches the maximum value, as the flow rate decreases and the power supply increases. The mobility of ions increases with the increase of current density, that is power supply increases, and it makes Cl^- active. On the other hand, production amount $\dot{m}_{AC}$ and production efficiency ζ_{AC} increase as the flow rate increases. Production amount $\dot{m}_{AC}$ increases as the input power P increases. In this case the mobility of ions becomes more active with input power increase like C_{AC}. However, η_{AC} and ζ_{AC} does not strongly depend on the input power. It depends on the flow rate. The solution should pass through the reaction area rapidly. This means that the flow rate plays an important role in higher production efficiency. Therefore, lower input power is desirable from an economic point of view.

Table 5. Relation of power supply and flow rate to C_{AC}, $\dot{m}_{AC}$ and ζ_{AC} (C_{NaCl} = 50,000 mg/l)

Flow rate $\dot{Q}$ [ml/s]	Current density I_s [A/m^2]	Power supply P [W]	Electrode interval d [mm]	Velocity u_{NaCl} [m/s]	Concentration of available chlorine C_{AC} [mg/l]	Production amount $\dot{m}_{AC}$ [mg/s]	Molar production efficiency ζ_{AC} [mol/(W·s)]
0.0358	600	9.9	1.0	7.16x10^{-4}	4,620	0.165	6.34x10^{-7}
		10.5	3.0	2.39x10^{-4}	5,100	0.183	6.60x10^{-7}
		11.2	5.0	1.43x10^{-4}	6,070	0.217	7.38x10^{-7}
	800	13.9	1.0	7.16x10^{-4}	5,220	0.187	5.13x10^{-7}
		14.6	3.0	2.39x10^{-4}	6,640	0.238	6.18x10^{-7}
		15.5	5.0	1.43x10^{-4}	7,030	0.252	6.19x10^{-7}
0.0551	600	10.0	1.0	1.10x10^{-3}	3,840	0.212	8.04x10^{-7}
		10.6	3.0	3.67x10^{-4}	5,180	0.285	1.03x10^{-6}
		11.3	5.0	2.20x10^{-4}	5,830	0.321	1.09x10^{-6}
	800	14.0	1.0	1.10x10^{-3}	4,460	0.246	6.67x10^{-7}
		14.8	3.0	3.67x10^{-4}	6,280	0.346	8.90x10^{-7}
		15.7	5.0	2.20x10^{-4}	6,840	0.377	9.12x10^{-7}
0.0775	600	10.2	1.0	1.55x10^{-3}	3,800	0.295	1.10x10^{-6}
		10.8	3.0	5.17x10^{-4}	4,540	0.352	1.24x10^{-6}
		11.3	5.0	3.10x10^{-4}	5,220	0.405	1.36x10^{-6}
	800	14.3	1.0	1.55x10^{-3}	4,630	0.359	9.53x10^{-7}
		15.0	3.0	5.17x10^{-4}	5,520	0.428	1.09x10^{-6}
		15.9	5.0	3.10x10^{-4}	6,010	0.466	1.11x10^{-6}
0.1141	600	10.2	1.0	2.28x10^{-3}	3,080	0.351	1.31x10^{-6}
		10.8	3.0	7.61x10^{-4}	4,010	0.458	1.61x10^{-6}
		11.4	5.0	4.56x10^{-4}	4,390	0.501	1.68x10^{-6}
	800	14.2	1.0	2.28x10^{-3}	3,390	0.387	1.03x10^{-6}
		15.0	3.0	7.61x10^{-4}	5,520	0.630	1.60x10^{-6}
		16.0	5.0	4.56x10^{-4}	5,140	0.586	1.39x10^{-6}

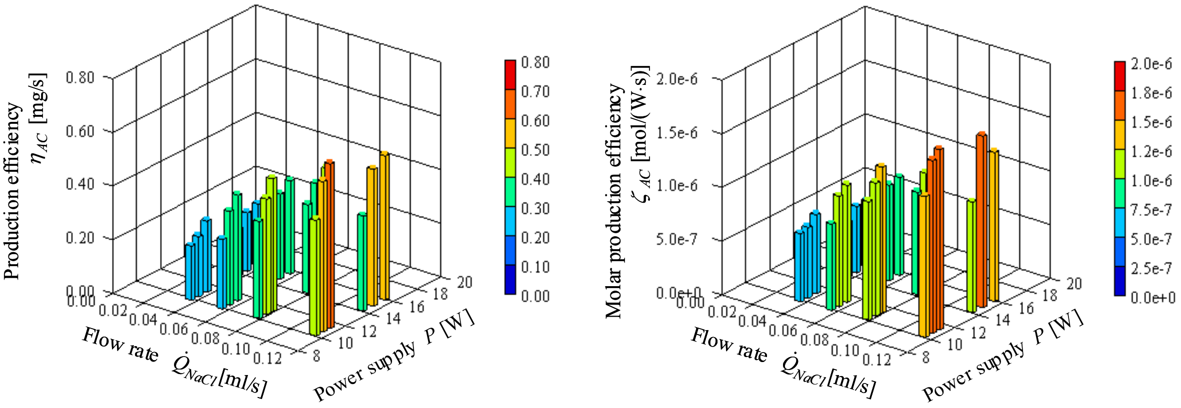
V. Conclusion

The ideal conditions for high efficiency hypochlorous acid water production were investigated experimentally using a narrow and parallel electrode plates without a barrier membrane. In this experiment, the NaCl concentration of the solution, current density supplied to the electrode plates, electrode plate interval and volume flow rate were all taken into account as experimental parameters. It was determined that production efficiency and molar production efficiency depend on the flow rate of the sodium chlorine solution, even though these are not strongly dependent on the input power. Higher concentration with higher production efficiency can be obtained at a higher flow rate. These results will be useful to produce chlorinated water, called hypochlorous acid water, with high concentration production, too.



(a) Concentration of available chlorine C_{AC}

(b) Production amount $\dot{m}_{AC}$



(c) Production efficiency η_{AC}

(d) Molar production efficiency ζ_{AC}

Figure 7. Relation of power supply and flow rate to C_{AC}, $\dot{m}_{AC}$, η_{AC} and ζ_{AC} ($C_{NaCl} = 50{,}000$ mg/l)

VI. Conclusion

The ideal conditions for high efficiency hypochlorous acid water production were investigated experimentally using a narrow and parallel electrode plates without a barrier membrane. In this experiment, the NaCl concentration of the solution, current density supplied to the electrode plates, electrode plate interval and volume flow rate were all taken into account as experimental parameters. It was determined that production efficiency and molar production efficiency depend on the flow rate of the sodium chlorine solution, even though these are not strongly dependent on the input power. Higher concentration with higher production efficiency can be obtained at a higher flow rate. These results will be useful to produce chlorinated water, called hypochlorous acid water, with high concentration production, too.

Nomenclature

C = Concentration [mg/l]
d = Electrode plate interval [mm]
F = Faraday constant (9.65×10^4 C/mol)
I = Current density [A/m^2]
K = Dissociation constant [mol/l]
M = Molecular weight [g/mol]
N = Number of moles [mol]
$\dot{m}$ = Production amount [mg/s]
P = Power supply [W]
$\dot{Q}$ = Volume flow rate [ml/s]

S = Reaction area [m^2]
t = Reaction time [s]
η = Production efficiency [-]
ζ = Molar production efficiency [mol/(W · s)]

Subscripts
AC = Available chlorine
e = electron
$HClO$ = hypochlorous acid
$NaCl$ = Sodium chloride
s = Electrode surface

References

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