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Removal of acetaldehyde in air using a wetted-wall corona discharge reactor

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Abstract

A vertical wetted-wall corona discharge reactor was used for removal of acetaldehyde in air. The reactor consists of a wire cathode sustained at the center of a cylindrical anode. Acetaldehyde laden air was fed either upward or downward through the wetted-wall reactor, in which water was circulated as a falling thin film on the inner wall of the anode. Ozone and short-lived species such as ions and radicals were generated in the reactor by gas corona. When some of these short-lived radicals drifted and reached the water film, reactive OH radical was produced in the water. Since gaseous acetaldehyde was readily absorbed into the water before the gas mixture entered the corona zone, decomposition of aqueous acetaldehyde by OH radical was considered as the main mechanism. O_3 oxidation did not play a significant role in the present condition. It was found that there are a minimum current and a maximum inlet concentration of gaseous acetaldehyde for highly effective decomposition of aqueous acetaldehyde and TOC, resulting in steady state operation. It was calculated that one electron removed approximately 13 molecules of acetaldehyde. In comparison with the deposition type, the wetted-wall type exhibited clearly higher removal efficiency and lower byproduct formation. In addition, the effect of gas flow direction was discussed.

Keywords: Corona discharge; Radicals; Gas purification; Water purification; Acetaldehyde

1. Introduction

Electrical discharge technique, for examples, dc corona discharge [1,2], pulse corona discharge [3,4], electron beam [5], and barrier discharge [6], has extensively been studied for gas purification. In dc corona discharge, low energy electrons are employed to remove toxic gases with high removal efficiency and low byproduct formation. The wetted-wall corona discharge reactor proposed by Sano et al. [2] can enhance the performance of the removal process. In this reactor, some of the negative ions produced by electron attachment are absorbed into a falling liquid film on the anode surface. This absorption of the ions improves the removal efficiency of soluble gas components. In addition, the anode self-cleaning action makes the wetted-wall reactor suitable for a long period of operation.

In fact, the wetted-wall corona discharge reactor has been used for water treatment [7]. Short-lived radicals, O, H, and OH, and ions, O^-, $O_2{}^-$, $O_3{}^-$, H^-, and OH^-, are produced in the gas phase by corona discharge [8–10]. Negative ions are naturally expected to reach the wetted-wall anode by Coulomb force. Ionic drift along the electric field is found to create an ion wind whose velocity is of several meters per second [11]. A portion of the uncharged radicals is expected to be carried to the anode by the ion wind. When the radicals and ions reach the water film surface and subsequently react with H_2O, aqueous OH radicals which possess a profoundly stronger oxidation power than ozone are produced [7,12,13]. The aqueous OH radical along with O_3 is expected to decompose aqueous organic compounds.

In the present work, a wetted-wall corona discharge reactor is used to purify acetaldehyde laden air. As one of the hazardous chemicals which are exhausted from diesel engines and waste incinerators as well as from building materials that cause the so-called sick house syndrome, acetaldehyde is chosen for a target compound because of its human and environmental effects. The treated gas and circulating water used for the wetted-wall are sampled for

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Nomenclature

$C_{i,g}$	influent concentration of gaseous acetaldehyde (mol-ppm)
$C_{o,g}$	effluent concentration of gaseous acetaldehyde (mol-ppm)
C_w	concentration of aqueous acetaldehyde (mol-ppm) or (mg L^{-1})
e	elemental charge (1.602×10^{-19} C)
I	corona discharge current (mA)
J	energetic efficiency (mol J^{-1})
N	Avogadro's number (6.02×10^{23} molecules mol^{-1})
N_e	the number of electrons produced by the corona discharge per unit time (mol s^{-1})
Q_g	gas flow rate (cm^3 min^{-1})
Q_l	water circulation rate (cm^3 min^{-1})
q_g	gas mole flow rate (mol s^{-1})
Re	Reynolds number (–)
t	time (s)
TOC	total organic carbon (mg L^{-1})
V	applied voltage (kV)
W	water volume (cm^3)
w	mole of water (mol)

Greek letters

ψ	removal extent (–)
η_e	electron efficiency (–)

chemical analysis to evaluate the performance of the reactor. Then the removal mechanism is proposed and discussed based on the experimental results.

2. Experimental

The experimental set-up is shown in Fig. 1. A stainless steel (SUS) wire cathode (0.34 mm diameter) is sustained along the center of a SUS cylindrical anode (34 mm inner diameter; 200 mm length). A dc high voltage of −8 to −13 kV is applied on the cathode to generate corona discharge. The effective length of axial corona discharge is fixed at 140 mm for all experiments. Standard acetaldehyde gas balanced with N_2 is mixed with O_2 and N_2 from gas cylinders and then fed either upward or downward through the reactor in one pass, whereas water, originally deionized, is circulated as a falling film on the inner surface of the grounded anode. The gas flow rate, water flow rate and volume of circulating water are 100 cm^3 min^{-1}, 1400 cm^3 min^{-1} and 1000 cm^3, respectively. The water film thickness, Reynolds numbers (Re) of water and gas streams in the absence of corona discharge are calculated as 0.4 mm, 870, and 4, respectively [14]. The temperature of the water is controlled at 10 °C by passing it through a heat exchanger unit.

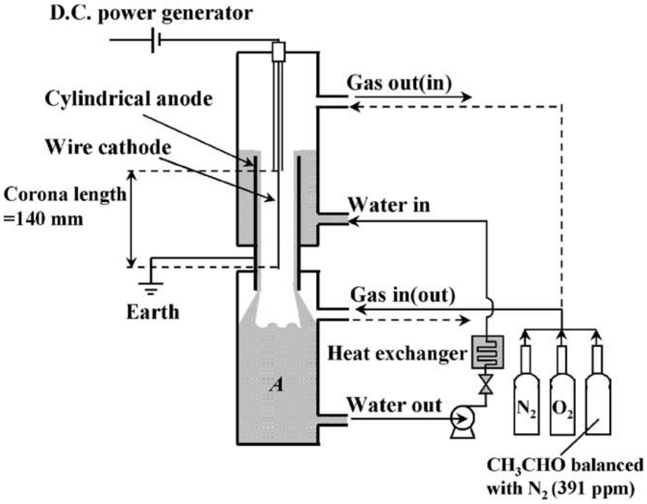

Fig. 1. Experimental set-up.

To determine the contribution of ozone to the decomposition of aqueous acetaldehyde, gaseous ozone is injected into the aqueous acetaldehyde mixture. O_2 (21 cm^3 min^{-1}) is fed to an ozone generator to produce ozone at approximately 10,000 ppm, and the ozone laden gas is bubbled into the water containing acetaldehyde around the bottom part of the wetted-wall reactor (A in Fig. 1). The aqueous acetaldehyde mixture is prepared by passing the N_2-balanced gaseous acetaldehyde (250 ppm by mole at 79 cm^3 min^{-1}) through the wetted-wall reactor. With these gas flow rates, the concentrations of all gas components at reactor inlet become similar to those found in the experiments with corona discharge.

The treated gas was analyzed by an FID gas chromatograph (Shimadzu, GC-9A) and ozone concentration was measured by the iodometric method. Meanwhile, the circulating water was analyzed by the FID gas chromatograph and a high performance liquid chromatograph (HPLC) with a UV-VIS detector (Shimadzu, SPD-10AVP). Total organic carbon (TOC) and pH were respectively monitored by a TOC analyzer (Shimadzu, TOC-5000) and a pH meter (Horiba, pH meter F-22).

3. Results and discussion

3.1. Gas purification by wetted-wall reactor

Fig. 2 shows the outlet concentration of gaseous acetaldehyde $C_{o,g}$ during the discharge operation. The influence of the corona discharge current, varied from 0.02 to 0.3 mA, on the purification of acetaldehyde laden air is investigated here. Even in the absence of corona discharge, $C_{o,g}$ initially drops to essentially zero because acetaldehyde is readily absorbed into fresh water. However, $C_{o,g}$ significantly rises with time as the water is incessantly contaminated with acetaldehyde.

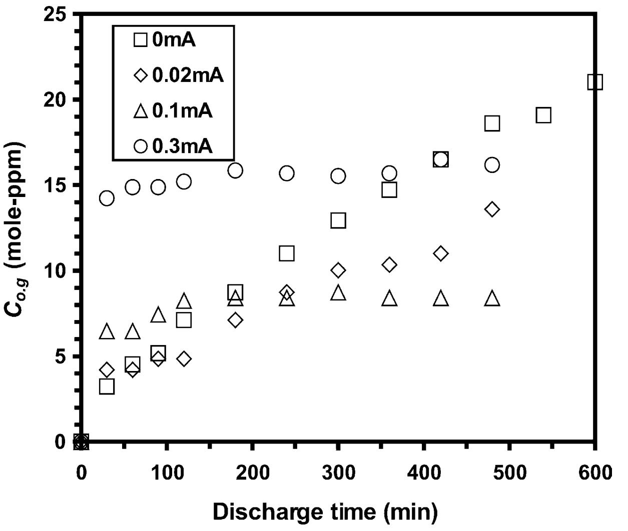

Fig. 2. Concentration of acetaldehyde in treated gas during corona discharge operation. $C_{i,g} = 200$ mol-ppm, $Q_g = 100\,cm^3\,min^{-1}$, $Q_l = 1400\,cm^3\,min^{-1}$, $W = 1000\,cm^3$, gas flow direction: upflow.

When the corona discharge is generated, for instance at the discharge current of 0.1 mA, $C_{o,g}$ continues to gradually increase at early stage but levels off at around 8 mol-ppm after 200 min. However, if the applied discharge current is lower than 0.1 mA, the effect of corona discharge becomes weak and $C_{o,g}$ does not level off at a constant value. It should be noted that the decomposition of the feed acetaldehyde takes place mainly in the aqueous phase as discussed in the latter section.

When the discharge current increases, the asymptotic value of $C_{o,g}$ is found to slightly increase. The result may be attributed to corona induced turbulence in the gas stream flowing inside the reactor, which affects the residence time distribution (RTD) of the gas in the reaction zone. When the ion wind induces turbulence, the RTD in the reactor becomes closer to that of a continuous stirred tank reactor (CSTR), which possesses a much broader RTD than a laminar flow model, or plug flow reactor (PFR), with the same mean residence time. This phenomenon is known to result in a lower conversion efficiency [14] and is the reason why a higher *I* yields a lower acetaldehyde removal efficiency.

When corona discharge is generated, electrons are emitted from the wire cathode and then drift to the anode. In the small region of high strength electric field adjacent to the wire cathode, electron impacts on gas molecules produce short-lived radicals such as O as shown in Eq. (1) [9,10]. During the drift process, low-energy electrons collide with gas molecules and electron attachment reactions are expected to take place, as shown in Eq. (2). In addition, ozone is produced, as in Eq. (3) [8].

$$O_2 + e^- \rightarrow O + O + e^- \quad (1)$$

$$O_2 + e^- \rightarrow O^- + O \quad (2)$$

$$O_2 + O \rightarrow O_3 \quad (3)$$

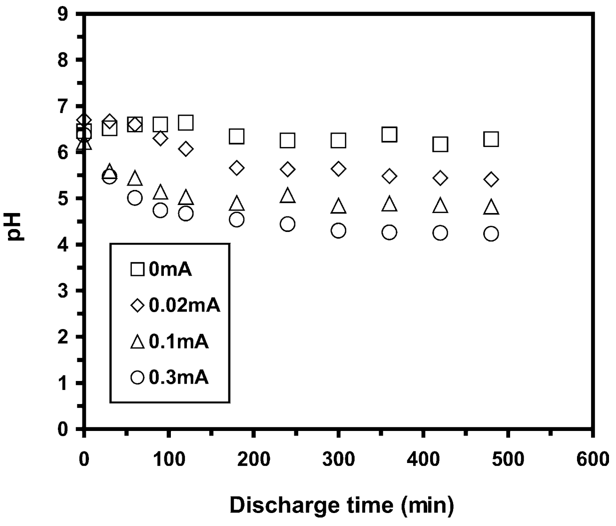

Fig. 3. pH of water against discharge time. $C_{i,g} = 200$ mol-ppm, $Q_g = 100\,cm^3\,min^{-1}$, $Q_l = 1400\,cm^3\,min^{-1}$, $W = 1000\,cm^3$, gas flow direction: upflow.

In the reactor, the gas stream also contains water vapor. Thus electron attachment to H_2O vapor should produce OH^- and H^-, whereas dissociation of H_2O should produce OH and H radicals [8,9]. Acetaldehyde not absorbed into the aqueous phase would be removed by the reactions with ions, radicals, and ozone generated by corona reactions in the gas phase as shown in Eqs. (4)–(8) [15–17].

$$OH + CH_3CHO \rightarrow CH_3CO + H_2O \quad (4)$$

$$HO_2 + CH_3CHO \rightarrow \text{product} \quad (5)$$

$$O + CH_3CHO \rightarrow \text{product} \quad (6)$$

$$O_3 + CH_3CHO \rightarrow \text{product} \quad (7)$$

$$O^- + CH_3CHO \rightarrow O^-[CH_3CHO]_m \quad (8)$$

Since the observed values of the $C_{o,g}$ shown in Fig. 2 depend mostly on the absorptivity of the gaseous acetaldehyde into the circulating water, $C_{o,g}$ is thought to depend on the pH of the water during the operation. In Fig. 3, the Ph values of the water during the discharge operation are plotted. Based on a starting pH value of around 6–7, they gradually decrease in around 200 min before stabilizing at around pH 4.5–5.5. This may be ascribed to the fact that HNO_3 is produced during the discharge operation [7,12]. It should be noted that the final value and decreasing rate of the pH depend on the applied corona discharge current. To investigate the influence of pH on the absorption rate, the initial pH of the circulating water was varied from 4 to 6.5 by adding HNO_3. In the absence of discharge current, acetaldehyde laden air is passed through the wetted-wall reactor and $C_{o,g}$ is measured at adequate time intervals. Influence of pH on the absorptivity of acetaldehyde is shown in Fig. 4. In these experiments, the observed difference in the acetaldehyde absorption rate is negligible in this pH range. Therefore it

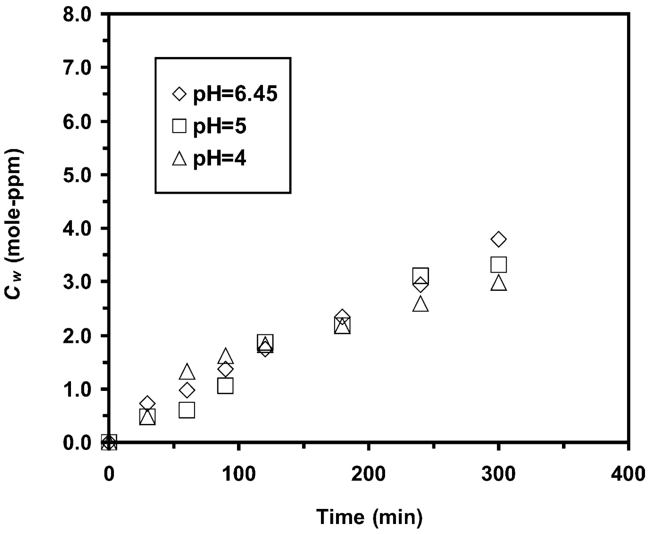

Fig. 4. Influence of pH on absorptivity of acetaldehyde. $C_{i,g} = 200$ mol-ppm, $Q_g = 100\,cm^3\,min^{-1}$, $Q_l = 1400\,cm^3\,min^{-1}$, $W = 1000\,cm^3$, gas flow direction: upflow.

may be considered that the absorptivity of acetaldehyde is not significantly affected by the pH of the water during the operation.

3.2. Analysis of the circulating water

The reaction scheme of production of OH radical is shown in Eqs. (9)–(11) [18]. The OH radical along with O_2H and ozone are expected to contribute to the decomposition of acetaldehyde in water as shown in Eq. (12) [18–20].

$$O_{(gas)}{}^{-} + H_2O \rightarrow OH_{(aq)}{}^{-} + OH_{(aq)} \qquad (9)$$

$$O_{2(gas)}{}^{-} + H_2O \rightarrow OH_{(aq)}{}^{-} + O_2H_{(aq)} \qquad (10)$$

$$O_{(gas)} + H_2O \rightarrow OH_{(aq)} + OH_{(aq)} \qquad (11)$$

$$OH_{(aq)}{}^{-}, O_2H_{(aq)}, O_{3(aq)} + CH_3CHO_{(aq)} \rightarrow \text{product} \qquad (12)$$

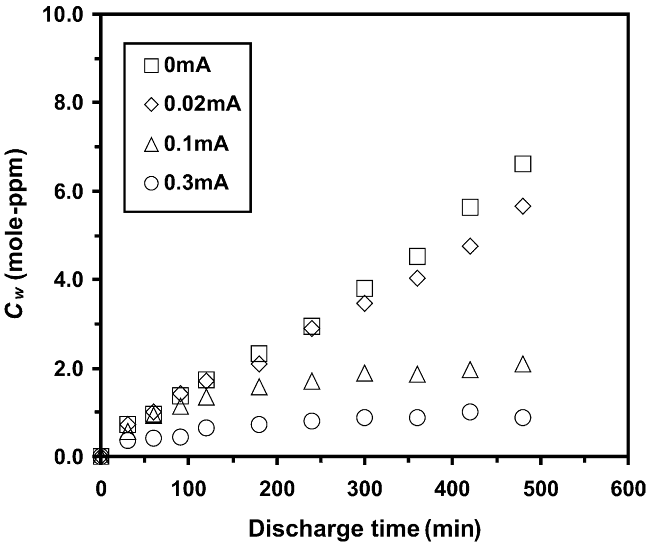

Fig. 5. Concentration of acetaldehyde in circulating water during corona discharge operation. $C_{i,g} = 200$ mol-ppm, $Q_g = 100\,cm^3\,min^{-1}$, $Q_l = 1400\,cm^3\,min^{-1}$, $W = 1000\,cm^3$, gas flow direction: upflow.

In Fig. 5, the concentrations of aqueous acetaldehyde C_w at various discharge currents are depicted against corona-discharge time. As expected, C_w increases with time in the absence of corona discharge. When corona discharge is generated, the increase in C_w is retarded by its decomposition. The rate of decomposition of the aqueous acetaldehyde becomes greater as I increases. It may be ascribed to the fact that, when I increases, more radicals, ions, and ozone are produced by the higher density of electrons. Similar to the effluent concentration of gaseous acetaldehyde shown in Fig. 2, C_w becomes stabilized after about 200 min at 0.1 and 0.3 mA, but keeps increasing at 0.02 mA. These results indicate that a current higher than 0.1 mA is required to stop the accumulation of the aqueous acetaldehyde. In other words, if the discharge current is too low, stable purification of acetaldehyde laden air cannot be achieved because the unceasing accumulation of the aqueous acetaldehyde inhibits the gas-phase removal of acetaldehyde by gas absorption. It should be noted that, according to overall mass balance of acetaldehyde in the blank test, the deposit of acetaldehyde on the wall of anode is negligible.

Fig. 6a shows the concentrations of liquid-phase TOC and of aqueous acetaldehyde during the discharge operation for

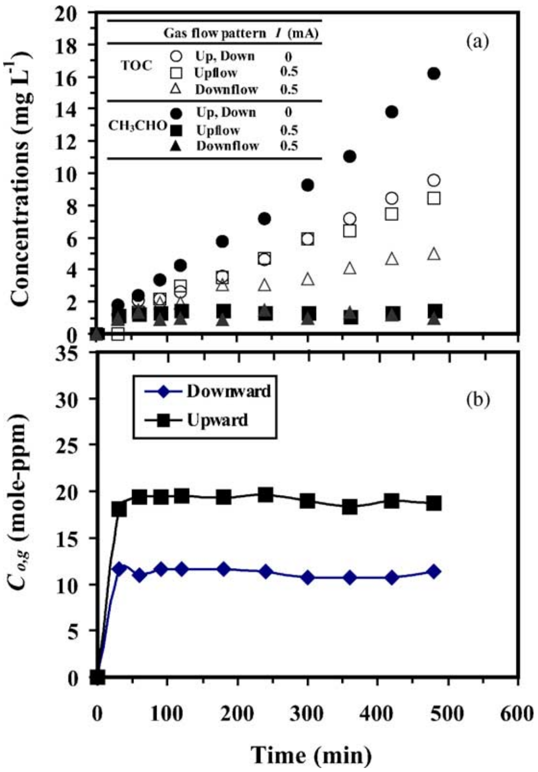

Fig. 6. Influence of gas flow directions on TOC and aqueous acetaldehyde concentration. $C_{i,g} = 200$ mol-ppm, $Q_g = 100\,cm^3\,min^{-1}$, $Q_l = 1400\,cm^3\,min^{-1}$.

the cases of both upflow and downflow. The current 0.5 mA was used here, which is the optimized value to degrade stable organic compounds in water under the present dimension of the reactor, as previously reported [7]. When corona discharge is not generated, both TOC and acetaldehyde concentrations increase with time due to gas absorption. As expected, their values in either case of upflow and downflow are essentially the same. When corona discharge is generated, the increases of TOC and acetaldehyde are significantly attenuated. However, though C_w becomes stabilized, TOC continues to gradually increase. This result indicates that some byproducts which are more stable than acetaldehyde are accumulated in the water. To identify the byproducts, the circulating water is analyzed with the GC and HPLC. The detected byproduct has the same retention time with acetic acid whereas the presence of formaldehyde is detected but rather negligible. Acetic acid is a common byproduct of the oxidation treatment of aqueous acetaldehyde [21].

The kinetic rate constants of acetaldehyde and acetic acid toward ozone and radical OH in water are listed in Table 1. Based on the rate constants, it is considered that acetic acid is more stable than acetaldehyde. To back up our assertion, we carried out experiment on the decomposition of aqueous acetic acid. One liter of aqueous acetic acid (50 mg L^{-1}) was decomposed in stationary air in the wetted-wall reactor with $I = 0.5$ mA and $Q_l = 1400\,\text{cm}^3\,\text{min}^{-1}$. The result shows that about 20% of TOC is slowly degraded in 8 h. Since O_3 is produced up to 2000–2500 ppm in our reactor, the contribution of O_3 toward the decomposition of aqueous acetaldehyde was also evaluated experimentally, as explained in the experimental section. The result explores that decomposition of aqueous acetaldehyde by O_3 does not play a significant role in the present condition.

To avoid the non-stop accumulation of TOC in the circulating water, the influent concentration of gaseous acetaldehyde $C_{i,g}$ was intentionally decreased. Fig. 7 shows the change in TOC concentration at various $C_{i,g}$. Here the current is maintained at 0.5 mA and the gas flow direction is upward. The result shows that, when $C_{i,g}$ is decreased, the increasing rate of TOC is reduced. The value of TOC becomes stabilized when $C_{i,g}$ is 50 mol-ppm. To maintain a stable TOC level, the generation rates of the aqueous acetaldehyde and its byproducts must equal their decomposition rates. It should be noted that the maximum $C_{i,g}$ to avoid the non-stop accumulation of TOC would depend on the experimental conditions such as corona current, gas flow rate and flow direction, and water flow rate.

Table 1
Kinetic rate constants of decomposition of aqueous acetaldehyde and aqueous acetic acid toward ozone (k_{O_3}) [22] and OH radical (k_{OH}) [20]

Substance	k_{O_3} (M^{-1} s^{-1}) (non-dissociated form)	k_{OH} (M^{-1} s^{-1})
Acetaldehyde	1.5	7.3×10^8
Acetic acid	3×10^{-5}	1.6×10^7

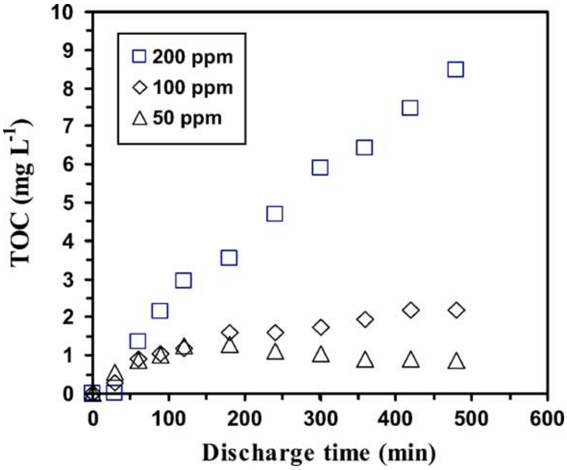

Fig. 7. TOC concentration in water against discharge time at various inlet concentrations of gaseous acetaldehyde. $I = 0.5$ mA, $C_{i,g} = 200$ mol-ppm, $Q_g = 100\,\text{cm}^3\,\text{min}^{-1}$, $Q_l = 1400\,\text{cm}^3\,\text{min}^{-1}$, gas flow direction: upflow.

3.3. Influence of gas flow direction

According to Fig. 6a, the downflow shows obviously higher decomposition rate of aqueous acetaldehyde and TOC than the upflow. To explain this result, the concentration profile of gaseous acetaldehyde along the reactor length in the absence of corona discharge was investigated experimentally, as shown in Fig. 8. Apparently significantly more unabsorbed gaseous acetaldehyde remains to penetrate deeper into the corona zone in the case of downflow than upflow due to the present structure of the reactor. Therefore, when gas flow direction is upward, the removal of acetaldehyde is dominated by the reaction in the water phase because most gaseous acetaldehyde is absorbed into water before it reaches the corona zone. On the other hand, when the gas flow direction is downward, the removal reactions also take place significantly in the gas phase. It is logical to consider that the combined decomposition of gaseous and aqueous acetaldehyde improves the purification performance of the reactor.

In Fig. 8, it should be noted that the concentration profile of gaseous acetaldehyde in the reactor was obtained in a condition without corona discharge. When corona discharge occurs, the gas-phase zone which is disturbed by ion-wind should be expanded to outside the corona zone (ion drift zone) because of convective effect. Therefore, it should be reminded that the excessive discharge current causing significant gas turbulence can raise the outlet concentration of acetaldehyde as explained in the previous section although most of the acetaldehyde can be absorbed in water before it reaches the corona zone.

O_3 is known to be produced from O radical and O_2 in the gas corona zone. If reaction of O with acetaldehyde does take place, then the observed O_3 concentration should be lowered. According to the ozone analysis, the effluent concentrations of ozone in the cases of upflow and downflow

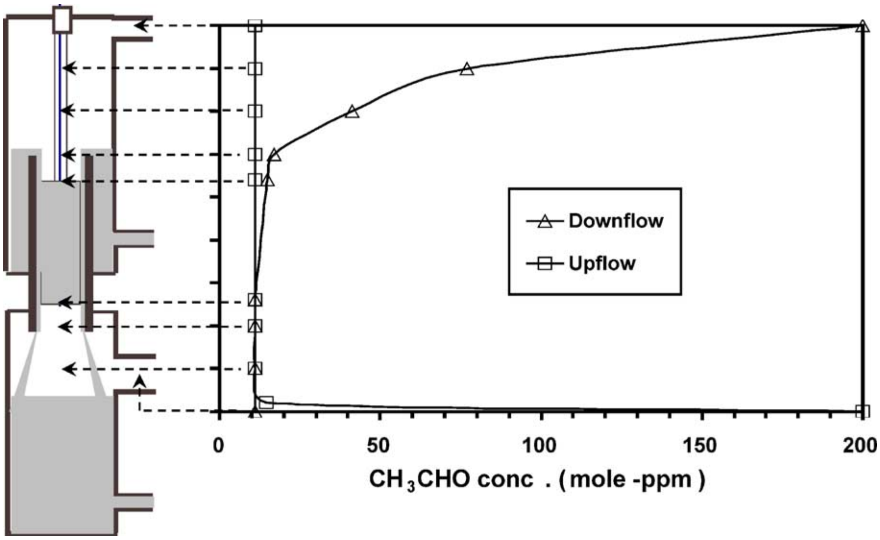

Fig. 8. Acetaldehyde concentration profile in gas stream along the reactor. $I = 0\,\text{mA}$, $C_{\text{i,g}} = 200$ mol-ppm, $Q_{\text{g}} = 100\,\text{cm}^3\,\text{min}^{-1}$, $Q_{\text{l}} = 1400\,\text{cm}^3\,\text{min}^{-1}$, $W = 1000\,\text{cm}^3$, time = 4 h.

are 2.2×10^3 and 1.6×10^3 ppm, respectively. The results of gas-phase O_3 analysis confirm that the reaction between O radical and gaseous acetaldehyde takes place to a significant extent when the gas is fed downward.

As seen in Fig. 6b, the downflow shows higher removal extent of gaseous acetaldehyde than the upflow. This is because the absorption of gaseous acetaldehyde into the water can be accelerated by the enhanced water purification when the gas flow direction is downward. In addition, the gas reactions should improve the removal of acetaldehyde.

3.4. Comparison of removal extent and byproduct formation between wetted-wall type and deposition type

The removal extent of acetaldehyde ψ, defined by Eq. (13), is plotted against corona discharge current, I, in Fig. 9.

$$\psi = \frac{C_{\text{i,g}} - C_{\text{o,g}}}{C_{\text{i,g}}} \tag{13}$$

where $C_{\text{i,g}}$ is the influent concentration of gaseous acetaldehyde and $C_{\text{o,g}}$ is the time-averaged effluent concentration of acetaldehyde during steady state operation. The deposition type reactor [1] used here consists of a cylindrical anode and a coaxial wire cathode of the same dimensions as the wetted-wall reactor. Since this reactor does not have the wetted-wall, acetaldehyde is removed from the gas stream solely by gas-phase corona discharge reaction. Compared with the deposition type, the wetted-wall type exhibits a clearly higher removal extent when the discharge current is low. This is because the absorption of gaseous acetaldehyde enhanced the removal extent. When gas downflow is applied, the removal extent in the wetted-wall reactor is higher than that of deposition reactor at corona currents less than 0.5 mA.

To investigate the influence of water vapor on the removal of acetaldehyde, we conducted experiments on the removal of gaseous acetaldehyde in the deposition-type corona discharge reactor in the absence and in presence of water vapor. Our results confirm that water vapor enhances the removal efficiency. When water vapor exists in the corona zone, related gaseous radicals and ions are produced as explained in Section 3.1. These radicals and ions are shown experimentally to contribute to the removal reaction of acetaldehyde.

Fig. 10 compares the GC chromatograms of gas analysis between the wetted-wall type and deposition type corona discharge reactors. Chromatograms a, b, c, and d stand, respectively, for the influent and effluent gas stream of the

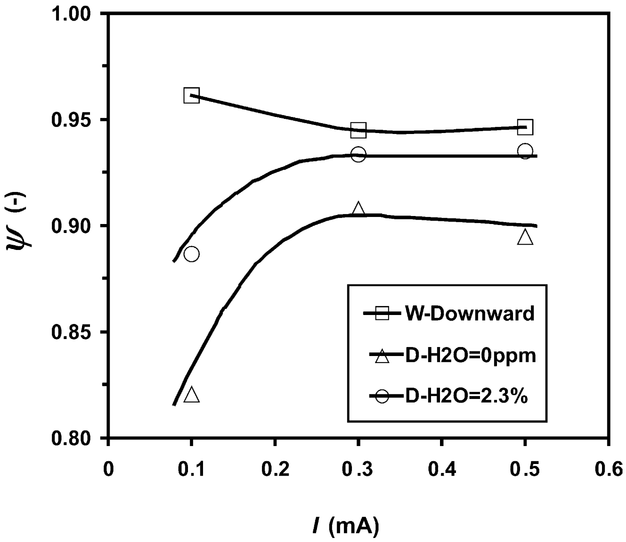

Fig. 9. Removal extent of gaseous acetaldehyde against corona discharge current. W: wetted-wall type, D: deposition type, $C_{\text{i,g}} = 200$ mol-ppm, $Q_{\text{g}} = 100\,\text{cm}^3\,\text{min}^{-1}$.

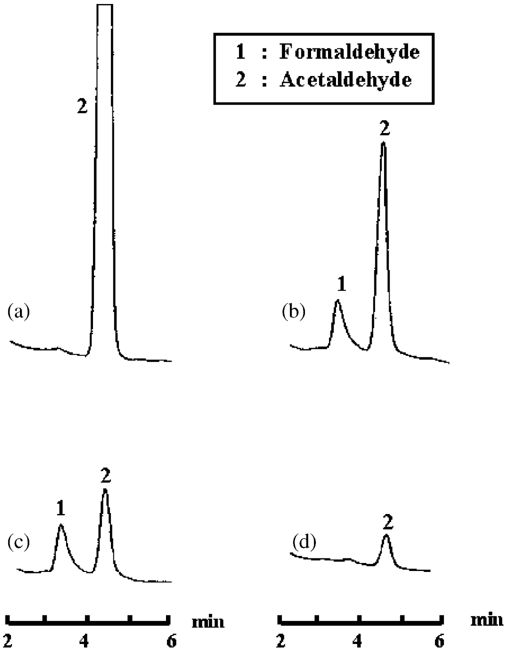

Fig. 10. GC chromatograms of gas analysis: (a) influent gas stream, (b) effluent gas streams of the deposition-type reactor in the absence of water vapor, (c) the effluent stream of the deposition type in the presence of 2.3% water vapor, and (d) the effluent stream of the wetted-wall reactor. $I = 0.1\,\text{mA}$, $C_{i,g} = 200\,\text{mol-ppm}$.

deposition-type reactor in the absence of water vapor, the effluent gas stream of the deposition-type reactor in the presence of 2.3% water vapor, and the effluent gas stream of the wetted-wall reactor. The only significant detected byproduct from the deposition type reactor has the same retention time as formaldehyde, whereas other byproducts are negligible. On the other hand, the formaldehyde which is detected in the deposition type reactor becomes negligible in the wetted-wall type. It should be noted that, though highly soluble, formaldehyde is not detected in the water after the corona discharge operation. This result may be ascribed to the following. In the deposition reactor, removal reactions of acetaldehyde take place in the gas phase, which produce formaldehyde as a byproduct. In contrast, formaldehyde is not detected significantly in both the gas and water phases in the wetted-wall reactor. This suggests that the main decomposition reaction which takes place in the water phase does not produce formaldehyde. Instead, it produces mainly acetic acid as byproduct.

3.5. Energy utilization: electron efficiency and energetic efficiency

The electron efficiency η_e and energetic efficiency J, defined by Eqs. (14) and (15), are calculated to evaluate the removal efficiency in the wetted-wall reactor.

$$\eta_e = \left(q_g(C_{i,g} - C_{o,g}) - \frac{w\,dC_w}{dt}\right)\frac{N}{N_e} \tag{14}$$

$$J = \frac{q_g(C_{i,g} - C_{o,g}) - (w\,dC_w/dt)}{IV} \tag{15}$$

where q_g is the total gas mole flow rate; $C_{i,g}$, inlet concentration of gaseous acetaldehyde; $C_{o,g}$, outlet concentration of gaseous acetaldehyde; w, mole of water; C_w, concentration of aqueous acetaldehyde; t, discharge time; N, Avogadro's number; N_e, the number of electrons produced by corona discharge per unit time; I, discharge current; and V, applied voltage. N_e is obtained from the discharge current as in Eq. (16)

$$N_e = \frac{I}{e} \tag{16}$$

where e is the elementary charge. η_e and J are based on the combined decomposition of gaseous and aqueous acetaldehyde by corona discharge. η_e represents the average number of acetaldehyde molecules removed by one electron, and J represents how many moles of acetaldehyde are degraded by unit energy. The values of η_e and J obtained at steady state when $dC_w/dt = 0$ are shown in Table 2. Obviously the wetted-wall type attains a higher η_e and J than the deposition one, revealing that the wetted-wall reactor is more promising for acetaldehyde removal.

Table 2
The electron efficiency η_e and energetic efficiency J of removal of acetaldehyde in air at steady state

Reactor type	η_e (−)	J ($10^{-9}\,\text{mol J}^{-1}$)
Dry deposition	11.0	12.1
Wetted-wall (upflow)	12.8	14.0
Wetted-wall (downflow)	12.9	14.0

$C_{i,g} = 200\,\text{mol-ppm}$, $I = 0.1\,\text{mA}$, $Q_g = 100\,\text{cm}^3\,\text{min}^{-1}$, $Q_l = 1400\,\text{cm}^3\,\text{min}^{-1}$, $W = 1000\,\text{cm}^3$.

Compared with other high voltage discharge systems with our results, some reports showing higher energetic efficiencies can be found. For example, Mizuno et al. shows $1.87 \times 10^{-7}\,\text{mol J}^{-1}$ using non-thermal pulsed plasma combining with TiO_2 [23]. However, it should be noted that the experimental condition used in their group is different from our work. For example, the inlet concentration used in their work is 1 ppm, whereas the energetic efficiency in our work is obtained with the inlet concentration 200 ppm. If extremely low concentration is used, the energy requirement is expected to be decreased since the discharge current and applied voltage can be decreased to lower values. In addition, the absolute efficiency can be further improved when some operational parameters are changed, for example the absorptivity of target components, pH in water, and discharge patterns such as non-thermal plasma. The advantage of our work is that there are no significant gaseous byproducts in treated gas. At this stage, rather than pursuing the highest removal efficiency among conventional methods, it is important to notice that this study proposes the potential ability of the wetted-wall discharge reactor for simultaneous purification of gas and water to degrade organic compounds in both phases.

4. Conclusions

A wetted-wall corona discharge reactor was applied to purify acetaldehyde laden air. It was elucidated that acetaldehyde is readily absorbed into the circulating water before the gas stream enters the corona zone and the aqueous acetaldehyde can effectively be decomposed by this reactor. The TOC degradation is slower than the decomposition of aqueous acetaldehyde since acetic acid is produced as the final byproduct, which is more stable than aqueous acetaldehyde. It was found that there is a minimum current around at 0.1 mA and a maximum influent concentration of gaseous acetaldehyde around at 50 mol-ppm for effective decomposition of aqueous TOC. An excessive current causes a slight decrease in the removal extent because the induced gaseous turbulence broadens the residence time distribution and reduces the effectiveness of gas-phase corona reactions. As for the effect of gas flow direction, the downflow yielded a higher removal efficiency and faster TOC decomposition rate than the upflow. It was evaluated that approximately 13 molecules of acetaldehyde are removed by one electron, and 7×10^7 J are required to remove 1 mol of acetaldehyde. In comparison with the deposition type, the wetted-wall type exhibits a clearly higher removal extent and less byproduct formation when the discharge current is low. The optimized discharge current is 0.1 mA for the removal of gaseous acetaldehyde and 0.5 mA for water treatment. The recommendable current lies in the range of 0.1–0.5 mA, though it depends on the individual purposes.

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References

[1] H. Tamon, H. Mizota, N. Sano, S. Shulze, M. Okazaki, New concept of gas purification by electron attachment, AIChE J. 41 (1995) 1701–1711.

[2] N. Sano, T. Nagamoto, H. Tamon, M. Okazaki, Removal of iodine and methyl iodide in gas by wetted-wall reactor base on selective electron attachment, J. Chem. Eng. Japan 29 (1996) 59–64.

[3] S. Masuda, Y. Wu, T. Urabe, Y. Ono, Pulse corona induced plasma chemical process for $DeNO_x$, $DeSO_x$, and mercury vapor control of combustion gas, in: Proceeding of the International Conference on Electrostatic Precipitation, Pavoda, Italy, 1987, pp. 667–676.

[4] T. Yamamoto, K. Ramanathan, P.A. Lawless, D.S. Ensor, J.R. Newsome, N. Planks, G.H. Ramsey, Control of volatile organic compounds by an ac energized ferroelectric pellet reactor and a pulsed corona reactor, IEEE Trans. Ind. Appl. 28 (1992) 528–534.

[5] K. Kawamura, Simultaneous removal of NO_x and SO_x by electron beam, Kagaku Kogaku. 53 (1989) 820–821.

[6] C.R. Mclarnon, U.K. Mathur, Nitrogen oxide decomposition by barrier discharge, Ind. Eng. Chem. Res. 39 (2000) 2779–2787.

[7] N. Sano, D. Yamamoto, T. Kanki, A. Toyoda, Decomposition of phenol in water by cylindrical wetted wall reactor using direct contact of gas corona discharge, Ind. Eng. Chem. Res. 42 (2003) 5423–5428.

[8] J.L. Moruzzi, A.V. Phelps, Survey of negative-ion-molecule reactions in O_2, CO_2, H_2O, CO, and mixtures of these gases at high pressures, J. Chem. Phys. 45 (1966) 4617–4627.

[9] R. Peyrous, P. Pignolet, B. Held, Kinetic simulation of gaseous species created by an electrical discharge in dry or humid oxygen, J. Phys. D: Appl. Phys. 22 (1989) 1658–1667.

[10] J.F. Loiseau, C. Monge, R. Peyrous, B. Held, C. Coste, Numerical simulation of ozone axial and radial distribution in a cylindrical oxygen-fed ozonizer, J. Phys. D: Appl. Phys. 27 (1994) 63–73.

[11] A. Yabe, Y. Mori, K. Hijikata, EHD study of the corona wind between wire and plate electrodes, AIAA J. 16 (1978) 340–345.

[12] W.F.L.M. Hoeben, Pulsed Corona-induced Degradation of Organic Materials in Water, Technische Universiteit Eindhoven, Netherland, 2000.

[13] A.K. Sharma, G.B. Josephson, D.M. Camaioni, S.C. Goheen, Destruction of pentachlorophenol using glow discharge plasma process, Environ. Sci. Technol. 34 (2000) 2267–2272.

[14] R.B. Bird, W.E. Stewart, E.N. Lightfoot, Transport Phenomena, Wiley, New York, 1960.

[15] N.I. Butkovskaya, D.W. Setser, Infrared chemiluminescence study of reaction of hydroxyl radical with acetaldehyde and the secondary reactions of acetyl radical with NO_2, OH, and H, J. Phys. Chem. A 104 (2000) 9428–9435.

[16] A. Tomas, E. Villenave, R. Lesclaux, Reaction of the HO_2 radical with CH_3CHO and $CH_3C(O)O_2$ in the gas phase, J. Phys. Chem. A 105 (2001) 3505–3514.

[17] N. Sano, H. Nagamoto, T. Tamon, T. Suzuki, M. Okazaki, Removal of acetaldehyde and skatole in gas by a corona-discharge reactor, Ind. Eng. Chem. Res. 36 (1997) 3783–3791.

[18] N. Sano, T. Kawashima, J. Fujikawa, T. Fujimoto, T. Kitai, T. Kanki, A. Toyoda, Decomposition of organic compounds in water by direct contact of gas corona discharge: influence of discharge conditions, Ind. Eng. Chem. Res. 41 (2002) 5906–5911.

[19] D. Hayashi, W.F.L.M. Hoeben, G. Dooms, E.M. Van Veldhuizen, W.R. Rutgers, G.M.W. Kroesen, Influence of gaseous atmosphere on corona-induced degradation of aqueous phenol, J. Phys. D: Appl. Phys. 33 (2000) 2769–2774.

[20] G.V. Buxton, C.L. Greenstock, W.P. Helman, A.B. Ross, Critical review of rate constants for reaction of hydrated electron, hydrogen atoms and hydroxyl radicals ($^{\bullet}OH/^{\bullet}O^-$) in aqueous solution, J. Phys. Chem. Ref. Data 17 (1988) 693–694.

[21] D.J. Jacob, E.W. Gottlied, M.J. Prather, Chemistry of a polluted boundary layer, J. Geophys. Res. 94 (1989) 12975–13002.

[22] L. Bruno, A.R. David, R.B. Deborah, Ozone in Water Treatment, Lewis Publishers Inc., Chelsea, Michigan, 1991.

[23] A. Mizuno, Y. Kisanuki, M. Noguchi, S. Katsura, S.H. Lee, Y.K. Hong, S.Y. Shin, J.H. Kang, Indoor air cleaning using pulsed discharge plasma, IEEE Trans. Ind. Appl. 35 (1995) 1284–1288.