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High-temperature simultaneous removal of acetaldehyde and ammonia gases using corona discharge

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Abstract

The electrostatic precipitator (ESP) is the most prevalent application for corona as ion source. ESP is widely used in industrial dust collection and home ventilation systems for cleaning gas streams. However, ESP has not been used to remove gas pollutants from a gas stream. Electron attachment is one of the promising techniques for gas purification. Electron attachment reactions occur when low-energy electrons generated in a corona-discharge reactor are captured by electronegative impurities such as acetaldehyde and fine particles to produce negative ions and charged particles. The negative ions and charged particles drift in the electric field to the anode (reactor wall) and are removed by deposition there. Moreover, O^- , O radical, ozone, and OH^- are also generated when the carrier gas is humid air. These anions and radicals also contribute to the removal efficiency. Though acetaldehyde (CH_3CHO) and ammonia (NH_3) are emitted at high temperature from a crematory furnace, there is insufficient report on the effect of high temperature and simultaneous removal. The present research experimentally investigates the technical feasibility of using corona discharge reactions to treat high-temperature exhaust gas containing acetaldehyde and ammonia. The simultaneous removal efficiency of these gases from nitrogen and air is obtained experimentally. The experimental results reveal that the presence of oxygen does significantly enhance the overall removal efficiency. Similarly, it is found that the presence of water vapor also enhances the removal efficiency of the target gases from the nitrogen–oxygen mixture. The generation of undesirable byproducts such as NO_x is also investigated. The present technique should be applicable to the simultaneous removal of different nanoparticles at high efficiency.

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Keywords: Corona discharge; Gas purification; High temperature

1. Introduction

Nowadays, air pollution in Thailand has become one of the most serious environmental problems. There are numerous sources of air pollutants such as heavy concentration of vehicles that release a large amount of toxic and obnoxious emission. One of the unexpected ubiquitous air pollution problems in Thailand is the emission gas from the crematoria during cremation rites. There are nearly 30,000 temples nationwide, including approximately 300 temples

with crematory furnaces in Bangkok Metropolitan Area alone. Various malodorous gases and particulates are emitted during cremation, causing frequent complaints from vicinal communities. Typically, the crematory gas is emitted from a stack to the atmosphere without adequate treatment. Only a few temples have installed furnaces with effective after-burning systems.

In 1995, a novel gas purification method using electron attachment was proposed [1]. The gas impurities are ionized by collisions with electrons produced, for example, in a corona discharge between a wire cathode and a cylindrical anode. The resulting negative ions drift to the anode and deposit on it. Previous studies have revealed that gases of high electron affinity, for example, sulfur compounds, halogens, malodorous gases, VOCs and CFCs are

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Notation

A	cross-sectional area (m^2)	θ	mean residence time (s)
C	concentration (ppm)	$\langle v \rangle$	superficial velocity (m/s)
E	electric field strength (V/m)	<i>Subscript</i>	
N	gas density (mol/m^3)	in	inlet of reactor
SV	space velocity (h^{-1})	out, 0 mA	outlet of reactor at zero discharge current
t_r	ratio of residence times in the corona discharge zone (–)	out, any mA	outlet of reactor at non-zero discharge current
V	discharge voltage (V)		

selectively removed by the proposed method [1–8]. Though acetaldehyde and ammonia are emitted at high temperature from a crematory furnace [9–11], there are insufficient reports on the effect of high temperature and simultaneous removal. In this study a corona discharge reactor is employed to simultaneously remove acetaldehyde (CH_3CHO) and ammonia (NH_3) from humid air from room temperature up to 300°C .

2. Experimental setup

Fig. 1 shows a schematic diagram of the experimental apparatus. The deposition-type corona-discharge reactor consists of a SUS tube, 3.7 cm inner diameter and 80 cm length, as the anode. The cathode is a 0.5-mm stainless-steel wire suspended from a silicone plug at the top of the reactor and straightened along the central axis of the vertical anode by a small weight. A high-voltage DC generator (Matsusada, HAR-30N5) is utilized to supply a steady stream of low-energy electrons to the corona-discharge reactor. To control the reactor temperature, six infrared

heating lamps (200 V, 700 W each) are installed lengthwise around the outside perimeter of the reactor. The temperature control unit consists of a temperature controller (FENWAL, AR-24L) and a thyristor power regulator (Shimaden, PAC15C003081-NO). The desired concentrations of acetaldehyde (CH_3CHO), NH_3 , and/or coexisting O_2 are adjusted by diluting standard gases with pure N_2 carrier gas. An additional O_2 /moisture trap is installed on the N_2 gas line to ensure the removal of trace O_2 and moisture in the carrier N_2 and diluent gas. The influent concentrations of CH_3CHO and NH_3 are 150 and 1000 ppm, respectively. Similarly, the concentrations of coexisting O_2 and H_2O are varied between 0–20% and 0–10,500 ppm, respectively. Inlet and outlet concentrations of CH_3CHO in the gas mixture are analyzed using a gas chromatograph (Shimadzu Corp., GC 9A) equipped with a flame ionization detector (FID). Concentrations of NH_3 are analyzed using another gas chromatograph (Shimadzu Corp., GC 14A and 14B) equipped with a thermal conductivity detector (TCD). The concentrations of O_3 and NO_x byproducts can separately be detected with appropriate gas detector tubes (GASTEC Co., Ltd).

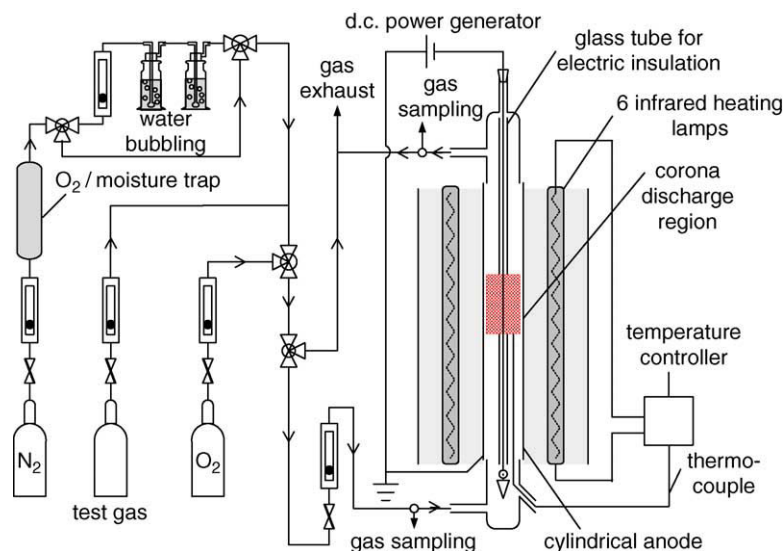


Fig. 1. Experimental apparatus.

3. Results and discussion

3.1. Definition of removal efficiency (by discharge effect only)

To exclude any possible effect of adsorption at low temperature and thermal decomposition at high temperature, the removal efficiency is defined as Eq. (1)

$$\text{Removal efficiency} = \frac{(C_{\text{out},0 \text{ mA}} - C_{\text{out at any mA}})}{C_{\text{out},0 \text{ mA}}} \quad (-) \quad (1)$$

Here, $C_{\text{out},0 \text{ mA}}$ (ppm) and $C_{\text{out at any mA}}$ (ppm) are the outlet concentration when applying no current and the outlet concentration when applying non-zero current, respectively. At steady state, the equation of continuity requires that $\rho_1 \langle v_1 \rangle A_1 = \rho_2 \langle v_2 \rangle A_2$. Since $A_1 = A_2$, and ρ is a function of the gas temperature, the gas velocity at high temperature T_2 will be faster than its velocity at room temperature T_1 . In other words, the mean residence time $\theta_2 = (V_r / \langle v_2 \rangle A_2)$ of this gas at T_2 is shorter than $\theta_1 = (V_r / \langle v_1 \rangle A_1)$ at room temperature. Here, V_r is the effective volume of the corona discharge reactor.

3.2. Influence of temperature on removal mechanism

Rigorously speaking, the relevant reactions contributing to the removal of CH_3CHO , NH_3 are affected not only by the change in electron energy level but also by other effects of the elevated temperature. To consider the reaction mechanism, one must take byproduct formation into account. As for the gaseous byproducts, O_3 and NO_x were, respectively, detected mainly in the low and high temperature ranges. However, when T is further increased above 300°C , O_3 concentration becomes negligible [12]. This is because O_3 is unstable at high temperature [13,14]. Therefore, oxidation by O_3 should play a negligible role in the high temperature range.

Contrary to O_3 formation, it is known that production of NO_x by the discharge process is favored at high temperature. This is also confirmed in our experiments. While the outlet concentration of NO_x was negligible at room temperature, its concentration gradually increased with temperature and reached 300 ppm at 400°C . Since NO_x formation can be attributed to the reaction of discharge-induced N radicals with O_2 [15,16], N radicals should also contribute to the removal of CH_3CHO and/or NH_3 from both N_2 and air at high temperatures. In addition, in the removal from air, there should be the extra effect of O radicals produced by electron impact to O_2 and by O_3 decomposition [13,17,18]. Therefore, the removal efficiency from air at high temperatures should be enhanced by O radicals, though O_3 oxidation is not effective. In fact Peyrous et al. [13] simulated the concentrations of O_3 and O radicals in pulsed corona discharge in the presence of O_2 , and showed that temperature elevation brings about higher O radical concentration and lower O_3 .

When H_2O is present in the gas stream, H^- , OH^- and a few O^- anions are expected to be produced by dissociative electron attachment to H_2O molecules at low temperature [19,20]. The selectivity for these ionic products should depend on the gas temperature and electron energy. At high temperature, electron detachment would become significant so that radicals of O, H, and OH may play a more important role than their anionic counterparts. These radicals are also expected to contribute to the removal of the target gases. More specifically, OH is believed to dissociate NH_3 to produce an aminogen radical (NH_2) and H_2O [21].

In non-thermal corona discharge in the air at room temperature, electrons are sometimes captured by O_2 to form negative ions, O^- , O_2^- , O_3^- , and clusters via electron attachment. The reversed electron detachment process, however, becomes significant at high temperature, causing ion clusters to become unstable [22]. The previous articles indicate that the corona discharge reactor plays host to electron attachment reactions and relevant ion cluster formation at room temperature [4,6]. However, because of high temperature effects such as electron detachment and radical formation, electron attachment and ion clustering would be less and less important as the temperature rises.

In the case of NH_3 removal, the removed NH_3 was converted to needle-like bright crystals of NH_4NO_3 , which were observed inside the reactor and the gas line after it. It is reported [23] that NH_3 does not react with O_3 to directly form NH_4NO_3 . Instead, the following consecutive reactions are mentioned: $2\text{NH}_3 + 4\text{O}_3 = \text{NH}_4\text{NO}_2 + \text{H}_2\text{O}_2 + 4\text{O}_2$; $\text{NH}_4\text{NO}_2 + \text{H}_2\text{O}_2 = \text{NH}_4\text{NO}_3 + \text{H}_2\text{O}$. Our result is also consistent with published reports that NH_4NO_3 solid is produced by corona treatment of humid air containing NH_3 [21,24,25]. The mechanism for NH_4NO_3 formation in the high temperature range is not clear but it may be considered that NH_3 , H_2O and NO_x as well as N and H radicals could react to form NH_4NO_3 .

3.3. Simultaneous removal of acetaldehyde and ammonia

As shown in Figs. 2 and 3, we see that, in the case of simultaneous removal of CH_3CHO and NH_3 from pure N_2 , the removal efficiency of both CH_3CHO and NH_3 increases with temperature up to 100°C because of the electron attachment reactions and relevant ion cluster formation [4,21]. Above 100°C , the tendency slightly reverses because the ion clusters are not stable and the mean residence time of the gas mixture inside the reactor is reduced at high temperature. N radicals should also contribute to the removal of CH_3CHO at high temperatures [15,16]. However, the effect of N radicals is not so high.

3.3.1. Effect of temperature and coexisting O_2 on the simultaneous removal of CH_3CHO and NH_3 from N_2

Fig. 2 shows the effect of O_2 on the simultaneous removal efficiency of CH_3CHO and NH_3 from N_2 . The inlet concentration of CH_3CHO and NH_3 are 150 and 1000 ppm,

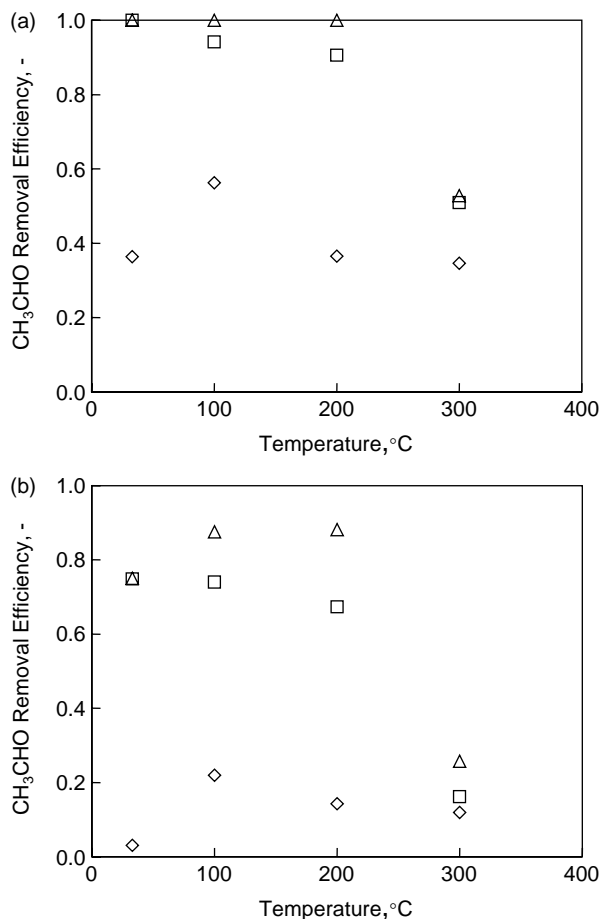


Fig. 2. Effect of coexisting O₂ on the simultaneous removal of CH₃CHO and NH₃ from N₂; C_{in, acetaldehyde} = 150 ppm, C_{in, ammonia} = 1000 ppm, I = 0.3 mA, SV = 55.8 h⁻¹ at room temperature: (◇) pure N₂, (□) O₂ 10%, (Δ) O₂ 20%.

respectively, while the current is 0.3 mA. It is found that the presence of O₂ has a significant enhancement effect on the simultaneous CH₃CHO removal efficiency in Fig. 2(a). As temperature increases, the CH₃CHO removal efficiency remains nearly 100% from room temperature to 200 °C because of the effect of O₃ and O⁻ anion at low temperatures [13,17,18]. In contrast, the CH₃CHO removal efficiency tends to significantly decrease above 200 °C because of less O₃ generated from O₂ and reduction of the mean residence time of the gas mixture inside the reactor at high temperature. This can be attributed to the fact that O₃ is produced from O₂ by the corona discharge reaction and is quite stable at room temperature. At room to moderate temperatures, electron attachment reactions contribute to, and relevant ion cluster formation enhances, the removal of numerous electro-negative compounds [4,21]. N and O radicals should also contribute to the removal of CH₃CHO from both N₂ and air at high temperatures.

In Fig. 2(b), at 20% O₂, the NH₃ removal efficiency increases with temperature up to 200 °C then dramatically decreases up to 300 °C. As mentioned previously, O₃ and

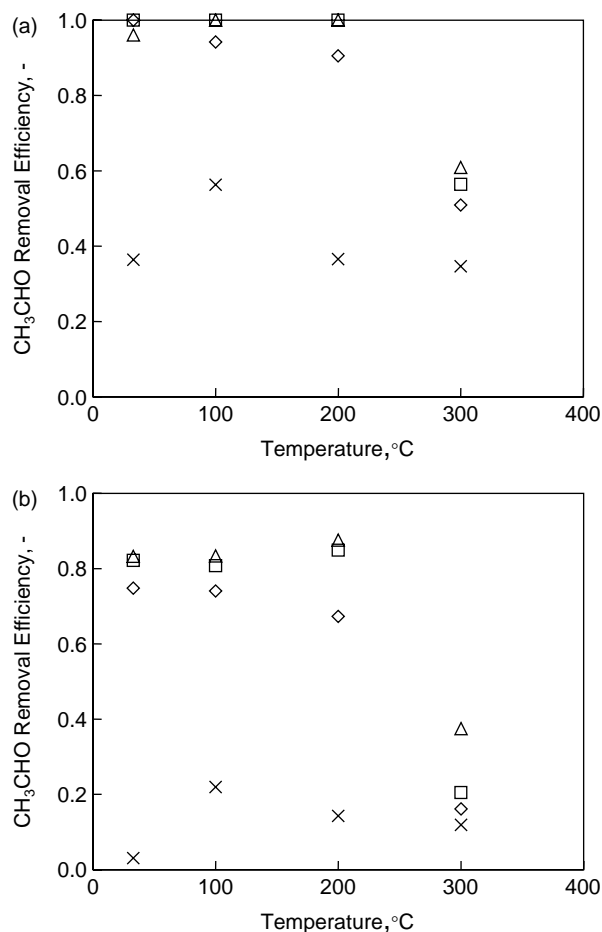


Fig. 3. Effect of H₂O on the simultaneous removal of CH₃CHO and NH₃ from N₂-O₂(10%); C_{in, acetaldehyde} = 150 ppm, C_{in, ammonia} = 1000 ppm, I = 0.3 mA, SV = 55.8 h⁻¹ at room temperature: (◇) H₂O 0 ppm, (□) H₂O 5250 ppm, (Δ) H₂O 10,500 ppm, (×) pure N₂.

O⁻ anion should contribute to the removal of NH₃ at low temperature whereas, at high temperatures, N and O radicals should contribute to the removal of NH₃. However, the effect of radicals on the removal efficiency is not as strong as O₃, O⁻ anion and ion cluster effect. In contrast, the NH₃ removal efficiency at lower O₂ concentration (10%) decreases with the temperature up to 300 °C because of less O₃ generated from O₂ compared to the cases of higher O₂ concentration.

3.3.2. Effect of temperature and H₂O on the simultaneous removal of CH₃CHO and NH₃ from N₂-O₂

Fig. 3 shows the combined effect of O₂ and H₂O on the simultaneous removal efficiency of CH₃CHO and NH₃. Compared to the case of only coexisting O₂, the simultaneous removal efficiency of both CH₃CHO and NH₃ is enhanced by the presence of H₂O because H⁻, OH⁻ and a few O⁻ anions contribute to the removal of both gases at low temperature. At high temperature, O₃ generated from O₂, and H⁻, OH⁻ and a few O⁻ anions from H₂O are unstable. Coupled with the negative effect of reduced

residence time, the simultaneous removal efficiency of both CH_3CHO and NH_3 decreases significantly at 300 °C.

In Fig. 3(a), we see that, as the temperature increases, the removal efficiency of CH_3CHO remains nearly 100% from room temperature up to 200 °C because of the effect of H^- , OH^- and a few O^- anions. Above 200 °C, the removal efficiency of CH_3CHO significantly decreases with temperature. At high temperature, the removal of CH_3CHO from humid air depends on the effect of O, H, OH and N radicals. As mentioned previously, NO_x formation can be attributed to the reaction of discharge-induced N radicals with O_2 [15,16]. Since N radicals are consumed by their reaction with H_2O at high temperatures, the CH_3CHO removal efficiency at high temperatures decreases in the presence of H_2O .

In Fig. 3(b), it is found that, as the temperature increases, the removal efficiency of NH_3 slightly increases from room temperature to 200 °C. However, the removal efficiency drastically drops at 300 °C. As mentioned above, H^- , OH^- and a few O^- anions and O_3 should contribute to the removal of NH_3 at low to moderate temperature. At 300 °C, the tendency reverses because of the combined effect of a decrease in the mean residence time and relatively lower activity of the radicals.

4. Conclusion

The present research is focused on the investigation of the effect of high temperature on the simultaneous removal of acetaldehyde (CH_3CHO) and ammonia (NH_3) from humid air using a corona-discharge reactor. It is found that the presence of O_2 significantly enhances the simultaneous removal efficiency of CH_3CHO and (NH_3) because of the contribution of O_3 and O^- anion at low temperature, and N and other radicals at high temperature. As in the case of $\text{N}_2\text{-O}_2$, the additional presence of water vapor in $\text{N}_2\text{-O}_2$ mixtures generally has a favorable effect on the simultaneous removal efficiency of CH_3CHO and NH_3 because of the effect of O_3 , H^- , OH^- and a few O^- anions at low temperature and N, O, H and OH radicals at high temperature. However, the removal efficiency of both cases decreases above 200 °C because the effect of radicals on the removal efficiency is not as strong as O_3 , H^- , OH^- , O^- anion and their ion clusters. In addition, there is the negative effect of reduced residence time. The present technique should be applicable to the simultaneous gas–solid separation of nanoparticles with high efficiency.

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