Electrostatic Separation of Carbon Dioxide by Ionization in Bifurcation Flow

Takao Ito[*], Yoshio Otani and Norikazu Namiki

Graduate School of Natural Science & Technology, Kanazawa University, 2-40-20 Kodatsuno, Kanazawa 920-8667, Japan.

Abstract

Carbon dioxide is one of the major green house gases as well as impurities in process gases used for various manufacturing industries. In the present work, our recently developed ionization separator (Ito *et al.*, *Ind. & Eng. Chem. Res., 42*, 5617-5621, 2003) was applied to the separation of carbon dioxide from inert gases. As a result, it was found that carbon dioxide can be separated mostly in the form of anion although some fraction of carbon dioxide decomposes by the soft X-ray irradiation. The maximum efficiency of electrostatic separation of carbon dioxide was 14% when helium stream contains 2.4 ppm of carbon dioxide at the applied voltage of 600V and the separation efficiency was decreased with increase in the inlet concentration. The dependency of separation efficiency on the applied voltage was qualitatively explained by the separation model that accounted for the electrical migration, the generation and the neutralization of anions and cations formed from carbon dioxide.

Keywords: ionization, soft X-ray, electrical migration, carbon dioxide, separation.

1. Introduction

Reduction of carbon dioxide (CO_2) concentration in air and other gases has been of great concern in various fields; e.g., control of indoor air quality by ventilation (Nabinger *et al.*, 1994, Persily, 1997), green house effect (Hansen *et al.*, 1981), contamination control of process gases for semiconductor manufacturing (Briesacher *et al.*, 1991).

Adsorption of CO_2 with molecular-sieves and other adsorbents has been a common method for reducing CO_2 concentration. However, it requires the replacement or regeneration of adsorbents for maintaining the adsorption performance and the high pressure drop across the packed bed is a great issue when considering the energy consumption for gas purification. Metallic getter alloys are also used for gas purification (Briesacher *et al.*, 1991). Although this method removes trace impurities

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[*] Corresponding author. Tel: +81 76 234 4813; Fax: +81 76 234 4829.

E-mail address: itoh@ aerosol.ch.t.kanazawa-u.ac.jp

Nomenclature

A = cross-sectional area of separator $[m^2]$

C = concentration of carbon dioxide in outlet flow $[m^{-3}, ppm]$

C_{in}= inlet concentration $[m^{-3}, ppm]$

$C_{V=0V}$ = outlet concentration at applied voltage of 0 V $[m^{-3}, ppm]$

C^* = relative concentration = $C/ C_{V=0V}$ [-]

D = diffusion coefficient $[m^2/s]$

e = elementary unit of charge $[C]$

E = electrical field strength $[V/m]$

G = generation rate $[m^{-3}s^{-1}]$

G_i = generation rate of reactive primary ions $[m^{-3}s^{-1}]$

I = ion current $[A]$

J = diffusion flux $[m^{-2}s^{-1}]$

L = distance between anode and cathode $[m]$

N_i = number concentration of ions $[m^{-3}]$

Q_{in} = inlet flow rate $[l/min]$

R = depletion rate $[m^{-3}s^{-1}]$

t = time $[s]$

u = gas flow velocity $[m/s]$

v = drift velocity of ion $(=ZE)$ $[m/s]$

V = voltage $[V]$

x = coordinate system $[m]$

Z_c = electrical mobility of CO_2 cation $[m^2V^{-1}s^{-1}]$

Z_a = electrical mobility of CO_2 anion $[m^2V^{-1}s^{-1}]$

α_c = net depletion rate constant of CO_2 cation $[s^{-1}]$

α_a = net depletion rate constant of CO_2 anion $[s^{-1}]$

β_c = net generation rate constant of CO_2 cation $[s^{-1}]$

β_a = net generation rate constant of CO_2 anion $[s^{-1}]$

κ = recombination coefficient of ion $[m^3/s]$

<Subscript>

Cn= CO_2 neutral molecule

Cc= CO_2 cation   Ca= CO_2 anion

such as O_2, H_2O, CO and CO_2 in carrier gas, it requires high temperatures to react these species with the getter.

Recently, behaviors of ions, radicals and molecular species have been studied for the purpose of gas cleaning and purification from gas stream by corona discharge (Ohkubo *et al.*, 1994), surface discharge (Oda *et al.*, 1997) and electron beam injection (Hirota *et al.*, 1995), etc. However these methods using high discharge energy are not effective for lowering the concentration of CO_2.

Ionization is the process where electrically neutral atoms or molecules acquire either positive or negative electrical charge. In aerosol researches, ions are most frequently used to charge aerosol particles for electrical mobility classification (Knutson and Whitby, 1975). Gaseous molecules in the atmosphere are ionized by irradiation with radioactive source, electrical discharge, and combustion, etc. Radioactive-ray irradiation or corona discharge initially generates primary positive ions and free electrons. The free electrons readily attach to electronegative species in air to form negative ions. Since the primary positive and negative ions are unstable, secondary ionization due to ion-molecule reaction occurs by the collisions between ions and neutral species. The ion-molecule reaction is influenced by the external electric fields.

We have proposed a new ionization separation technique which utilizes selective ionization of contaminant species and electrostatic migration of ions in bifurcating flow. The contaminant species removed by this method are toluene (Ito *et al.*, 2002a, Ito *et al.*, 2003) and ethanol vapor (Ito *et al.*, 2002b). By this method, the contaminant species that have lower ionization potential and higher proton affinity compared to carrier gas becomes positive ions by the irradiation with α-ray or soft X-ray, and are electrostatically separated from the carrier gas. Negatively charged impurities generated by electron attachment can also be separated from carrier gas by applying an electrical field (Tammon *et al.*, 1995). However, most of the molecular species in the atmosphere are ionized into both polarities. For instance, CO_2 can take both forms of cation and anion, such as $(CO_2)_x^+$, $(CO_2)_xCO^+$, $(CO_2)_xO^+$, $(CO_2)_xC^+$, $(CO_2)O_2^+$, $(CO_2)_x^-$, $(CO_2)_xO^-$ (x=1,2,3,…) according to Alger and Rees (1976). Nevertheless, since the generation rate and concentration of each ion species formed from CO_2 are different (Stamatovic *et al.*, 1985), it might be possible to separate CO_2 in a form of cation or anion by ionization followed by electrostatic separation.

In this paper, our ionization separation technique was applied to CO_2. We studied the separation performance of CO_2 in various carrier gases and the influence of CO_2 concentration on the separation efficiency, and discussed the separation mechanisms via the ion-molecule reaction kinetics of CO_2 in bipolar ionization field.

2. Experimental apparatus and procedure

Fig. 1 shows the ionization aeron separator, whose structure is basically the same as used previously (Ito *et al.*, 2002a), except that the improvement was made to avoid contamination from structural
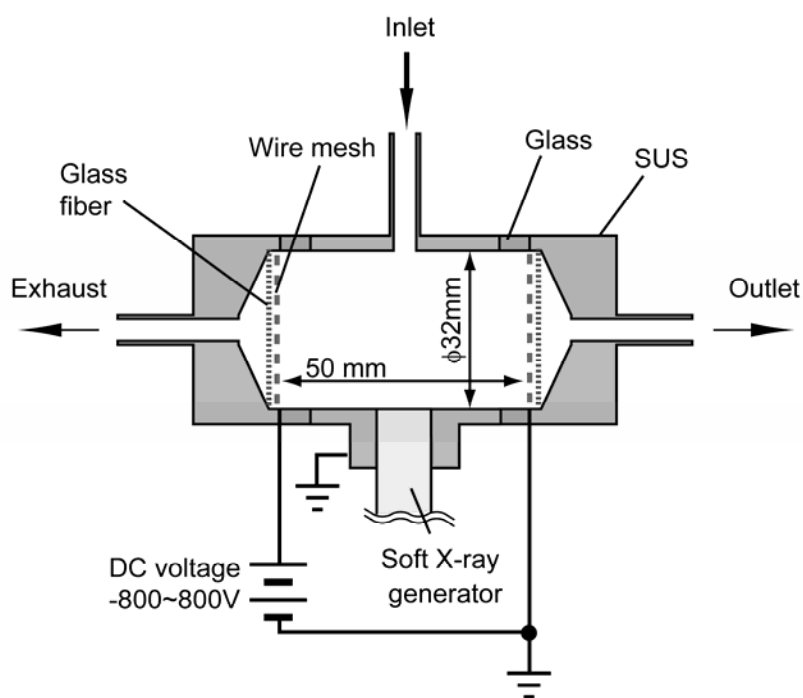
Figure 1. Schematic of ionization-separator with Soft X-ray generator and wire mesh electrodes.
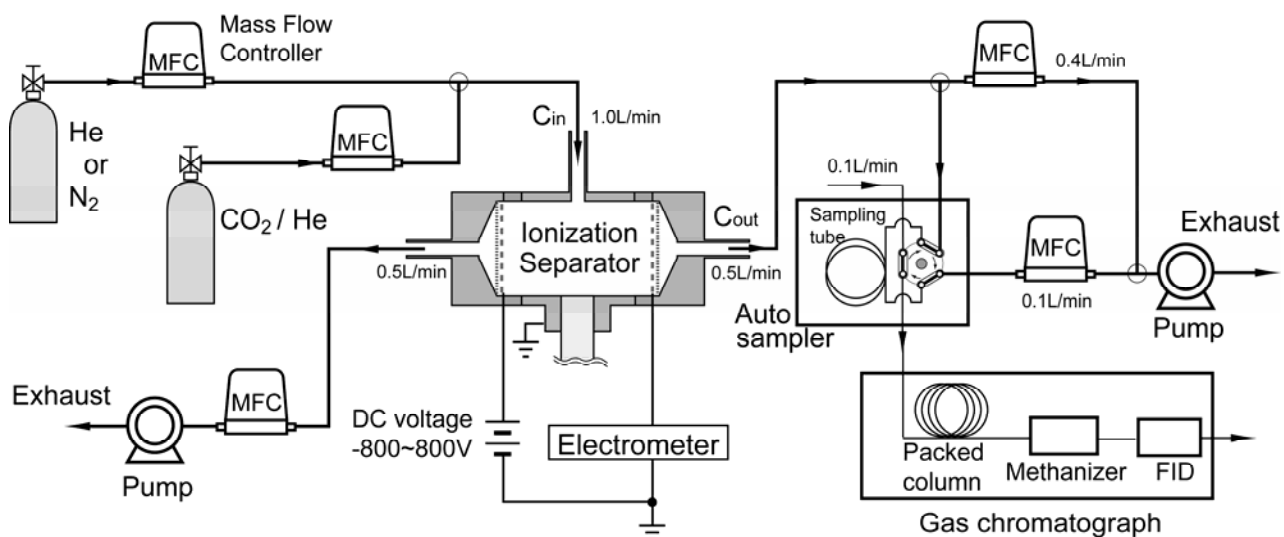
Figure 2. Flowchart of experimental setup.

Table 1. Ionization potential, proton affinity and electron affinity of various gases

Component	Ionization potential	Proton affinity	Electron affinity
	[eV]	[kJ/mol]	[eV]
He	24.587	178	unstable
N_2	15.59	494.5	unstable
O_2	12.07	422	0.451
H_2O	12.612	697	-
CO_2	13.78 [$(CO_2)_2^+$]	548	>2 [$(CO_2)_n^-$]

materials by exchanging metal (SUS316) and quartz glass for plastics. Flow containing CO_2 is split into two ones, while the flow is being irradiated with soft X-ray from photoionizer (Model L6941, Hamamatsu Photonics, Japan; energy: 3.0-9.5 keV) under an electric field. The soft X-ray emitter has a dose of 4.7×10^{-6} Gy/s at a distance of 1 m away from the source. The production rate of primary ions in the ionization separator is estimated to be in the order of 10^{16} m^3/s when the generation of an ion pair requires the energy of 34 eV in air (Ito *et al.*, 2003). In the separator, CO_2 molecules are positively and negatively ionized as a result of ion-molecule reactions, and migrate towards either cathode or anode by the applied electric field. The ionization potential, proton affinity and electron affinity of CO_2 compared to other gases are listed in Table 1.

Fig. 2 shows the experimental setup. High purity helium (>99.9999%) and nitrogen (>99.9999%) are used as a carrier gas. The carrier gas is mixed with standard gas of nitrogen containing CO_2 (0.103%) to obtain a given concentration, and introduced into the ionization separator at the flow rate of 1.0 L/min. Inlet flow is equally divided into two outlet ones. The outlet concentrations of CO_2 are measured by sampling the gas by auto-sampler (GL Sciences Inc.: GSS-5000AH) followed by the determination with a gas chromatograph with FID (Shimadzu, GC-17A), converting CO_2 into methane by a methanizer (GL Sciences Inc.: MT221).

3. Experimental Results

The decomposition of CO_2 may occur by the irradiation with soft X-ray in the ionization-separator. Hatherly and Codling (1995) reported the dissociative ionization of CO_2 by soft X-ray irradiation ($CO_2 \rightarrow CO^+ + O^+$ or $O^+ + C^+ + O^+$). Therefore the decomposition of CO_2 by the irradiation in the ionization-separator should be taken into account. Consequently, we measured the change in outlet concentration of CO_2 without applying an electric field. Fig. 3(a) shows the change in CO_2
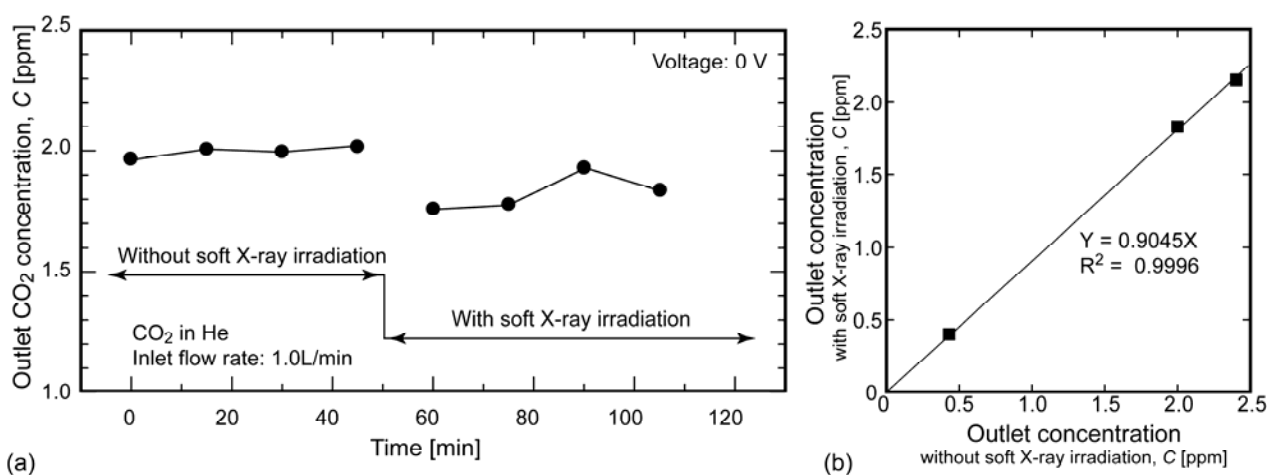
Figure 3. Decomposition of CO_2 by soft X-ray irradiation in the ionization-separator. (a) Change in the CO_2 concentration with soft X-ray irradiation. (b) Outlet concentration with soft X-ray irradiation vs. outlet concentration without soft X-ray irradiation.
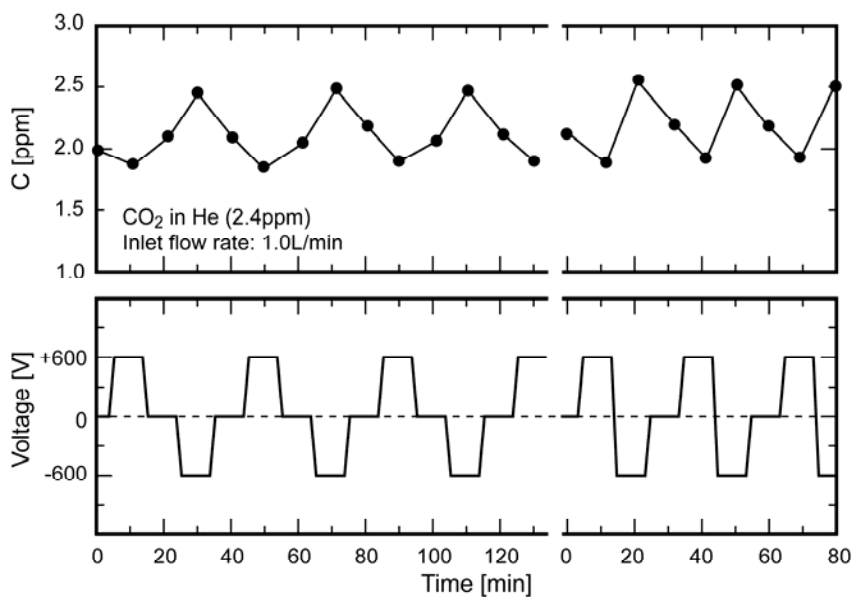
Figure 4. Change in the CO_2 concentration with applied voltage of +600V and -600V.
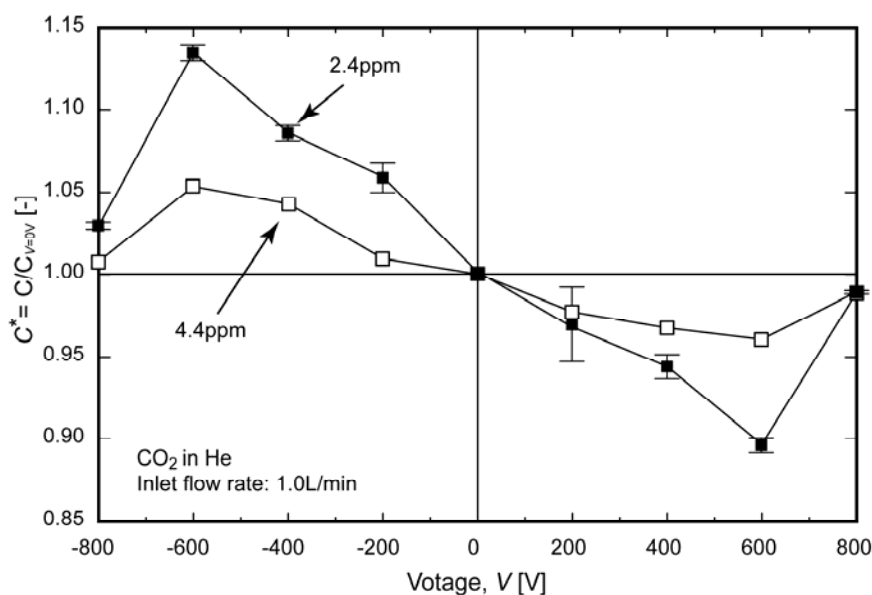
Figure 5. Change in the relative concentration of CO_2 as a function of applied voltage.

concentration when the irradiation of soft X-ray is initiated. As seen in the figure, CO_2 concentration is reduced from 2.0 ppm to 1.8 ppm due to the irradiation with soft X-ray. The relationship between inlet concentration and outlet concentration is shown in Fig. 3(b). The outlet concentration of CO_2 is decreased by about 10% due to the photo-dissociation for the residence time of gas in the separator of 2.4 s.

In order to evaluate the separation performance of ionization-separator for CO_2 taking into account the decomposition during the separation, we defined the relative CO_2 concentration from the separator as $C^* = C/C_{V=0}$, where C is the outlet concentration with soft X-ray irradiation and voltage application, and $C_{V=0}$ is the outlet concentration with the irradiation without the application. Fig. 4 shows the change in CO_2 concentration at the grounded outlet of ionization-separator with the voltage application under the irradiation of soft X-ray. Helium flow containing 2.4 ppm CO_2 was introduced into the separator at the flow rate of 1.0 L/min. CO_2 concentration was measured at the outlet of grounded electrode while varying the voltage between +600V and -600V as shown in the figure. As seen in the figure, CO_2 concentration is slightly higher than the inlet one of 2.4 ppm when the ground electrode is anode. It is considerably lower than when it is cathode, and follows the change in applied voltage, indicating that the separation of CO_2 does occur even though the dissociation of CO_2 takes place. Furthermore, we can see from it that the migration of CO_2 in a form of anion is dominant in the separator whereas toluene vapor was separated in the form of cation (Ito *et al.*, 2002a, 2003).

Fig. 5 shows the influence of applied voltage on the relative concentration of CO_2 at the inlet CO_2 concentrations of 2.4 and 4.4 ppm. The relative CO_2 concentration has a minimum and maximum at the voltage of 600V and -600V for both inlet concentrations. The voltage corresponds with that at which the maximum separation of toluene was attained in the previous work (Ito *et al.*, 2003). Fig. 6 shows the relative CO_2 concentration at the anode and cathode as a function of inlet CO_2
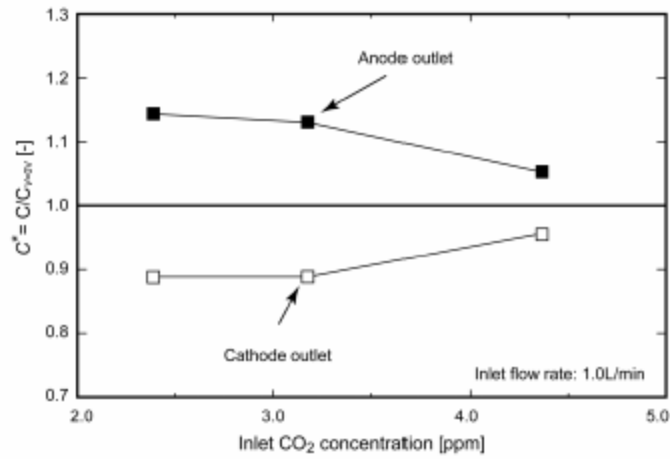
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Figure 6. Influence of inlet CO_2 concentration on relative one with appliedvoltage of 600V.
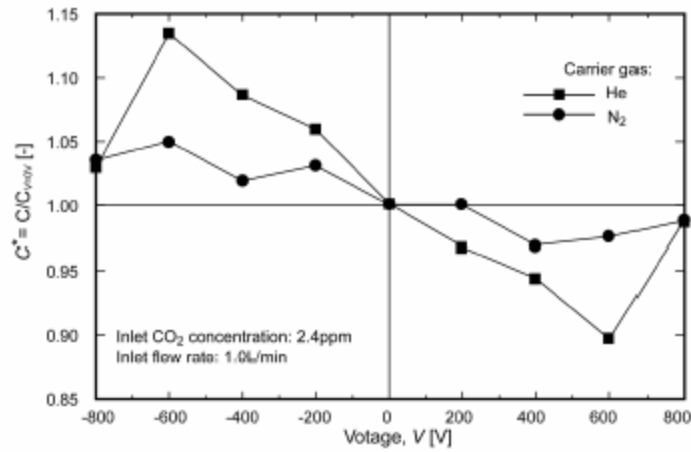

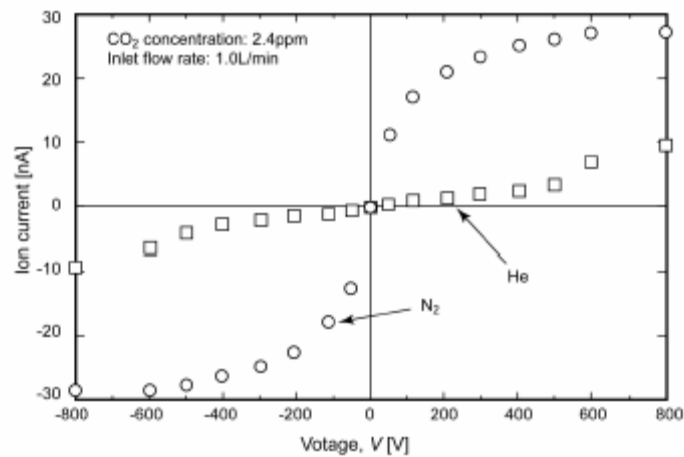**Figure 7.** Influence of carrier gas on relative CO_2 concentration.

Figure 8. Change in the ion current in ionization-separator with applied voltage.

concentration at the applied voltage of 600 V. Relative CO_2 concentration at the anode increases as the inlet one decreases, while that at the cathode decreases. The dependency of relative concentration on the inlet CO_2 one is similarly reported for toluene in our previous work (Ito *et al.*, 2003). However, the dependency on the inlet concentration is less pronounced compared to toluene.

The influence of carrier gas on relative CO_2 concentration was studied by using helium and nitrogen. Fig. 7 compares the relative CO_2 concentrations helium and nitrogen at the inlet CO_2 one of 2.4 ppm. Relative CO_2 concentration in helium is higher than that in nitrogen because ion-molecule reaction of CO_2 was probably enhanced due to higher mobility of ions in helium than that in nitrogen. The mobility of ions in helium is about three times the mobility in nitrogen (Bricard and Pradel, 1966). The ion currents in the ionization-separator for both carrier gases were measured with an electrometer, and are shown in Fig. 8 as a function of applied voltage. The ion current in nitrogen steeply increases with the voltage and reaches constant value at a voltage of 600V. On the contrary, the ion current in helium slightly changes with the applied voltage, and is only one-third of the current in nitrogen. Because helium has a smaller proton affinity and a higher ionization potential compared to nitrogen, helium molecules are difficult to become cations compared to nitrogen ones. The concentration of generated ions, *N*, was calculated by $I = e \kappa N^2 AL$ (Liu and Pui, 1974), where *e* is the elementary unit of charge, κ is the recombination coefficient of ions (1.5×10^{-12} m³/s in helium, 2.0×10^{-12} m³/s in nitrogen, Glushchenko *et al.,* 1988), *AL* is the effective volume in separator. The concentrations of ions calculated from the saturation current are 3.13×10^{13} molecules/m³ in helium and 4.66×10^{13} molecules/m³ in nitrogen. Since 1 ppm of CO_2 concentration corresponds to 2.5×10^{19} molecules/m³, the number of generated ions measured by the electrometer is six orders of magnitude lower than that of CO_2 molecules. The following two explanations are plausible to explain the discrepancy between the ion concentration and CO_2 concentration: 1) the ion concentrations determined from the saturation current are not correct because many ionic species and electrons are involved in the transfer of electrical charge; 2) the CO_2 depletion mechanism is more complicated than just ionization-separation.

4. Discussion

The separation mechanism of ionization separator was discussed in our previous work by using a simple transport model, which employed one-dimensional convective diffusion equations for positively charged species and neutral species. In this paper, we applied the same separation model while including the transport of anions of CO_2 as well as the transport of cations. It was assumed that the net generation rates of CO_2 cations and anion are proportional to the number of neutral CO_2 molecules and that the depletion rates of CO_2 anions and cations are proportional to the numbers of CO_2 cations and anions without the decomposition of CO_2. The mass balance of CO_2 molecules and CO_2 ions, as shown in Fig. 9, yields the following one-dimensional convective diffusion equations.
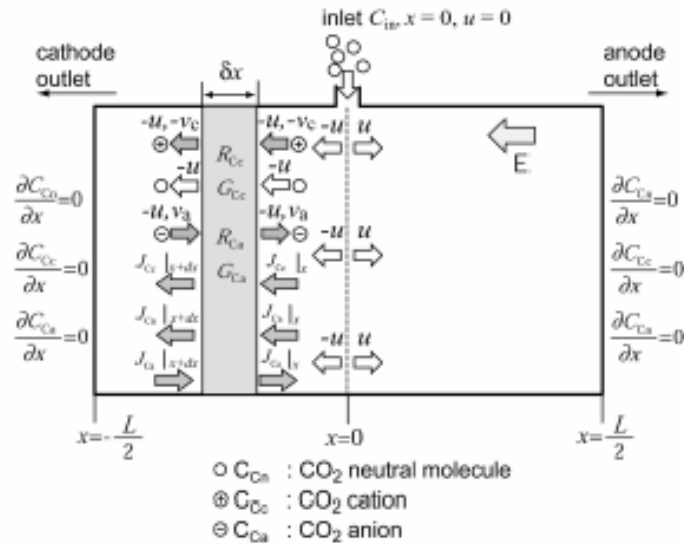
Figure 9. Change in the ion current in ionization-separator with applied voltage.

$$\begin{cases} \dfrac{\partial C_{Cn}}{\partial t} = u\dfrac{\partial C_{Cn}}{\partial x} + \alpha_c C_{Cc} + \alpha_a C_{Ca} - (\beta_c + \beta_a)C_{Cn} + D_{Cn}\dfrac{\partial^2 C_{Cn}}{\partial x^2} \quad (1) \\[3mm] \dfrac{\partial C_{Cc}}{\partial t} = (u - Z_c E)\dfrac{\partial C_{Cc}}{\partial x} - \alpha_c C_{Cc} + \beta_c C_{Cn} + D_{Cc}\dfrac{\partial^2 C_{Cc}}{\partial x^2} \quad (2) \\[3mm] \dfrac{\partial C_{Ca}}{\partial t} = (u + Z_a E)\dfrac{\partial C_{Ca}}{\partial x} - \alpha_a C_{Ca} + \beta_a C_{Cn} + D_{Ca}\dfrac{\partial^2 C_{Ca}}{\partial x^2} \quad (3) \end{cases}$$
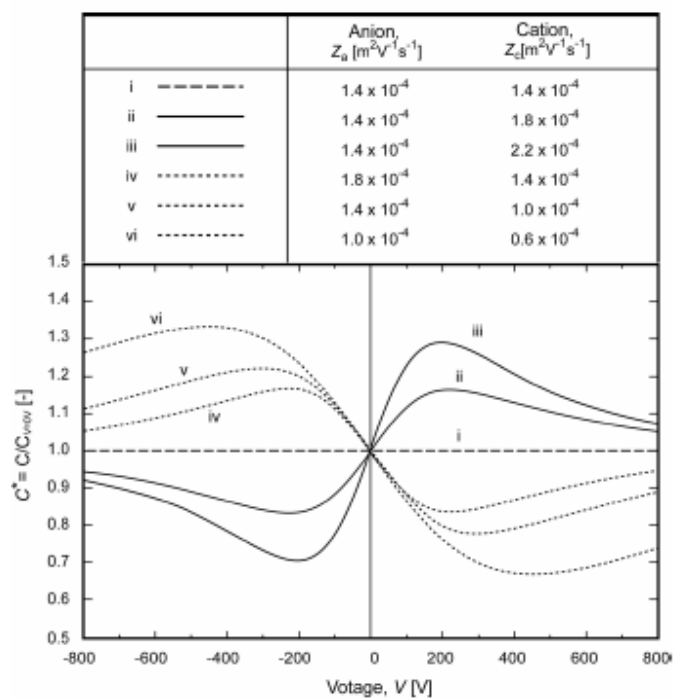
where C is the concentration, u is the gas flow velocity, D is the diffusion coefficient, α is the net depletion rate constant of CO_2 ions, β is the net generation rate constant of CO_2 ions, and Z and E are the electrical mobility of CO_2 ions and the electrical field strength, respectively. ZE is equal to the ion drift velocity, $ZE = v$.

The boundary conditions for the ionization-separator imposed for solving Eqs. 1, 2 and 3 are

$$\frac{dC_{Cc}}{dx} = 0, \quad \frac{dC_{Ca}}{dx} = 0, \quad \text{at } x = \frac{L}{2}, \quad x = -\frac{L}{2} \qquad \text{(Separator outlets)}$$

$$C_{Cn} = C_{in}, \quad \text{at } x = 0 \qquad \text{(Separator inlet)}$$

where L is the distance between the anode and the cathode. The following constraint for the mass balance is also adopted.

$$\frac{Q_{in}}{2}\left[C_{Cn} + C_{Cc} + C_{Ca}\right]_{x=\frac{L}{2}} + \frac{Q_{in}}{2}\left[C_{Cn} + C_{Cc} + C_{Ca}\right]_{x=-\frac{L}{2}} = C_{in}Q_{in}$$

		Anion, Z_a [$m^2V^{-1}s^{-1}$]	Cation, Z_c[$m^2V^{-1}s^{-1}$]
i	-------	1.4×10^{-4}	1.4×10^{-4}
ii	———	1.4×10^{-4}	1.8×10^{-4}
iii	———	1.4×10^{-4}	2.2×10^{-4}
iv	··········	1.8×10^{-4}	1.4×10^{-4}
v	··········	1.4×10^{-4}	1.0×10^{-4}
vi	··········	1.0×10^{-4}	0.6×10^{-4}

Figure 10. Change in the calculated dimensionless concentration of CO_2 as a function of applied voltage at various electrical mobility of CO_2 ion. $\beta_a = \beta_c = \alpha_a = \alpha_c = 500$ s^{-1}.

		Anion		Cation	
		β_a [s^{-1}]	α_a [s^{-1}]	β_c [s^{-1}]	α_c [s^{-1}]
i	-------	500	500	500	500
ii	———	800	500	500	500
iii	—·—·—	800	650	500	500
iv	—··—··—	500	500	300	500
v	··········	500	500	300	400
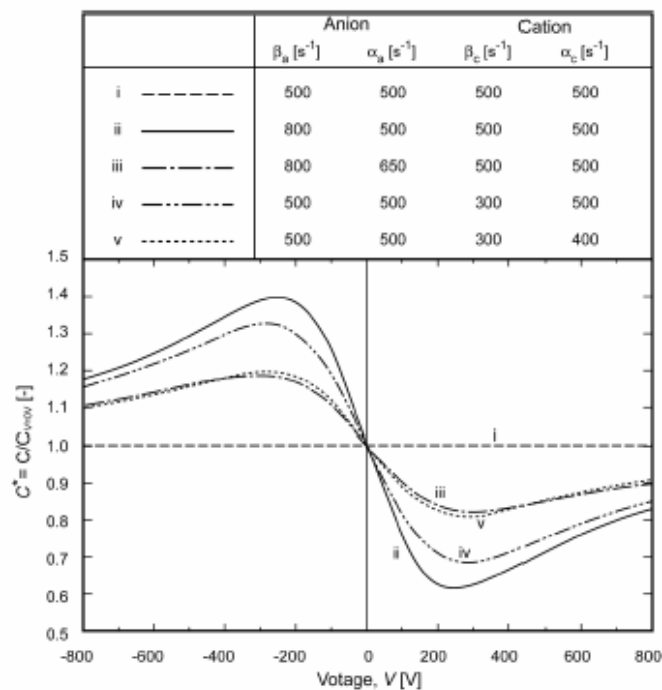
Figure 11. Influence of net generation and depletion rate constant on calculated concentration. $Z_a = Z_c = 1.4$ m^{2}V^{-1}s^{-1}.

The rate constants of α and β and electrical mobility of CO_2 ions are varied in the order of 10^2 to 10^3 referring to the previous work (Iinuma, 1991). We assigned the diffusion coefficient of CO_2 in helium at 293 K to be 6.2×10^{-5} m^2/s according to Poling *et al.* (2000).

The dimensionless concentration of CO_2 is calculated by solving Eqs. 1, 2 and 3 to see how the difference of electrical mobility between CO_2 cation and CO_2 anion influences CO_2 separation. Fig. 10 shows the dimensionless CO_2 concentration at the outlet as a function of applied voltage for various electrical mobilities while keeping the other parameters unchanged. No separation occurs when the mobilities of CO_2 anions and cations are the same ($Z_c = Z_a$). However, when the mobility of cations is different from that of anions, which is conceivable, ionic species generated from CO_2 are transported to the cathode and anode at a different velocity, causing the separation of CO_2 with a maximum at a given applied voltage in all cases. When the electrical mobility of CO_2 cations is larger than that of anions ($Z_c > Z_a$), CO_2 is concentrated at the cathode, whereas CO_2 concentration is higher at anode when the inequality is reversed ($Z_c < Z_a$). Therefore, when the species to be separated are ionized in both polarities, the separation occurs by the difference between electrical mobility of cations and anions originated from the species. The extent of separation is more pronounced when the difference is larger. Furthermore, it is known from Fig. 10 that the decreases in cation's and anion's moblities lead to the increase in separation efficiency together with the shift of peak voltage toward higher one.

The calculated curves of dimensionless concentration are symmetric with respect to the origin point of V=0 and C*=1 for all mobilities although it was observed in Fig. 5 that the experimental curves are asymmetric with respect to the point. The asymmetry in the experimental curve of dimensionless concentration may result from the decomposition of CO_2 ions after separation because the decomposition of neutral CO_2 does not lead to the asymmetry in the concentration curves.

We also investigated the influence of net generation rate and net depletion rate constants when the electrical mobilities of cations and anions are the same. The results are shown in Fig. 11. The calculated curves are symmetric with respect to the origin point for various combinations of α and β. Therefore, the asymmetry observed in the experimental curve as a function of applied voltage may result from the change in α and β with the applied voltage other than the decomposition of CO_2. The larger net generation rate constant of CO_2 anions (compare curve i with ii), or the smaller net generation rate of cations (compare curve i with iv) causes the higher dimensionless outlet concentration. We also see from Fig. 12 that the increase in net depletion rate constant of CO_2 anions (compare curve ii with iii) or the decrease in net depletion rate constant of cations (compare curve iv with v) leads to the reduction in dimensionless concentration of CO_2.

What follows from the calculations is that the separation efficiency of CO_2 is a function of the electrical mobilities of anions and cations and the depletion and generation rate constants of CO_2 ions, and that there exists an optimal voltage to separate CO_2. Furthermore, the asymmetry in the dimensionless outlet concentration plotted as a function of applied voltage results from the changes in net generation and depletion rate constants with the voltage as well as the decomposition of CO_2.

The experimental separation efficiency is no better than 15% as shown in Figs. 4 to 7. Therefore, further improvement is required in order to use the separator as a practical tool for CO_2 contamination control. The CO_2 separation efficiency can be improved by increasing CO_2 anion generation rate as well as avoiding the ion depletion. This might be achieved by using more intensive ionization source and improving the structure of separator so as to make the flow in it well-defined bifurcating one without stagnant regions in the separator.

5. Conclusions

The following findings are obtained through the application of ionization separator to CO_2 separation.

i) CO_2 molecules in a helium and nitrogen can be separated mostly in the form of anion with the ionization separator although some fraction of CO_2 is decomposed by the soft X-ray irradiation.

ii) The CO_2 concentration relative to that without applied voltage increases with decreasing the inlet concentration as found in the separation of toluene of our previous works.

iii) Maximum relative concentration of CO_2 is 1.14 in helium carrier gas containing 2.4 ppm CO_2 at an applied voltage of 600V.

iv) The dependency of relative concentration on the applied voltage is qualitatively explained by the separation model that accounts for the transports of CO_2 cations and anions as well as neutral molecules together with the generation and depletion of these species.

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