Polarity Effect in DC Breakdown Voltage Characteristics of Pressurized Carbon Dioxide up to Supercritical Conditions

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Abstract—This paper deals with the effect of the polarity and gap geometry on dc breakdown voltage characteristics of a point-to-plane gap in supercritical carbon dioxide (SC CO_2) that is required to design a plasma reactor. In the experiments, the gap width d and the curvature radius of the point tip r were changed in the range of 80 to 300 μm and 50 to 170 μm respectively, and the CO_2 state was controlled within the gas, liquid, and SC phases. The experimental results showed that a remarkable polarity effect appeared under certain gap conditions. As a result, it was found that negative polarity and a higher stressed electrode are desired conditions for the dc plasma reactor since an active corona supplying rich chemical radicals appears in SC CO_2 under such conditions.

Index Terms—Carbon dioxide, corona discharge, DC discharge, mist, plasma reactor, polarity effect, supercritical (SC) fluid.

I. INTRODUCTION

SUPERCRITICAL carbon dioxide (SC CO_2) has been regarded as an attractive fluid in a wide range of fields such as chemistry, heat engineering, medicine, and pharmacy because of its unique characteristics and high potential in materials processing at about room temperature [1]–[8]. With regard to electrical discharges in SC CO_2, Ito and Terashima [9] studied the electric discharge characteristics in SC CO_2 using a gap of microorder, and observed the unique phenomenon in which the local minimum of the breakdown voltage (BDV) appeared near the critical point. Additionally, the authors have studied prebreakdown phenomena of a nonuniform field gap in SC CO_2 in order to develop a plasma reactor using SC CO_2 in which the synergistic effect of the reactive species such as electrons, ions and radicals by the plasma and unique characteristics of SC CO_2 might be expected. In a previous paper [10], the authors reported experimental results on the breakdown characteristics of negative dc point-to-plane gap in compressed CO_2 up to the SC pressure.

Generally, the breakdown events depend on the electrode geometry, the state of the background fluid, and the polarity of a higher stressed electrode. Studies on the effect of such parameters on the BDV characteristics are required to understand the breakdown mechanism and for design of the plasma reactor. A few researches have been carried out in order to explore the polarity effects in CO_2 gas [11], [12]. With respect to the polarity effects in SC fluids, no research has been done on SC CO_2 although interesting data have been reported for SC helium [13].

In this paper, we have focused on exploring the polarity effect in dc breakdown characteristics of pressurized CO_2 up to the SC state under various electrode geometries. Additional studies regarding the polarity effects in CO_2 liquid are conducted as well.

II. EXPERIMENTAL SETUP AND METHOD

The schematic diagram of the experimental setup is shown in Fig. 1(a). The test chamber, which is adjustable up to the pressure of 30 MPa, is the same one as used in the previous study [10].

All breakdown experiments are conducted with a point-to-plane geometry. The gap width d and curvature radius of the point tip r are adjusted in the range of 80 to 300 μm and 50 to 170 μm, respectively, to investigate the effect of electrode geometry. The tip radius is determined from the measured radius of the circle which includes the point tip as depicted in Fig. 1(b). Since erosion of the point tip caused by the arc discharge current for breakdowns under a given electrode geometry and fluid temperature cannot be disregarded when the gap width is narrow, a current limiting circuit, (xiii) and (xiv) in Fig. 1(a), is included.

Setting of the CO_2 state is done by the same method performed in previous experiments [10]. That is, after the test chamber is evacuated, liquid CO_2 is transferred from a CO_2 container to the chamber with a high pressure pump. The temperature T and pressure P inside the chamber are controlled by a heater installed outside of the chamber and a backpressure regulator, respectively.

A series of BDV measurements is done by changing the pressure from high pressure (12 MPa) to low pressure (0.1 MPa) at two desired temperatures of 313 K and 298 K as shown along the thick solid lines in Fig. 2 where the fluid state changes from SC phase to gas at 313 K and liquid to gas at 298 K, respectively.
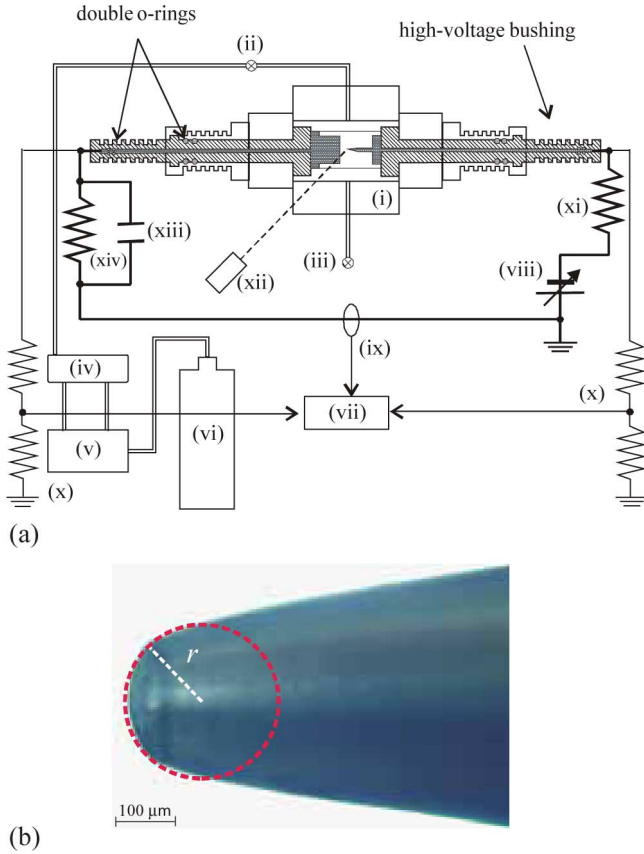
Fig. 1. Schematic diagram of the experimental setup and photograph of the tip of point electrode; (a) experimental setup: (i) test cell, (ii) CO_2 inlet, (iii) CO_2 outlet, (iv) syringe pump, (v) cooling system, (vi) CO_2 container, (vii) digital oscilloscope, (viii) dc power source, (ix) current transformer, (x) high voltage probe, (xi) charging resistors, 2 MΩ (xii) PM (xiii) capacitor, 17 nF and (xiv) damping resistor, 2 MΩ, (b) Typical point electrode and the configuration of curvature radius.
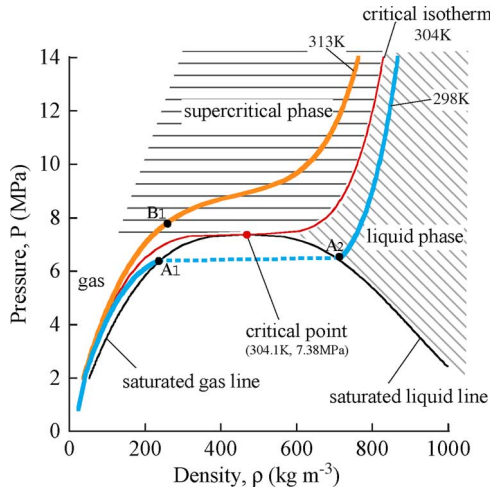

Fig. 2. CO_2 pressure-density diagram.

In order to minimize the influence of point tip erosion at the observed polarity effect, the positive and negative BDVs were alternately measured five times, respectively under a given temperature and pressure. After the pressure was set to the next step value, similar BDV measurements were repeated.

The measurement method of the breakdown and corona onset voltages (COVs) is basically the same as performed in previous experiments [10]. DC voltage is applied to the point electrode and increased normally at the rate of 2.5 kV/s by using a function generator (SG-4115, IWATSU, Japan) and an amplifier (HAR-50R0.6, MATSUSADA Precision, Inc., Japan). In the following explanation of the results, the polarity is indicated by the polarity of the point electrode. Although the discharge current, discharge light and acoustic emission were detected as the signal of corona discharge, the light measurement through the Sapphire glass window of the chamber by a photomultiplier (PM: No.722, ATAGO BUSSAN Co., Ltd., Japan) gave the highest sensitivity. Then, the PM diagnostic setup was utilized for the measurement of corona inception voltage in the same way as in previous experiments [10].

III. RESULTS AND DISCUSSION

Before presenting the experimental results, previous results using a negative point-to-plane electrode system [10] are summarized briefly to aid in later discussions.

In CO_2 gas at the temperatures of 298 K, 305 K, and 313 K, breakdown seemed to occur without a preceding stable corona discharge and the BDV increased with the gas density. The estimated BDVs, according to streamer theory [14], [15], agreed well with experimental values in the density region of 0.1 to 30 kg $\cdot$ m^{-3}.

The characteristics of the BDV versus density showed a different behavior in the medium state: the slope was largest in the gas phase and decreased with temperature in the liquid and SC phases and the measured BDV tended to scatter widely in the SC, especially in the higher density region. Corona discharge light was observed clearly in the SC phase, only.

A. BDV and COV

1) COV: In the gas and SC phases, the corona light intensity I_{ph} and applied voltage V_{app} are recorded simultaneously to measure the COV V_C and BDV V_B as shown in Fig. 3. Because the sensitivity of the PM has been increased, the oscillogram shows the superposition of the thermal white noise on the light signal which is illustrated by the solid line on the trace. As in the previous experiments with a negative point electrode, corona light is clearly observed only at the negative tip in the SC CO_2 as shown in Fig. 3(a). In this paper, the generation of a corona discharge is rarely recognized at the negative polarity in gas as shown in Fig. 3(c) and shown by the x-symbol in Fig. 4(a). The whole gap was irradiated by UV light produced by a deuterium lamp through the Sapphire glass window of 40 mm thickness and 15 mm aperture from the outside of the chamber to examine the effect of initial free electrons on the corona discharge events, but no clear change in the oscillogram of I_{ph} is recognized.

2) BDV: BDVs for both polarities are measured as a function of density as shown in Fig. 4, where the COVs are also plotted. In the figure, the average of the five measured BDVs and their deviation are illustrated by an open symbol and vertical bar. The following conclusions are drawn from the
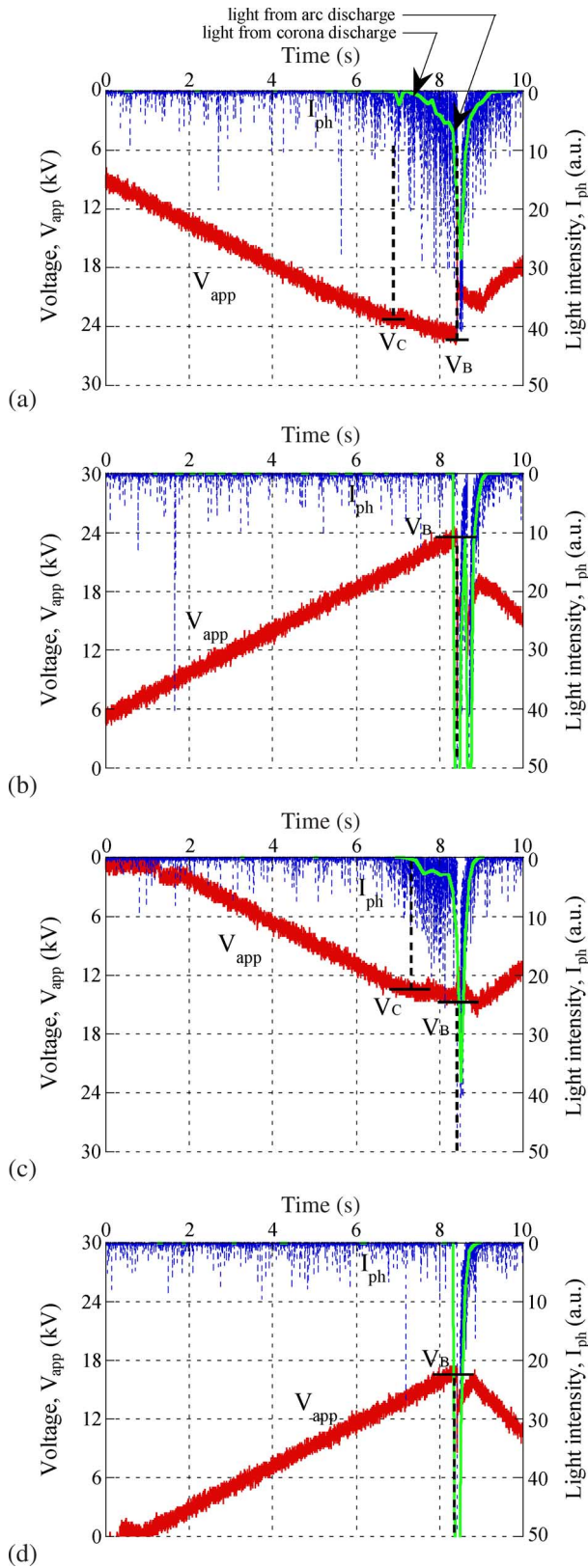
Fig. 3. Oscillograms of applied voltage V_{app} and discharge light emission I_{ph}; (a) negative polarity in SC phase (T : 313 K, P : 9.4 MPa, d : 250 μm, r : 80 μm), (b) positive polarity in SC phase (T : 313 K, P : 8.7 MPa, d : 250 μm, r : 80 μm), (c) negative polarity in gas (T : 313 K, P : 6.4 MPa, d : 250 μm, r : 80 μm), (d) positive polarity in gas (T : 313 K, P : 5.7 MPa, d : 250 μm, r : 80 μm).
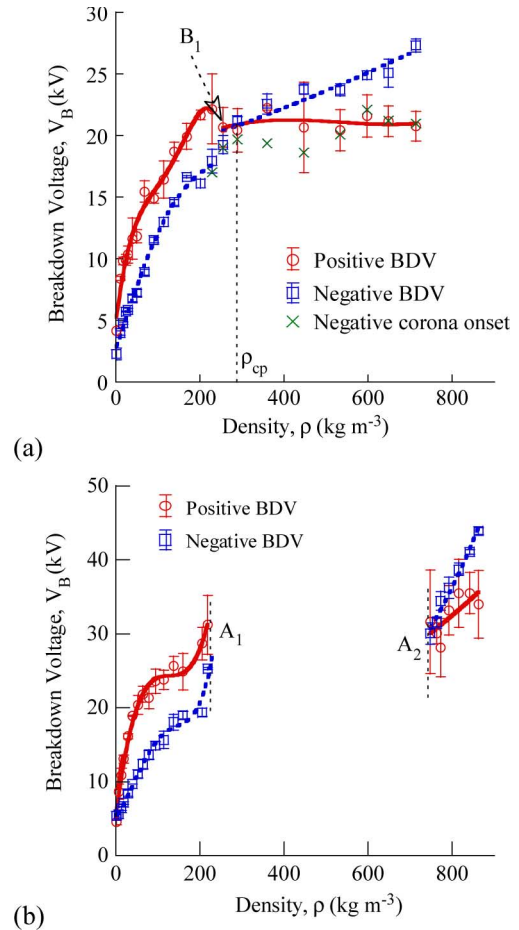
Fig. 4. Dependence of BDV on CO_2 density; (a) T : 313 K, d : 250 μm, r : 80 μm, (b) T : 298 K, d : 250 μm, r : 120 μm.

experiments performed under the conditions of $d = 250$ μm, $r = 80$ μm, and $T = 313$ K and of $d = 250$ μm and $r = 120$ μm and $T = 298$ K.

1) In the gas phase, the negative BDV is lower than the positive BDV at a given gas density. The slope of the BDV as a function of density changes with density and polarity: For a negative polarity, with increasing density the increasing rate of BDV decreases in the low density region and increases in the high density region, and for positive polarity, the BDV changes with density in a similar manner as negative polarity at 298 K but at 313 K it is curved convexly in all density regions. It could be argued that the negative BDV increases significantly near the saturation state at 298 K noted by the point A1 in Fig. 2 and the sub-SC state at 313 K noted by the point B1 in Fig. 2, but in contrast with this, the positive BDV at 313 K tends to show complete saturation as shown in Fig. 4(a).

2) At 298 K, both positive and negative BDVs change discontinuously at the phase shift from gas to liquid which is indicated by A1 and A2 in Figs. 2 and 4(b). The negative BDV rises about 20% there, but the positive BDV decreases slightly.

3) In liquid phase at 298 K, both the positive and negative BDVs increase with density and the slope of the negative BDV is larger than for positive BDV.

4) At 313 K, the discontinuity in the BDV curves near the sub-SC region which is indicated by B1 in Figs. 2 and 4(a), occurs for both polarities. The negative BDV rises but the positive one decreases stepwise.

5) In the SC phase, negative breakdown occurs after the preceding corona discharge, as stated before, and the negative BDV increases with density. However, the positive average BDV is almost independent on the density. The two characteristic curves of the average BDVs cross at a certain density ρ_{cp} as shown in Fig. 4(a).

6) In all experiments, the measured BDV tends to scatter widely in the liquid and SC phases and the deviation for positive BDVs is much larger than for negative BDV. It should be noted that the lowest BDV for positive polarity is somewhat close to the negative BDV in gas.

3) Discussion:

a) Gas: According to the above experimental results, in CO_2 gas a preceding corona is not clearly recognized and a positive BDV is higher than negative BDV. It is unclear why positive polarity should result in a higher BDV than the negative case. According to streamer theory [14], [15], if the breakdown is initiated at the instant of corona initiation, the polarity effect in the BDV should not appear.

The polarity effect in CO_2 gas up to 2 MPa using a standard lightning impulse voltage has been investigated in the power industry with a large coaxial cylindrical electrode system (70 mm in outer diameter, 170 mm in inner diameter). These experiments showed that positive BDV had a higher value than negative BDV at gas pressures from 0.4 to 1.0 MPa. However, it tended to saturate with the increase of the gas pressure, and at 2 MPa was equivalent to that of negative BDV. The variations in BDV were only large (exceeding 7%) for positive polarity at 2 MPa. The authors pointed out that this was caused by the electrode surface conditions rather than by the gas properties [11].

We believe that corona discharge might occur in the gas at positive polarity but the light emission is too weak to be detected by the PM measurement system. This is because the light detection is done through an observation window consisting of thick sapphire glass. If the glowlike corona studied by Hermstein [16], [17] appears, the corona light might be too weak to be detected by the measurement system and the BDV will be raised by the corona stabilization effect. Moreover, the scatter of the positive BDV data is large compared with that of negative polarity with the lowest positive BDV sometimes indicating a value close to the negative BDV which suggests the existence of the corona stabilization effect for positive polarity. Further investigations are needed to explain the cause of the above discrepancy in the BDV characteristics. Since the corona observation system in this paper needs to be improved, we will deal with the BDV characteristics in the following.

As stated in previous studies [10], negative BDV in low density gas up to 30 kg · m^{-3} could be estimated using streamer theory, but in the higher density region the estimated BDV deviated from experimental results due to the following reasons: 1) the experimental conditions are out of the effective region of E/P for estimation of the effective Townsend ionization coefficient; 2) the state of gas in the higher density region deviates from that of an ideal gas therefore the equation of state for the gas used in the theory could not be applied in the high density region; and 3) the formation of CO_2 clusters will contribute to a rise in the BDV.

The drastic increases in the negative BDV near saturation at 298 K and the sub-SC state at 313 K in Fig. 4 may be explained by electrostriction. When a high voltage is applied to a nonuniform field gap in saturation or sub-SC gases, the gases in the region of the highest electric field will condense by electrostriction. That is, the polar atoms are pulled to the regions of the highest field strength whenever a field is nonuniform. If we treat CO_2 gas as an ideal gas at which the density is proportional to pressure, we can calculate the field strength necessary to compress the vapor up to its saturation pressure P_s where a vapor mist may condense from the equation [18]

$$kT \bullet \ln\left(\frac{P_s}{P_o}\right) = f(\varepsilon_o \alpha_i) \bullet \left(E_{max}^2 - E_o^2\right) \tag{1}$$

where k, $f(\varepsilon_o \alpha_i)$, P_s, and P_o are Boltzmann's constant, a constant determined by the polarization of the CO_2 molecule, the saturation pressure and the ambient pressure, respectively, and E_{max} and E_o are the field strength at the tip of point electrode and in the gap space on the gap axis, respectively. Once the vapor mist is formed, the BDV will increase due to the mist [19], [20], and the effective density of the mixture of gas and mist increases again. It should be pointed out that this process may raise the negative BDV of the gas near the transition state from saturated gas to saturated liquid phase or sub-SC fluid. On the other hand it is believed that the existence of an unstable corona at positive polarity as assumed above would have less influence on the effect of the mist at sub-SC state.

SC CO_2: As stated in conclusion 5) of Section III-A2, negative BDV depends on density but positive BDV is almost independent of density in the SC phase. No research on the polarity effect in BDV characteristics in SC CO_2 has been conducted. The strong density dependence of negative BDV is probably related to the formation of a low-density region due to electron injection from the cathode that will suppress breakdown through the higher density region surrounding the low-density region and trigger a corona discharge. The corona discharge in the low-density region then produces a corona stabilization effect, i.e., that is the strong density dependence of negative BDV.

On the other hand, the weak density dependence of positive BDV may be based on the generation of a positive streamer, which progresses rapidly and gives a short spike signal of corona light on the oscillogram. It is likely not to have been depicted on an oscillogram with a very slow sweep trace because this signal is a very short impulse and hidden by the thermal noise in Fig. 3(b). Ishii and Noguchi [13] have studied the polarity effect in SC helium which has thermodynamically similar properties as those of SC CO_2, that is, high solubility,
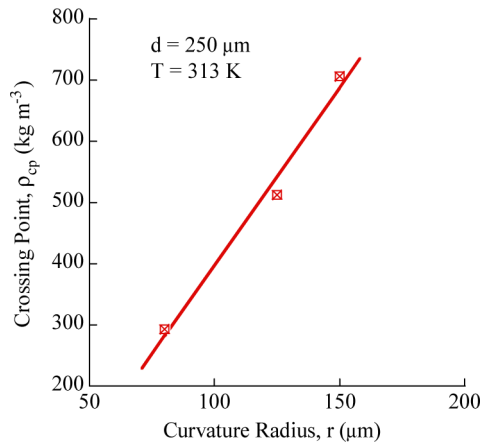
Fig. 5. Density at the crossing point ρ_{cp} versus point electrode curvature radius (T : 313 K, d : 250 μm).

low viscosity, and high diffusivity. According to their experiments, positive BDV voltage gradually increased with helium density even if corona was initiated before breakdown. On the other hand, negative BDV increased with density gradually and is lower than positive BDV in the density region $\rho < 30$ kg/m^3. In the higher density region of $30 < \rho < 83$ kg/m^3, negative BDV increased remarkably due to corona stabilization and is higher than positive BDV. Similar breakdown mechanisms as in the SC helium are supposed to occur in SC CO$_2$. CO$_2$ is a weakly electronegative gas and negative ion could be formed in it; on the other hand He is nonelectronegative gas and the electron bubble which has a low mobility compared with the electron mobility is formed in high density He. These differences in negative carrier behavior should be taken into account in the discussion of discharge mechanism in SC CO$_2$ and He at discussion.

B. Effect of the Gap Geometry on BDV at 313 K

1) Effect of the Point-Tip Radius r on BDV: DC BDVs for both polarities are measured as a function of CO$_2$ density when the gap width d and point-tip radius r are changed at a temperature of 313 K. The outline of the BDV characteristics is basically the same as that of Fig. 4 for both polarities. However, the value of ρ_{cp} increases linearly with r as shown in Fig. 5.

In Fig. 6, the BDVs are depicted as a function of r for four different densities. In the gas state of $\rho = 100$ and 200 kg/m^3, BDV tends to increase linearly with r but it shows a linear increase and then reaches saturation in the SC phase, $\rho = 400$ and 600 kg/m^3. These tendencies are independent of polarity.

2) Effect of the Gap Width d on BDV: In Fig. 7, the BDVs are depicted as a function of d for four densities. BDV increases with gap width d. As stated previously, negative BDVs are lower for the whole range for gap width d in the gas state of $\rho = 100$ and 200 kg/m^3, but becomes higher in the SC phase of $\rho = 400$ and 600 kg/m^3 and at wider gap widths as compared with positive BDV.

3) Discussion: When designing a plasma reactor using SC fluid, it is desirable that the voltage range for stabilizing corona discharges is wide, which will produce an abundant supply of reactive species for chemical reactions. From this viewpoint and
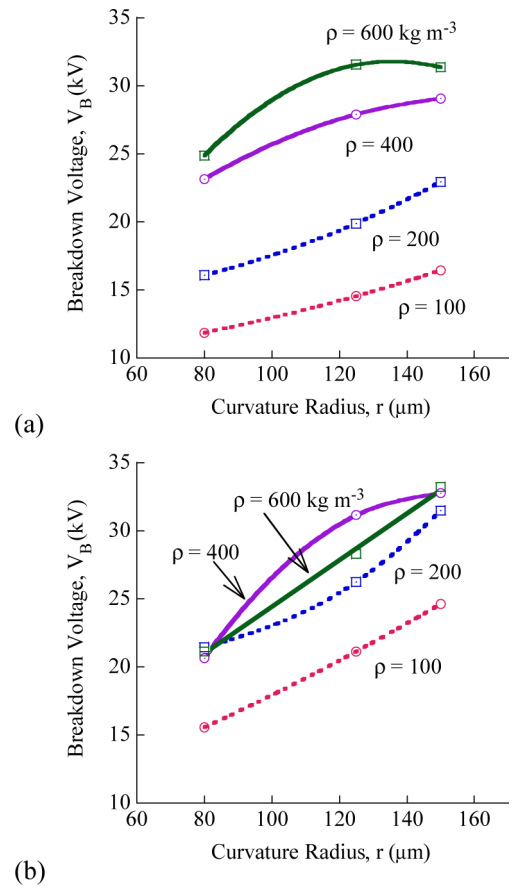

(a)

(b)

Fig. 6. BDV versus point electrode curvature radius for four CO$_2$ densities (T : 313 K, d : 250 μm); (a) negative polarity, (b) positive polarity.

the experimental results shown by Figs. 6 and 7, the following can be recommended for the polarity, gap geometry, and state of the fluid for the plasma reactor if we use dc voltage as a power supply.

1) Use negative polarity since it is easy to generate a stable corona within a wide voltage range and the BDV shows little deviation.
2) Use a small curvature radius for the high stressed electrode since it helps provide a stable negative corona.
3) Apply a wider gap width in the range of the presented experiments since a higher voltage may be applied to the gap. The desired gap width will then exist to produce the dense radical species for chemical reaction since the corona discharge can develop in the wider gap even if a higher voltage is applied. Determining desired gap width and estimating the volume fraction of the reactor filled with radicals by corona would be a future research subject.
4) Use higher fluid density since the voltage range providing a stable corona increases with an increase in density in SC CO$_2$ as shown in Fig. 4(a).

It is noted from Figs. 6 and 7 that the BDV in SC CO$_2$ is sensitive to the radius of the point electrode tip and less sensitive to the gap spacing. This may be due to the following reasons. In order to build up the complete breakdown, at least two conditions of the generation of initial electron and
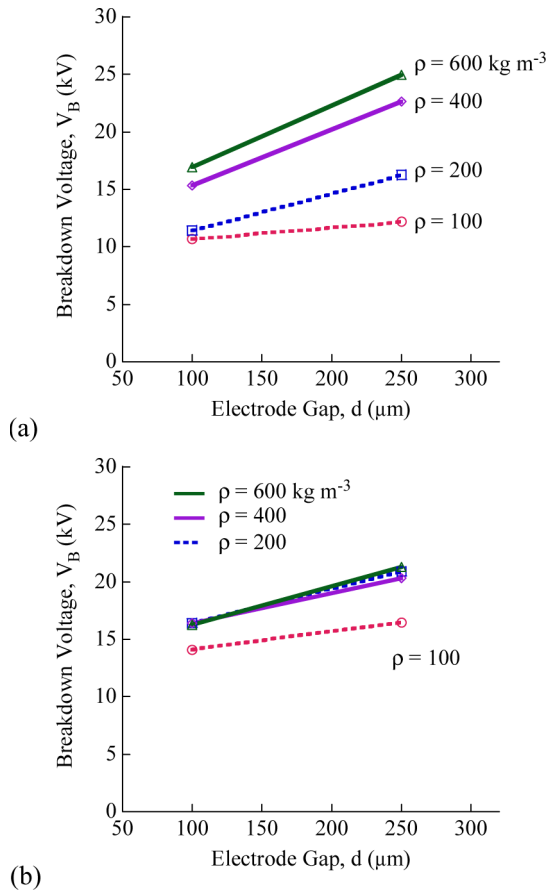
Fig. 7. BDV versus gap width for four CO_2 densities at a temperature of 313 K, ($d : 250$ μm, $r : 80$ μm) and ($d : 100$ μm, $r : 100$ μm); (a) negative polarity, (b) positive polarity.

the formation of discharge channel like a streamer should be satisfied. In the present experimental conditions, the voltage for the generation of initial electron around the point electrode is thought to be higher than the voltage for the development of the discharge channel. The electric field at the point surface that closely relates to the generation of initial electron is more sensitive to the point radius than to the gap spacing, as shown by (1) in the previous paper [10].

IV. CONCLUSION

DC BDV characteristics of point-to-plane gaps in SC CO_2 have been investigated experimentally to determine the design criteria for a plasma reactor using SC CO_2. Efforts focused on the effects of the polarity and gap geometry on the gap are studied to determine the desired conditions for the plasma reactor. The results can be summarized as follows.

1) In the gas state, negative BDV is lower than positive BDV and seems to be affected by the electrostriction effect in the higher density region near the saturation or sub-SC state.
2) In the gas state, positive BDV shows a wide scatter and its lowest value at a given density is sometimes close to that for negative BDV. It is believed that positive corona discharge might appear before breakdown although mea-

surements with a PM were not able to detect any light emission from the corona.
3) Near the phase transition points from gas to liquid or gas to subcritical phase, the characteristics of BDV versus density show a discontinuity at both negative and positive polarities.
4) In SC CO_2, the slope of BDV versus density for negative polarity is larger than that for positive polarity and both characteristic curves of BDV versus density cross each other at a certain density ρ_{cp} which increases with tip radius for higher-stressed electrode.
5) Negative discharges in SC CO_2 with a higher density are better for a plasma reactor than positive discharges.
6) A sharper tip of a higher stressed electrode is recommended for a plasma reactor but the desired gap width should be researched further to produce the most radicals.

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