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DC BREAKDOWN VOLTAGE OF CARBON DIOXIDE MEDIUM UNDER NEEDLE TO PLANE ELECTRODE *

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Abstract

This paper reports the experimental results on the corona onset voltage and breakdown voltage by positive and negative dc discharges in carbon dioxide medium within the pressure range of 0.1 to 15MPa under the needle to plane electrode. From the experimental results of dc discharge, negative corona discharges are observed more clearly in liquid and supercritical phase than in gas phase of carbon dioxide. However, in our experimental condition, positive corona discharge was not found for dc discharges. The breakdown mechanism in liquid can be classified into two categories on account of the bubble-triggered formation.

I. INTRODUCTION

Supercritical fluid (SCF) has received increasing importance in a variety of fields due to its high diffusivity (like gas) with improved mass transfer rates and high solubility (like liquid), and the operation can be manipulated by changing temperature and pressure. Carbon dioxide (CO_2) is mostly used as SCF because of its low critical temperature 304.1K and low critical pressure 7.38MPa, having significant physical as well as transport properties. Those properties of carbon dioxide make it widely used as an extracting solvent of various useful ingredients, reaction solvent and reaction catalyst [1, 2]. On the other hand, electric discharges have well known properties of high reactivity and high chemical activity. Over the past few decades, a considerable number of studies have been conducted on the research of electric discharge contributing to an environmental problem such as the decompositions of harmful gases, removal of toxic compounds namely dioxin [3-5]. However, very few attempts have been made at about the reaction of the plasma production in SCFs. Several studied have been made on the plasma production in supercritical carbon dioxide ($SCCO_2$) [6, 7]. The research of discharge phenomena in SCFs is an undeveloped field, and the prebreakdown phenomena are not well explained. Therefore, it is an important and attractive job to study of electric discharge plasma production in SCFs, and possibly offers significant and new ideas in future industrial fields. The objective of this work is to study the prebreakdown phenomena and breakdown characteristic in CO_2 medium at the experimental conditions from atmospheric to supercritical condition in order to development of a new chemical reactor that can be used for the production of discharge plasma in $SCCO_2$.

II. THERMODYNAMIC STATE OF CO_2

Figure 1 was obtained from the numerical calculations of the physical properties of CO_2 using equation of state [8], shows the different phases of CO_2 with the parameters such as density, pressure and temperature. The figure contains some density isotherm lines and regions. The isotherms below the critical temperature and pressure in lower density region are termed as saturated gas lines and that of in higher density region are saturated liquid lines. In the higher density region, the liquid phase is divided into two regions such as higher pressurized liquid region located above the critical pressure (P>Pc) and lower pressurized liquid region located below the critical pressure (P<Pc).

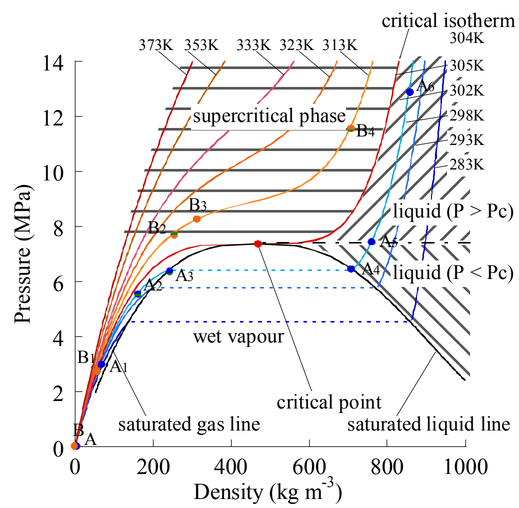

Figure 1. Pressure-density diagram of carbon dioxide

This experiment was carried out at 298K and 313K. The density isotherm at 298K indicates that the density of CO_2 increases with increase in pressure from gas to liquid phase, and that of at 313K the density increases from gas to supercritical phase. The figure also illustrates how the

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density at different regions such as lower pressured liquid, higher pressured liquid and SCF regions of CO_2 medium changes while it is locally heated at the tip of needle during the production of discharge plasma. In the region of lower pressurized liquid, the state at the tip of a point seems to shift in the direction where the density of CO_2 decreases with the Joule's heat from electric discharge plasma. In other words, lower pressurized liquid phase seems to move into gas phase through wet vapor region where the gas and liquid molecules are in fog condition. On the other hand, in the region of higher pressurized liquid, the same tendency is observed, but the higher pressurized liquid phase moves to SCF phase. This point of view deserves careful attention.

III. EXPERIMENTAL APPARATUS

A schematic diagram of the experimental setup is shown in Figure 2. The test reactor is made of stainless steel: SUS316 having compressive strength of 30MPa, the maximum temperature of 573K and the total volume of reaction cell of 1,300mL. Power lead is introduced through the center of long bushing made of peak resin, and the annular space is sealed off with double o-rings.

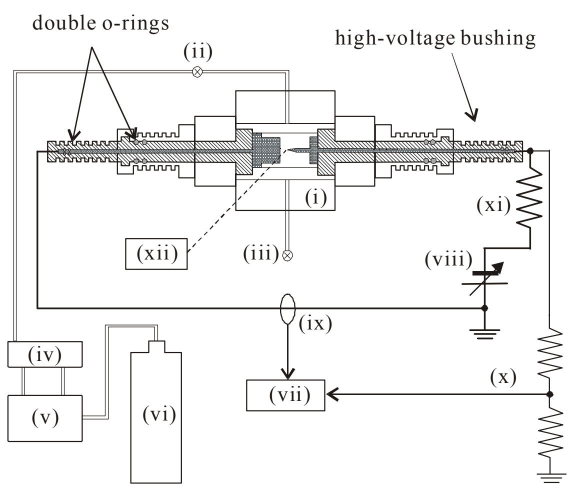

Figure 2. Schematic diagram of the experimental setup: (i) test cell, (ii) CO_2 inlet, (iii) CO_2 outlet, (iv) syringe pump, (v) cooling system, (vi) CO_2 container, (vii) digital oscilloscope, (viii) dc power source, (ix) current transformer, (x) high voltage probe, (xi) damping resistance, 2MΩ and (xii) photo multiplier

Liquid CO_2 from a cylinder with a siphon attachment was passed through a cooling head of high-pressure pump to the test reactor. A thermocouple and a backpressure regulator controlled the temperature and the pressure of the test reactor, respectively. After attaining the experimental temperature and pressure, it was kept for a night for steady state of carbon dioxide medium in test reactor. The experimental temperatures were 298K and 313K. At constant temperature, the experiment was performed at different pressures in descending direction. The negative/positive dc voltage from a high voltage stable dc power supply was applied to needle electrode at a rate of applied voltage 2.5 kV/s. The signal of corona light intensity measured by photo multiplier tube (PMT) goes into the oscilloscope of the shield room.

The relation of breakdown voltage to each pressure was converted to the relation of breakdown voltage vs. corresponding density because the ionization phenomenon in media is deeply related to mean free path that is inversely proportional to density of medium, it was numerically calculated by using the equation of state [8] at each experimental pressure. That is, the experimental results for breakdown voltages were shown in respect with the density changes of CO_2.

IV. RESULTS AND DISCUSSION

A. Prebreakdown phenomena and corona onset voltage

Figure 3a shows the typical negative corona onset voltage V_C and breakdown voltage V_B in supercritical phase at 313K. The discharge light intensity I_{ph} and the applied voltage V_{app} were obtained by additional experiments using a short gap of about 80μm and a high increase rate of voltage.

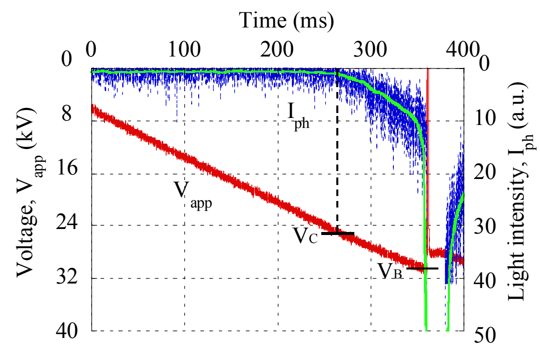

Figure 3a. Typical negative corona onset and breakdown voltages in supercritical phase at 313 K

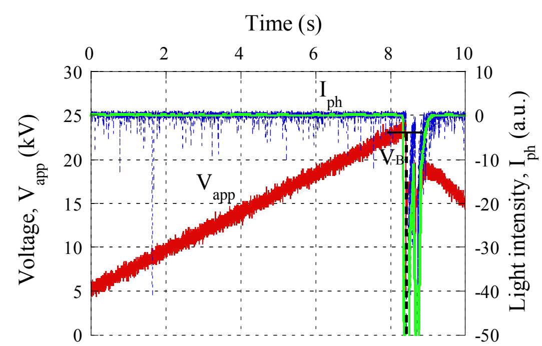

Figure 3b. Typical positive corona onset and breakdown voltages in supercritical phase at 313K

The corona light intensity is increased with the applied voltage. Once an electric discharge form shifts to arc from corona discharge, simultaneously with a dielectric breakdown, luminescence intensity is rapidly increasing and is surpassing the measuring range of an oscilloscope.

In our experimental condition, negative corona discharge has been observed in both supercritical and liquid phases, but it has not been so stable in gas phase. On the other hand, positive corona discharge has not been observed in any phases shown in Figure 3b.

B. Breakdown characteristic using negative dc

The measurement of breakdown voltages by negative dc discharge has been investigated using the electrodes gap 200 μm with a curvature radius of about 35 μm, and the increasing rate of the applied voltage was constant. As shown in figure 4, the line traced by the dotted line indicates the density dependence of breakdown voltages at 298 and 313K, and the points of A-A_1-A_2-A_3-A_4-A_5-A_6 and B-B_1-B_2-B_3-B_4 correspond with that in the figure 1, respectively. The density in media was calculated at the experimental conditions by the equation of state [8].

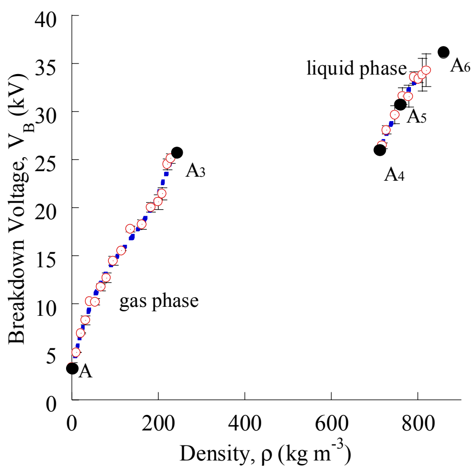

Figure 4(a). Breakdown voltage vs. density by negative dc discharge at 298 K

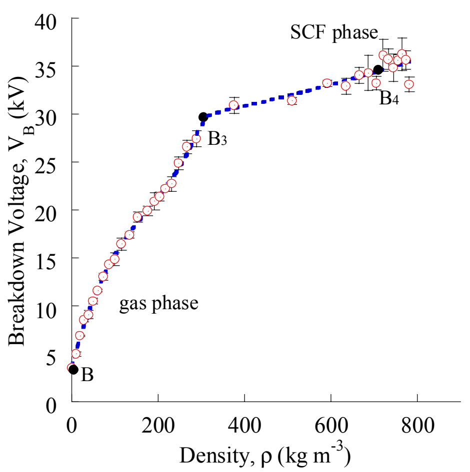

Figure 4(b). Breakdown voltage vs. density by negative dc discharge at 313 K

In figure 4a, breakdown voltage in gas phase conforms to the density changes of carbon dioxide up to point A_3 as in figure 1 then gas phase changes to liquid phase at point A_4 that goes to A_5 is the boundary of low-pressure liquid and high-pressure liquid, and attain high-pressure liquid phase at point A_6. According to this situation, breakdown voltage increased uniformly within the low-liquid region but beyond A_5 where the phase is high-pressure liquid, the increasing rate is comparatively lower than the value obtained in low-pressure liquid region A_4-A_5. However, in figure 4b, the gas phase moves to supercritical phase at point B_3 followed by B_4 without passing through liquid phase with increase in density of CO_2 at the temperature 313K. The increasing rate of breakdown voltages from B_3 to B_4 is different even in comparison with gas phase.

From the experimental results, it was observed that breakdown voltage curve is slightly gentler for higher temperature in liquid phase whereas the increasing rate of breakdown voltage in a supercritical phase is smaller than that in liquid phase. The explanation of the breakdown phenomena in supercritical phase is still ambiguous, it may be due to the peculiar characteristics of SCFs such as high diffusibility, thermal conductivity, cluster and so on, where the effect of bubble triggering is not effective. In contrast, breakdown voltage in lower pressurized liquid was influenced by vapor bubble locally generated, it is caused by suddenly heating liquid near the tip of needle due to ionization that will start with the electron emission from the cathode accompanying prebreakdown process. In higher pressurized liquid and supercritical phase, the same situation cannot happen.

C. Breakdown characteristic using positive dc

The measurement of breakdown voltages has been carried out using a different gap and curvature radius against negative dc discharge: Both the gap length and the curvature radius of the electrode are about100μm. In case of positive dc discharge shown in figure 5, it turned out that the fluctuation of breakdown voltages in liquid and supercritical phase is larger than the negative one.

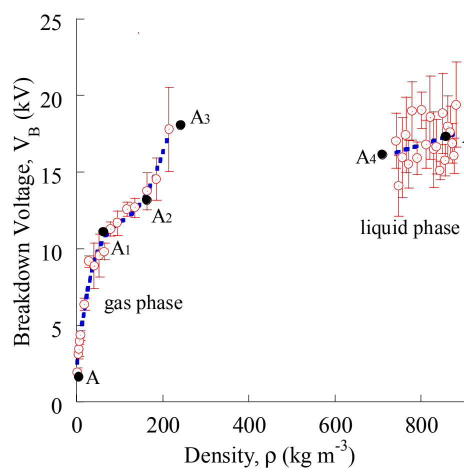

Figure 5(a). Breakdown voltage vs. density by positive dc discharge at 298 K

In both of gas phases at 298 and 313K, the line traced of breakdown voltage seems to make stairs in the area of A_1-A_2-A_3 and B_1-B_2-B_3. It might be inferred from the effect of the short gap. In contrast with the case of negative dc discharge, the slope of breakdown voltages in liquid and supercritical phase remains roughly flat in the area of A_4-A_6 and B_3-B_4 shown in figure 5.

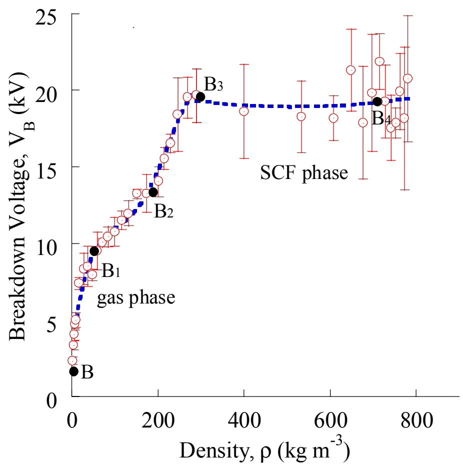

Figure 5(b). Positive breakdown voltage at 313 K

D. Polarity effect of dc discharge

In order to study the polarity effect in non-uniform electrode, the experiment by positive and negative dc discharges has been simultaneously performed as trial data, which is in the experimental condition which corona discharge generates certainly, as shown in figure 6.

Normally, breakdown voltage of positive dc discharge is higher than breakdown voltage of negative one. However, in our experimental condition, the following results were obtained: It was found that breakdown voltage of positive discharge and negative discharge is crossed by about 270kg/m^3 of the density, which can exist of stable negative corona in a supercritical phase.

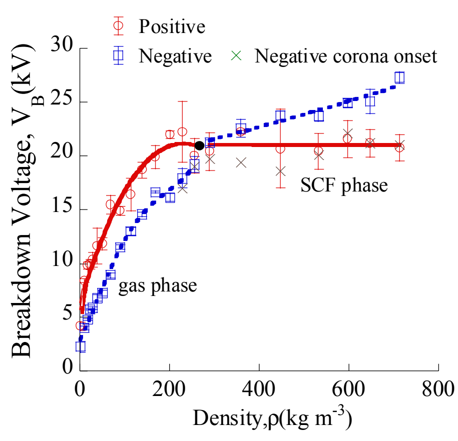

Figure 6. Positive and negative breakdown voltage by the polarity effect (trail data) at 313 K

V. SUMMARY

The breakdown phenomena and breakdown voltage characteristics of positive and negative dc discharges were investigated in carbon dioxide medium using needle to plane electrode. The following results were obtained:

1) Negative corona discharge is observed in both supercritical and liquid phases, but it is not so stable in gas phase. On the other hand, positive corona discharge is not observed in any phases.

2) Breakdown voltages are fluctuated significantly at the same experimental conditions in the case of positive discharge, whereas no such fluctuation is observed in the case of negative discharge.

3) The breakdown mechanism in lower-pressure liquid for negative discharge can be explained by the bubble-triggered effect that is not applicable for the higher-pressure liquid as well as supercritical phase.

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