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PULSED AND DC DISCHARGES IN SUPERCRITICAL CARBON DIOXIDE*

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Abstract

This paper reports the experimental results on the breakdown voltage and phenomena in carbon dioxide medium at 298, 304 and 373 K and within the pressure range of 0.1 to 12.0/20.0 MPa under the point-to-plane electrode using negative dc and pulsed discharge. From the experimental results of negative dc discharge, corona discharges with preceding as well as complete breakdown are observed more clearly in liquid and in supercritical phase than in gas phase of carbon dioxide. The calculated electric field intensity on the tip of point electrode at the corona onset voltage is about 450 MV/m; it suggests that corona is triggered by the field emission of electron. The breakdown mechanism of liquid phase can be classified into two categories in comparison with critical pressure of medium. On the other hand, in the experimental result of pulse electric discharge, the time delay of pulse forming and the relevance of the medium density were found.

I. INTRODUCTION

The point of view of green chemistry, the concern with the union of supercritical fluids (SCFs) and discharge plasma has been growing for the last several years. SCFs used as extraction as well as reaction media have received increasing attention in a variety of field due to the following features: a) the attractive properties of it (which are neither typical gases nor typical liquids but having intermediate properties) such as liquid like densities and gas like viscosities coupled with enhanced diffusion coefficients b) provide high solubility, improved mass transfer and high heat transfer is achieved by the large thermal conductivity [1, 2].

Among the industrial applications of SCFs, carbon dioxide (CO_2) is mostly preferred because it is safe, noncombustible, inexpensive, nontoxic, easily available, and has low critical temperature and pressure. On the other hand, the production of electric discharge plasma in atmosphere [3] or underwater [4] will generate various reactive species such as high-electrons, ions and radicals those may enhance chemical reaction in media. To put it more concretely, it is surely a very attractive idea for the production of electric discharge plasma combines with supercritical fluids technology. It may offer a possibility of new horizon in reaction fields near future.

Ito et al. studied the electric discharge characteristics in supercritical carbon dioxide using minute gap of micron order, and observed the unique phenomenon in which the local minimum of breakdown voltage appeared near the critical point, and presumed that this may be due to the cluster formation in supercritical carbon dioxide [5]. Lock et al. studied the characteristic of dielectric breakdown for CO_2 media by applying pulsed voltage with 100ns pulse width in a coaxial cylinder electrode system. In her study, it is demonstrated that the dropping of breakdown voltage to about one third of conventional Paschen's law estimated due to the inhomogeneity of carbon dioxide density near critical point [6].

Figure 1 shows a typical photograph of the arc discharge (thermal plasma) produced by our experimental apparatus using a Blumlein type Pulse Forming Network (B-PFN) at 10.1 MPa and 323 K. For instance, the light emitted from discharge plasma directly indicates its chemical activity.

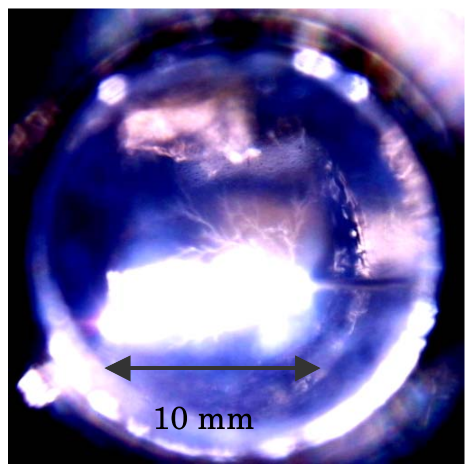

Figure 1. Typical photograph of arc discharge in CO_2

The purpose of this study is to explore discharge phenomena in carbon dioxide media using negative dc and pulsed discharge. Because of the development of a plasma reactor under supercritical fluids, knowledge of prebreakdown phenomena and breakdown characteristics in pressurized carbon dioxide up to supercritical condition is indispensable. The final goal of this research is the industrial application of discharge plasma in supercritical fluids medium.

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II. EXPERIMENT APPROACH AND EXPERIMENTAL CONDITION

In both pulsed and negative dc discharge, a schematic diagram of the base experimental setup is shown in Figure 2a. In cases where pulse electric discharge was generated, B-PFN was introduced into the input side of feedthrough. Test reactor is made of stainless steel (SUS316) having the maximum temperature 573 K, compressive strength of 30 MPa, the total volume 1,300 mL and an inspection window through which continuous monitoring was performed. Power lead was introduced through the center of long bushing made of peak resin, and the annular space was sealed off with double o-rings placed outer surface of it to prevent air leakage. The CO_2 used as an electric discharge atmosphere was connected with the test cell through the cooling system and pump. The B-PFN consists of 7 stages with winding ratio of pulsed transformer 1: 3, as shown in Figure 2b, which was used in case of pulsed discharge. The circuit parameters of B-PFN have a capacitor (1.7 nF) and an inductor (0.2 μH).

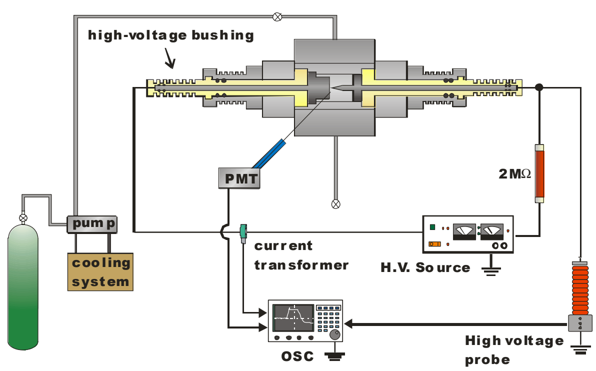

Figure 2a. Schematic diagram of the experimental setup

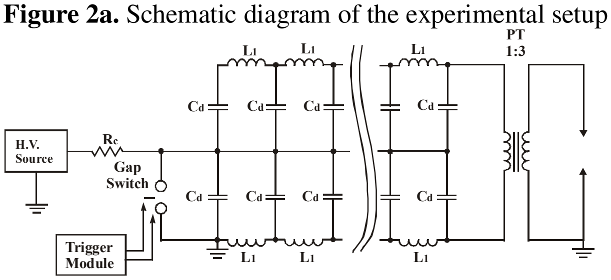

Figure 2b. The circuit of B-PFN with trigger module is connecting to high voltage power supply.

In case of negative dc discharge, the corona light intensity of the tip of the point electrode and applied voltage within electrodes were simultaneously measured using photo multiplier tube (PMT) in order to observe prebreakdown phenomena of electric discharge. A series of measurements in our experiment were carried out controlling pressure decreased gradually in the test cell to each experimental pressure that was controlled by backpressure regulator keeping constant temperature. The experimental conditions are shown in Table 1.

Table 1. Experimental conditions

Exp. condition	Exp.#1	Exp.#2
Applied source:	Negative dc discharge	Pulsed discharge
Electrode:	Point - Plane	Point - Plane
Gap length:	0.20 mm	10.0 mm
Temperature:	298K (Gas-Liquid)	304/373K (Gas-SCF)
Pressure range:	0.1 - 12.0MPa	0.1 - 20.0MPa

III. RESULTS AND DISCUSSION

The measurement of breakdown voltage has been carried out using two kinds of power supplies namely negative dc and pulse discharge. And also we have performed the observation of luminescence from corona discharge in order to investigate prebreakdown phenomena. In this work, the discharge phenomenon in CO_2 media has been illustrated under the three points: the prebreakdown phenomena, the breakdown characteristic using negative dc, and the breakdown characteristic using pulsed discharge. But at first, it is better to demonstrate how the phase changes as a function of molecular density in order to explain the electric discharges phenomenon.

Figure 3 shows thermodynamic state of CO_2 named the pressure-density changes of carbon dioxide within the temperature range 283 - 373 K. These curves were calculated using the equation of state [7], to demonstrate the characteristics of electric discharges. The bold lines in this figure indicate the experimental temperatures of 298 and 304, 373 K for the production of negative dc and pulsed discharge, respectively. The point corresponding to the critical temperature 304 K and critical pressure 7.38 MPa is the critical point. In order to understand breakdown phenomena of electric discharges, the liquid phase to be considered as low-pressurized liquid and high-pressurized liquid phases those are separated by the critical pressure line.

Another important fact is the ionization phenomenon in gas, liquid and super fluidity media is deeply related to a mean free path, and a mean free path is inversely proportional to density. From this figure, it is observed that the characteristics of curves above the critical isotherm and below it are different with change in temperature. The area between the right-upper side of saturated curve and the right side of critical isotherm is liquid but it would be better to mention low-pressure liquid and high-pressure liquid below and above the critical pressure line, respectively, on the viewpoint of electric discharges. This distinction is very useful to explain the mechanism of breakdown process. In the experimental condition of temperature constancy, if electric discharge is generated between electrodes, the phase state at the tip of a point locally can be changed the state by the electric discharge. For example, X_1 may be shifted to X_2-X_3-X_4, and X_5 to X_6. Therefore, it seems that the phase change may be occurred locally, as shown figure 3.

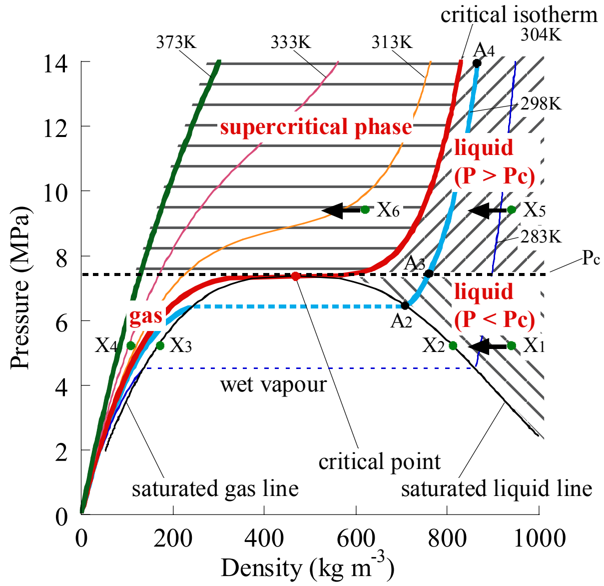

Figure 3. Pressure-density diagram of carbon dioxide

A. Prebreakdown phenomena

From the results in case of exp.#1, we have observed the phenomena of prebreakdown process. Figure 4 shows the typical oscillogram of corona onset V_c and breakdown voltage V_b at 13MPa, 298K (liquid phase). Corona light intensity and applied voltage are indicated by the blue line and red line on the trace, respectively.

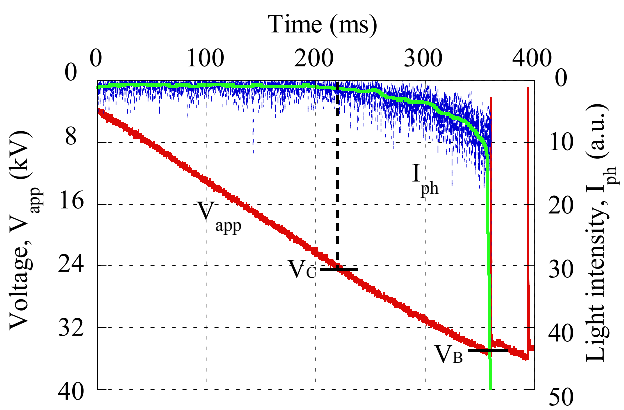

Figure 4. Typical negative corona onset at 13MPa, 298K (Liquid phase)

Negative corona discharge was stable in liquid and supercritical phase, the corona was observed before complete breakdown. According to our experiment, the corona onset voltages in the liquid and supercritical phase are almost unrelated to the state and density of a medium. The calculated electric field intensity on the tip of point electrode at the corona onset voltage is about 450 MV/m, which is enough to initiate the field emission. In these results, it suggests that corona is triggered by the field emission of electron.

B. Breakdown characteristic using negative dc

Figure 5 shows the density dependence of breakdown voltages at 298K in the area beyond pressure range 6.5MPa where areas indicate liquid phase while the density was calculated from the experimental conditions by the equation of state [7]. The increasing rate of the breakdown voltage in liquid phase is similar to the characteristics in gas phase. However, it is noted that the clear discontinuity appears near the boundary point at critical pressure Pc where the point of A_2, A_3 and A_4 corresponds to points as shown in figure 3.

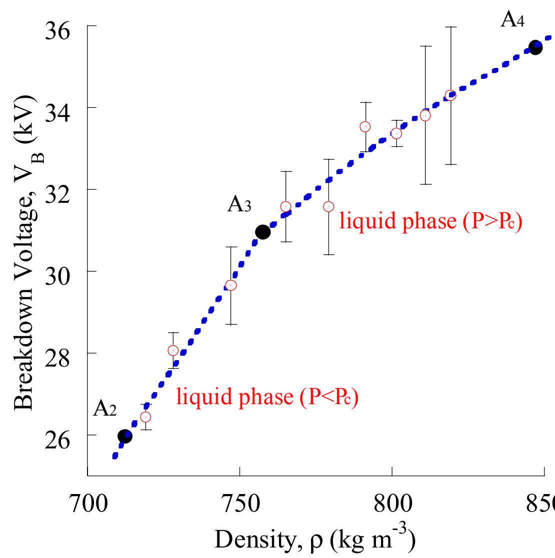

Figure 5. Negative breakdown voltage at 298 K in the area beyond pressure range 6.5MPa

It is very interesting phenomenon that the lower pressured liquid the state moves from the liquid to the gas, while at higher pressured liquid the state moves to the supercritical one. That is, the bubble generation may occur only in the lower pressured liquid. Briefly, Ionization may start with the electron emission from the cathode; in the lower pressured liquid, the bubble is generated due to the power by the emitted electrons and a partial discharge in the bubble will precede the complete breakdown. Breakdown voltage is influenced by vapor bubble generated that is caused by the gradually heating of liquid near the tip of point by electron emission followed by the decrease of density. In this case, partial breakdown was initiated due to presence of vapor bubbles in media, and finally, a complete breakdown was occurred having similar characteristics to that in gas phase. However, the process of vapor bubble generating was not occurred in supercritical phase because of different physical and transport properties with compare to gas and liquid. This figure revealed that the breakdown voltage curve raised steeper in low-pressure liquid phase than high-pressure liquid phase. The low-pressure curve was similar to the curve obtained in gas phase whereas high-pressure curve conforms to supercritical phase curve. Therefore, the mechanism of breakdown process in low-pressure and high-pressure phases is different where the breakdown process in low-pressure liquid phase may be influenced by bubble-triggering effect, but it is very difficult to consider that breakdown process is influenced by bubble triggering effect in high-pressure liquid phase.

C. Breakdown characteristic using pulsed discharge

In case of exp.#2, the pressure dependability of pulse generation was investigated using B-PFN. It revealed that the pulsed width is changed with each pressure. This clearly shows that the rising time of pulse formation and peak voltage are almost constant except at 0.1 MPa.

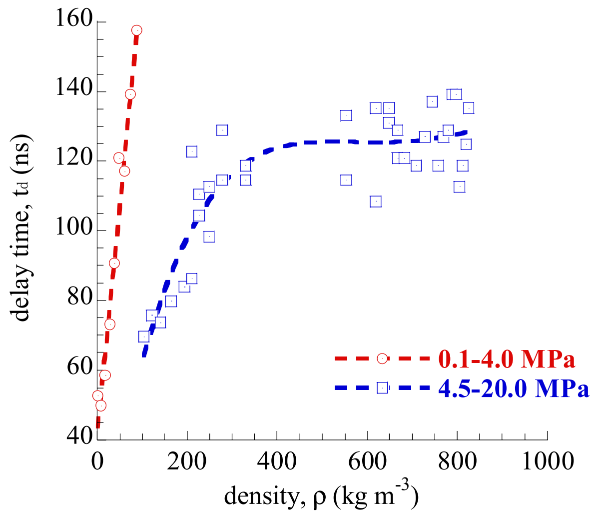

Figure 6a. The relation of delay time vs. density at 304K

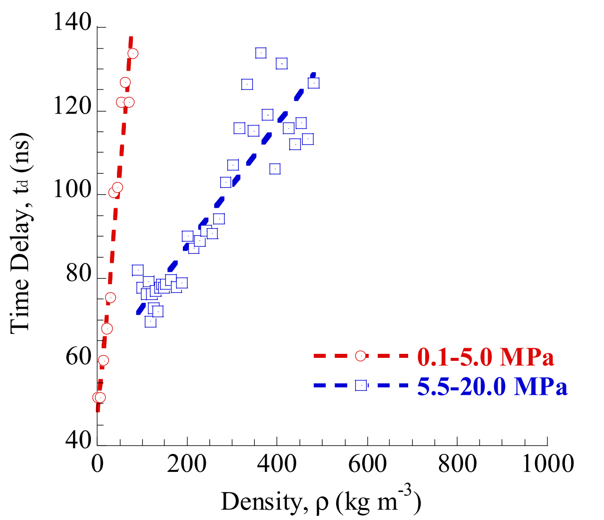

Figure 6b. The relation of delay time vs. density at 373K

From the viewpoint of above situation, it may say that Full Width at Half of Maximum peak voltage is termed as delay time of dielectric breakdown. Figure 6a, b shows the delay time of the dielectric breakdown to each density that was calculated using the equation of state [7]. In the beginning, the delay of dielectric breakdown increased until the density of about 90 kg/m^3 with the increase in CO_2 density, and decreased rapidly from 160 ns to 70 ns to density changes as shown in figure 3. This sudden fluctuation of delay time at this region (about 4-5MPa around) is not clear now. The same tendency was observed in the case of 373 K. It is reasonable to conclude that intimate correlativity is between the propagation of streamer and the density of media.

IV. SUMMARY

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

In case of negative dc discharge, corona discharge was stably observed in liquid and supercritical phase, whereas it was unstable in gas phase. The breakdown mechanism of liquid phase can be classified into two categories, bubble-triggered breakdown in pressures lower than critical pressure Pc and non bubble-triggered breakdown in pressures higher than critical pressure Pc, respectively from the results of negative dc discharge.

In case of pulsed discharge, the breakdown voltages were measured by changing pressure from 20 MPa to 0.1 MPa at desired temperatures 305 K and 373 K. As a consequence, it seems reasonable to conclude that there is intimate correlativity in the streamer formation and the density change in medium state.

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