Anomalous Plasma Temperature at Supercritical Phase of Pressurized CO_2 after Pulsed Breakdown Followed by Large Short-Circuiting Current

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ABSTRACT

The relation between breakdown characteristics and plasma temperature of a pulsed arc discharge in highly pressurized CO_2 was investigated up to a supercritical phase. A transient arc discharge was generated by applying nanosecond pulsed voltage with a rising rate of 0.7 kV/ns to a point-to-plane gap of 1 mm. The breakdown voltage, arc current, and consumption energy increased with the CO_2 density in the gas phase. However, they were constant at the CO_2 densities in the supercritical phase. The plasma temperature determined from the blackbody radiation ranged from 8000 to 12000 K and its dependence with respect to the CO_2 density was all similar to the breakdown voltage, arc current, and consumption energy characteristics except under the critical density of CO_2 (469 kg/m^3). Although the breakdown voltage, arc current, and consumption energy in supercritical phase were also constant against the CO_2 density, the plasma temperature demonstrated a local decrease around the critical density. The anomaly of the plasma temperature is consistent with the calculated result of the isochoric specific heat of CO_2 which has a local maximum around the critical density.

Index Terms — **Supercritical fluids, pulsed power, breakdown, pulsed arc discharge, optical emission spectroscopy.**

1 INTRODUCTION

RECENTLY, plasmas in supercritical (SC) fluids have been studied for use as an arc extinguishing medium [1, 2] and have drawn attention as a novel chemical synthesis method, e.g. phenol polymerization [3], metallic oxide nanomaterials [4], carbon nanomaterials [5], and nanodiamonds [6]. Understanding breakdown characteristics in the SC-fluids is of importance for the design of high-pressure chambers. In the supercritical fluid condition, a medium exceeds specific critical pressure and

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temperature. Consequently, an antagonistic action between a molecular attraction and a thermal agitation under a condition close to its critical condition causes an increase in density fluctuation which often affects breakdown characteristics in SC-fluids. A local decrease of breakdown voltage appears under SC conditions close to critical condition of medium for DC breakdown in SC-Xe, CO_2 and H_2O at the 1 μm gap [7], pulsed breakdown in SC-air at 25 μm gap [8], and DC breakdown in SC-He at 3 μm gap [9, 10]. However, the anomaly of breakdown voltage around critical condition disappears under longer electrode gap, e.g. 0.5, 1 and 4 mm gaps in SC-He under DC voltage application [11-13], 200 μm and 250 μm gaps in SC-CO_2 under DC voltage application [14-16], and 140 μm and 10 mm gaps in SC-CO_2 under pulsed voltage application [3, 17].

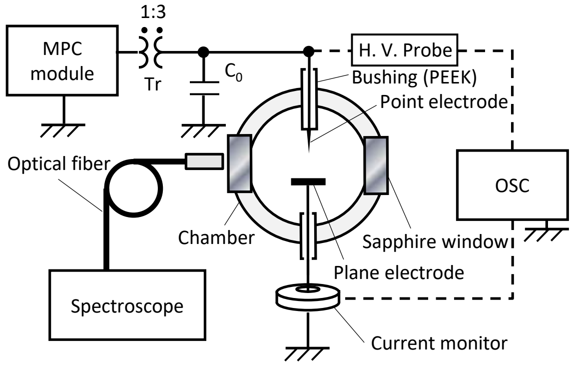
Elucidation of plasma behavior in SC-fluids plays a significant role in the development of power switches and chemical synthesis. Thus, few studies have focused on measurement of plasma spectrometry in high-pressure CO_2 including SC phase. Optical emission spectroscopy (OES) for discharges in SC-CO_2 was conducted by means of steady high-frequency plasma [18, 19]. In these reports, plasma temperature was approximately 3600–7000 K. Critical anomaly of plasma has not been confirmed under these experimental conditions. T. Kato, et al. reporting on results of OES with pulsed laser ablation plasma in SC-CO_2 [20], noted that emission intensity of atomic oxygen OI at 496 nm decreases with increasing pressure of CO_2 except around the critical point. The peak emission of OI around the critical condition of CO_2 (7.4 MPa and 304.5 K) shows a high intensity irrespective of the decreasing trend of the emission intensity of OI with increasing the pressure of CO_2. However, little is available on a transient arc plasma in SC-fluids.

In this paper, pulsed breakdown characteristics and plasma temperature were investigated to confirm critical anomalies of arc plasma in SC-CO_2. Relation between plasma temperature and physical properties of SC-CO_2 are discussed by the calculated isochoric specific heat of CO_2.

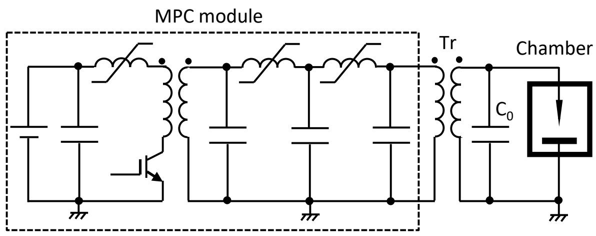
2 EXPERIMENTAL SETUP AND PROCEDURE

2.1 EXPERIMENTALCIRCUIT AND OBSERVATION SYSTEM

Figure 1a and b show discharge observation system and our experimental circuit (MPC3010S-25LP, SUEMATSU DENSHI Co. Ltd.). A point-to-plane electrode was adopted to facilitate breakdown within nanosecond order in high-density medium. The material of the needle electrode was tungsten, which has a high melting point. The gap distance inside the high-pressure chamber was set to approximately 1 mm. Nanosecond positive pulsed voltage was applied to the point-to-plane electrodes with a magnetic pulse compression circuit via a boosting pulse transformer. Voltage and current were measured with a high voltage probe (EP-100K, Nissin Pulse Co., Ltd. Japan) and a current monitor (MODEL 6585, Pearson Electronics Inc. USA), respectively. The emission spectra from the pulsed arc discharge were recorded with a multi-channel spectrometer (Glacier X, B&W Tek. Inc.: wavelength range: 350 - 1050 nm, slit width: 10 μm) which has an instrumental resolution of 1.1 nm. The calibration of spectral radiance was performed in advance. The integration time of the spectrometer was set at 3 s. The temperature in our high-pressure chamber was kept at 306 K while the pressure was varied between 1.5 to 8.0 MPa ($\rho = 27.9 – 614$ kg/m^3).



(a) Experimental setup overview



(b) Experimental circuit including MPC module

Figure. 1. Schematic diagram of experimental setup and observation system.

2.2 EXPERIMENTAL CONDITIONS

Figure 2 shows pressure variation of CO_2 as a function of the medium density of CO_2 at the given constant temperature. The condition with the temperature of 304.1 K and pressure of 7.38 MPa that is indicated as a filled circle is the critical point of CO_2. All experiments ranging from 27.9 to 613.7 kg/m^3 were done under the equivalent medium temperature of 306 K. The isothermal line of 306 K drastically varies around the critical point.

3 RESULTS AND DISCUSSION

3.1 BREAKDOWN VOLTAGE AND ARC CURRENT CHARACTERISTICS

Understanding of breakdown characteristics is important because it is closely related to the plasma behavior. Figure 3 shows a typical voltage and current waveforms under SC phase at 347 kg/m^3 when the needle to plane electrode was bridged. Pulse current of several hundred ampere began to flow at the moment of the breakdown. The voltage value immediately before the sudden decrease in voltage which occurs simultaneously with initiation the pulse current was defined as BDV. In addition, the first peak of pulse current was defined as an arc current. Figure 4 shows BDV and arc current as a function of the medium density of CO_2. Each data point is mean value of 5 measurements. Error bars indicate standard deviation of the shot-to-shot variation. Description of "medium density" instead of "pressure" is used to understand a discharge phenomenon that is related to a mean free path under a given field. Provided that equivalent voltage rise rate of 0.7 kV/ns was applied irrespective of the medium density. In the gas phase, the breakdown voltage increases with medium density up to 200 kg/m^3. The value of arc current depends on the BDV because the circuit becomes a series inductance-capacitance-resistance circuit. On the other hand, BDV and arc current are almost constant irrespective of medium density in the sub-critical and SC phases. Error bars in sub- and SC phase are greater than that of the gas phase. The variations of error bar of BDV is presumed to be caused by the CO_2 clustering. Because the experimental results of negative dc BDVs in high-pressure CO_2 above 30 kg/m^3 under non-uniform field are disagreement with a theoretical BDV estimation on the basis of streamer theory [14]. The difference of their experiment and calculation indicates that the CO_2 clustering affects the effective Townsend first ionization coefficient.

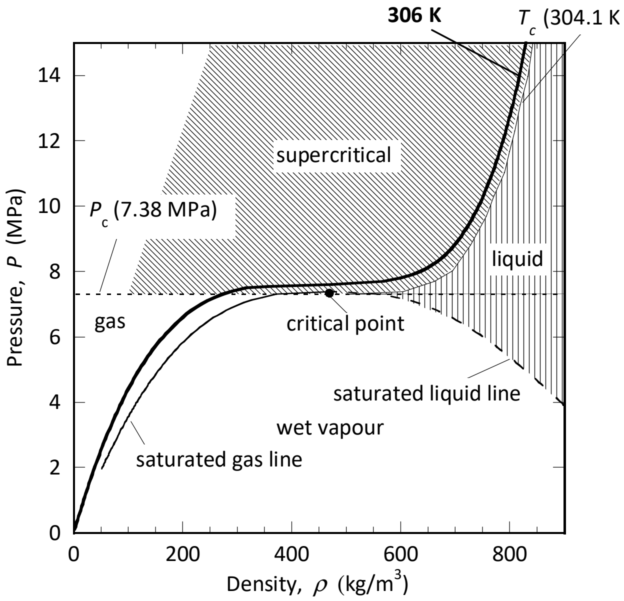


Figure 2. Phase diagram of carbon dioxide.

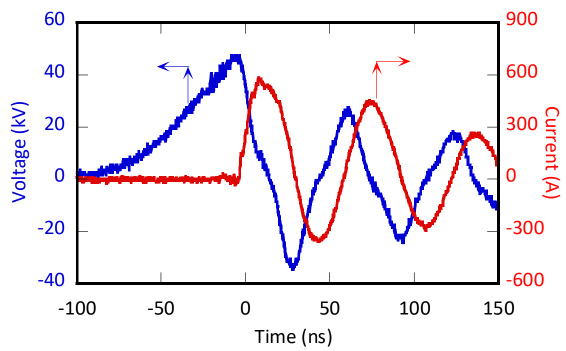


Figure 3. Voltage and current waveforms (ρ = 347 kg/m^3). The first peaks of voltage and current are determined as "breakdown voltage" and "arc current", respectively.

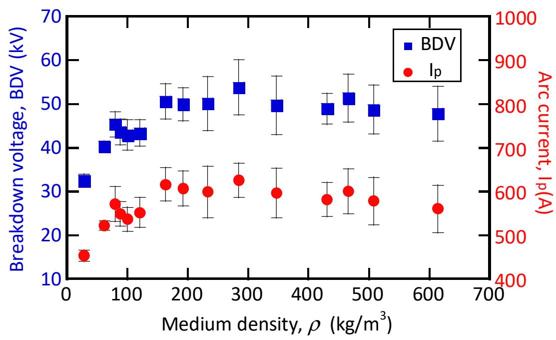


Figure 4. Pulsed breakdown voltage and pulse arc current as a function of the medium density of CO_2

3.2 CONSUMPTION ENERGY OF PULSED ARC DISCHARGES

A consumption energy is normally equal to the integral of a product of voltage and current, however, resistance and inductance of the circuit should be considered on a nanosecond phenomenon. In this study, a consumption energy of the arc discharge E_{arc} is calculated by following equation:

$$E_{arc} = \int_0^t \left(v(t) - Ri(t) - L\frac{di(t)}{dt} \right) \times i(t)dt \tag{1}$$

and

$$L = \frac{T_0^2}{4C\left(\ln \left| \frac{i_{s1}}{i_{s2}} \right|^2 + \pi^2 \right)}, \tag{2}$$

$$R = \frac{4L \ln \left| \frac{i_{s1}}{i_{s2}} \right|^2}{T_0}, \tag{3}$$

where $v(t)$ and $i(t)$ are voltage and current obtained from experiments; R and L are the resistance and inductance in the circuit; C is the capacitance of the capacitor in parallel with the needle-to-plane electrode; and T_0, i_{s1}, and i_{s2} are the period, first peak and second peak of the dumped oscillation current when the needle-to-plane electrode was shorten. Provided that if the condition of $R^2 < 4L/C$ holds true under R-L-C series circuit, the formulas (2) and (3) can be used. The calculated L and R by formulas (2) and (3) were 1.1 μH and 3.5 Ω, respectively. The dependence of E_{arc} calculated by the formula (1) on the medium density is similar to the characteristics of voltage and current waveforms, as shown in Figures 4 and 5.

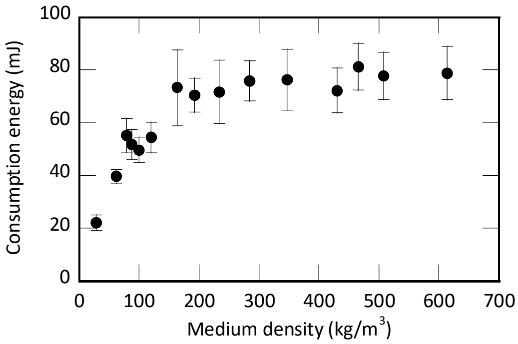


Figure 5. Consumption energy as a function of the medium density of CO_2.

3.3 VALIDATION OF LOCAL THERMAL EQUILIBRIUM AND DEPENDENCE OF SPECTROSCOPIC CHARACTERISTICS ON MEDIUM DENSITY OF CO_2

We estimated a time interval required for the discharge in SC phase to reach local thermal equilibrium (LTE) condition (τ_{LTE}) in this study. A collision frequency between electrons and ions ν_{ie} can be expressed as follows:

$$\nu_{ie} = n_i \sigma_{ie} v_{eth}, \tag{4}$$

and

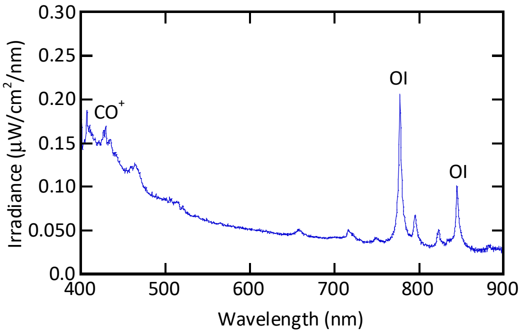
$$v_{eth} = \sqrt{\frac{8kT}{\pi m_e}}, \tag{5}$$

where n_i is the particle density (m^{-3}), σ_{ie} is the Coulomb collision cross-section (m^2), v_{eth} is the mean velocity of electrons (m/s), k is the Boltzmann constant (J/K), and m_e is the mass of an electron (kg). We calculated ν_{ie} on the basis of the estimated temperature of approximately 1 eV, which will be indicated in sub-section 3.4. The value of v_{eth} was calculated ~ 10^6 m/s by the formula (5). According to [21], the σ_{ie} is estimated to be ~ 10^{-17} m^2. In this experiment, the n_i is in the range from 10^{25} to 10^{27} m^{-3}. Consequently, the ν_{ie} is calculated to be 10^{14}-10^{16} Hz. Here, the ratio of a mass of a heavy particle m_b to m_e should be taken into account to estimate the τ_{LTE} as follows:

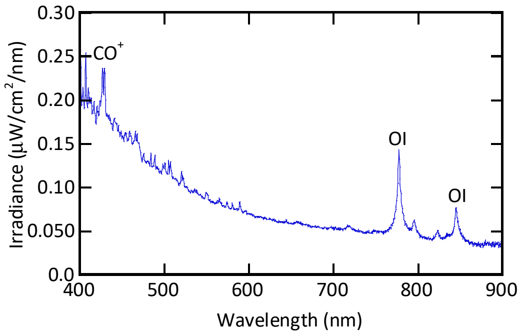
$$(\tau_{LTE})^{-1} = \nu_{ie} \frac{m_b}{m_e}. \tag{6}$$

The calculation result of m_b/m_e is approximately 10^4, e.g. an atomic oxygen. Consequently, the τ_{LTE} can be estimated in the range form 1 ps to 1 ns. In this study, the discharge condition is supposed to reach to the LTE immediately after the moment of breakdown because of the quite short τ_{LTE}. The establishment of the LTE made the heavy particle temperature T_h (K) equal to the electron temperature T_e (K) and allowed us to use the plasma temperature T (K) (~ T_h ~ T_e).

Figures 6a and b show OES of the pulsed arc discharge in the gas phase (61.2 kg/m^3) and SC phase (466.1 kg/m^3), respectively. The collective lens was focused on the needle tip. The intensity of the emission indicates a spectral-radiance (μW/cm^2/nm). Both spectra are characterized by continuum radiation due to plasma recombination of free-bound



(a) gas phase



(b) supercritical phase

Figure 6. Optical emission spectra of pulsed arc discharge under high pressure CO_2. (a) high pressure gas phase at 61.2 kg/m^3. (b) supercritical phase at 466.1 kg/m^3.

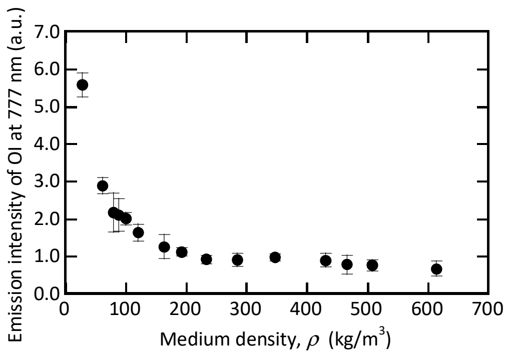


Figure 7. Variation of the emission intensity of OI (777 nm) as a function of the medium density of CO_2.

transition and free-free transition (bremsstrahlung). On the other hand, the three spectra are also characterized and are resulted from dissociation of CO_2, such as positive carbon monoxide ion CO^+ at 427 nm and atomic oxygen OI at 777 and 845 nm. The spectrum indicates the pulsed arc discharge leads to active CO_2 dissociation and O excitation. To confirm a dependence of line intensity on the medium density of CO_2, the largest line at 777 nm was measured as a function of the medium density, as shown in Figure 7. The error bars indicate standard deviation of shot-to-shot variations. The density dependence of light emission clearly changed around the 200 kg/m^3. In the gas phase (27.9 - 200 kg/m^3), the intensity decreases with medium density. Meanwhile, it is almost constant independent of medium density including the SC phase (above 345 kg/m^3). Although the arc current increases with medium density from 27.9 to 200 kg/m^3 (see Figure 4), the intensity of OI decrease with the medium density. This is because a near-resonant collision owing to increase of pressure of the CO_2 is supposed to be dominant compared with an increase of the arc current [20].

3.4 PLASMA TEMPERATURE OF PULSED ARC DISCHARGE

As stated in sub-section 3.3, the plasma temperature T can be defined for the present arc discharge. Figure 8 shows the result of OES of the pulsed arc discharge plasma acquired in SC-CO_2 (347 kg/m^3) and simulated continuous spectrum. Under such high pressure, the continuous spectrum of the arc is estimated from the Planck's blackbody radiation law according to the following equation [22]:

$$M(\lambda, T) = \frac{c_1}{\lambda^5\left(\exp\left(\frac{c_2}{\lambda T}\right)-1\right)}, \tag{7}$$

where the first radiation constant $c_1 = 3.7415\times10^{-16}$ Wm^2, the second radiation constant $c_2 = 1.4388\times10^{-2}$ mK, and λ = light wavelength in nm. T in formula (7) indicates the surface temperature of the arc. In this study, the surface temperature is treated as the plasma temperature of the arc discharge. The simulated blackbody emission spectrum obtained as a function of T by the method of least squares is indicated by a dashed line (T = 11200 K) as shown in Figure 8.

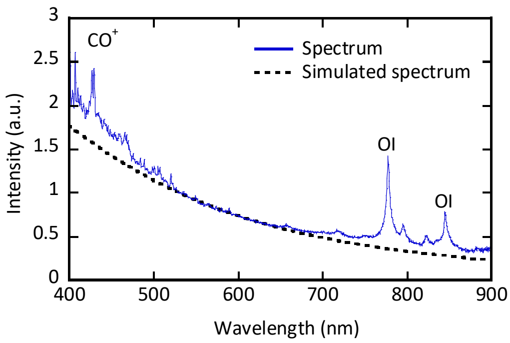


Figure 8. Optical emission spectra of pulsed arc discharge measured at (ρ = 347 kg/m^3) and simulated spectrum on the basis of a Planck's blackbody radiation law.

Figure 9 shows the T as a function of the medium density. The error bars indicate the standard deviation of the shot-to-shot variation. The T has local minimum around the critical density ρ_C (469 kg/m^3), although the value of BDV, I_p, and E_{arc} are constant irrespective of the medium density in the range of SC phase. The local minimum of T is presumed to be caused by the characteristic of isochoric specific heat C_v of CO_2, which has a local maximum at critical density. Here, the relation between T and C_v is discussed by a heat transfer. In general, a basic formula of heat transfer in a material is expressed as

$$Q = C_v\frac{\partial T}{\partial t} - div(K\cdot gradT), \tag{8}$$

where Q is the joule heat (W/m^3) and K is the thermal conductivity (W/(mK)). As indicated in sub-section 3.3, the τ_{LTE} was 1 ps to 1 ns in this study. As a consequence, the second term of the right side in formula (8) can be neglected because the time scale of the first term of (8) depending on the τ_{LTE} is 5-8 orders of magnitude less than that of the second term. Here, Equation (8) can be approximated as follows:

$$Q \approx C_v\frac{dT}{dt}. \tag{9}$$

Equation (9) can be integrated from zero to τ_{LTE} under an assumption that the C_v is constant for sake of simplicity because the variation of C_v is presumed to be small in comparison with the variation of T,

$$T_{LTE} = \frac{q_{LTE}\tau_{LTE}}{C_v} + T_0, \tag{10}$$

where q_{LTE} is joule heat per unit volume within τ_{LTE}, T_{LTE} is the increased temperature of arc discharge within τ_{LTE}, and T_0 is the initial temperature. The $q_{LTE}\tau_{LTE}$ means injection energy into the discharge channel. From the formula (10), the C_v bears an inverse relation to the rising ratio of T_{LTE}. The calculation result with C_v of CO_2 at 306 K [23] as a function of medium density is shown in Figure 9. The value of C_v has a local maximum around the critical density, called a critical anomaly. The calculation result implies that the local maximum of C_v causes the local minimum of T. On the other hand, the E_{arc} in SC phase is constant irrespective of medium density even though the T has the local minimum value close to the critical density. The disagreement between E_{arc} and the decrease of T at critical density is presumed to be caused by an anomaly of C_v. We have inferred that the energy which does not contribute to the rise of T was consumed by producing reactive plasma close to the critical density, because the production of carbon nanostructured material using a microplasma with dielectric barrier discharge close to the critical density of CO_2 is larger than that of under other SC conditions [24].

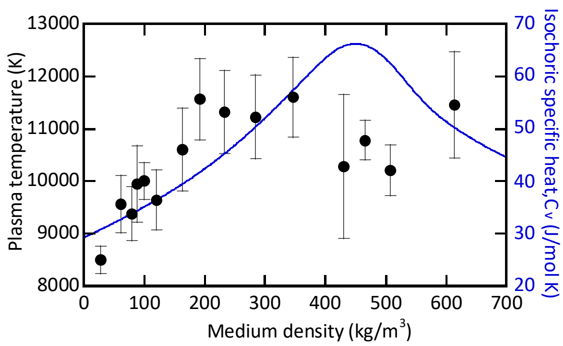


Figure 9. Plasma temperature and isochoric specific heat of CO_2 as a function of medium density of CO_2.

4 CONCLUSION

Nanosecond pulsed arc discharge plasma in high pressurized CO_2 including in the supercritical (SC) phase was investigated by spectroscopic analysis. A comparative investigation of the breakdown voltage, arc current, consumption energy, and plasma temperature was performed with varying CO_2 density. The obtained results are summarized as follows:

(1) A change in the CO_2 density dependence of breakdown voltage, arc current, and consumption energy appeared around the subcritical phase when going from gas to SC phase. In the gas phase, they increase with CO_2 density. Meanwhile, in the SC phase, they are almost constant irrespective of CO_2 density.

(2) The optical emission spectra were characterized by continuum radiation and several line spectra, such as positive carbon monoxide ion CO^+ at 427 nm and atomic oxygen OI at 777 and 845 nm independent of CO_2 density. However, the intensity of OI at 777 nm decreases with CO_2 density in the gas phase and keeps constant low intensity in SC phase.

(3) Plasma temperature T was estimated by fitting blackbody radiation on the basis of Planck's law. The dependence of T on CO_2 density is similar to breakdown voltage, arc current consumption energy except for the condition of close to the critical density. The T has the local minimum value close to the critical density. This anomaly is explained by calculation result of isochoric specific heat, which has a local maximum around critical density.

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