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Transient-Spark Discharge in $N_2/CO_2/H_2O$ Mixtures at Atmospheric Pressure

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***Abstract*—Transient spark, which is a novel type of streamer-to-spark transition discharge, was investigated in $N_2/CO_2/H_2O$ mixtures. It has a pulsed character with short (∼100 ns) pulses of high current amplitude (∼1 A) with repetitive frequencies of some kilohertz, and it generates nonequilibrium plasma. Emissions of N_2 first and second positive systems, CN violet system, NH, and atomic N, O, and C lines were detected. Two distinct discharge modes at low and high frequencies with different properties were found.**

***Index Terms*—Emission spectroscopy, non-thermal plasma, transition discharges.**

THE $N_2/CO_2/H_2O$ mixtures representing a model prebiotic atmosphere of the Earth and a simplified flue gas from the natural gas combustion were studied recently [1]–[3]. We focused on the decomposition of CO_2 and the formation of organic species by transient spark (TS) in this mixture [3]. We used a novel type of streamer-to-spark transition discharge identified as "spontaneously pulsing transition discharge" or "TS," which has already been successfully applied for volatile organic compounds abatement [4].

The TS is initiated by a streamer, which transforms to the spark pulse due to the discharging of an internal capacity of the discharge chamber $C_{\rm int}$. When $C_{\rm int}$ is discharged, the current reaches a high pulse (∼1 A) amplitude, and the voltage drops to zero due to the fall on the external resistance R. Then, $C_{\rm int}$ is recharged by a growing potential on the stressed electrode. As soon as $C_{\rm int}$ is charged enough again, it triggers a new pulse. This process repeats with a typical repetitive frequency f of 0.5–15 kHz. The value of f increases with the growing applied voltage $U_{\rm o}$. Above a certain value of f, the TS may transform to a pulse-less glow-discharge regime [4], [5], with a constant current above 1 mA.

The experiments were carried out at room temperature and atmospheric pressure with N_2 and CO_2 gas flows of 0.32 l/min. The concentration of CO_2 (c_{CO_2}) in the N_2 buffer gas varied from 0 to 100 vol.%. Water was directly present in the discharge chamber. The low-voltage planar copper electrode was submersed. A stainless-steel needle was used as a high-voltage electrode. The distance between the HV electrode and the water surface was 6 mm.

The UV–VIS spectra were obtained by an optical system, leading to a two-channel emission spectrometer Ocean Optics SD2000 (200–1100 nm; resolution of 0.6–1.7 nm). The photographs of the discharge were taken by a digital camera Nikon E4300 with manually adjustable aperture and exposure time.

We studied the electrical and optical characteristics of TS in our mixture as a function of f and c_{CO_2}. As the CO_2 concentration grew from 0 to 100 vol.%, the threshold voltage for TS increased almost linearly from about 7 to 13.5 kV. As a result, the applied voltage required to achieve the desired f grew with the increasing c_{CO_2}. Further increase of $U_{\rm o}$ led to a monotonous increase of f.

In N_2, the discharge was contracted to a single dominant channel [Fig. 1(a)], and when f reached approximately 7 kHz, the TS converted to the pulse-less glow regime. In CO_2-containing mixtures, perhaps due to the electronegative character of CO_2, the transition to the glow regime did not occur in the studied frequency range. The discharge "spatial spread" increased with c_{CO_2} [Fig. 1(b) and (c)]. In CO_2-containing mixtures, the frequency became less stable above ∼3 kHz. With the same $U_{\rm o}$, the discharge could operate with several different values of f. This is related to the decrease of the discharge voltage $U_{\rm dis}$ and the appearance of pulses with a lower current. The TS randomly switched between high and low current pulses. However, as f increased, the low current pulses started to dominate, and the discharge contracted to smaller volumes [Fig. 1(d)–(f)].

The apparent transition to the high-frequency regime with low current pulses can be also observed in the obtained emission spectra. At low frequencies, CN emission dominated over N_2 second positive system, and atomic lines (C, N, and O) were clearly visible, whereas at 9 kHz, atomic lines disappeared, and N_2 first and second positive systems dominated in the spectra [Fig. 1(g)]. Moreover, the gas temperature in the discharge channel increased from around 600 K to 1300 K. The rotational temperature, i.e., gas temperature, was obtained by fitting the experimental spectra of the N_2 second positive system with the simulated ones by Specair program [6].

The explanation of this phenomenon requires further research on detailed kinetic mechanisms involved. However, the results obtained so far lead us to the following reasoning. With increasing f, more energy is deposited in the gas, thereby leading to the increased gas temperature, thus the decreased gas

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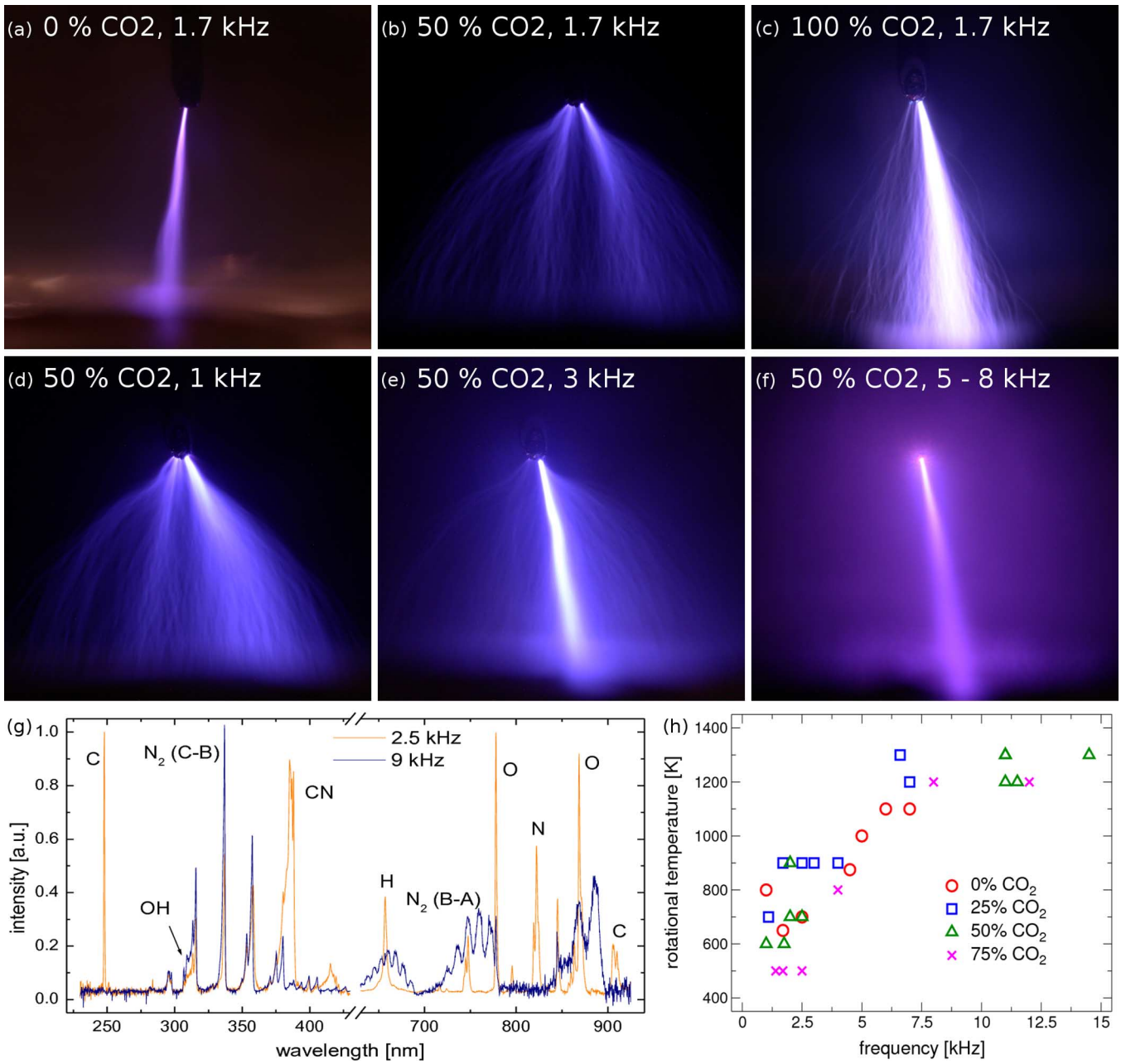

Fig. 1. Photographic images of TS in various gas mixtures and at different frequencies (exposure times: (a)–(c) 1 s and (d)–(f) 4 s). Obtained emission spectra of low- and high-frequency regimes and calculated rotational temperatures as a function of frequency.

density N. Since some threshold reduced electric field E/N is needed to initiate the TS pulse, E may be now lowered. This was indeed observed by the decrease of U_{dis}. Lower E results in a possible decrease of the mean electron energy, leading to the lower production rate of atomic and molecular radicals compared with the production rate of N_2 excited species.

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