LIF DIAGNOSTICS OF NO MOLECULES IN ATMOSPHERIC-PRESSURE DC STREAMER CORONAS USED FOR NO_X ABATEMENT

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1 Introduction

Recently, the streamer coronas have been proved to be one of the most efficient atmospheric-pressure non-thermal plasma methods used for NO_x removal from the flue gases. In order to optimize the non-thermal plasma processing of the flue gases, it is important to study directly in the plasma reactor the discharge-induced plasma-chemical processes responsible for NO_x removal. Laser-induced fluorescence (LIF) is a useful diagnostics method for in-situ observation of the phenomena occurring during the NO_x removal process (e.g., for monitoring the spatial and temporal behaviours of the species involved) [1-5].

The main objective of the presented work was LIF visualization of NO density (two-dimensional distribution) and monitoring the process of NO_x removal in various regions of the streamer corona discharge reactor through which a simulator of flue gas (air/NO) flowed.

2 Experimental

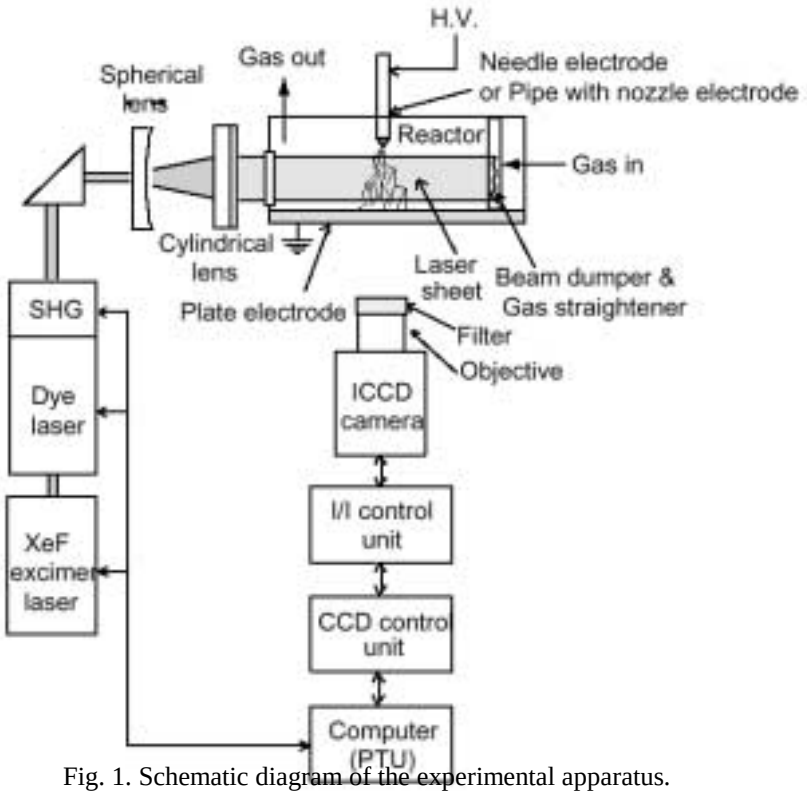

Fig. 1. Schematic diagram of the experimental apparatus.

Our investigation was aimed at measuring NO density distribution in the high-repetition dc corona discharge between a 3-cm gap electrode arrangement using LIF technique. This was an essential difference to the previous investigations [1-5] in which the LIF monitoring of NO molecules was carried out either after the single transient discharge or between two discharge pulses with a relatively long interval between them (low-repetition pulsed discharges, e.g. 10 Hz).

The laser induce fluorescence on NO ($X^2\Pi(v''=0) \leftarrow A^2\Sigma^+(v'=0)$) transition at 226nm was chosen for monitoring ground-state NO molecules. This transition was induced by irradiation of the NO molecules with UV laser pulses generated by a laser system consisted of a XeF excimer laser, dye laser and BBO crystal (Fig. 1). The laser pulses from the XeF excimer laser (Lambda Physik, Compex 150, tuned at 351 nm) pumped a dye laser (Lambda Physik, Scanmate) with Coumarin 47 as a dye, which generated the laser beam of a wavelength tuned around 450 nm. Then, the tuned dye laser beam pumped a BBO crystal in which the second harmonic radiation of a wavelength correspondingly tuned around 226 nm was generated. The 226-nm UV laser beam pulses of energy of 0.8-2 mJ and duration of about 20 ns were transformed into the form of the so-called laser sheet (width of 1 mm, height of about 30 mm) which passed between the electrodes with a 30-mm gap. LIF signal emitted at 90 degree to the laser sheet was imaged onto a gated ICCD camera (La Vision, Flame Star II) and two-dimensional distribution of NO molecules density around the discharge gap was recorded and them processed by a computer.

In the reactor either a pipe with a nozzle electrode or conventional needle electrode was used as the stressed electrode. The grounded electrode was a stainless-steel plate. The stainless-steel pipe had a diameter of 5mm, and the nozzle had an outer and inner diameter of 1.5 mm and 1 mm, respectively. The needle electrode was made of a brass rod (6 mm in diameter), the end of which had a tapered profile with the tip of a radius of curvature of 0.5 mm. dc high voltage with positive polarity was applied between the electrodes. Two corona discharge modes, glow corona or streamer corona could be generated in the gap. The discharge mode could be controlled by adjusting the tip of the stressed electrode and operating voltage. An air/NO mixture flowed along the reactor with a rate of 0.2-1 L/min. Concentrations of NO and NO_2 molecules at the inlet and outlet of the reactor were measured with a NO_x monitor (Hodakatest, Tests 33). This allowed the calibration of LIF signal in absolute units of NO concentration. The experiment was carried out at room temperature and atmospheric pressure.

3 Results

The typical LIF spectrum of NO molecules is shown in Fig. 2. The laser wavelength was scanned through NO ($X^2\Pi(v''=0) \leftarrow A^2\Sigma^+(v'=0)$) transitions in the range of 225-227 nm. In all further LIF measurements, the laser wavelength was fixed at 226.192 nm in order to obtain an intensive NO LIF signal. A linear relationship between NO LIF intensity at 226.192 nm and NO concentration was observed up to NO concentration of about 250 ppm. This linear relationship was then used for calibration of NO concentration in absolute units (ppm).

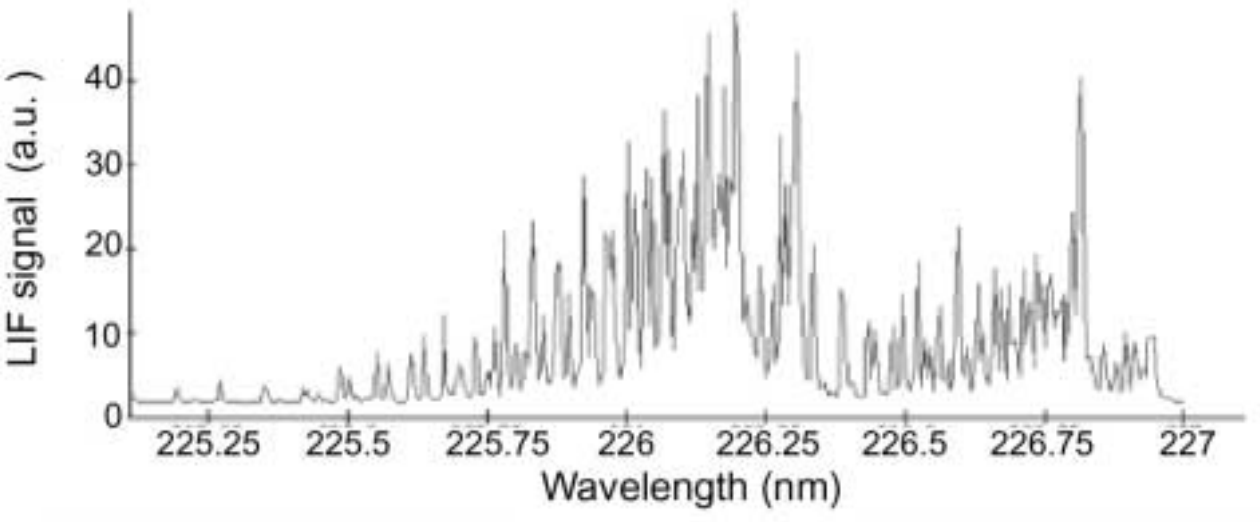

Fig. 2. LIF spectrum of NO molecule. Operating gas: NO (300ppm)/air.

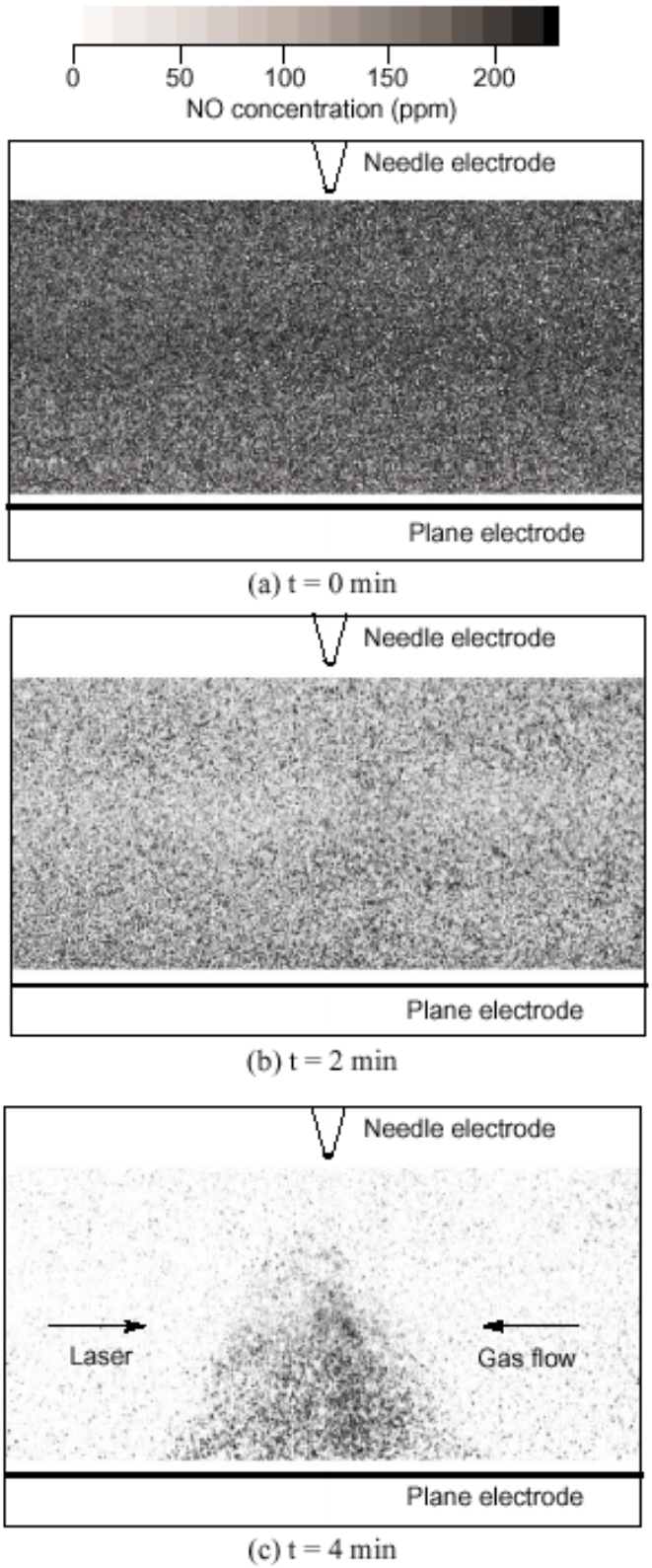

Fig. 3. Time evolution of the NO LIF image during the streamer corona discharge in the closed system. Time t=0 min and t=4 min indicate the start of the discharge and the steady-state, respectively. Operating gas: NO (200ppm)/air, flow rate 1L/min, applied voltage: 21kV.

In the case of streamer corona discharge mode, typical streamer repetition rate is about 5kHz. The streamer repetition rate increases with increasing applied voltage. The light emitted by the streamers can interfere the LIF measurements of NO concentration around the discharge zone. mainly because LIF signal is weaker than the discharge emission. However, although the streamer repetition rate in the dc corona discharge is much higher than in the pulsed coronas used in the previous investigations [1-5], the coincidence of the regular streamers with LIF signal was not the obstacle which might hinder the LIF measurements. A real obstacle for the LIF measurements can become streamers induced by the UV laser sheet pulses passing the electrode gap. As we also observed before [6], each laser pulse induced a streamer which appeared about 20-300 ns (depending on the position of the laser sheet in the gap) after the laser pulse and lasted over about 350 ns. Fortunately, in our measurements the LIF signals appeared almost immediately after the laser pulse and lasted over about 30 ns, while the laser induced streamers usually started later. Due to it, no coincidence between the laser pulse and the induced streamer occurred, and the LIF measurement was possible if an appropriate adjusting the recording delay and gate time of the ICCD camera was made.

Figs.3 (a-c) show two-dimensional distributions of NO concentration around the electrode gap in the reactor during dc corona discharge processing of a dry NO (200 ppm)/air mixture flowing with a velocity of 2 mm/s along the reactor. At time (t=0) the dc corona discharge started in the flowing gas uniformly polluted with NO [Fig. 3(a)]. After the start of the corona discharge, NO LIF signal became weaker compared that before the discharge. This indicated a decrease of NO concentration, i.e. NO removal from the processed gas mixture. However, astonishingly the NO concentration decrease was observed not only in the close vicinity of the streamer but also in both upstream and down stream regions of the reactor [Figs. 3 (b)]. After about 4 minutes a steady-state established (4-5 minutes is the residence time of the operating gas in the reactor) and practically NO molecules were found only in the region below the needle electrode [Fig.

3(c)]. However, their concentration was much lower than their initial concentration at t=0. It is worth underlining that in the steady-state NO molecules were removed also in the upstream region of the reactor.

The astonishing decrease of the concentration of NO molecules not only at the plasma region formed by the streamers but also in both upstream and downstream regions of the reactor may suggests that electrohydrodynamic flow (EHD) enhanced the NO removal in the corona discharge reactor used in this study. Since the operating gas flow velocity is low (2mm/s), the ionic wind (electric wind) could become dominant and caused the secondary flow (Fig. 4) that had a positive effect on the NO removal.

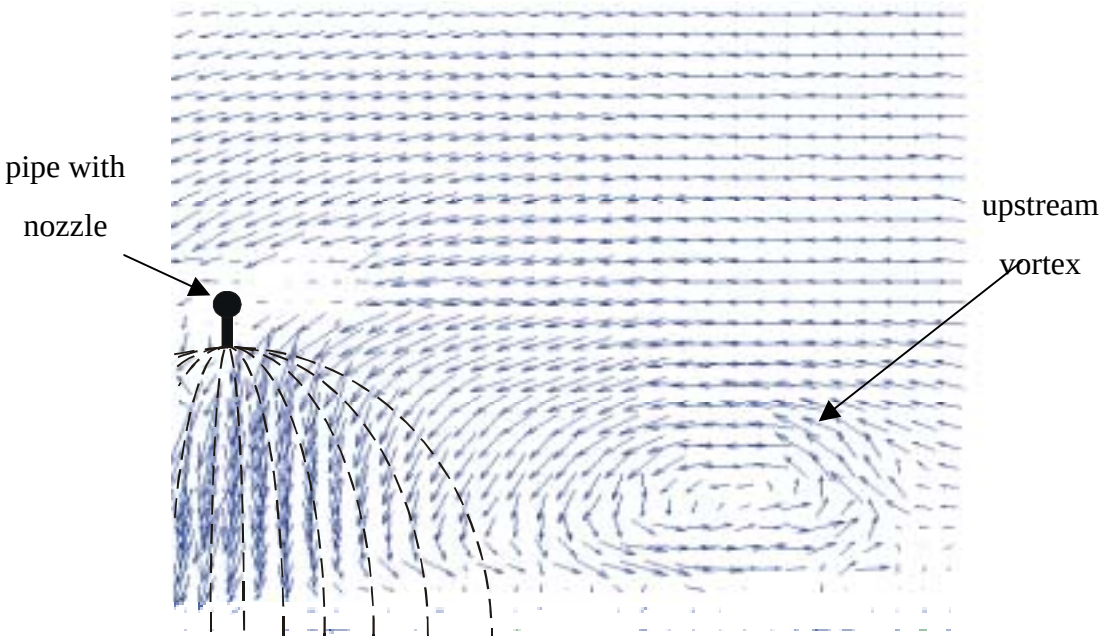

Fig.4. Velocity field of the gas flow at the upstream side of pipe with nozzle electrode during corona discharge. Operating voltage +35 kV, current 120 mA, gas mean velocity 0.56 m/s, lower plate electrode grounded.

4 Acknowledgement

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5 References

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