Transport of NH$_4$NO$_3$ Aerosols in a Corona Radical Injection Reactor

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(Received September 11, 2003; Accepted January 9, 2004)

In this paper results of the Particle Image Velocimetry (PIV) investigation of the transport of NH$_4$NO$_3$ aerosols produced in a corona radical injection (CRI) reactor during NO$_x$ removal process are presented. The PIV investigation, in which NH$_4$NO$_3$ aerosols were employed as tracers, showed that NH$_4$NO$_3$ aerosols were transported and distributed in the CRI reactor by the EHD secondary flow (ionic wind). The flow structures generated in the CRI reactor are similar to those found in other corona discharge reactors. The deposition of NH$_4$NO$_3$ aerosols in the CRI reactor was not homogeneous. The highest density of the deposited NH$_4$NO$_3$ aerosols was on the plate electrode surface where the corona discharge streamers terminated. This was caused by a high velocity of NH$_4$NO$_3$ aerosols on their way from the nozzle electrode to the plate electrode.

1. Introduction

Over the last ten years investigations carried out in laboratories and pilot plants showed that removal of NO$_x$ from flue gases by corona discharges may be very efficient$^{1,2}$. Many recent investigations have been focused on the performance improvement of the corona discharge processing by optimizing the power source$^3$, using various gaseous additives$^{4-6}$ or combining the corona discharge processing with other methods$^{7-9}$.

Among many corona discharge types, the most efficient in NO$_x$ removal is a corona radical shower (CRS) or corona radical injection (CRI)$^{10-14}$. In the CRI reactor, a radical precursor gas is introduced into the main flow of the NO$_x$ polluted gas. The radicals produced by the corona enhance NO$_x$ removal. Mostly ammonia is employed as a radical precursor.

NH$_3$ is introduced through a hollow needle electrode into the corona discharge zone where NH$_3$ molecules dissociate to NH$_2$ and NH radicals. These radicals assist both reduction and oxidation of NO$_x$ molecules. NO$_x$ molecules are reduced as follows$^{13,15}$:

1. NO + NH$_2$ → N$_2$ + H$_2$O, \( k = 2.1 \times 10^{11} \text{ cm}^3 \text{ s}^{-1} \)
2. NO + NH → N$_2$ + OH, \( k = 4.7 \times 10^{11} \text{ cm}^3 \text{ s}^{-1} \)
3. NO$_2$ + NH$_2$ → N$_2$O + H$_2$O, \( k = 2 \times 10^{11} \text{ cm}^3 \text{ s}^{-1} \)
4. NO$_2$ + NH → N$_2$O + OH, \( k = 1.6 \times 10^{11} \text{ cm}^3 \text{ s}^{-1} \)

OH radicals produced in reactions (2) and (4) oxidize NO and NO$_2$ into nitric acids in the following reactions:

5. NO + OH + M → HNO$_2$ + M, \( k = 4 \times 10^{12} \text{ cm}^6 \text{ s}^{-1} \)
6. NO$_2$ + OH + M → HNO$_3$ + M, \( k = 2.6 \times 10^{16} \text{ cm}^6 \text{ s}^{-1} \)

Once HNO$_x$ molecules are formed, ion induced aerosol particle formation will be initiated$^{15,16}$ in the form of heterogeneous nucleation, together with aerosol particle surface reactions with adsorbed ammonia to form ammonium nitrate NH$_4$NO$_3$ aerosol:

HNO$_3$ + NH$_3$ → NH$_4$NO$_3$.

The record NO$_x$ removal energy yield of the CRI method is 40-250 g/kWh at NO$_x$ removal of 95-100%.

This work was aimed at studying the production and movement of NH$_4$NO$_3$ aerosols produced in a corona radical shower reactor. The study was carried out using the Particle Image Velocimetry (PIV) technique. NH$_4$NO$_3$ aerosols were expected to be tracers sufficiently good for the PIV measurements. In addition to the fundamental interest in the NH$_4$NO$_3$ aerosol behaviour in the corona radical shower reactors, this investigation is related to the topical question - where NH$_4$NO$_3$ aerosols are produced, in the discharge region or outside it.

Key words: corona radical injection, secondary flow, PIV, streamer, NH$_4$NO$_3$ aerosol

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2. Experimental set-up

A schematic of the experimental apparatus is shown in Fig. 1. The CRI reactor was an acrylic rectangular reactor (100 mm x 125 mm x 700 mm). A brass hollow needle (2 mm outer diameter, 1.6 mm inner diameter) was used as a CRI electrode. The grounded electrode was a stainless-steel plate. The distance between the electrodes was either 30 mm or 50 mm. DC high voltage with positive polarity was applied through a 10 MΩ resistor to the CRI electrode.

Two gas flows, the main and the additional, were established in the reactor similarly like in the experiment of the CRS14, 16. The main gas (dry air with NO) flowed along the reactor with a flow rate of 3 L/min. The additional gas [dry air with NH₃ and Ar (1.0 %)] was injected through the hollow needle into the main gas flow with a flow rate of 0.5 L/min. The concentration of NO as well as NH₃ in the experiment of Kanazawa et al.16 was 200 ppm. However, at these concentrations of NO and NH₃ the density of NH₄NO₃ aerosols produced in our experiment was too low to employ them as tracers in the PIV measurements. Thus, we increased NO and NH₃ concentrations to 0.5 %.

FTIR analysis of a liquid sample made by solving white powder produced in the reactor proved that no other aerosols than NH₄NO₃ existed in the system. Also the ion-chromatography analysis of the sample shows the NH₄⁺ and NO₃⁻ ions14. The size of the aerosols, as laser light reflection measurement showed, was below 1 µm.

The PIV equipment consisted of a twin second harmonic Nd-YAG laser system (λ=532 nm, pulse energy 50 mJ), imaging optics (a cylindrical telescope), CCD camera, image processor (Dantec PIV 1100), and PC computer. The laser sheet of a thickness of 1 mm, formed from the Nd-YAG laser beam by a cylindrical telescope was introduced into the CRI reactor, perpendicularly to the plate electrode. The NH₄NO₃ particle images were recorded by the Kodak Mega Plus ES 1.0 CCD camera, which captured two images with a time separation of 600 µs. The CCD camera active element size was 1008×1018 pixels. The captured images were transmitted by the Dantec PIV 1100 image processor to the PC computer for digital analysis. The final result of this analysis were maps of the velocity field of NH₄NO₃ aerosols.

When the interelectrode distance was 30 mm, the velocity field maps presented in this paper are composed of several adjacent velocity fields (2 or 4 fields), each of an area of 25 x 25 mm. At the interelectrode distance of 50 mm, each velocity field map is a single field of an area 50 x 50 mm. All the presented velocity field maps resulted from the averaging of 15-25 measurements, i.e. the velocity maps are time-averaged. Measurement error of calculating velocity values is up to 4 %.
3. Results

The corona discharge generated in the CRI reactor was not uniformly distributed around the hollow needle edge. The corona discharge is shown in Fig. 2 as a narrow conical streamer starting from one point at the hollow-needle edge the position of which changed irregularly.

The main consequence of such nature of the corona discharge is that velocity field patterns of NH₄NO₃ aerosols measured by PIV are not symmetric in relation to the hollow needle. The asymmetry increases with increasing distance between the hollow needle and plate electrode.

We observed that without the high voltage applied to the hollow needle electrode, NH₄NO₃ aerosols were not produced in the reactor. Their production started when the glow corona discharge mode was established. However, the efficient production of NH₄NO₃ aerosols started in the streamer corona discharge mode, when an optimum voltage was set as in (15). This optimum voltage was 24 kV and 33 kV at an interelectrode distance of 30 mm and 50 mm, respectively. In both cases the corona discharge current was about 300 μA. All the PIV measurements were carried out at the optimum voltage.

When interelectrode distance was 30 mm, the bulk NH₄NO₃ aerosols move in the entire measurement area more or less perpendicularly to the plane electrode with the highest velocities in the discharge region just below the hollow needle (up to 4 m/s, Fig. 3). Reynolds (Re) and electrohydrodynamical (Nₑₓₑ) numbers estimated for the region just at the hollow needle outlet are 550 and 7×10³, respectively. Since the ratio $Nₑₓₑ/Re^2$ is much higher than 1, transport of NH₄NO₃ aerosols is controlled by the EHD-induced secondary flow. As the flow field pattern shown in Fig. 3 resulted from averaging of 20 individual measurements, lasting 6 s in total, the position of the streamer on the nozzle could randomly vary.

Fig. 2. Typical (we chosen them from a number of photos so that to show different points on the hollow needle edge from which corona streamer developed) temporal images of the downstream area around the hollow-needle during corona discharge. Exposure time of each image was 133 ns. Interelectrode distance 30 mm. The main flow comes from the right side. NH₄NO₃ aerosol is shown.
Transport of NH$_4$NO$_3$ Aerosols in a Corona Radical Injection Reactor (Marek Kociś et al.)

Fig. 3. Flow velocity field downstream of the hollow needle in the CRI reactor. Main gas [air:NO (0.5%)] flow rate - 3 L/min, additional gas [air:Ar (1.0%):NH$_3$ (0.5%)] flow rate - 0.5 L/min. The distance from the hollow-needle to the plate electrode - 30 mm. Applied voltage and corona discharge current - 24 kV and 300 µA, respectively (x - horizontal distance from the position of the hollow-needle electrode).

As it is shown in Fig. 3, NH$_4$NO$_3$ aerosols flow first along the corona discharge and then along the plate electrode outwards. The relatively fast flow along the corona discharge disturbs electrohydrodynamically the main flow$^{15}$ in the reactor (Re=8, $N_{Re}=5.9 \times 10^7$), causing formation of large vortices, both downstream and upstream of the hollow needle, as seen in Figs. 4a and 4b. The vortices are formed about 75 mm from the hollow needle. The similar vortices were found in a needle-to-plate corona discharge reactor through which dry air (without NO and NH$_3$) flowed$^{17}$.

The bulk NH$_4$NO$_3$ aerosols circulates in the whole measurement area towards the corona discharge region, following the electrohydrodynamically disturbed flow pattern. It gives impression of sucking NH$_4$NO$_3$ aerosols present in the reactor by the fast flow directed towards the corona discharge region. After the experiment, it was found that NH$_4$NO$_3$ aerosols deposited the whole inner surface of the CRI reactor and also outer reactor ducts.

At the distance between the hollow needle and plate electrode of 50 mm the asymmetry of velocity field patterns of NH$_4$NO$_3$ aerosols in relation to the hollow-needle is well pronounced (Fig. 5). The highest velocities of NH$_4$NO$_3$ aerosols downstream of the hollow needle electrode are twice than those upstream (4 m/s against 2 m/s, respectively).

4. Conclusions

In this paper results of the PIV measurements of the transport of NH$_4$NO$_3$ aerosols produced in the CRI reactor during NO$_x$ removal process are presented for the first time.

The results showed that:
- NH$_4$NO$_3$ aerosols can be employed as tracers for PIV measurements;
- NH$_4$NO$_3$ aerosols are transported and distributed in the CRI reactor by the EHD secondary flow (ionic wind);
- Flow structures generated in the CRI reactor are similar to those found in other corona discharge reactors;
- Deposition of NH$_4$NO$_3$ aerosols in the CRI reactor was not homogeneous. The highest density of deposited NH$_4$NO$_3$ aerosols was on the plate electrode surface below the nozzle electrode. This was caused by a high velocity of NH$_4$NO$_3$ aerosols in the corona discharge region, directed more or less perpendicularly to the plate electrode.

From the obtained results it is difficult to answer where NH$_4$NO$_3$ aerosols were produced: in the corona discharge region or outside it. NH$_4$NO$_3$ aerosols were present in the entire measurement area, also very close to the hollow needle outlet. This means that they might be either produced in the corona discharge region or transported to the vicinity of the nozzle by the electrohydrodynamically generated vortices. Thus, the question on the place of NH$_4$NO$_3$ aerosols origin is still open. It must be noticed that the PIV measurements of the velocity field were carried out only in one plane, passing through the hollow needle, parallel to the reactor side walls. To know the comprehensive flow velocity field pattern in the reactor, three dimensional map of the velocity field has to be measured.
Fig. 4. Flow velocity fields in a distance of 50-100 mm from the hollow needle in the CRI reactor: a) downstream, and b) upstream. Main gas [air:NO(0.5%)] flow rate - 3 L/min, additional gas [air:Ar(1.0%):NH₃(0.5 %)] flow rate - 0.5 L/min. The distance from the hollow-needle to plate electrode - 30 mm. Applied voltage and corona discharge current - 24 kV and 300 μA, respectively. x - horizontal distance from the position of the hollow-needle electrode.

Fig. 5. Flow velocity field in the CRI reactor: a) downstream, and b) upstream. Main gas [air:NO(0.5%)] flow rate - 3 L/min, additional gas [air:Ar(1.0%):NH₃(0.5 %)] flow rate - 0.5 L/min. The distance from the hollow-needle to the plate electrode - 50 mm. Applied voltage and corona discharge current - 33 kV and 300 μA, respectively. x - horizontal distance from the position of the hollow-needle electrode.
References


