Soot Particle Re-entrainment in a Corona Discharge Reactor*

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Among the various types of diesel after-treatment device, the corona discharge reactor may be considered as a powerful tool for trapping submicron particles. But after precipitation on the electrodes occurs, the re-entrainment of particles is severe and often causes low or negative precipitation efficiency. Experiments were performed to investigate the effect of an applied voltage on the re-entrainment of soot particles from the electrodes. A co-annular laminar diffusion flame burner was used as the soot generator. When a highly negative voltage was applied, exfoliation of the deposited soot particles and an increase in concentration of particles smaller than approximately 150 nm were observed. Turbulence induced from the negative tuft corona and sputtering caused particle re-entrainment from the corona wire and from plates as well. Under soot laden combustion gas, a streamer corona often occurred in the wire-cylinder reactor. Because of its transient nature, streamer corona violently increased the concentration of reentrained particles and CO gas.

Key Words: Re-entrainment, Corona Discharge Reactor, Flame Soot, Submicron Particles, Self-Cleaning, Corona-Induced Oxidation

1. Introduction

Plasma-based traps of particulates in application to diesel after-treatment may need periodic regeneration, which can be achieved in a thermal or non-thermal mode. The plasma reactor can be operated occasionally in the arc discharge mode to thermally oxidize the particles. In Ref.(1), this mode is referred to as the self-cleaning phase of the electrostatic muffler. Thermal oxidation of the particulates precipitated on the electrode surface is achieved in very localized regions of high temperature near the tip of the arc microdischarges. The plasma reactor can also be operated in a non-thermal corona discharge mode to provide continuous oxidation of the particulates by radicals. Masuda et al.⁽²⁾ have observed a self-cleaning effect in the corona mode that correlates with the production of CO and CO₂.

Penetrante et al.⁽³⁾ have mentioned that the non-thermal oxidation is presumably accomplished by the O and OH radicals, or by negative hydronium cluster ions, [(H₂O)_nO₂⁻].

Particulate trapping in a plasma reactor can be accomplished by electrostatic precipitation applied to diesel after-treatment. Corona-type reactors operating as electrostatic precipitators for diesel soot have been reported⁽¹⁾⁻⁽⁴⁾. Corona discharge is a low energy electrical discharge with non-thermal ionization that takes place in the vicinity of an electrode of sufficiently low radius of curvature, in a medium the pressure of which is close to atmosphere. Corona discharges exist in several forms, depending on the polarity of the field and the electrode's geometrical configurations. For a wire-cylinder or a wire-plate electrode configuration, corona generated at a positive wire electrode may appear as a tight sheath around the electrode (glow corona) or as a streamer moving away from the electrode (streamer corona)⁽⁵⁾. Corona generated at a negative wire electrode may take the form of a general, rapidly moving glow or may be concentrated into small active spots called tufts (tuft corona).

Regardless of corona discharge mode, however,

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Fig.1 Experimental set-up.

re-entrainment(resuspension) of the particulates deposited on the electrodes makes it complicated to understand the physical and chemical behavior of the trapped particulates. Among the particulates, the carbon soot particles cause serious re-entrainment problems due to high electrical conductivity($3 \times 10^4 \Omega^{-1} \text{m}^{-1}$), resulting in a low or negative collection efficiency^{(6),(7)}. In this paper, re-entrainment of soot particles is experimentally studied using measurements of numbers and sizes of the reentrained particles for various applied voltages. Effects of a negative and a positive corona on the re-entrainment of particles are discussed.

2. Experiment

As shown in Fig. 1, the experimental set-up consists of a particle generation section, a flow-mixing chamber, and a corona discharge reactor. A co-annular laminar diffusion flame burner is used as the soot generator. The burner consists of two concentric brass tubes of 11.1 mm and 101.6 mm i.d., with ethylene(C_2H_4 ; 240 ml/min) flowing through the central tube and air(40 l/min) through the outer passage, which contains screens and 3.0 mm glass beads⁽⁸⁾. A 250 mm long pyrex glass cylinder is used as a chimney to shield the flame from laboratory air currents. Figure 1 also shows a picture of soot agglomerates composed of primary particles that are approximately 20~50 nm in size taken with an SEM photograph of the particles collected in an isopore filter. Two types of corona discharge reactors are used; wire-to-plate type(reactor ①) and wire-to-cylinder (reactor ②). The former is used for particle measurements and the latter for combustion gas measurements. In reactor ①, three pairs of

wire-to-plate electrodes are connected in parallel. Each wire-to-plate reactor consists of a wire(smooth surface nickel wire 0.2 mm in diameter, or twisted shape alloy copper wire of 1.1 mm) and two copper plates(each 3 cm in length). The wire is connected to a DC power supply(positive/negative; maximum voltage = 20 kV) and the plates are grounded. The distance between the grounded plates in each wire-to-plate reactor is 3 cm. The wire-to-cylinder type reactor(reactor ②), installed right after the exit of the pyrex chimney and thus at a relatively high temperature(~500 K), is used without dilution to investigate the effect of a corona discharge on the conversion of solid carbon to gas phase species, especially CO. The wire-to-cylinder reactor consists of a wire(same as the wire used in reactor ①) and a stainless steel cylinder(20 cm in length). The distance between the wire and the cylinder is 2.5 cm. Changes in concentration of CO(g) is measured by a combustion gas analyzer.

The concentration of soot particles emitted from the C_2H_4 flame is so high that a dilution system is necessary for size and concentration measurements with reactor ①. Soot laden combustion gas mixes with clean air that passes through a HEPA filter. Background airborne particle concentration of clean air is approximately 30 particles/ cm^3 . As the particle/gas mixture flow passes through two stages of mixing baffle, flow straightening plates, a honeycomb, and a mesh in the mixing tunnel, the concentration of the soot particles is nearly uniform over the cross-sections and the flow velocity is approximately 0.5 m/s. Soot particles are isokinetically sampled at 50 cm downstream from the wire, and transported to a SMPS system. The sampling probe is constructed based on Ref.(9), so particle loss at the inlet of the sampling probe is negligible. The SMPS(Scanning Mobility Particle Sizer) is based on the principle of electrical mobility and is used to measure particle size distributions(from 20 to 700 nm) and concentrations⁽¹⁰⁾. The SMPS consists of a DMA(Differential Mobility Analyzer) and a CPC(Condensation Particle Counter; TSI 3022A). The DMA classifies particles along their mobilities after neutralizing their naturally born charges, after which the CPC counts the number of particles grown in size by condensation through alcohol pool.

3. Results and Discussion

3.1 With Reactor ① configuration

Effects of a negative and a positive corona on the re-entrainment of soot particles were investigated with measurements of numbers and sizes of the reentrained particles by utilizing reactor ①. The voltage applied to the wire electrode ranged from 0 to ± 13.5 kV, above

which sparks were created leading to inconsistent data for any further sampling. Within this voltage range, positive and negative corona showed characteristics of a glow and tuft corona, respectively.

Figures 2 and 3 show that the size distribution of soot particles was almost a log-normal distribution (Geometric Mean Diameter: 280 nm, Geometric Standard Deviation: 1.63), when there was no external field. When a high voltage was applied between the two electrodes enough to cause a corona discharge, through which a soot-laden flow passes, most of the charged soot particles were expected to be deposited on the plate electrodes but some may have been deposited on the wire electrode⁽¹¹⁾. Figures 2 and 3 show that the overall particle concentration, measured downstream to the reactor ①, decreases as the applied voltage increases up to 12 kV, for both cases of positive and negative polarities. For the applied voltage of +13.5 kV, it is interesting to note that the concentration of particles smaller than approximately 50 nm increases compared to the case of +12 kV. Figure 3 shows similar trends for particles smaller than approximately 150 nm, when the applied voltage becomes -13.5 kV. This may be due to re-entrainment of particles trapped on the wire or the plate electrodes. It seems that the re-entrainment for a negative corona is higher than that for a positive one, since a negative corona increases the flow turbulence level significantly beyond that measured without the corona discharge⁽¹²⁾.

Figure 3 also shows that for the applied voltage of -13.5 kV, the concentration of particles larger than approximately 300 nm increases compared to the case of -12 kV. These large particles were expected to be soot particles that were deposited and agglomerated on the plate(collecting) electrodes. Thus, experiments were carried out to study the re-entrainment of particles deposited only on the plate. For this purpose, clean air containing no soot was forced to flow between the wire(smooth surface nickel wire of 0.2 mm in diameter) and the plate electrodes. Initially the wire was clean and the plates were covered with soot particles. Then a high negative voltage was applied between the electrodes. Figure 4 shows that the degree of re-entrainment increases as the applied voltage increases. Figure 4 also shows that when the applied voltage is -13.5 kV, the concentration of reentrained particles increases with particle size. The reentrained particles are expected to have resulted from flow instabilities associated with the presence of corona winds induced by the discharge⁽¹²⁾.

Based on the results shown in Figs. 2-4, it is expected that the increase in concentration of particles smaller than 50 nm or 150 nm, is mainly due to the reentrained particles from the wire. To check this, photo pictures of the wire electrode located in a soot

laden flow were taken for different experimental conditions.

Fig.2 Effect of positive applied voltage on size distribution.

Fig.3 Effect of negative applied voltage on size

Fig.4 Re-entrainment from soot agglomerated plate.

Fig.5 Effect of corona polarity on self-cleaning of wire.

Figure 5 shows the pictures for a positive and a negative corona, taken after 30 minutes of experiments. Figure 5 also shows a picture of the original clean wire for the purpose of comparison. The wire under a negative corona discharge seems to be relatively clean; while the positive corona wire has been slightly changed during the same time elapsed. Corona induced self-cleaning of wire has been discussed by Masuda et al.⁽²⁾. They have insisted that soot particles deposited on the corona wire are converted to CO or CO₂ gas in a negative corona. It is the corona-induced oxidation (so called self-cleaning) that have resulted from the active oxidizing species generated in a corona process.

Based on the results discussed so far, the local cleaning of the corona wire may be caused either by the corona-induced oxidation or by particle detachment from the wire, especially for the case of a negative corona. Small soot particles can be easily detached from the wire due to the sputtering effect resulting from highly energizing corona tuft⁽¹³⁾. Small metal particles, originated and exfoliated from wire material, may get included with the reentrained particles due to the sputtering effect⁽¹⁴⁾. It is required to investigate details of the corona-induced oxidation and the particle detachment. This can be accomplished by simultaneous measurements of gaseous species and particles. However, for the configuration of reactor ①, measurements of CO variation were not detected.

As Hobbs et al.⁽¹⁴⁾ have previously reported, there may be metal fragment particles detached from the highly energized corona discharge electrode. Then, there must be some differences for different wire materials and wire surface conditions. Degree of re-entrainment was investigated (a) by changing the wire configuration (material, size, and surface condition), and (b) by monitoring the particle concentration change. Clean air containing no soot

was again forced to flow between the wire and the plate electrodes. Initially the wire and the plate were

Fig.6 Effect of applied voltage on size distribution.

Fig.7 Effect of reactor operating time on particle re-entrainment.

Fig.8 Effect of reactor operating time on particle re-entrainment.

clean. Alloy copper wire (twisted shape wire of 1.1 mm in diameter) was used as a substitute for the nickel wire (smooth surface wire of 0.2 mm in diameter), used for the experiments that produced the results shown in Figs 2-5. Results obtained with the alloy copper wire in a clean environment are summarized in Figs. 6 and 7. Applied voltages used in the results shown in Fig. 6 and 7 were purposely chosen different from those for Fig. 2 and 3. For example, a voltage of -14.3 kV applied to the alloy copper wire in a clean environment produced an electrical current of 0.45 mA, which was also obtained with the same voltage in the previous experiments. Figure 6 implies that for the applied voltage of -14.3 kV, particles smaller than 150 nm are reentrained to the flow.

It is considered that these particles must be metal particles exfoliated from the wire by sputtering. It is interesting that the voltage of $+15$ kV, also resulting in a current of 0.45 mA, caused no particle re-entrainment at all. Figure 7 shows that the degree of re-entrainment diminishes as time passes. For the purpose of comparison, Fig. 8 shows the results obtained with the smooth surface nickel wire in the soot free condition, for the applied voltage of -15.8 kV resulting in the same electrical current. With the smooth nickel wire, the degree of re-entrainment is much less than the case with the twisted copper wire. This implies that the re-entrainment from a wire is dependent upon the wire surface conditions (roughness, protrusion and etc).

3.2 With Reactor ② configuration

Corona-induced oxidation of soot particles can be checked with measurements of combustion gas (CO , CO_2) concentrations. When the soot laden combustion gas emitted from a flame was introduced without dilution air into the wire-to-cylinder reactor (reactor ②), gas temperature was about 500 K, and it was possible to measure in-situ CO concentration change. When the alloy copper wire was used, streamer coronas were produced for positive applied voltages greater than $+15$ kV. Figure 9 shows that a streamer spreads over the reactor and makes the reactor bright on the inside. For the case of a negative tuft corona the reactor looks dark. A positive streamer is produced when the positive ion density is high enough to extend the region for corona initiation into the electrode gap⁽⁵⁾. It should be noted that a corona is easily produced near sharp protruded edges of the wire. Since the twisted wire has large protrusion spots, the streamer corona causes continuous soot deposition on the wire surface, as shown in Fig. 10. The continued deposition further increases the protrusion areas, which makes it easy for the re-entrainment and exfoliation to take place. Kanazawa et al.⁽¹³⁾ have

reported that smoke deposited wire are exfoliated in a positive streamer corona. Experiments were carried out with the smooth surface wire to confirm the effect of wire surface conditions on streamer generation. As expected, no streamer corona discharge was observed for repeated experiments. Figure 11(a) and (b) are SEM photographs of aluminum foils, which were wrapped on the reactor wall for applied voltages of $+15$ kV and -15 kV, respectively.

As shown in Fig. 11(a), in the case of a streamer

Fig.9 Photographs of the (a) streamer corona and (b) tuft corona in the wire-to-cylinder reactor.

Fig.10 Exfoliation of the soot deposit.

Fig.11 SEM photographs for aluminum foil wrapped in the reactor wall.

Fig.12 CO gas concentration change.

corona, the surface of aluminum foil was locally burnt with soot by its highly energized streamer heads, which might increase the surface temperature of the foil above a few 100 K⁽⁵⁾. On the other hand, Fig. 11(b) shows that the surface state of aluminum foil did not change in the case of a tuft corona.

The corona-induced oxidation occurred in both cases of streamer and tuft discharge. Figure 12 shows that when the negative applied voltage is high enough, the concentration of CO increases slightly. But when the positive streamer corona is produced, it abruptly increases. The increase of CO concentration may be due to corona-induced oxidation of carbon soot particles, deposited on electrodes or laden in flow. Another possibility is the decomposition of CO₂⁽⁵⁾, which needs to be studied further.

4. Conclusion

Soot particle re-entrainment in a corona discharge process was investigated using measurements of size distribution and CO concentration for different reactors and wire surface conditions.

When the applied voltage was positive, there was only a slight change in size distribution. But in a negative voltage (approximately 9kV/cm), re-entrainment occurred for particles smaller than 150 nm and greater than 300 nm. The latter was due to the soot particles agglomerated on the plates, and reentrained by corona-induced turbulent flow near the plates. The former might be due to soot particles in the case of nickel wire by ion sputtering or corona-induced oxidation, which can be inferred from the following results. Particles smaller than 150 nm might be metal fraction particles in the case of the alloy copper wire. But when the nickel wire was used, the re-entrainment of metal fraction particles was negligibly small. When a negative voltage was applied, self-cleaning occurred on the surface of the soot deposited corona wire. This might be due to the ion sputtering effect or corona-induced oxidation. CO concentration increased in the wire-to-cylinder reactor in both cases of positive and negative voltage. Especially for the case when the applied voltage was positive, streamer corona occurred with a violent increase of particle re-entrainment and CO concentration.

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