Pulsed Dielectric Barrier Discharge Reactor for Diesel Particulate Matter Removal

S. Yao, M. Okumoto, K. Madokoro, and T. Yashima
Chemical Research Group, Research Institute of Innovative Technology for the Earth, Kyoto 619-0292, Japan

E. Suzuki
Dept. of Fine Material Engineering, Shinshu University, Ueda 386-8567, Japan

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A dielectric barrier discharge (DBD) reactor, driven by a pulsed corona surface discharge (PCSD), was developed for the removal of particulate matter (PM) in an undiluted exhaust gas of a diesel engine. The DBD reactor consisted mainly of alumina (Al_2O_3) plates and metal meshes covered on the surfaces of Al_2O_3 plates. The PCSD was carried out with a pulse power supply at atmospheric pressure and the temperature of exhaust gases. The energy efficiency for PM removal was 26.5 μg/J at maximum and 1 μg/J with 42% PM removal. The construction of the DBD reactor that promoted PM deposition on Al_2O_3 surfaces improved energy efficiency. The constants in PM removal model were given. © 2004 American Institute of Chemical Engineers *AIChE J,* 50: 1901–1907, 2004

Keywords: particulate matter (PM) removal, pulsed plasma, corona surface discharge, dielectric barrier discharge (DBD) reactor

Introduction

Worldwide emission standards of diesel particulate matter (PM) and nitric oxides (NO_x) from diesel engines have become increasingly restrictive (Zelenka et al., 1996). Many efforts have been made to reduce emissions of PM and NO_x into the atmosphere. There is a limit for engine improvement, given that the emission of NO_x and PM has a trade-off relation; that is, a diesel engine working with a lower NO_x emission results in a higher PM emission; thus aftertreatment of NO_x and PM is required. Many reports focused on using diesel particulate filters (DPF) or catalyst-based DPF (CB-DPF) to remove PM and using fuels or urea to reduce NO_x (Andersson et al., 2002; Gilot et al., 1997; Koltsakis et al., 1997; Neeft et al., 1996). Another effective method for NO_x emission reduction is to use a high rate of exhaust gas recirculation (EGR). Wagner et al. (2000) reported that a 44% EGR ratio can reduce NO_x emission by 90%, although excess PM is emitted because of EGR. Additional aftertreatments are required to reduce 97% PM emission to the same PM emission level with that without EGR. Plasma technology has potential to reduce this excess PM emission (Chae et al., 2001; Chang, 1993; Chang et al., 1991; Dorai and Kushner, 1999; Dorai et al., 2000; Eliasson, 1991; Franick and Bykowski, 1994; Herling et al., 2000; Larkin et al., 1998; Lepperhoff et al., 1999; Liu et al., 1996; Matsui et al., 2001; Okubo and Yamamoto, 2002; Penetrante, 1993; Penetrante et al., 1999; Tamon et al., 1995, 1998; Thanyachotpaiboon et al., 1998; Thomas et al., 2000; Vercammen et al., 1997).

One of our recent reports (Yao et al., 2004) demonstrated that PM from a diesel engine could be removed using a pulsed corona surface discharge (PCSD) and a tubular dielectric barrier discharge (DBD) reactor. The PM removal rate is a function of PM concentration in diluted exhaust gases and plasma energy injection. Because the corona discharges occur over the dielectric barrier surfaces of Al_2O_3, the adsorption of PM on the corona discharge surfaces is important; especially at a plasma energy injection higher than 2 W (to a 1.07 L/min

Correspondence concerning this article should be addressed to S. Yao at yao@rite.or.jp.

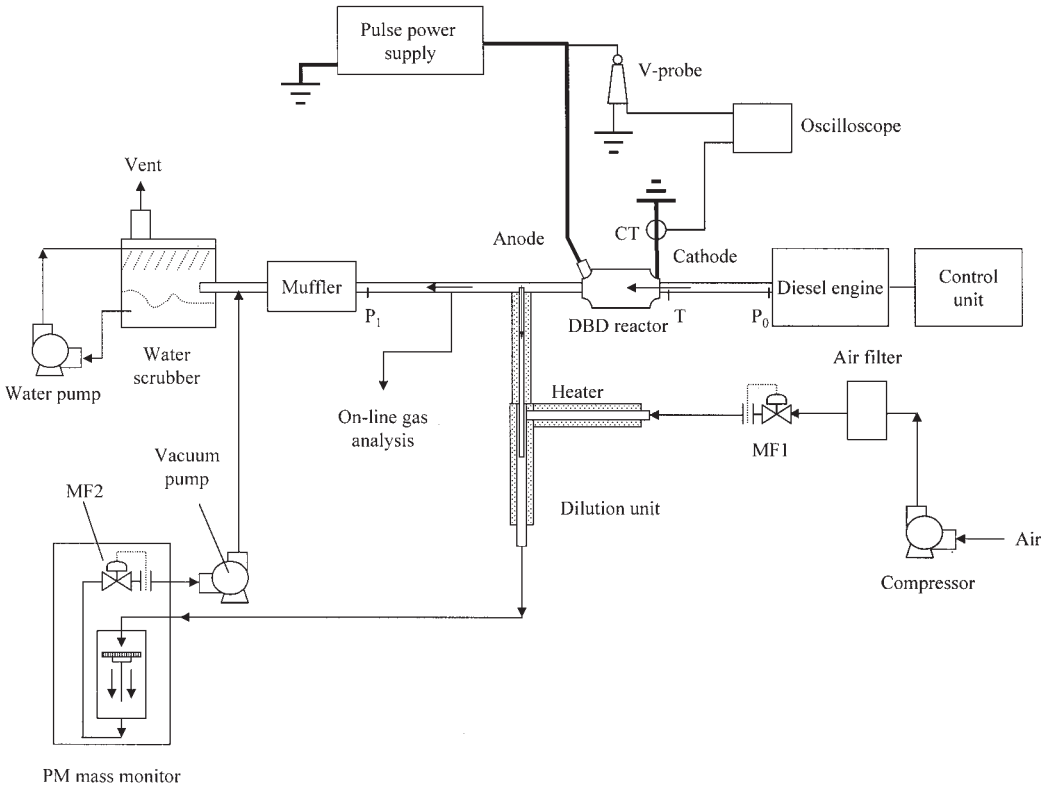

Figure 1. Diesel engine and its dynamic system, pulse power supply, dielectric barrier discharge (DBD) reactor and the voltage and current measuring system, and NO/NO_2 and gas composition analysis systems.

diluted exhaust gas flow), both the adsorption of PM on corona discharge surfaces and plasma energy input contribute to PM removal. The best result shows that to treat PM emission of 2.78×10^{-4} g/s (1 g/h) from a diesel engine using the tubular DBD reactor (only one surface used), a 3.4-kW output of the pulse power supply is required to achieve an 89% PM removal. This energy requirement should be reduced if such a PM removal system (mainly a pulse power supply and a DBD reactor) is designed for installation in, for example, diesel trucks.

For the scale-up of the plasma PM removal system, the kinetic characteristics of the DBD reactor are important. Dorai and Kushner (2002) calculated the soot oxidation in the presence of O_2 and NO_2. They found that the energy efficiency for PM removal (W-value) has a level of 3.3 μg/J at an energy deposition (a ratio of the amount of discharge energy to the volume of exhaust) of 30 J/L for multiple-pulse discharges. This energy efficiency increases with increasing energy deposition.

In this study, a DBD reactor was developed for PM removal from whole exhaust gases of a diesel engine. The structure of the DBD reactor was designed to promote PM adsorption on Al_2O_3 surfaces and to improve energy efficiency. The PM removal characteristics using the DBD and the pulse power supply were investigated. The constants in PM removal model were given.

Experimental Setup

Figure 1 shows the experimental system, including a diesel engine, a hydrodynamometer test cell equipped with a performance measurement system, and a gas analysis system, which was described in detail elsewhere (Yao et al., 2004). A DBD reactor was installed in the exhaust pipeline 1.5 m downstream of the exhaust point of the engine. The temperature of the exhaust gases from the diesel engine was measured with a thermocouple (T in Figure 1) 0.05 m upstream of the reactor. Two pressure sensors (P_0 and P_1, VPRN, 0–100 kPa, Valcom) were used to measure the pressure drop resulting from the use of the DBD reactor. A portion of the exhaust gas (0.65 L/min) from 0.05 m downstream of the DBD reactor was diluted 4× with air at 150°C using a dilution unit. The air-diluted exhaust gas was then cooled to room temperature and sent to a PM mass monitor (R&P TEOM 1105; Rupperecht and Patashnick) for PM emission rate (g/h) measurement.

The main property of the diesel fuel (Japan Energy, Tokyo) was specific gravity at 15°C: 0.8351 g/cm^3, cetane number: 55.0, flash point: 76°C, and sulfur: 0.002 wt %. The exhaust gas from the diesel engine was analyzed on-line using a GC-103 gas chromatograph (Ohkura, FID; a methanizer converting CO and CO_2 to CH_4 was equipped before FID) for CO and CO_2 analyses; AGC280 gas chromatograph (Ohkura, TCD) for O_2 and N_2 analyses; and NO/NO_2/NO_x analyzer (ECL-88US, Yanaco) for NO, NO_2, and NO_x analyses.

Construction of the DBD reactor is shown in Figure 2. Eight sample Al_2O_3 plates ($150 \times 150 \times 2$ mm^3) were used for discharges. Three samples of iron mesh [110×110 $mm^2 \times 0.8$ mm (thickness), expanded metal mesh with Katsurada original pattern and an aperture ratio of 74%] were inserted between

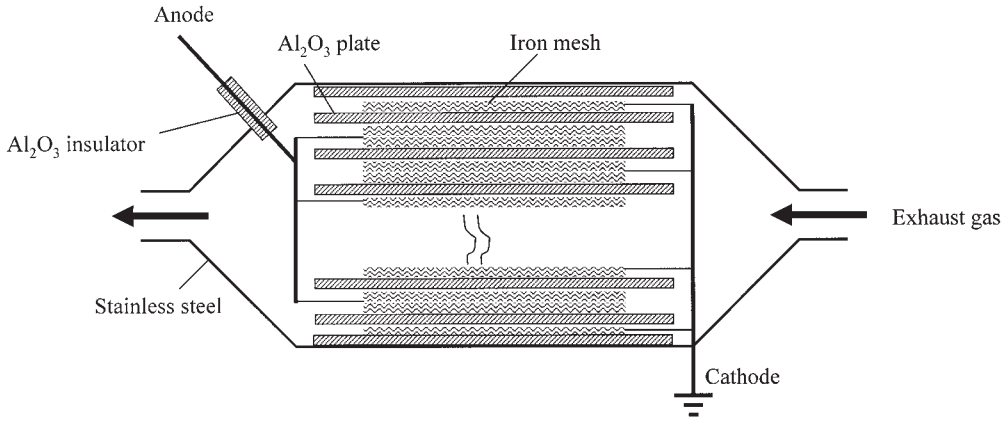

Figure 2. Structure of the DBD reactor.

each two of the Al_2O_3 plates. The total area of the Al_2O_3 surface holding iron meshes was 0.194 m^2. About half of the DBD reactor space was used in this study; the remaining reactor space was sheltered with a copper plate. Metal meshes were connected with copper wires that were used as an anode or cathode. Glass spacers were used to control all exhaust gases passing through the mesh zone and to inhibit electric shorts between anode and cathode or the wall of the reactor. Two 2-m silicone rubber insulated wires (CSA type TV-30, Kurabe) were used to connect the DBD reactor and a pulse power supply (DP-30K10, Peec) that were driven by a 24 direct current power supply. The pulse power supply could generate a pulse peak voltage of 30 kV of a rise time level of 10^{-6} s and a pulse repetition of 10 kHz at maximum. The discharge voltage and cathode current were measured with a voltage probe (EP-50K, Peec) and a current transformer (Model 2-1.0, Strangenes), respectively. The analogue signals from the voltage probe and current transformer were recorded with a digital phosphor oscilloscope (TDS 754D, Tektronix). The energy injection P_d (in watts) from the pulse power supply to the DBD reactor was calculated from the waveforms of voltage and cathode current using Eq. 1.

$$P_d = F \sum_i [V_i I_{c_i}(t_{i+1} - t_i)] \tag{1}$$

where F is pulse frequency (in hertz); V_i and I_{c_i} are voltage (in volts) and cathode current (in amperes), respectively, at discharge time t_i in seconds. The general energy consumption P_t (in watts) was calculated from the product of the input voltage (24 V) and current that was measured with a multimeter (Tektronix TX3 True RMS MultiMeter).

PM removal X was calculated using Eq. 2.

$$\text{PM removal } X = \frac{\text{PM emission rate without plasmas} - \text{PM emission rate with plasmas}}{\text{PM emission rate without plasmas}} \times 100\% \tag{2}$$

The discharge energy efficiency η_d of PM removal (in g/J or g/kWh), on the basis of the energy injection P_d, was defined according to Eq. 3.

$$\eta_d = \frac{\text{PM emission rate} \times \text{PM removal } X}{P_d} \quad \text{(g/J)}$$
$$= \frac{\text{PM emission rate} \times \text{PM removal } X}{P_d} \times 3.6 \times 10^6 \quad \text{(g/kWh)} \tag{3}$$

The general energy efficiency η_t of PM removal (in g/J or g/kWh), on the basis of the general energy consumption P_t, was defined according to Eq. 4.

$$\eta_t = \frac{\text{PM emission rate} \times \text{PM removal}}{P_t} \quad \text{(g/J)}$$
$$= \frac{\text{PM emission rate} \times \text{PM removal}}{P_t} \times 3.6 \times 10^6 \quad \text{(g/kWh)} \tag{4}$$

Results and Discussion

Typical waveforms of discharge voltage and current

The typical waveforms of discharge voltage and cathode current are shown in Figure 3 at a fixed pulse frequency of 2 kHz. The pulse voltage increased to a peak voltage of 7.6 kV within about 1.8×10^{-6} s. The cathode current increased to a peak value of 1.2 A simultaneously, whereas voltage increased and dropped to zero within a time level of 1×10^{-5} s. This discharge current involved the current for PCSD and the current for charging stray capacitors on the DBD reactor side.

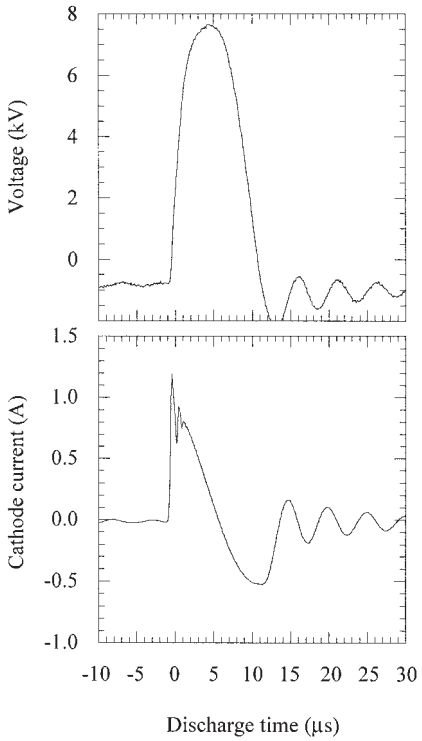

Figure 3. Typical waveforms of discharge voltage and cathode current at 2 kHz.

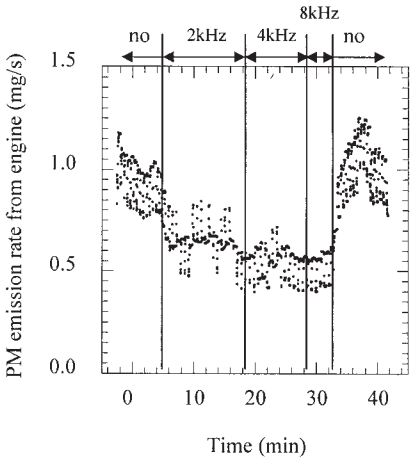

Figure 4. Typical results of PM removal without or with pulsed discharges at 7.8-kV peak voltage and various pulse frequencies.

PM removal

Whole exhaust gases from the diesel engine at an engine rotation speed of 1.03 krpm and power output of 3.8 kW passed through the DBD reactor. The volume flow rate of the exhaust gases was estimated to be 0.014 m^3/s {[1030 (rpm) $\times$ 0.0008 m^3/rpm]/60} at room temperature and atmospheric pressure. The composition of the exhaust gas was O_2, 14%; N_2, 77%; CO_2, 4.15%; H_2O (estimated as the same as CO_2), 4.15%; CO, about 50 ppm; NO_x, 80 ppm. No changes in the composition of the exhaust gas were found with or without plasma discharges. The pressure drop attributed to the use of the DBD reactor was about 1 kPa. The temperature of the exhaust gases was 450 K, indicating that the temperature of the DBD reactor was not higher than 450 K. Figure 4 shows typical PM emission rates with or without plasma discharges. The average PM emission rate (based on whole exhaust gases) was 9.36×10^{-4} g/s without plasma discharges and decreased to a level of 6.5×10^{-4}, 5.53×10^{-4}, and 5.39×10^{-4} g/s with plasma discharges at a peak voltage of 7.0 kV and a pulse frequency of 2, 4, and 8 kHz, respectively.

Energy injection is an important factor influencing PM removal. Their relation is shown in Figure 5. PM removal increased with increasing energy injection, which was consistent with that reported by the authors (Yao et al., 2004). PM removal is a function of PM adsorption on Al_2O_3 surface and plasma energy injection, according to Eq. 5

$$X = 1 - \exp[-(m_1 m_2 P_d)/(m_1 + m_2 P_d)] \qquad (5)$$

where $m_1 = (k_1 - k_2)(s/V)\tau$, $m_2 = (k_3/\lambda)(s/V)\tau$, k_1, and k_2 (in m/s^2) are adsorption and desorption constants of PM on Al_2O_3 surface, respectively; k_3 is a reaction rate constant of PM oxidation under plasma discharge conditions; s, V, τ, and λ are, respectively, the discharge surface area of the DBD reactor, volume flow rate of the exhaust to the DBD reactor, residence time of the exhaust gases in the DBD reactor, and the ratio of PM concentration in the exhaust gases to PM concentration on the discharge surface area at adsorption/desorption equilibrium. The ratio of $\gamma = m_1/(m_2 P_d)$ (in J m/s^3) corresponds to the net PM deposition ability to the PM plasma removal ability. The γ values are also illustrated in Figure 5. Under experimental conditions, γ values were lower than 3.5, indicating that both PM adsorption on the Al_2O_3 surface and PCSD contribute to PM removal.

The parameters shown in Eq. 5 are listed in Table 1, together with the comparison with our previous results using a small DBD reactor. The ratio of k_3/λ was 5.18×10^{-2}, close to the previous result of 7.70×10^{-2}. The net PM deposition on the Al_2O_3 surface ($k_1 - k_2$) was 5.73, improved 52-fold compared with the 0.109 value of the previous study, which suggested that the construction of the DBD reactor in this study increased the deposition rate of PM.

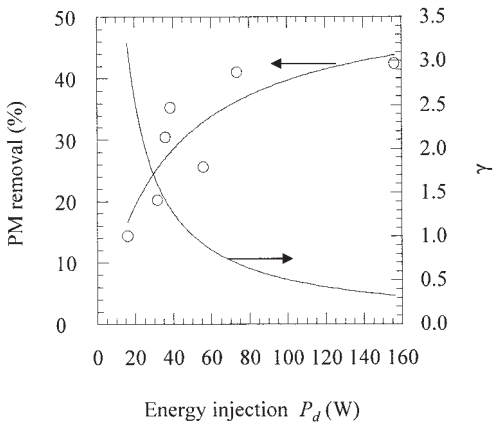

Figure 5. PM removal as a function of energy injection.

Table 1. Experimental Conditions and Constants in the PM Removal Model

Factor	This Study	Previous Study
Reactor type	DBD plates, two side surfaces used	DBD tube, out side surface used
Exhaust V, m^3/s	0.02	1.8×10^{-5}
Temperature, K	450	300
Pressure (P_1), atm	1	1
Rise time of pulse power supply	1 μs	20 ns
Discharge area S, m^2	0.194	2.67×10^{-3}
Residence time τ, s	0.017	0.258
m_1, (–)	0.96	4.2
m_2, W	0.0129	2.0
$\frac{S}{V}\tau$, s^2/m	0.168	38.6
$(k_1 - k_2) = \frac{m}{\left(\frac{S}{V}\tau\right)}$, m/s^2	5.73	0.109
$\frac{k_3}{\lambda} = \frac{m_2}{\left(\frac{S}{V}\tau\right)}$, $J\,m/s^3$	7.7×10^{-2}	5.18×10^{-2}

*Calculated as the area of the Al_2O_3 surface holding the iron meshes.

Energy efficiency for PM removal

The discharge energy efficiencies (η_d) of PM removal at various energy injections are illustrated in Figure 6. At a low energy injection P_d of 15.8 W, the discharge energy efficiency was 26.5 μg/J. The energy efficiency of PM removal obviously decreased when the energy injection increased to 60 W and gradually to 1.46 μg/J at 156.3 W (P_d) (Figure 6a). This energy efficiency was close to that of 3.3 μg/J at an energy deposition of 30 J/L using a pulse discharge given by Dorai and Kushner (2002) and that of activated carbon oxidation (2.8 μg/J) using pulsed spark discharges given by the authors (Yao et al., 2001). The adsorption time of PM on the Al_2O_3 surface affected the energy efficiency. In this study, the increase in energy injection was controlled by increasing pulse frequency. The increase in pulse frequency resulted in a decrease in adsorption time of PM on the Al_2O_3 surface. For example, in a second discharge duration, the total discharge time was about 10 ms (1% of 1 s) at 1 kHz, but 80 ms (8% of 1 s) at 8 kHz. This finding agreed with our previous results that the PM removal saturated at a pulse frequency higher than 2 kHz.

Because of the energy loss in the pulse power supply, the general energy efficiency (η_t) of PM removal was 10.9 μg/J at 38.4 W, and decreased to 0.67 μg/J at 341 W (Figure 6b). Here the energy loss in the pulse power supply accounted for about 50%. Such an energy loss included electricity consumption in the support parts in the pulse power supply, such as indication lamps. This energy loss can be reduced to less than 10%, as reported by Yan (2001).

Improvement of energy efficiency

The energy efficiency of PM removal was improved more than 10-fold, compared with that using a tubular pulsed corona surface reactor (Figure 7), under conditions shown in Table 1. This energy efficiency improvement is mainly attributed to the increase in PM adsorption on the Al_2O_3 surface promoted by the construction of the DBD reactor. The high PM concentration and high reaction temperature could also contribute to PM removal, given that PM removal is a function of PM concentration and energy injection.

The energy efficiency improvement using the pulse power supply of a 1-μs rise time used in this study is higher than that of a 20-ns rise time used in our previous study. This finding implied that the use of the pulse power supply having a longer rise time could reduce installation cost of the PM removal system because the pulse power supply of a longer rise time is cheaper than that of a shorter rise time.

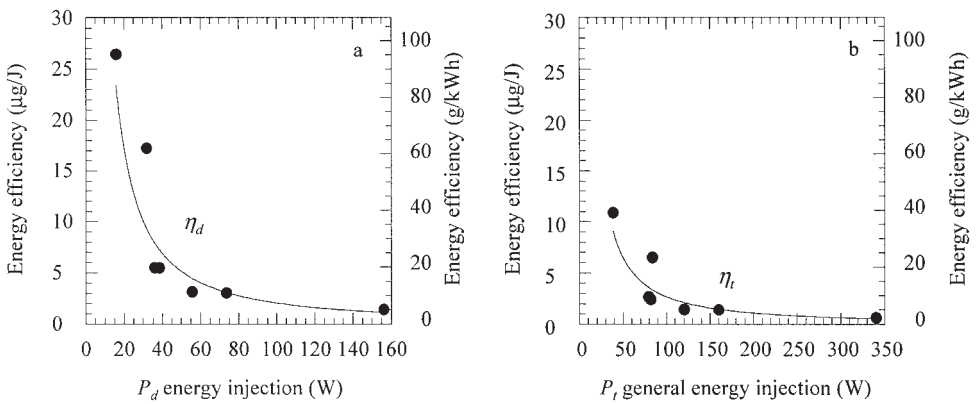

Figure 6. Energy efficiency of PM removal as a function of energy injection.

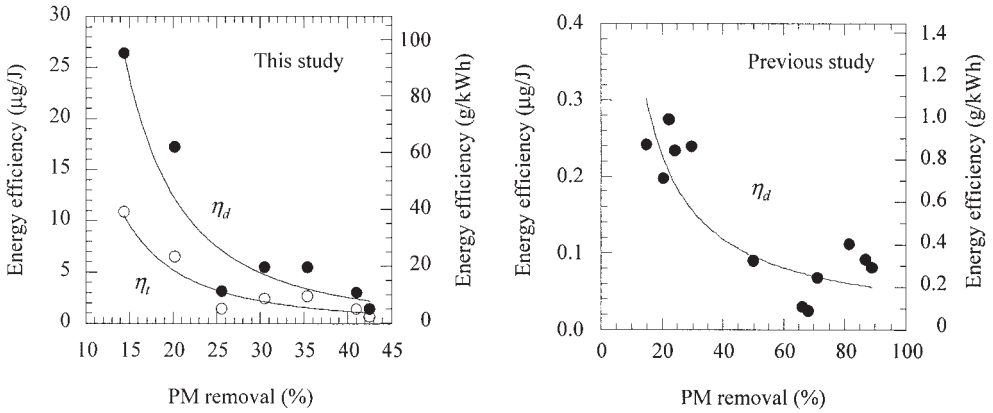

Figure 7. Comparison of energy efficiency with our previous results using a tubular DBD reactor.

NO_x removal

The reduction of NO_x is suggested as follows in a pulsed-plasma discharge (Dorai and Kushner, 2002)

$$e + N_2 \rightarrow N + N + e \quad (6)$$

$$N + NO \rightarrow N_2 + O \quad (7)$$

where e represents energized electrons by pulsed-plasma discharges.

Reactions of NO with such as O atoms and OH radicals result in formation of HNO_3 (Eqs. 8–11), which contribute to NO_x removal (Dorai et al., 2000)

$$e + O_2 \rightarrow O + O + e \quad (8)$$

$$e + H_2O \rightarrow OH + H + e \quad (9)$$

$$NO + O + M \rightarrow NO_2 + M \quad (10)$$

$$NO_2 + OH + M \rightarrow HNO_3 + M \quad (11)$$

We recently reported that NO_x could be reduced by the pulsed corona surface discharges by a mechanism of NO_x reduction to N_2 and O_2, and most possibly to HNO_2 and HNO_3 because of the NO oxidation with active oxygen species such as O and O_3 (Yao et al., 2004). In this study, no changes in NO and NO_2 concentrations were observed. These differences might be attributable to the reaction conditions, such as pulse voltage, oxygen concentration, and gas temperature. For example, in the oxygen-rich exhaust gases it might be more difficult at 450 K, than at room temperature, for NO to be oxidized to NO_2, which is further removed after reaction with OH or H_2O to give HNO_2 and HNO_3. The PM concentration in undiluted exhaust gases is higher than that in diluted exhaust gases, which also explains why NO_2 formation from NO is difficult, given that the composition of NO_x depends on PM (soot) (Dorai and Kushner, 2002).

Conclusions

Undiluted PM from a diesel engine could be removed using PCSD and a DBD reactor. The energy efficiency for PM removal was improved more than 10-fold compared with that reported in our previous study. The discharge energy efficiency was about 1 μg/J with 42% PM removal. The constants in the PM removal model had been given, which were very important for the scale-up of the DBD reactor. Using the PM removal model (Eq. 5), the energy injection P_d is estimated to be 220 W [about 0.4% of 54 kW (maximum output) of the diesel engine] for a 90% PM removal, if the residence time of the exhaust gases in the DBD reactor and discharge surface areas could be magnified by 2 times.

Although there are a number of factors that should be investigated in greater depth, such as the influence of the exhaust gas temperature, concentrations of water and oxygen in exhaust gases, pressure drop resulting from the use of the reactor, and the corrosion problem of metal meshes, we anticipate that the problem of PM can be solved by using our PM removal system.

Acknowledgments

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